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# Dietary exposure to total and inorganic arsenic via rice and rice-based products consumption

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ABSTRACT

Arsenic (As) is a toxic metal known for its carcinogenic potential, especially the inorganic species. In turn, it has been extensively reported that the major route of exposure is the diet, being rice and rice products one of the food groups with the highest arsenic concentrations. This study was aimed at determining the concentrations of total arsenic (total As) and inorganic arsenic (InAs) in rice and rice products. Furthermore, the dietary exposure and health risks for infant and adult population were also estimated. Brown varieties of rice showed higher arsenic concentrations than white rice. Regarding the dietary exposure to As by groups of population, toddlers and infants presented the highest exposure to total As, but unlike the rest of population groups, the main contributor was organic arsenic. Focusing on the contribution of each food item, rice represents the major contributor to InAs exposure by the adult population, while baby cereals and breakfast cereals are the most important contributors for infant exposure. Anyhow, none of the population groups exceeded the lower limit of the $BMDL_{01}$ range (from 0.3 to 8.0 μg/kg body weight/day) set by EFSA in any of the three exposure scenarios (high, mean, and low) hereby considered. Finally, consumption of white rice varieties, as well as rice washing before cooking, are recommended in order to minimize the exposure to arsenic.

*Keywords:* arsenic; dietary exposure; rice; rice products; risk assessment

# 1. Introduction

Arsenic (As) is a metalloid that generally acts as a metal. It is widely distributed in the Earth's crust and can react with oxygen, chlorine and sulphur to form inorganic compounds, and with carbon and hydrogen to form organic compounds (Jomova et al., 2011). Arsenic is on the top 1 on the substance priority list of the Agency for Toxic Substances and Disease Registry (ATSDR, 2017).

The natural release of inorganic arsenic (InAs) to the environment is caused by the alteration and erosion of rocks and soil, where it is present as arsine, arsenite, arsenate, and oxide. In turn, anthropogenic sources include mining, metallurgical activity, use of pesticides, and combustion of different materials, such as coal and wood. The incineration of domestic and urban waste is also an important source (Margallo et al., 2015; Rovira et al., 2018, 2015).

The major route of exposure to arsenic is diet, while other pathways, like dermal and inhalation are less important (Chung et al., 2014). Organic arsenic is mainly found in products of marine origin, such as fish and seafood. This food group contains foodstuffs with the highest concentrations of the organic form of arsenic: dimethylarsenic (DMA) (Taylor et al., 2017). In fact, fish, crustaceans, molluscs and other aquatic animals have the ability to metabolize arsenic and accumulate it as DMA. DMA has a notable lower toxicity than the inorganic forms, being primarily excreted through the urine (Hughes, 2002).

On the other hand, InAs is found in water in certain geographical areas, and in rice and rice products (Hojsak et al., 2015). In addition, it should be highlighted that the inorganic forms of arsenic are the most toxic. The International Agency for Research on Cancer (IARC) catalogues arsenic as a known carcinogen, category 1 (carcinogenic to humans, with sufficient epidemiological evidence), while the US Environmental Protection Agency (EPA) classifies it in group A (human carcinogen, with enough evidence obtained from epidemiological studies) and establishes a risk of cancer through oral exposure (EPA, 2016; IARC, 2012). A number of studies have shown that intake of InAs can increase the risk of developing cancer of lung (Hubaux et al., 2013), skin (Bailey et al., 2009), bladder (Bailey et al., 2012), liver (Sung et al., 2012) and kidney (Yuan et al., 2010), among others.

Based on the fact that arsenic is one of the most dangerous trace elements for human health, in 2009 the European Food Safety Authority (EFSA) re-evaluated the toxicity and the health effects of InAs. According to human lung cancer data, the EFSA proposed to use the lowest limit of the 95 percentile of the experimental dose, associated with a 1 % of incidence or extra risk ($BMDL_{01}$). As a result, a range from 0.3 to 8.0 µg/kg body weight/day was set (EFSA, 2014). In 2010, the Joint FAO/WHO Expert Committee on Food Additives (JECFA) established a $BMDL_{0.5}$ of 3 µg/kg body weight/day (with an interval between 2 and 7 µg/kg body weight/day). Moreover, the provisional tolerable weekly intake (PTWI) established for InAs of 15 µg/kg body weight/week was withdrawn (FAO/WHO, 2010).

The last total diet study (TDS) conducted in Catalonia (Spain) in 2017 showed that rice is one of the food groups containing the highest concentrations of arsenic and InAs (González et al., 2019). Since the content of organic and InAs in rice and rice products might vary, the Catalan Agency of Food Safety (ACSA) has launched a study aimed at: i) determining the concentration of this toxic trace element in these foodstuffs; ii) evaluating the exposure and the potential risks for the children and the adult population.

## 2. Materials and methods

### *2.1. Sampling*

In September 2018, rice and rice products were purchased in different stores located in Reus and Tarragona (Catalonia, Spain). Rice samples counted with 7 rice varieties: white long rice from Spain; white long rice from Asia (India/Pakistan); white round rice from Spain; white round rice from Asia (Japan); brown long rice from Spain; brown long rice from Asia (India/Pakistan) and brown round rice from Spain. Pre-cooked rice was also included, but a geographical differentiation was not considered. Rice products included rice flour, rice grits, bread with cereals other than wheat, rice cakes, Chinese noodles, rice cookies, breakfast cereals containing rice, rice desserts, rice milk, baby cereals containing rice, baby food with fish and rice, and baby food with chicken and rice. Once in the laboratory, composite samples were prepared with 121 individual samples of rice and rice products. Each composite contained 7 individual samples of the 7 rice varieties, and 6 individual samples of 12 rice products. Each composite included samples from different locations and stores. Food samples were mixed and homogenized with a domestic shredder and stored until further analysis.

### 2.2. *Analysis of arsenic*

The sample treatment to analyse the content of total As has been recently reported by González et al. (2019). Briefly, 9 ml of HNO3 diluted 1/3 (65% Suprapur, E. Merck, Darmstadt, Germany: Purified water; 1:2) and 0.5 ml of H2O2 (30% Suprapur, E. Merck, Darmstadt, Germany) were added to 0.5 g of sample. Subsequently, a microwave digestion was performed up to a final temperature of 200 ºC for a total program time of 30 minutes. Finally, purified water was added to the extracts until a final volume of 30 ml.

On the other hand, InAs was determined using 0.25 g of sample and 10 mL of an acidic-oxidant solution (0.2% w/v nitric acid + 1% w/v hydrogen peroxide). Sample extraction lasted for 1 h up to a final temperature of 95 °C (Llorente-Mirandes et al., 2012). InAs was determined by means of liquid chromatography (HPLC, Agilent Series 1100) coupled to the ICP-MS was used. For Total organic arsenic content species, an inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7700x, with collision cell) was used. The microwave system for Total As digestion and InAs extraction, was an Ethos One from Millestone.

Quality control/quality assurance (QC/QA) of the analytical method was ensured by analyzing standards and blanks every batch of samples, plus spiked samples and reference materials (from several FAPAS interlaboratory proficiency tests). Both methods are accredited under ISO 17025.

### 2.3. *Dietary exposure assessment*

The dietary exposure to total As and InAs was calculated by groups of population according to age: toddlers (6 – 11 months), infants (12 – 36 months), children (3 – 9 years), adolescents (10 – 17 years), young adults (18 – 39 years), adults (40 – 64 years), seniors (65 – 74 years) and pregnant women. The mean body weight for each population group was obtained from the literature (Table 1).

Data on food consumption were obtained from the Spanish National Food Survey in Children and Adolescents (ENALIA) and the Spanish National Food Survey in Adults, Elderly and Pregnant Women (ENALIA2). Both surveys were conducted by the Spanish Agency of Consumption, Food Safety and Nutrition (AECOSAN) (AECOSAN, 2016a,b).

For calculations, when the concentration of total As or InAs in the analysed composites were under the respective limit of detection (LOD), that value was assumed to be one-half of that limit (ND=1/2LOD).

Risk assessment was conducted considering three exposure scenarios, which were considered taking into account that brown round rice from Spain contained the highest concentration of InAs, while the lowest levels corresponded to pre-cooked rice. These 3 simulated exposure scenarios were: a) high exposure, considering that the rice consumed is brown round rice from Spain; b) mean exposure, based on the mean value of all varieties; and c) low exposure, considering that only pre-cooked rice is consumed.

## 3. Results and discussion

Mean concentrations of total As and InAs are summarized in Table 2. In general, brown varieties of rice contained higher concentrations of total As and InAs than white rice. Specifically, the highest levels of total As and InAs were found in the brown round rice from Spain (229 and 190 µg/kg, respectively). This difference between brown and white rice is most probably due to the removal of the bran during the polishing of white rice, where most of the arsenic is accumulated. These findings are in agreement with other studies (Althobiti et al., 2018; Chen et al., 2015; Lee et al., 2018; Yim et al., 2017).

On the other hand, few variations in the concentrations of total As and InAs are found when the origin of the rice is considered. Spanish rice has a slightly higher amount of total As and InAs than Asian rice: 169 vs 141 µg/kg for total As, and 122 vs 92 µg/kg for InAs, which means a difference of 17 and 25%, respectively, between both origins.

The concentrations of total As and InAs among rice from Asia were also different. Asian white, long rice was harvested in India and Pakistan, while white, round rice was from Japan. In terms of As, Japanese white rice was more contaminated than Indian/Pakistani white rice. Thus, total As and InAs content was 55% and 33% lower in rice from India and Pakistan than that from Japan, respectively. On the other hand, Asian brown long rice was also harvested in India and Pakistan. As expected, a difference of 57% and 53% were observed between brown and white long rice for total As and InAs.

The concentrations of total As and InAs were also dependent on the variety as well as the grain length. The average content of total As and InAs in long grains were 142 and 99 µg/kg, respectively. In turn, concentrations of total As and InAs were 177 and 122 µg/kg, respectively, in round grains. Hence, round rice contained a concentration about 30% higher of As (both total As and InAs) than that of long grains.

Interestingly, pre-cooked rice contains less than the half of the concentration of As and InAs contained in dry rice. In fact, pre-cooked rice has already been boiled, and consequently, the content of arsenic has been partially removed. Indeed, it has been reported that simply washing the rice before cooking, or boiling it with plenty of water, could reduce the arsenic content up to 60% (Althobiti et al., 2018; Jitaru et al., 2016; Gray et al., 2015).

Regarding rice products, rice flour and rice cakes showed the highest concentration of total As (208 and 173 µg/kg, respectively) and InAs (112 and 135 µg/kg, respectively). In contrast, the lowest levels of both total As and InAs were found, by far, in the baby food with chicken and rice (9.1 and 3.1 µg/kg, respectively). Anyhow, none of the analysed rice-based food products exceeded the maximum InAs level, 0.3 mg/kg, set by the European Commission (EC) (EC, 2015)

There is a huge difference between the concentration of total As in baby food containing fish and rice and that of baby food made with chicken and rice (163 vs 9.1 µg/kg, respectively). This finding is in agreement with the fact that fish and seafood is the food group with the highest arsenic content (González et al., 2019; Perelló et al., 2014). Regarding InAs, the EC established in 2015 a limit of 100 µg/kg for InAs for the rice intended for the production of food for infants and young children (EC, 2015). None of the baby products exceeded that threshold, being levels actually well below: 4.0 µg/kg for baby food with fish and rice, and 3.1 µg/kg for baby food with chicken and rice (Table 2).

The estimated dietary intake of total As and InAs by the adult population of Catalonia is shown in Table 3. Baby products were excluded because they are not consumed by the adult population (AECOSAN, 2016b). As expected, rice is the major contributor to As and InAs exposure (1.61 and 1.11 µg/day, respectively). It means that rice accounts with more than the 80% of the total exposure to both arsenic species. In fact, rice is the most consumed food item with relatively high amount of arsenic species among all the rice foodstuffs under study.

Figure 1 depicts the estimated dietary exposure to InAs and organic arsenic by population groups. Toddler and infants were the population group presenting the highest exposure to total As (4.08 and 3.99 µg/day, respectively). Looking into the contribution of each arsenic species, the main contribution comes from organic arsenic (57% and 55%, respectively) in toddler and infants. In contrast, remaining groups of population (children, adolescents, young adults, adults, seniors and pregnant women) were more exposed to InAs than organic Arsenic, being the former responsible for up to 68% of total exposure.

As aforementioned, fish and seafood are the major contributors to the total As exposure. Hence, calculating the total dietary exposure to As considering the two main contributors would provide a comprehensive approach. With this purpose, data recently published by González et al., (2019) – together with the present exposure findings – has been used. Thus, the estimated exposure to total As considering rice, rice-based products, fish and seafood by population groups is as follows: toddlers, 64 µg/day; infants, 191 µg/day; children, 134 µg/day; adolescents, 173 µg/day; young adults, 94 µg/day; adults, 128 µg/day; seniors, 138 µg/day; and pregnant women, 102 µg/day.

The contribution of each food item to the total exposure to InAs is depicted in Figure 2. Baby cereals and breakfast cereals containing rice, are the highest contributors to InAs exposure for toddlers and infants, while rice is the major contributor to InAs exposure for the rest of population groups.

The comparison between the estimated dietary exposure for the three scenarios and the $BMDL_{01}$ is shown in Figure 3. The average dietary intake of InAs was estimated in 0.02 µg/kg bw/day for the adult population. Even though the highest exposure corresponded to the toddlers (0.22 µg/kg bw/day), none of the population groups exceeded the established threshold in any of the considered scenarios.

For comparison purposes, the concentrations of the total As and InAs in rice from various countries are summarized in Table 4. The high variability among the results of these studies suggest that the content of arsenic does not depend on the region or either the continent. In Europe, levels of total As range from 76 (Spain) to 400 µg/kg (Italy), while in Asia, total As content varies from 56 (Bangladesh) to 872 µg/kg (Iran). In turn, in Oceania, total As concentrations found in Australia vary between 283 and 438, while in America, As concentrations are notably different from Canada (28 µg/kg) to Argentina (858 µg/kg). Similar findings were found for InAs, as its content in rice varies among countries and within continents. This variability is mainly attributed to the fact that the content of arsenic in the soil and in the irrigation water is directly related with the content of arsenic in the harvested rice (Sofuoglu et al., 2014), regardless the socioeconomic status of the country.

Finally, data regarding the intake of InAs in different countries are presented in Table 5. In general terms, the current dietary exposure to InAs for the Catalan population is lower than those reported by the scientific literature in other countries. However, the contribution of rice to the total As exposure is in the upper part of the range in comparison with those levels of total As reported in other studies. As expected, Asian countries show a higher exposure to InAs mainly because rice is highly consumed since it is an essential food in their dietary habits (Lin et al., 2015; Naito et al., 2015).

## 4. Conclusions

In this study, the concentrations of total As and InAs in rice and rice-based products, as well as the dietary exposure considering different population groups and exposure scenarios, have been assessed. Brown varieties of rice presented higher concentrations of total As and InAs than white rice varieties, being the variety "brown, round rice from Spain" the one with the highest concentration of total As and InAs. In general, Spanish rice contained more total As and InAs than Asian rice. However, the difference was not significant, and it could be more due to the differential content of As and InAs in different rice varieties rather than geographical variations. According to the scientific literature, the concentrations of total As and InAs vary considerably among countries and continents. Regarding rice products, rice flour and rice cakes were the food items with the highest content of total As and InAs.

The average dietary intake of InAs was estimated in 0.02 µg/kg bw/day for the adult population. This value is below the threshold set by EFSA (0.3 µg/kg bw/day). According to present findings, none of the population groups surpassed this safety limit, even though under a high exposure scenario. Furthermore, it is in the lower range when compared with data from other studies.

Finally, although the current results show a low-risk exposure to arsenic, consumption of white rice varieties, as well as a pre-rinsing before cooking, is recommended.

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## 5. References


AECOSAN, 2016a. Encuesta nacional de alimentación en la población infantil y adolescente. Resultados sobre datos de consumo. Agencia Española de Consumo, Seguridad Alimentaria y Nutrición. Ministry of Health, Madrid, Spain Available at: http://www.aecosan.msssi.gob.es/AECOSAN/web/seguridad_alimentaria/ampliacion/enalia.htm, Access date: 10 March 2020.

AECOSAN, 2016b. Encuesta nacional de alimentación en población adulta, mayores y embarazadas. Resultados sobre datos de consumo. Agencia Española de Consumo, Seguridad Alimentaria y Nutrición. Ministry of Health, Madrid, Spain Available at: http://www.aecosan.msssi.gob.es/AECOSAN/web/seguridad_alimentaria/subdetalle/enalia_2.htm. Access date: 10 March 2020.

ATSDR, 2017. ATSDR's Substance Priority List. Agency for Toxic Substances and Disease Registry Available online at: https://www.atsdr.cdc.gov/SPL/. Access date: 10 March 2020.

Althobiti, R.A., Sadiq, N.W., Beauchemin, D., 2018. Realistic risk assessment of arsenic in rice. Food Chem. 257, 230–236.

Bailey, K., Xia, Y., Ward, W.O., Knapp, G., Mo, J., Mumford, J.L., Owen, R.D., Thai, S.F., 2009. Global gene expression profiling of hyperkeratotic skin lesions from inner Mongolians chronically exposed to arsenic. Toxicol. Pathol. 37: 849-859.

Bailey, K,A., Wallace, K., Smeester, L., Thai, S.F., Wolf, D.C., Edwards, S.W., Fry, R.C., 2012. Transcriptional Modulation of the ERK1/2 MAPK and NF-κB Pathways in Human urothelial cells after trivalent arsenical exposure: implications for urinary bladder cancer. J. Can. Res. Updates 1: 57-68.

Carrascosa, A., Fernández, J.M., Fernández, A., López-Siguero, J.P., López, D., Sánchez, E., Colaborador, y Grupo, 2010. Estudios de crecimiento. [in Spanish] Available at: http://www.estudiosdecrecimiento.es/estudio-transversal.html. Access date: 10 March 2020.

Chen, H.L., Lee, C.C., Huang, W.J., Huang, H.T., Wu, Y.C., Hsu, Y.C., Kao, Y.T., 2015. Arsenic speciation in rice and risk assessment of inorganic arsenic in Taiwan population. Environ. Sci. Pollut. Res. 23, 4481–4488.

Chung, J.Y., Yu, S. Do, Hong, Y.S., 2014. Environmental source of arsenic exposure. J. Prev. Med. Public Heal. 47, 253–257.

Ciminelli, V.S.T., Gasparon, M., Ng, J.C., Silva, G.C., Caldeira, C.L., 2017. Dietary arsenic exposure in Brazil: The contribution of rice and beans. Chemosphere 168, 996–1003.

Cubadda, F., D'Amato, M., Aureli, F., Raggi, A., Mantovani, A., 2016. Dietary exposure of the Italian population to inorganic arsenic: The 2012–2014 Total Diet Study. Food Chem. Toxicol. 98, 148–158.

Djahed, B., Taghavi, M., Farzadkia, M., Norzaee, S., Miri, M., 2018. Stochastic exposure and health risk assessment of rice contamination to the heavy metals in the market of Iranshahr, Iran. Food Chem. Toxicol. 115, 405-412

EC, 2015. European Commission (EU) 2015⁄1006 of 25 June 2015 amending Regulation (EC) No 1881/2006 as regards maximum levels of inorganic arsenic in foodstuffs. Official Journal L 161, 26.6.2015.

EFSA, 2014. Dietary exposure to inorganic arsenic in the European population. EFSA J. 12. https://doi.org/10.2903/j.efsa.2014.3597

EPA, 2016. Arsenic compounds. Natl. Cent. Environ. Assessment, Off. Res. Dev. Washington, DC, USA.

FAO/WHO, 2010. JOINT FAO/WHO Expert Committee On Food Additives, Seventy-second meeting Rome, 16–25 February 2010 1–15.

Gavilanes-Terán, I., Cano-Lamadrid, M., Idrovo-Novillo, J., García-García, E., Veloz-Mayorga, N., Erazo-Arrieta, R., Burló, F., Cruz-Paca, F., Carbonell-Barrachina, Á.A., 2019. Inorganic arsenic content in Ecuadorian rice-based products. Food Addit. Contam. - Part A Chem. Anal. Control. Expo. Risk Assess. 36, 922–928.

González, N., Calderón, J., Rúbies, A., Timoner, I., Castell, V., Domingo, J.L., Nadal, M., 2019. Dietary intake of arsenic , cadmium , mercury and lead by the population of Catalonia , Spain : Analysis of the temporal trend. Food Chem. Toxicol. 132, 110721.

Gray, P.J., Conklin, S.D., Todorov, T.I., Kasko, S.M., 2015. Cooking rice in excess water reduces both arsenic and enriched vitamins in the cooked grain. Food Addit. Contam. Part A Chem. Anal. Control Expo. Risk Assess. 33, 78-85

Guillod-Magnin, R., Brüschweiler, B.J., Aubert, R., Haldimann, M., 2018. Arsenic species in rice and rice-based products consumed by toddlers in Switzerland. Food Addit. Contam. - Part A Chem. Anal. Control. Expo. Risk Assess. 35, 1164–1178.

Halder, D., Biswas, A., Šlejkovec, Z., Chatterjee, D., Nriagu, J., Jacks, G., Bhattacharya, P., 2014. Arsenic species in raw and cooked rice: Implications for human health in rural Bengal. Sci. Total Environ. 497–498, 200–208.

Hojsak, I., Braegger, C., Bronsky, J., Campoy, C., Colomb, V., Decsi, T., Domellöf, M., Fewtrell, M., Mis, N.F., Mihatsch, W., Molgaard, C., Van Goudoever, J., 2015. Arsenic in rice: A cause for concern. J. Pediatr. Gastroenterol. Nutr. 60, 142–145.

Huang, Y., Wang, M., Mao, X., Qian, Y., Chen, T., Zhang, Y., 2015. Concentrations of inorganic arsenic in milled rice from China and associated dietary exposure assessment. J. Agric. Food Chem. 63, 10838–10845.

Hubaux, R., Becker-Santos, D.D., Enfield, K.S., Rowbotham, D., Lam, S., Lam, W.L., Martinez, V.D., 2013. Molecular features in arsenic-induced lung tumors. Mol. Cancer 12: 20.

Hughes, M.F., 2002. Arsenic toxicity and potential mechanisms of action. Toxicol. Lett. 133, 1–

16.
IARC, 2012. Arsenic, metals, fibres, and dusts. A review of human carcinogens. Iarc Monogr. 100, 407–443.
INE, 2001. Institut Nacional d'Estadística. Peso medio de la población por países, sexo, periodo y edad. Available at: http://www.ine.es/jaxi/tabla.do?path=/t25/p442/e01/l0/&file=02006.px&type=pcaxis, Access date: 10 March 2020.
Islam, S., Rahman, M.M., Islam, M.R., Naidu, R., 2017. Geographical variation and age-related dietary exposure to arsenic in rice from Bangladesh. Sci. Total Environ. 601–602, 122–131.
Jitaru, P., Millour, S., Roman, M., El Koulali, K., Noël, L., Guérin, T., 2016. Exposure assessment of arsenic speciation in different rice types depending on the cooking mode. J. Food Compos. Anal. 54, 37–47.
Jomova, K., Jenisova, Z., Feszterova, M., Baros, S., Liska, J., Hudecova, D., Rhodes, C.J., Valko, M., 2011. Arsenic: toxicity, oxidative stress and human disease. J Appl Toxicol. 31, 95-107.
Kollander, B., Sand, S., Almerud, P., Ankarberg, E.H., Concha, G., Barregård, L., Darnerud, P.O., 2019. Inorganic arsenic in food products on the Swedish market and a risk-based intake assessment. Sci. Total Environ. 672, 525–535.
Kwon, J.C., Nejad, Z.D., Jung, M.C., 2017. Arsenic and heavy metals in paddy soil and polished rice contaminated by mining activities in Korea. Catena 148, 92–100.
Lee, S.G., Kim, J., Park, H., Holzapfel, W., Lee, K.W., 2018. Assessment of the effect of cooking on speciation and bioaccessibility/ cellular uptake of arsenic in rice, using in vitro digestion and Caco-2 and PSI cells as model. Food Chem. Toxicol. 111, 597-604.
Lin, K., Lu, A., Wang, J., Yang, Y., 2015. The arsenic contamination of rice in Guangdong Province, the most economically dynamic provinces of China: arsenic speciation and its potential health risk. Environ Geochem Health 37, 353-361
Llorente-Mirandes, T., Calderón, J., López-Sánchez, J.F., Centrich, F., Rubio, R., 2012. A fully validated method for the determination of arsenic species in rice and infant cereal products. Pure Appl. Chem. 84, 225–238.
Londonio, A., Morzán, E., Smichowski, P., 2019 Determination of toxic and potentially toxic elements in rice and rice-based products by inductively coupled plasma-mass spectrometry. Food Chem. 284, 149-154
López-Sobaler, A.M., Aparicio, A., Aranceta-Bartrina, J., Gil, A., González-Gross, M., Serra-Majem, Ll, Varela-Moreiras, G., 2016. Overweight and general and abdominal obesity in a representative sample of Spanish adults: findings from the ANIBES study. BioMed Res. Int. 8341487 2016.
Ma, L., Wang, L., Jia, Y., Yang, Z., 2016. Arsenic speciation in locally grown rice grains from Hunan Province, China: Spatial distribution and potential health risk. Sci. Total Environ.

557–558, 438–444.

Margallo, M., Taddei, M.B.M., Hernández-Pellón, A., Aldaco, R., Irabien, Á., 2015. Environmental sustainability assessment of the management of municipal solid waste incineration residues: A review of the current situation. Clean Technol. Environ. Policy 17, 1333–1353.

Martínez, M.A., Rovira, J., Prasad Sharma, R., Nadal, M., Schuhmacher, M., Kumar, V., 2017. Prenatal exposure estimation of BPA and DEPH using integrated external and internal dosimetry: a case study. Environ. Res. 158, 566–575.

Meharg, A.A., Williams, P.N., Adomako, E., Lawgali, Y.Y., Deacon, C., Villada, A., Cambell, R.C.J., Sun, G., Zhu, Y.G., Feldmann, J., Raab, A., Zhao, F.J., Islam, R., Hossain, S., Yanai, J., 2009. Geographical variation in total and inorganic arsenic content of polished (white) rice. Environ. Sci. Technol. 43, 1612–1617.

Munera-Picazo, S., Ramírez-Gandolfo, A., Burló, F., Carbonell-Barrachina, Á.A., 2014. Inorganic and total arsenic contents in rice-based foods for children with celiac disease. J. Food Sci. 79, 122–128.

Naito, S., Matsumoto, E., Shindoh, K., Nishimura, T., 2015. Effects of polishing, cooking, and storing on total arsenic and arsenic species concentrations in rice cultivated in Japan. Food Chem. 168, 294-301

Narukawa, T., Matsumoto, E., Nishimura, T., Hioki, A., 2014. Determination of sixteen elements and arsenic species in brown, polished and milled rice. Anal. Sci. 30, 245-250

Perelló, G., Llobet, J.M., Gómez-Catalán, J., Castell, V., Centrich, F., Nadal, M., Domingo, J.L., 2014. human health risks derived from dietary exposure to toxic metals in Catalonia, Spain: Temporal trend. Biol. Trace Elem. Res. 162, 26–37.

Rahman, M.A., Rahman, M.M., Reichman, S.M., Lim, R.P., Naidu, R., 2014. Arsenic speciation in australian-grown and imported rice on sale in Australia: Implications for human health risk. J. Agric. Food Chem. 62, 6016–6024.

Rovira, J., Nadal, M., Schuhmacher, M., Domingo, J.L., 2018. Concentrations of trace elements and PCDD/Fs around a municipal solid waste incinerator in Girona (Catalonia, Spain). Human health risks for the population living in the neighborhood. Sci. Total Environ. 630, 34–45.

Rovira, J., Vilavert, L., Nadal, M., Schuhmacher, M., Domingo, J.L., 2015. Temporal trends in the levels of metals, PCDD/Fs and PCBs in the vicinity of a municipal solid waste incinerator. Preliminary assessment of human health risks. Waste Manag. 43, 168–175.

Rowell, C., Kuiper, N., Al-Saad, K., Nriagu, J., Shomar, B., 2014. A market basket survey of As, Zn and Se in rice imports in Qatar: Health implications. Food Chem. Toxicol. 70, 33-39.

Ruttens, A., Cheyns, K., Blanpain, A.C., De Temmerman, L., Waegeneers, N., 2018. Arsenic speciation in food in Belgium. Part 2: Cereals and cereal products. Food Chem. Toxicol.

118, 32-41.
Seo, M.N., Lee, S.G., Eom, S.Y., Kim, J., Oh, S.Y., Kwon, H.J., Kim, H., Choi, B.S., Yu, I.J., Park, J.D., 2016. Estimation of Total and Inorganic Arsenic Intake from the Diet in Korean Adults. Arch. Environ. Contam. Toxicol. 70, 647–656.
Sofuoglu, S.C., Güzelkaya, H., Akgül, Ö., Kavcar, P., Kurucaovalı, F., 2014. Speciated arsenic concentrations, exposure, and associated health risks for rice and bulgur. Food Chem. Toxicol. 64, 184-191.
Sommella, A., Deacon, C., Norton, G., Pigna, M., Violante, A., Meharg, A.A., 2013. Total arsenic, inorganic arsenic, and other elements concentrations in Italian rice grain varies with origin and type. Environ. Pollut. 181, 38–43.
Sung, T. I., Wang, Y.J., Chen, C.Y., Hung, T.L., Guo, H.R., 2012. Increased serum level of epidermal growth factor receptor in liver cancer patients and its association with exposure to arsenic. Sci. Total Environ. 424: 74-78.
Tattibayeva, D., Nebot, C., Miranda, J.M., Cepeda, A., Mateyev, E., Erkebaev, M., Franco, C.M., 2016. A study on toxic and essential elements in rice from the Republic of Kazakhstan: comparing the level of contamination in rice from the European Community. Environ. Geochem. Health 38, 85–98.
Taylor, V., Goodale, B., Raab, A., Schwerdtle, T., Reimer, K., Conklin, S., Karagas, M.R., Francesconi, K.A., 2017. Human exposure to organic arsenic species from seafood. Sci. Total Environ. 580, 266–282.
WHO, 2003. Child Growth Standards 1997-2003. Word Health Organization. Available at: http://www.who.int/childgrowth/standards/weight_for_age/en/, Access date: 10 March 2020.
Wong, W.W.K., Chung, S.W.C., Chan, B.T.P., Ho, Y.Y., Xiao, Y., 2013. Dietary exposure to inorganic arsenic of the Hong Kong population: Results of the first Hong Kong Total Diet Study. Food Chem. Toxicol. 51, 379–385.
Yim, S.R., Park, G.Y., Lee, K.W., Chung, M.S., Shim, S.M., 2017. Determination of total arsenic content and arsenic speciation in different types of rice. Food Sci. Biotechnol. 26, 293–298.
Yuan, Y., Marshall, G., Ferreccio, C., Steinmaus, C., Liaw, J., Bates, M., Smith, A.H., 2010. Kidney cancer mortality: fifty-year latency patterns related to arsenic exposure. Epidemiology 21: 103-108.

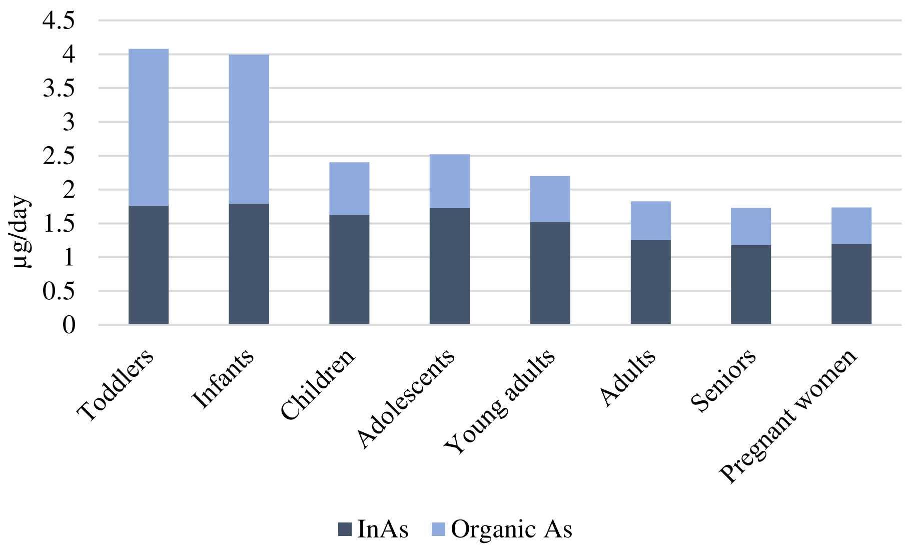


**Figure 1**. Dietary intake of total As and InAs by different population groups

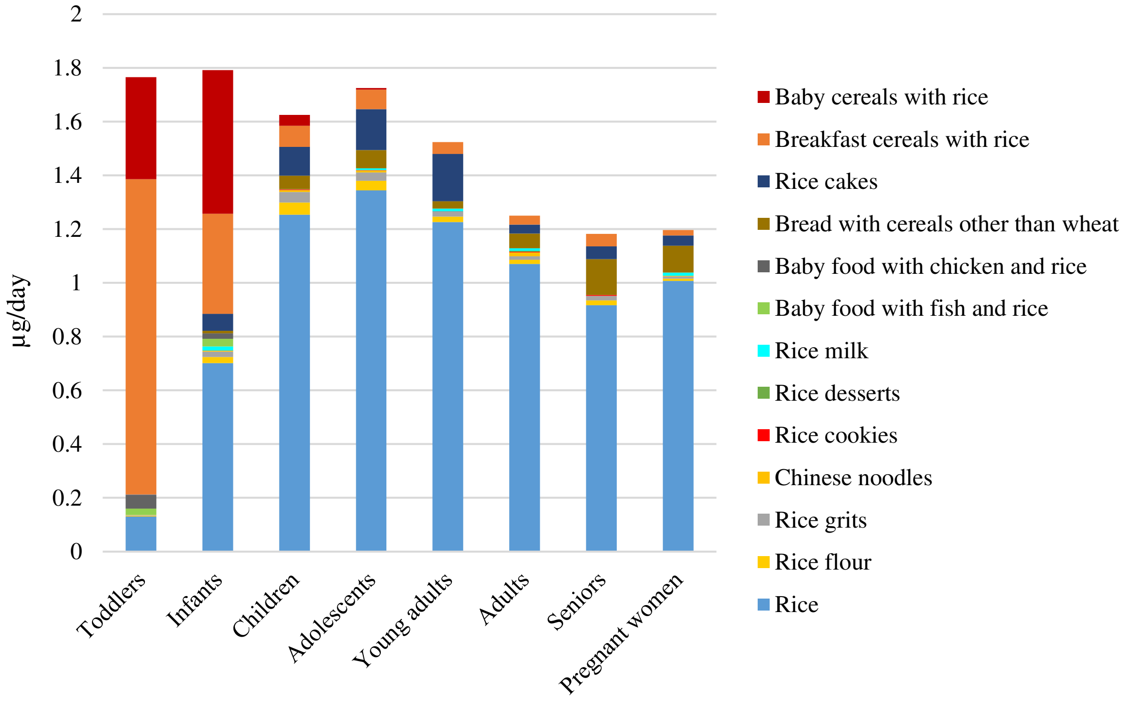


**Figure 2**. Contribution of individual food items to the exposure of InAs for different population groups.

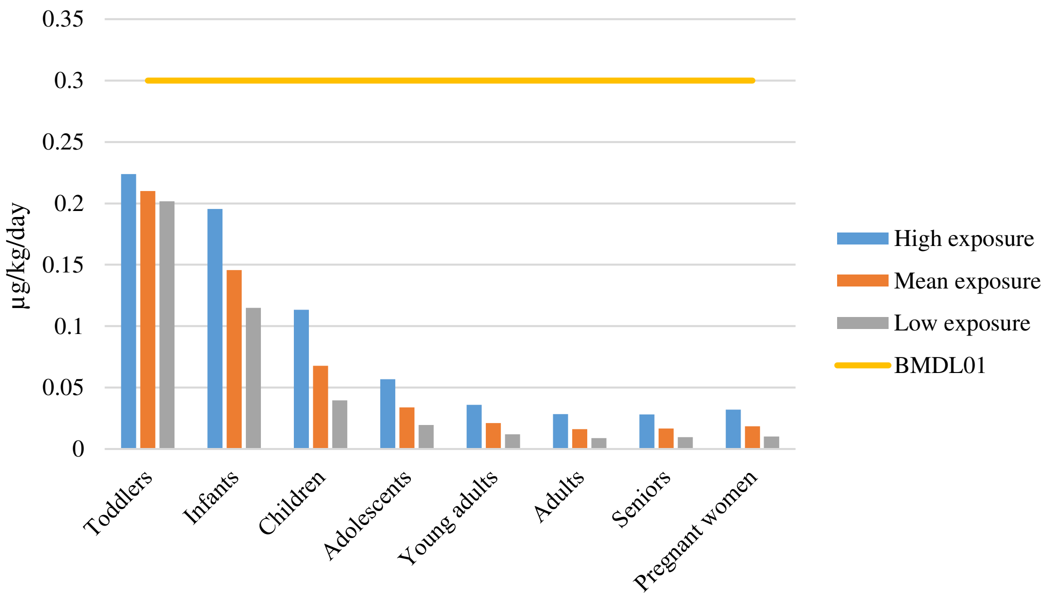


**Figure 3**. Dietary intake of InAs by various population groups in three exposure scenarios, and comparison to the safety level established by EFSA

**Table 1**

Population groups in this study

| Population group | Age (years) | Body weight (kg) | Reference |
|---|---|---|---|
| Toddlers | 6 – 11 months | 8.4 | WHO, 2003 |
| Infants | 12 – 36 months | 12.3 | Carrascosa et al., 2010 |
| Children | 3 – 9 | 24 | Carrascosa et al., 2010 |
| Adolescents | 10 – 17 | 51 | Carrascosa et al., 2010 |
| Young adults | 18 – 39 | 72 | López-Sobaler et al., 2016 |
| Adults | 40 – 64 | 77 | López-Sobaler et al., 2016 |
| Seniors | 65 – 74 | 70.5 | INE, 2001 |
| Pregnant women | - | 65 | Martínez et al., 2017 |

**Table 2**

Concentrations (µg/kg) of total As and InAs in marketed rice and rice products food items in Catalonia, Spain

| Food item | As | InAs |
|---|---|---|
| White long rice from Spain | 151 | 90 |
| White long rice from Asia (India/Pakistan) | 76 | 60 |
| White round rice from Spain | 132 | 87 |
| White round rice from Asia (Japan) | 169 | 89 |
| Brown long rice from Spain | 162 | 119 |
| Brown long rice from Asia (India/Pakistan) | 177 | 127 |
| Brown round rice from Spain | 229 | 190 |
| Pre-cooked rice | 73 | 47 |
| *Rice from Spain* | *169* | *122* |
| *Rice from Asian countries* | *141* | *92* |
| ***Mean rice*** | **146** | **101** |
| Rice flour | 208 | 112 |
| Rice grits | 162 | 100 |
| Chinese noodles | 65.3 | 45.8 |
| Rice cookies | 18.0 | 13.2 |
| Rice desserts | 18.1 | 9.3 |
| Rice milk | 16.0 | 13.1 |
| Baby food with fish and rice | 163 | 4.0 |
| Baby food with chicken and rice | 9.1 | 3.1 |
| Bread with cereals other than wheat | 72 | 49 |
| Rice cakes | 173 | 135 |
| Breakfast cereals with rice | 72 | 40 |
| Baby cereals with rice | 85 | 47 |

**Table 3**

Food consumption (g/day) and estimated dietary intake of total As and InAs (µg/day) by the adult population from Catalonia, Spain

| Food item | Consumption (g/day) | As (µg/day) | InAs (µg/day) |
|---|---|---|---|
| Rice | 11 | 1.61 | 1.11 |
| Rice flour | 0.15 | 0.03 | 0.02 |
| Rice grits | 0.15 | 0.02 | 0.02 |
| Chinese noodles | 0.15 | 0.01 | 0.01 |
| Rice cookies | 0.14 | 0.003 | 0.002 |
| Rice desserts | 0.18 | 0.003 | 0.002 |
| Rice milk | 0.69 | 0.01 | 0.01 |
| Bread with cereals other than wheat | 1.10 | 0.08 | 0.05 |
| Rice cakes | 0.68 | 0.12 | 0.09 |
| Breakfast cereals with rice | 0.98 | 0.07 | 0.04 |
| **TOTAL** | **15.3** | **1.96** | **1.35** |

**Table 4**

Concentration (µg/kg) of total As and InAs in commercial rice grain from different countries.

| Country | Total As | Inorganic As | Reference |
|---|---|---|---|
| Spain | 76 – 169 (white rice)<br>162 – 229 (brown rice) | 60 – 90 (white rice)<br>119 – 190 (brown rice) | Present study |
| Italy | 400 | 271 | Rahman et al., 2014 |
| Italy | 180 to 280 | 80 to 110 | Sommella et al., 2013 |
| Kazakhstan | 240 | 360 | Tattibayeva et al., 2016 |
| Spain | 280 | 250 | Tattibayeva et al., 2016 |
| Portugal | 360 | 180 | Tattibayeva et al., 2016 |
| Belgium | 216 (white rice)<br>200 (brown rice)<br>148 (Thai rice)<br>61 (basmati rice)<br>108 (wild rice) | 172 (white rice)<br>167 (brown rice)<br>77 (Thai rice)<br>30 (basmati rice)<br>80 (wild rice) | Ruttens et al., 2018 |
| Bangladesh | 126 | 11 to 502 | Islam et al., 2017 |
| Bangladesh | 56 | 68 | Rahman et al., 2014 |
| India | 92 | 94 | Rahman et al., 2014 |
| India | 283 | 194 | Halder et al., 2014 |
| Japan | 239 | 208 | Narukawa et al., 2014 |
| China | 116.5 | 90.9 | Huang et al., 2015 |
| China | 129.4 | 111.8 | Ma et al., 2016 |
| Pakistan | 82 | 83 | Rahman et al., 2014 |
| Iran | 91 to 872 | - | Djahed et al., 2018 |
| Qatar | 41.3 to 169 | - | Rowell et al., 2014 |
| Thailand | 172 | 168 | Rahman et al., 2014 |
| Taiwan | 116.6 (white rice)<br>215.5 (brown rice) | 70 (white rice)<br>110 (brown rice) | Chen et al., 2015 |
| Korea | 247 | - | Kwon et al., 2017 |
| Korea | 112 to 161 (white rice)<br>157 to 260 (brown rice) | 76 to 130 (white rice)<br>103 to 158 (brown rice) | Lee et al., 2018 |
| Australia | 283 (white rice)<br>438 (brown rice) | 165 – 177 (white rice)<br>178 – 276 (brown rice) | Rahman et al., 2014 |
| USA | 250 | 100 | Meharg et al., 2009 |
| Canada | 28 | 19 | Rahman et al., 2014 |
| Argentina | 67 – 858 | - | Londonio et al., 2019 |

**Table 5**

Recent data (2013-2019) on the dietary intake of InAs and percentage of rice contribution to the total exposure in various countries.

| Country | InAs (µg/kg bw/day) | Food items | Reference |
|---|---|---|---|
| Spain | 0.02 (82%) | Rice, rice flour, rice grits, Chinese noodles, rice cookies, rice desserts, rice milk, bread with cereals other than wheat, rice cakes, breakfast cereals, baby cereals with rice, baby food with rice | Present study |
| Spain | 0.61 to 0.78* | Rice-based pasta, rice-based bread, rice-based breakfast cereals, rice-based chocolate wafers, rice-based biscuits | Munera-Picazo et al., 2014 |
| Hong Kong | 0.22 (45.2%) | Cereals and their products, vegetables and their products, legumes, nuts and seeds, fruits, meat and meat products, eggs and their products, fish and seafood, dairy products, fats and oils, alcoholic beverages, non-alcoholic beverages, mixed dishes, snack foods, sugars and confectionery, condiments, sauces and herbs | Wong et al., 2013 |
| Qatar | 0.26 | Rice | Rowell et al., 2014 |
| Korea | 0.2 (54%) | Grains, potatoes, sugars, pulses, seeds, vegetables, mushrooms, fruits, meats, eggs, fish and shellfish, seaweeds, milks, oils, beverages, flavourings | Seo et al., 2016 |
| Italy | 0.074 to 0.086 (36%) | Cereals and cereal products, pulses, vegetables, potatoes and tubers, fruit, meat and meat products, fish and seafood, milk and milk products, oils and fats, eggs, alcoholic beverages, sweet products, water and non-alcoholic beverages | Cubadda et al., 2016 |
| Brazil | 0.188 (78.7%) | Beans, beef, cabbage, carrot, chicken breast, coffee, egg, fish, garlic, lettuce, milk, pasta, potatoes, rice, tomato | Ciminelli et al., 2017 |
| Bangladesh | 0.38 to 1.92 | Rice | Islam et al., 2017 |
| Switzerland | 0.044* | Rice cereals, milk rice, baby food dry form, baby food ready-to-use form, rice cracker, rice drinks, rice grains, rice dishes | Guillod-Magnin et al., 2018 |
| Belgium | 0.04 (54%) | Rice, breakfast cereals with rice, pasta, bread, breakfast cereals without rice | Ruttens et al., 2018 |

| | | | |
|---|---|---|---|
| Spain | 0.04 (42%) | Meat and meat products, fish and seafood, vegetables, tubers, fruit, eggs, milk and dairy analogues, dairy products, bread and cereals, pulses, oils, industrial bakery, sauces, chocolates, infant food | González et al., 2019 |
| Ecuador | 0.55* | Rice cakes, breakfast cereals, cereal bars, baby and children products, biscuit, milk and cereal products | Gavilanes-Terán et al., 2019 |
| Sweden | 0.056 (18%) | Cereals, fruits, sweets and condiments, fish, pastries, vegetables, potatoes, meat, egg, dairy products, beverages | Kollander et al., 2019 |

*Rice was not analysed in these studies, therefore, percentage of rice contribution is not provided.