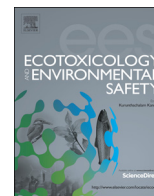




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The arsenic content in marketed seafood and associated health risks for the residents of Shandong, China

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ABSTRACT

Seafood is considered as the main source of arsenic in the human diet. In this study, we quantified the total arsenic content in 200 samples of 22 species collected from eight cities in Shandong, China. Subsequently, we evaluated the health risks associated with seafood consumption for three consumption scenarios based on the quantification of inorganic arsenic in three commonly consumed seafood species. The bivalves had the highest total arsenic concentrations in three categories of seafood (fish, shrimp, and bivalves) and the mean total arsenic concentrations ranged from 0.037 $\mu\text{g/g}$ ww in fish to 3.4 $\mu\text{g/g}$ in bivalves. The results suggested that organisms which had a closer relationship with sediments may accumulate more arsenic. Bivalves were the major contributor for the arsenic intakes in the seafood consumers. The margins of exposure (MOEs) estimated in the present work showed that there existed a health risk for the consumers. The carcinogen risks exceeded the acceptable range for life cancer risk. Our results suggested that more attention should be paid to the safety of seafood consumption, especially of benthic economic species and for special consumers.

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1. Introduction

Arsenic (As) is a ubiquitously distributed element, naturally present in the environment as a result of volcanic activity and weathering of As-containing minerals. Its abundance has been elevated by anthropogenic activities (e.g., pesticide use, burning of coal, and industrial activity) in the past century (Bissen and Frimmel, 2003; Smedley and Kinniburgh, 2002). Arsenic is involved in many complex chemical and biological processes, and present in a number of organic and inorganic forms in water, air, soil and food, including arsenite (As (III)), arsenate (As(V)), monomethylarsonic acid (MMA), dimethylarsinic acid (DMA), arsenobetaine, arsenosugars and arsenocholine, etc. (Hughes,

2002; Mandal and Suzuki, 2002; Tuzen et al., 2009a; Uluozlu et al., 2010).

The toxicity of arsenic to multicellular organisms is dependent on its chemical forms and oxidation states. Inorganic arsenic species (iAs), including As (III) and As (V), the most toxic forms due to their high bioavailability and toxicological effects, are classified as non-threshold, class (I) human carcinogens (ATSDR, 2007). When entering the food chain, iAs and their compounds will be progressively metabolized to less toxic forms via methylation (Reimer et al., 2010). Therefore, in the context of human health risk assessment, previous studies mainly focused on exposure to As in drinking water and airborne As (ACGIH, 2002; IARC Working Group, 2004; Kapaj et al., 2006), however, recent studies have demonstrated that As is routinely found in most foods (Borak and Hosgood, 2007). There is worldwide concern about dietary As exposure and the associated health risks with iAs intakes (Fontcuberta et al., 2011; Gilbert-Diamond et al., 2011; Halder et al., 2012, 2013), and has been emphasized in recent evaluations by the European Food Safety Authority (EFSA) and the Joint Food and Agriculture Organization of the United Nations/World Health Organization (FAO/WHO) Expert Committee on Food Additives (JECFA) (EFSA, 2009; FAO/WHO, 2010).

In 1989, a provisional tolerable weekly intake (PTWI) for iAs (15 $\mu\text{g/kg}$ body weight) was established by JECFA (FAO/WHO, 1989).

Abbreviations: ACGIH, American Conference of Governmental Industrial Hygienists; ATSDR, Agency for Toxic Substances and Disease Registry; BMD, benchmark dose; BMDL, the lower 95 percent confidence limit of BMD; EFSA, European Food Safety Authority; IARC, International Agency for Research on Cancer; IRIS, Integrated Risk Information System; JECFA, the Joint Food and Agriculture Organization of the United Nations/World Health Organization (FAO/WHO) Expert Committee on Food Additives; MOEs, margins of exposure; POD, point of departure; PTWI, provisional tolerable weekly intake

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However, recent studies showed that the PTWI for iAs was not conservative because lung and bladder cancer and a range of adverse effects had been reported at exposure levels previously thought to be safe (EFSA, 2009; Ferreccio et al., 2000; Kurtio et al., 1999). The EFSA has suggested the health risks for dietary As intakes should be assessed using margins of exposure (MOEs), a ratio calculated by comparing the dose at which a small but measurable adverse effect is first observed and the level of exposure to iAs (EFSA, 2009).

Seafood is the main source of As in the human diet. It was estimated that about 90 percent of As in U.S. diet was from seafood (Larsen and Berg, 2001). Shandong province lies on the east coast of China, which borders the Bohai Sea to the north, and the Yellow Sea to the southeast (Fig. 1). It is one of the most important provinces for seafood production and export. Total production of seafood in Shandong was up to 6.46 million tons in 2010, which contributed to approximately 20 percent of China's seafood production. Seafood is a common diet for the residents in the coastal cities of Shandong province. Fish, shrimp and bivalves are the most commonly consumed species. In 2011, it was estimated that export of seafood from Shandong province reached 0.78 million tons and accounted for about 20 percent of that from China. The main importing countries included Japan, the United States, the European Union and Korea (Qingdao Customs District People's Republic of China, 2011).

In the present work, we quantified the total As content of various seafood species from different cities in Shandong province. Our goal was to investigate As content in seafood in one of the main production regions in China. Furthermore, based on the quantification of inorganic arsenic in three commonly consumed seafood species, we assessed the health risks associated with seafood consumption for local residents with different consumption habits.

2. Materials and methods

2.1. Sampling and sample preparation

From July to September 2011, a total of 200 samples of fish ($n=81$), shrimp ($n=35$) and bivalves ($n=84$) were collected from the local markets (supermarkets, municipal markets, and other retail stores) along the eastern coastline of Shandong province (Fig. 1). The species of the collected samples (shown in Fig. 2) were chosen because they represented the main available seafood in the market and are commonly consumed by the residents. The sample size for each species was given in Fig. 2 and the species names are presented in Table S1.

The specimens were wrapped in aluminum foil, stored on ice and brought to the laboratory on the same day of collection. Muscular tissue (edible part) from each fish was homogenized and treated as one sample. The edible parts of 2–3 shrimps from each collection location were pooled and homogenized to form a compound sample. For bivalves, the soft tissues were removed and pooled (six

specimens per pool) and then homogenized as one sample. The homogenized samples were stored in a freezer at -20°C until they were analyzed.

2.2. Chemicals and reagents

All glassware was soaked with 10 percent v/v nitric acid (HNO_3) for 24 h and then rinsed three times with deionized water before use. Concentrated ultrapure nitric acid (HNO_3) and sulfuric acid (H_2SO_4) were used for tissue digestions. Standard reference materials (BCR 627, TORT-2, NIST 1566a, NMIIJ CRM 7503-a) were purchased from CRM/RM information center (Beijing, China), other chemicals of analytical purity or better were obtained from Sinopharm Chemical Reagent Corporation (SCRC Company, China). Deionized water ($>18.2\text{ M}\Omega\text{ cm}^{-1}$) was used for the preparations of reagents.

2.3. Determination of total As and iAs in the samples

The total As concentrations in the samples were determined according to the protocol described by Vilano and Rubio (2001). Briefly, the samples (approximately 1.0 g of fish and shrimps or 0.5 g of bivalves) were submitted to a microwave-assisted wet digestion using 10.0 mL of concentrated nitric acid and 5.0 mL of

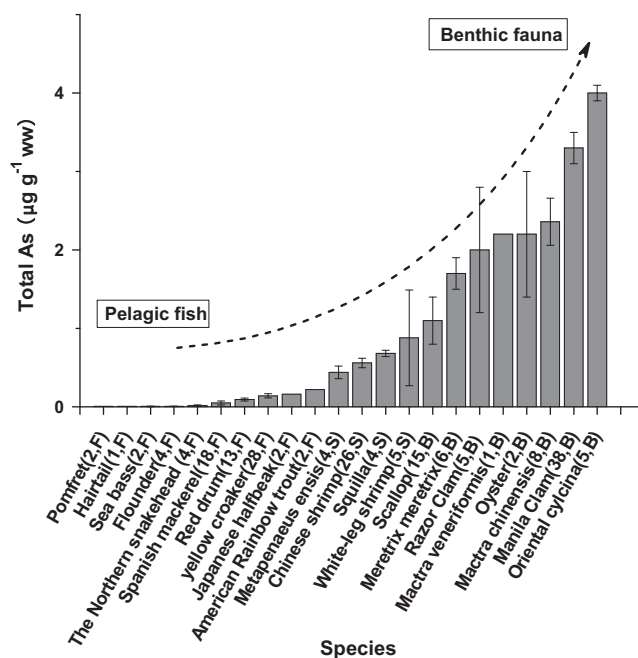


Fig. 2. The total As content for each sampling species. Standard error is shown for each species. The numbers in the parentheses are the replicates for each species and the capital letters indicate the food categories which the species belongs to. (F) Fish group; (S) shrimp group; and (B) bivalve group.



Fig. 1. Approximate of sampling cities in Shandong, China.

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