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Trace elements concentrations in squids consumed in Shandong Province China and their associated risks to the human health

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ABSTRACT

Determination of ten metal levels in 160 squid samples caught from offshore and the oceanic fishing locations by ICP-MS was made. The mean metal concentration in the squid muscles decreasing in the order of: Zn, Fe, Cu, As, Mn, Se, Cd, Pb, Cr and Ni. Metal concentrations in the squids were assessed for human uses according to provisional tolerable weekly intake (PTWI) and provisional tolerable daily intake (PTDI)·The estimated hazard quotients for the individual metals from squids were found in accordance to the following sequence: As > Cd > Cu > Se > Cr > Zn > Pb > Fe > Mn > Ni, with the greatest hazard upon human health coming primarily from As (1.34 < HQ < 1.73 in the Loliginidae from two offshore sampling sites and the Humboldt squid from the eastern Pacific Ocean). In regards to the heavy metal pollution of the coastal sea areas, the squids captured from offshore sites might pose a higher potential health risk to consumers compared to those from the ocean.

1. Introduction

Anthropogenically-derived trace elements are widely released into the marine environment. Large emission and contamination of trace elements are of concern at present because of the rapid economic growth and increasing population in recent years. As trace elements flow into marine systems, causing marine organisms to have continual exposure to low trace elements concentrations, it may result in bioaccumulation and eventually the subsequent transfer to man through the food chain. With the exception of occupational exposures, the main route of human exposure to trace elements is through their daily dietary, thus trace elements contamination of marine organisms is a serious threat to the human health (Sharma and Tripathi, 2008). Over the past decades, there has been an increasing interest in the determination of trace elements levels in the marine environment and further attention was paid to the measurement of contamination levels in public food supplies (Makedonski et al., 2017; Saha et al., 2016; Zaza et al., 2015).

The demand for fish protein has increased significantly due to the expansion of the global population within the past decades, allowing the demand for fishery products to exceed the traditional fish supply. Thus, alternative fish protein sources such as squids with short life-cycles have become one of the most important fishing targets and its total catches continues to increase (Cortes-Ruiz et al., 2016). Squids are antioxidant and anti-inflammatory mollusks that can supply high quality protein. They are renowned for their desirable and functional properties as well as their high nutritional value, making them a widely consumed seafood (Zhang et al., 2016). The acknowledgement of the commercial significance of squids to the world of fisheries is relatively recent, but growing in importance and recognition. The FAO yearbook Fishery and Aquaculture Statistics demonstrated that the capture production of squids, cuttlefishes and octopuses in 2014 was 4,779,091 t (FAO, 2016), with the output contributing to > 5% of total fishery production. Now squids support a major commercial fishing industry in China; a country that owns one of the largest fishing fleets for oceanic squids (Chen et al., 2008).

Over the past decades, concentrations of trace elements in fish have been studied extensively in various places around the world (Baharom and Ishak, 2015; Gu et al., 2015; Makedonski et al., 2017; Saha et al., 2016), however few reports are found on squids. The local residents of the Shandong province, located in the eastern coast of China, have a traditional custom of eating squids such as grilled squid, fried squid, and squid dumplings. To protect the diet safety and health of the local residents, it is essential to receive recognition in terms of the accumulation status and potential risk of trace elements in squids consumed in the Shandong Province, China. There are several analytical techniques

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can be taken for a metal concentration analysis in marine samples (Makedonski et al., 2017; Tuzen and Soylak, 2007; Zaza et al., 2015). This study specifically utilizes inductively coupled plasma mass spectrometry (ICP-MS) to determine the ten trace elements (Fe, Mn, Cr, Cu, Zn, Ni, As, Se, Cd, Pb) concentrations in the squids collected from four different fishing zones. Likewise, the human health risk assessment of residents' exposure to the trace elements present in squids is also of great significance, and several ways have been reported to assess the potential human health risks of seafood from their chemical exposures (Bourliva et al., 2017; Cherfi et al., 2014; de Souza et al., 2017). The Target Hazard Quotient (THQ) and the Estimated Daily Intake (EDI) were the chosen methods of assessment in this study to evaluate the health risk from exposure to the ten metals measured.

160 squid samples were collected and ten trace elements were determined by inductively coupled plasma mass spectrometry. The main objective of this study is: 1) to determine the concentrations of Fe, Mn, Cr, Cu, Zn, Ni, As, Se, Cd and Pb in squids captured from offshore and the ocean, 2) to assess the potential health risks of trace elements in squids from different fishing locations.

2. Materials and methods

2.1. Sample collection and preparation

The squid samples were collected from five sites (S1-S5) in the sea areas from June to December 2016 (Fig. 1). These sites represent the main fishing locations in which the Chinese catch edible squids for consumption. The samples studied are classed into four categories: (a) The Loliginidae (Loligo chinesis), captured from the offshore area of the Shandong Province, China (37°N-41°N, 117°E—121°E), are commonly known as pencil squids and are an aquatic family of the order Teuthida (squid). (b) The neon flying squid (*Ommastrephes bartramii*), sampled in the North Pacific(35°N-46°N,145°E-160°E) and often named the red flying squid, is a species of large flying squid in the family Ommastrephidae. (c) Illex argentines, captured from the southwestern Atlantic (35°S-55°S, 55°W-70°W), commonly known as the Argentine shotfin squid, is also a species of squid in the Ommastrephidae family. (d) The Humboldt squid (*Dosidicus gigas*), sampled in the eastern Pacific Ocean (5°S-30°S, 70°W-85°W), also known as jumbo flying squid, is a large squid living in the waters of the Humboldt Current in the eastern Pacific Ocean. Four categories can be grouped into offshore (category a) and ocean zones (category b, c, d) in this study. The (a) Loliginidae (Loligo chinesis) captured from the offshore area was purchased by our team from the Rizhao market and Penglai market within the Shandong Province, and the squid was caught by local fishermen in the offshore area. The ocean squid(b, c, d), was purchased from two shipping companies named Shandong Huiyang Group and Shandong Meijia Group. Upon buying their products, detailed information in regards to the squid samples was obtained, including the fishing area and fishing time.

All samples were transported to the laboratory in their original packaging, rinsed thoroughly with tap water, then with deionized distilled water, and finally dried between layers of clean tissue papers. The lengths and weights of the squids were then measured and recorded (Table 1). The moisture concentration of the samples was determined by drying them at 105 °C until a consistent weight was obtained (Nho et al., 2016). The dried samples were crushed in a blender (MR 350CA, Braun, Spain), placed in polyethylene bags, and stored at −20 °C until analysis.

2.2. Chemical analysis

Chemical analyses of the tissues were conducted as previously described (Yang et al., 2016). For a short period of time, the tissues were digested using HNO_3 (Superpure, Merck, Germany) in Teflon bombs. The concentrations of the ten trace elements (Fe, Mn, Cr, Cu, Zn, Ni, As, Se, Cd, Pb) as well as those in the tissues were determined by inductively coupled plasma mass spectrometry (ICP-MS, Thermo iCAPQ, Thermo Fisher Scientific, Bremen, Germany). The LOD of Fe, Mn, Cr, Cu, Zn, Ni, As, Se, Cd and Pb was 0.70, 0.045, 0.11, 0.10, 0.56, 0.045, 0.020, 0.13, 0.013, and 0.091 μg/L, respectively. Two certified reference Materials GBW10024(GSB-15) pectinid, GBW10050(GSB-28) prawn (Institute of Geophysical and Geochemical Prospecting, LangFang, China) were included in the QA/QC for data analyses (Table 2). The recoveries ranged from 90% to 110% and the differences were all within a range of 8%.

2.3. Risk assessment for human health

The human health risk assessment of the trace elements from fish consumption was estimated using the following equations:

$$EDI = \frac{C_{fish} \times CR}{BW}$$

$$THQ = \frac{EDI}{RfD}$$

$$TTHQ = THQ(toxicant1) + THQ(toxicant2) + \ldots\ldots + THQ(toxicantn)$$

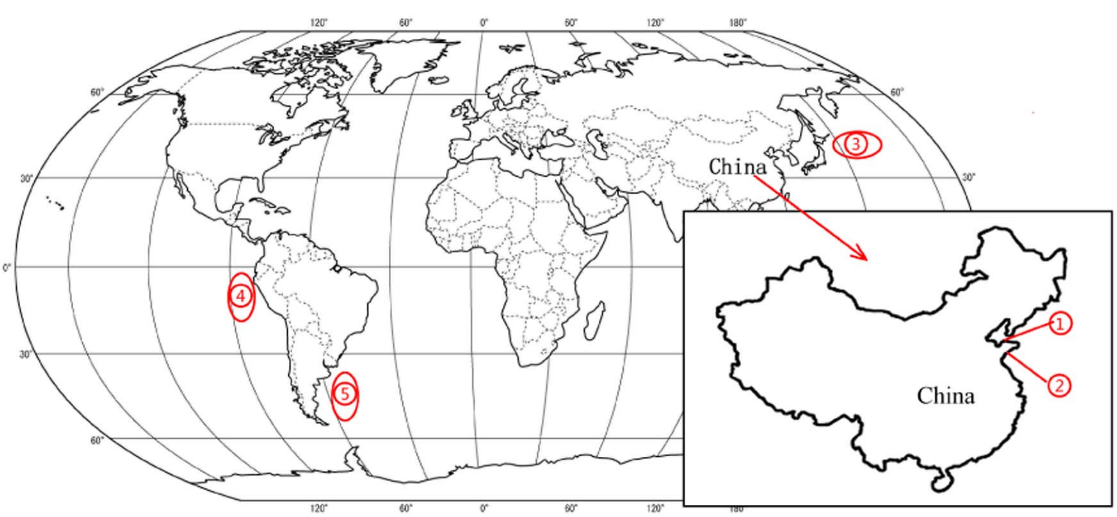

Fig. 1. Map of the fishing site locations, where the squids were collected, including two offshore areas: ①Penglai offshore area of Shandong Province ②Rizhao offshore area of Shandong Province, and three ocean areas: ③the North Pacific ④the southwestern Atlantic ⑤the eastern Pacific Ocean.