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Baseline

Arsenic and five metal concentrations in the muscle tissue of bigeye tuna (*Thunnus obesus*) in the Atlantic and Indian Oceans

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A B S T R A C T

White muscle concentrations of As, Cd, Cu, Fe, Se, and Zn were investigated in Atlantic- and Indian-bigeye tuna (BET) (*Thunnus obesus*) from 6 regions. As and Cd muscle concentrations were significantly higher in the Indian-BET than in the Atlantic-BET, whereas the Indian-BET caught in the waters off South Africa revealed the highest As, Se, and Zn muscle concentrations. Accordingly, multidimensional scaling separated them into two oceanic groups. Positive linear relationships between muscle Cd concentration and fork length (FL) were established in both oceans. For the other elements, only muscle-Fe and FL relationship was found in the Atlantic-BET. 10.3% of BET > 145 cm FL from both oceans possessed muscle Cd concentrations exceeding the food safety limit (0.1 μg g^{-1} wet weight) set by the European Commission. Increased Cd, Cu and Zn pollution was found in the Atlantic Ocean compared with previous data, with higher levels found in the Indian Ocean.

As a cosmopolitan species, the bigeye tuna (*Thunnus obesus*) live in the Pacific, Indian, and Atlantic Oceans. They inhabit tropical and subtropical areas of each ocean with limited gene exchange between the oceans (e.g. Alvarado Bremer et al., 1998; Durand et al., 2005). Therefore, their population in the northern and southern hemispheres of the same ocean is defined as the same stock (e.g. ICCAT, 1997; Shono et al., 2009; Hampton, 2010). They are an apex predator, exposed to various kinds of environmental contaminants through bioaccumulation and biomagnification processes in the food web (e.g. Gray, 2002; Barwick and Maher, 2003). Their wide distribution and key role in the ecosystem make them ideal sentinel organisms for global marine pollution biomonitoring studies (e.g. Marcovecchio et al., 1988, 1994; Vas, 1991), since they are known to accumulate high concentrations of metals in their tissues throughout their lifetime, therefore becoming an effective biological indicator (e.g. Ueno et al., 2004; Chen et al., 2011, 2017; Chen et al., 2014).

The bigeye tuna, *Thunnus obesus*, are a commercially important fisheries resource in the Atlantic, Indian, and Pacific Oceans. Their global capture production between 2012 and 2014 was roughly 400,000 metric tons per year (FAO, 2016). Since the 1970s, researchers have demonstrated bioaccumulation of heavy metals in the tissues of tuna (e.g. Menasveta and Siriyong, 1977; Besada et al., 2006; Chen et al., 2011; Chen et al., 2014; Raimundo et al., 2017), raising public awareness of the health threat deriving from the consumption of tuna (e.g. Storelli et al., 2010; Torres et al., 2016). To the best of our knowledge, information on the concentrations of trace elements other than mercury (Hg) in the muscle tissues of the worldwide bigeye tuna populations is currently limited.

Among the various heavy metal pollutants, Hg has been the element of most concern, and there have been extensive studies on the impacts of seafood consumption on human health (e.g. Burger and Gochfeld, 2004; Chen et al., 2011; Chen et al., 2014). However, arsenic (As) and cadmium (Cd) are ranked as the first and seventh, respectively, hazardous substances for human exposure in the 2015 Priority List of Hazardous Substances set by ATSDR (ATSDR, 2015). Furthermore, zinc (Zn), copper (Cu) and selenium (Se) are also on the list, ranking 75, 116, and 145, respectively, and are thus considered less toxic (ATSDR, 2015). However, with the growth of the world population driving rapid industrial development and raw material demands in all aspects of life, there is no avoiding use of these elements. For example, Zn and Cu are used in the production of wire, plumbing pipes, and sheet metal; are combined with other metals to make alloys such as brass and bronze; are used as coatings to prevent rust; and are used in dry cell batteries. Their compounds are also widely used in industry to make paint, rubber, dyes, wood preservatives, and ointments. Furthermore, Cu compounds are commonly used in agriculture to treat plant diseases, for

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water treatment to remove algal bloom, and as preservatives for wood, leather, and fabrics. Selenium (Se) is not only used in the electronics industry, but also as a nutritional supplement for both humans and animals; in the glass industry; as a component of pigments for various materials; and in the preparation of pharmaceuticals, pesticides, and fungicides (ATSDR, 2015). The increasingly unavoidable widespread use of these metals results in them ultimately passing through the inland waterways to the ocean, where they have elevated the metal concentrations (Pan and Wang, 2012; Wang et al., 2013), which may have adverse effects on the marine organisms. Thereafter, a negative impact on the entire marine ecosystem could well occur.

To safeguard the health of the marine environment and to ensure the safety of seafood consumption, it is therefore necessary to understand the levels of heavy metal contamination in marine food fish, especially those of high trophic levels. In this paper, bigeye tuna specimens collected from the Atlantic and Indian Oceans were used to analyze the white muscle tissue concentrations of six trace elements, namely As, Cd, Cu, Fe, Se, and Zn. The main goals were to compare the metal bioaccumulation levels of the tuna in different oceanic regions, to pinpoint their geographical distribution, to elucidate the relationship between the muscle concentration of the trace elements and BET size, and to evaluate the status of global metal pollution of marine environment.

In total, 87 bigeye tuna (BET), *Thunnus obesus*, specimens were collected by scientific observers on board Taiwanese longline fishing vessels operating in the Atlantic and Indian Oceans from 14 August 2005 to 18 July 2007. Specifically, 53 specimens were collected from the Atlantic Ocean (Gulf of Guinea (A1): $n = 25$, two subgroups (A1-1 and A1-2) were defined according to their muscle metal concentrations; off West Africa (A2): $n = 26$; off the Azores islands (A3): n = 2), and 34 specimens were collected from the Indian Ocean (Central Indian Ocean (I1): $n = 12$; off northern Madagascar (I2): $n = 6$; and off South Africa (I3): $n = 16$) (Fig. 1). All of the BET specimens collected were recorded with capture locations and measured fork length (FL, in cm), and their gender was also recorded by the scientific observers. Subsequently, a chunk of the caudal peduncle of each BET was cut off and kept deep-frozen (−40 °C) on board and shipped back to the mother port, Chiangcheng fishing port, Kaohsiung. They were then transported to the laboratory and kept at −20 °C until further analysis.

To obtain the tissues for metal analysis, firstly, the chunk of caudal peduncle was thawed and skinned, then the white muscle tissues without the tendon were collected. These pieces of muscle were then freeze-dried for further chemical analysis.

The metal analyses were performed following the method established in M.-H. Chen's lab. (Chen, 2002). Approximately 0.3 g of homogenized freeze-dried white muscle sample was used for the analysis. At the same time, the standard reference materials, DORM-2 (dogfish muscle, NRCC) from the National Research Council of Canada (NRCC) were used to verify the analytical quality.

The six elements were measured by different analytical methods according to the sample nature, availability of analytical resources, and budget constraints. Fe and Zn were measured by flame atomic absorption spectrometry (AAS) (Hitachi Z-5000, tube type: 7JO-8885). Cu and As were measured by graphite furnace AAS without and with a palladium modifier (Chen, 2002; Liu et al., 2015). Cd and Se concentrations were measured by ICP-MS (Inductively coupled plasma-mass spectrometer, Perkin-Elmer Elan). The recovery of the standard materials of DORM-2 with four replicates (vs. certified value), presenting as μg g^{-1} dry weight, were 18.58 ± 0.38 (vs. 18.0 ± 1.1) for As, 0.052 ± 0.03 (vs. 0.043 ± 0.008) for Cd, 2.64 ± 0.50 (vs. 2.34 ± 0.16) for Cu, 144.0 ± 19.4 (vs. 142.0 ± 10.0) for Fe, 1.17 ± 0.07 (vs. 1.4 ± 0.09) for Se, and 25.2 ± 3.7 (vs. 25.6 ± 2.3) for Zn. We used a conversion factor of 4 to divide the dry weight data for comparison with the literature in wet weight.

For the data analysis by statistics, SAS 9.4 (SAS Institute Inc.) was used to perform the statistical analyses. The Student's *t*-test (Zar, 1999) was used to test whether the Atlantic- and Indian-BET specimens had significant differences in their fork length and muscle elemental concentrations. One-way analysis of variance (ANOVA) was used to examine whether the concentrations of each element in the BET muscle tissue among the sampling regions (A1-1, A1-2, A2, I1, I2, and I3) were significantly different. Additionally, Duncan's multiple range test was used as the post hoc test. Simple linear regression (Zar, 1999) was used to assess the statistical significance of the relationship between the muscle elemental concentration and FL. Moreover, multidimensional scaling (MDS) of the statistical software "Primer 6" (Clarke and Gorley, 2006) was applied to distinguish the spatial representations of the elemental concentrations.

The gender difference has been presented in Chen's (2017) thesis. There were only 10 female vs. 19 male for Atlantic-BET, and 14 female vs. 13 male for Indian-BET available in comparison with the gender difference by ocean. No gender difference was found in the muscle concentrations of the 6 metals in either ocean, except that the Cu concentration of females (2.55 ± 0.59 μg g^{-1} dry weight) was significantly higher than that of males (1.89 ± 0.88 μg g^{-1} dry weight) in the Indian-BET (Student's *t*-test, $p < 0.05$).

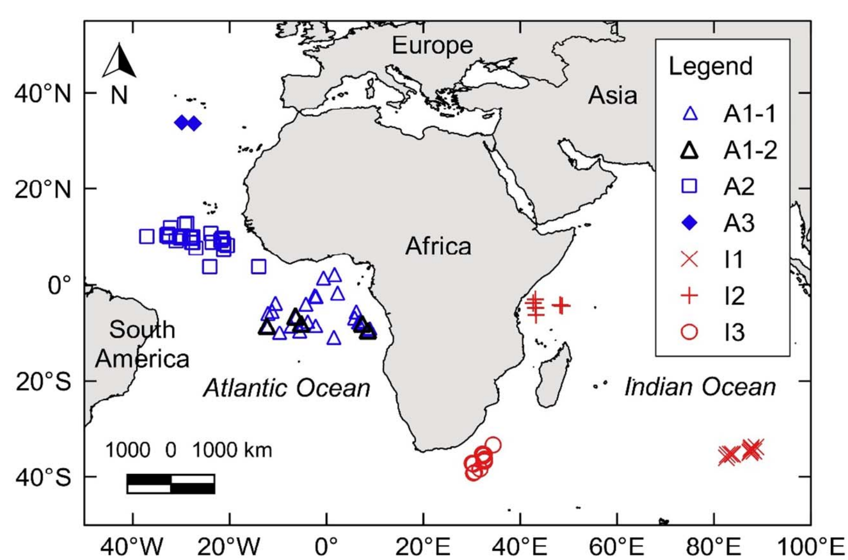

Fig. 1. Map showing the 6 sampling sites, namely Gulf of Guinea (including A1-1 and A1-2); off West Africa (A2); off the Azores islands; Central Indian Ocean (I1); off northern Madagascar (I2); and off South Africa (I3), of bigeye tuna (*Thunnus obesus*) in the Atlantic and Indian Oceans from14 August 2005 to 18 July 2007.