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^{210}Po Activity and Concentrations of Selected Trace Elements (As, Cd, Cu, Hg, Pb, Zn) in the Muscle Tissue of Tunas *Thunnus albacares* and *Katsuwonus pelamis* from the Eastern Pacific Ocean

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Abstract Daily mineral intake (DMI) of Cu and Zn, percentage weekly intake (PWI) of As, Cd, Hg, Pb, and doses of ^{210}Po were estimated by using their elemental concentration in muscle of two tuna species and the average tuna consumption in Mexico. Skipjack tuna *Katsuwonus pelamis* had significantly ($p<0.05$) higher levels of As (1.38 μg g^{-1} dw) and Cu (1.85 μg g^{-1} dw) than yellowfin tuna *Thunnus albacares*, whereas Pb concentrations (0.18 μg g^{-1} dw) were significantly ($p<0.05$) higher in *T. albacares*. The sequence of elemental concentrations in both species was Zn>Cu>As>Hg>Pb>Cd. In *T. albacares*, concentrations of Cd and Pb in muscle tissue were positively correlated ($p<0.05$) with weight of specimens, while Cu was negatively correlated. DMI values were below 10 %. PWI figures (<2 %) are not potentially harmful to human health. ^{210}Po concentration in *T. albacares* and *K. pelamis* accounts for 13.5 to 89.7 % of the median individual annual dose (7.1 μSv) from consumption of marine fish and shellfish for the world population.

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Introduction

Among aquatic resources, fish is a high-quality food for humans that has several advantages. From the perspective of health benefits, minerals, high-quality proteins, and omega-3 fatty acids are among the most attractive features [1]. A relevant difference between fish and other animal products is the content of essential unsaturated fatty acids as docosahexaenoic, eicosapentaenoic, and docosapentaenoic [2]. Such compounds have protective effects against coronary heart disease and thrombosis [3]. On the contrary, radionuclides, metals, metalloids, and other substances can affect human health through fish consumption. In the case of tuna, muscle tissue constitutes an important part from the nutritional point of view and also for representing a stable pool of trace metals and metalloids [4]. Regarding ^{210}Po, liver and caecal mass have been found to have high concentrations in tuna [5]. The yellowfin tuna *Thunnus albacares* is a pelagic active swimmer that travels long distances in a short time [6] and is commonly found in water temperatures ranging from 18 to 31°C. It constitutes an important fishery product that is widely consumed around the world, including Mexico. Diverse authors have concluded that food availability is a key factor related with the abundance and distribution of this species [7]. Another important commercial species of tuna is the skipjack tuna *Katsuwonus pelamis*. This tuna is distributed in waters of tropical and temperate regions in the Pacific, Atlantic, and Indian oceans [8]. Adult specimens of *K. pelamis* are more common in the isotherm of 15°C, while juveniles are confined to warmer waters (>25°C). The vertical distribution of *K. pelamis* ranges from the surface to 260 m depth. In addition to the above characteristics, these pelagic organisms are high-performance fish with high metabolic rates, and consequently, high food intake rates that enhance contaminant exposure [9]. Considering that these fish species are top predators and on the basis of their high mobility within the Eastern Pacific, they were selected for biomonitoring the occurrence of As, metals, and ^{210}Po in this wide geographical region. In this region, studies related to the occurrence of trace metals and radionuclides in tuna are scarce [5, 10–13]. Information concerning the levels of metals, metalloids, and radionuclides in the edible portion of tuna is relevant because of the potential implications on human health. In the present study, the daily mineral intake (DMI) of essential elements (Cu and Zn), percentage weekly intake (PWI) of toxic elements (As, Cd, Hg, and Pb), and doses of ^{210}Po were estimated according to their concentrations in muscle and average tuna consumption in Mexico.

Material and Methods

Fish were caught in diverse sites of the offshore waters of the Pacific Ocean and in front of the Baja California Peninsula (Fig. 1). Fish were supplied by a commercial fleet based in Mazatlán, Sinaloa (Mexico). Six skipjack (*K. pelamis*) and 14 yellowfin (*T. albacares*) specimens were captured during seining operations between June and July 2005. According to weight of specimens and considering the required mass of tissue for analyses, individual samples were used. Plastic containers, glassware, and all laboratory materials used for handling, transportation, and dissection of biota were thoroughly acid washed [14] in order to prevent contamination of samples. After taxonomic identification of specimens [15], total weight was registered. Fish dissection was conducted in the processing plant in order to get muscle from the median dorsal part of both sides of each animal. Samples were freeze-dried (Labconco Freeze-dry System, FreeZone 6) at 80× 10^{-3} mBar and −52°C (72 h), followed by manual grinding in an agate mortar with pestle (Fisher Scientific).

For metals and As, acid digestion (J.T. Baker concentrated nitric acid, trace metal grade) of duplicate subsamples was made by using Teflon vials with caps (Savillex™) at 120°C for 3 h [16]. For ^{210}Po, aliquots of 0.3 g of dried tissue

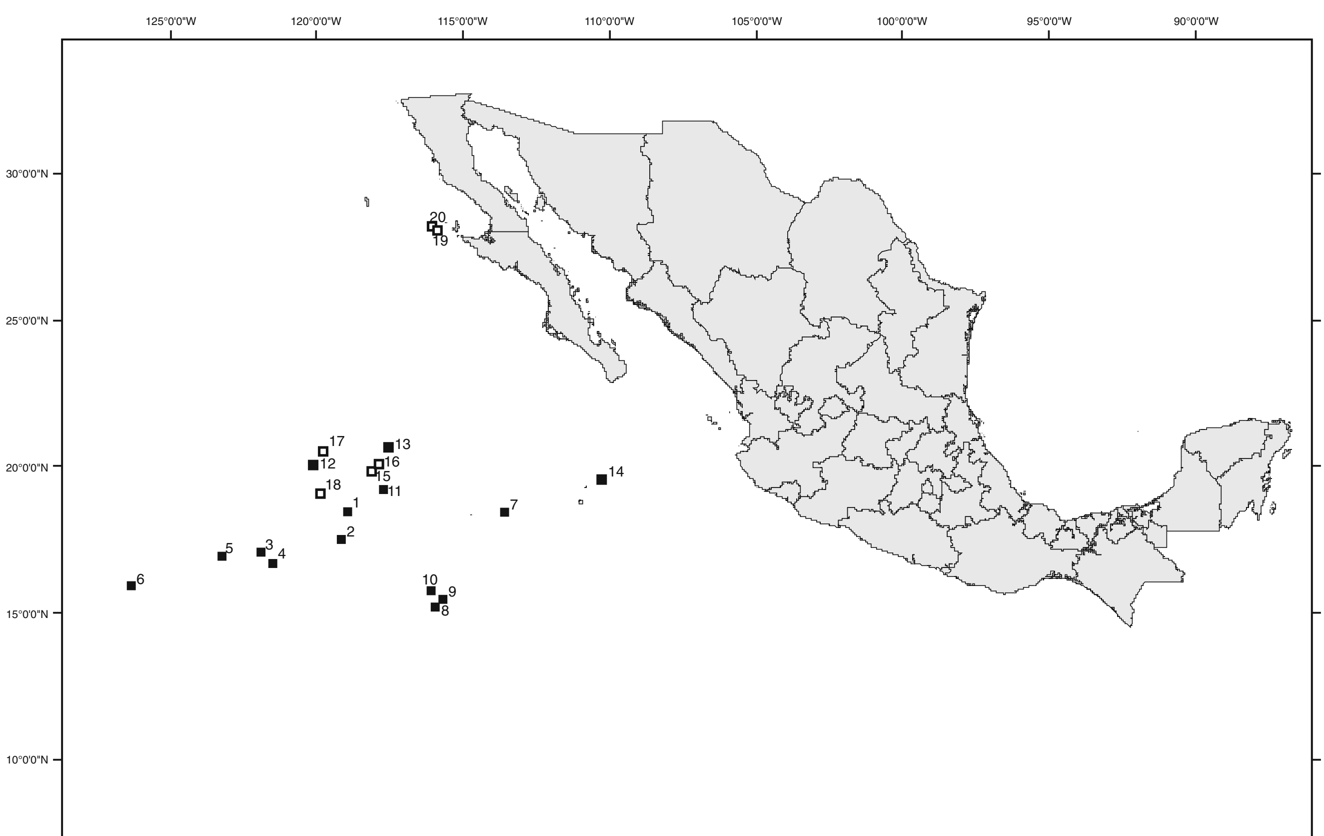

Fig. 1 Location of sampling sites of yellowfin tuna *T. albacares* (*solid squares*) and skipjack tuna *K. pelamis* (*empty squares*) in the Eastern Pacific Ocean

Table 1 Fresh weight (in kilograms) of tuna collected in the Eastern Pacific Ocean

Species	Common name	*n*	Weight mean (min–max)	Dates
T. albacares	Yellowfin tuna	14	15.7 (10.0–30.0)	June and July 2005
K. pelamis	Skipjack tuna	6	4.3 (3.0–5.0)	July 2005

n number of specimens

spiked with ^{209}Po as a yield tracer were digested in closed Teflon vials (Savillex) on a hot plate using 10 ml of concentrated HNO_3 at 150–180°C overnight. The residue was converted into a chloride salt by repeated evaporation with 12 M HCl and redissolution in 0.5 M HCl; 0.2 g of ascorbic acid was added to the solution. Po isotopes were deposited on a silver disk in contact with the acid solution overnight by using an orbital shaker at room temperature. Po activity was measured by alpha spectrometry using ORTEC silicon surface barrier detectors coupled to a PC running under Maestro™ data acquisition software. Blanks were run in parallel to correct for any contamination. The results are expressed in becquerels per kilogram.

Trace elements were analyzed by atomic absorption spectrophotometry (Varian SpectrAA220): Cu and Zn (flame), Cd and Pb (graphite furnace), Hg (cold vapor), and As (hydride generation). Elemental concentrations were expressed as micrograms per gram on a dry weight basis. Conversions of metal concentrations from dry weight (d.w.) to wet weight (w.w.) were made according to the equation: $\text{metal}_{\text{w.w}}=\text{metal}_{\text{d.w}}\times(100-\text{percent humidity})/100$ [17]. The quality of the analytical method was assessed by trace metal determinations of certified reference material consisting of fish muscle (DORM-3) parallel to the samples. Limits of detection (in micrograms per gram) of As, Cd, Cu, Hg, Pb, and Zn were 0.08, 0.002, 0.10, 0.012, 0.014, and 0.48, respectively. Measured concentrations in reference material were within certified intervals for all elements (recoveries ranged from 92 to 103 %). The coefficients of variation (percentage) of As, Cd, Cu, Hg, Pb, and Zn were 8, 12, 6, 7, 11, and 7 %, respectively. Regarding ^{210}Po, the minimum detectable activity was 0.3 Bq kg^{-1}.

In the context of public health, the DMI of Cu and Zn was determined according to [18]: $\text{DMI}=C\times 100/\text{RDA}$, where C is the metal concentration (in milligrams) in 100 g (wet weight) of the edible portion of fish (muscle); RDA (recommended dietary allowances) according to the Institute of Medicine of the National Academies of the United States (Cu, 900 μg day^{-1}; and Zn, 15 and 12 mg day^{-1} for males and females, respectively). In the case of toxic elements (As, Cd, Hg, and Pb), the PWI was calculated according to $26.7\times\text{Cmc}/\text{PTWI}$, where 26.7 results from an average *per capita* tuna consumption in Mexico of 1.44 kg per year [19]; Cmc is the maximum element content (milligrams per kilogram product); PTWI is provisional tolerable weekly intake for a 60-kg person (15 μg kg^{-1} body weight for As; 7 μg kg^{-1} body weight for Cd; 4 μg kg^{-1} body weight for Hg; 25 μg kg^{-1} body weight for Pb) [20]. The dose of ^{210}Po to tuna consumers was calculated by multiplying a dose coefficient of 1.2×10^{-6} Sv Bq^{-1} [21] by the ^{210}Po activity in fish by the consumption rate for tuna (1.44 kg $year^{-1}$) and the delay factor for radioactive decay between collection of specimens and consumption (half life 138.4 days). Significant differences in metal concentrations between species were defined by an unpaired Student's t test or a Mann–Whitney U test (for parametric and nonparametric data, respectively); elemental concentrations in muscle tissue were correlated with weight of specimens. Statistical analyses were made by using GraphPad Prism 4.0 (Graph Pad Software, San Diego, CA).

Results and Discussion

Dates of collection and total fresh weight of *T. albacares* and *K. pelamis* are provided in Table 1. Though no age or length data were available, from weight of specimens, it can be estimated that *T. albacares* (mean weight, 15.7 kg) organisms were represented by a mix of mature and juvenile yellowfin tuna (with a higher proportion of mature specimens) [22], while *K. pelamis* (mean weight, 4.3 kg) specimens were in an adult stage [15]. The sex of the specimens was not determined. Figure 2 shows the concentrations (micrograms per gram dry weight) of As and Cu in muscle

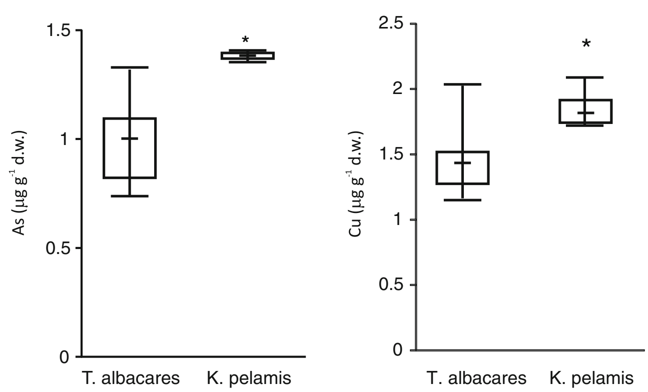

Fig. 2 Concentrations of As and Cu in muscle tissue of *T. albacares* and *K. pelamis* from the Eastern Pacific (*boxes with an asterisk* were significantly higher)

Table 2 Trace element concentrations (micrograms per gram wet weight) and ^{210}Po activity (becquerels per kilogram wet weight) in muscle of *T. albacares* and *K. pelamis*

Species	Weight (kg)	Length (cm)	Site	As	Cd	Cu	Hg	Pb	Zn	^{210}Po	Reference
T. albacares		96-145	Atlantic ocean	0.601	0.004	0.297	0.327	<0.002–0.018	4.82	–	[23]
Tuna[a]			Portugal coasts							5	[30]
T. albacares	21	107	Mozambique Channel	–	0.06	0.24	0.13	0.02	15.8	–	[9]
T. albacares	25	88	Reunion Island	–	0.06	0.52	0.30	0.01	42.1	–	[9]
T. albacares	13.6	86	Eastern Pacific	–	–	0.61	–	–	7.36	–	[13]
T. albacares	–	–	Gulf of Guinea, Ghana	–	–	–	0.06	–	–	–	[32]
T. albacares	–	–	Gulf of Mexico	–	–	–	0.18	–	–	–	[33]
T. albacares	15.7		Eastern Pacific	0.3158	0.006	0.462	0.076	0.057	5.39	0.45	This study
K. pelamis	9	68	Reunion Island	–	0.18	0.29	0.19	0.02	35.98	–	[9]
K. pelamis	–	–	Fish market, Japan	–	0.0175	0.61	–	0.19	5.25	–	[24]
K. pelamis	2.7	53	Eastern Pacific	–	–	1.073	–	–	6.757	–	[13]
K. pelamis	4.3		Eastern Pacific	0.585	0.055	0.78	0.104	0.03	11.18	1.76	This study

[a] *T. albacares*, *T. obesus*, *T. alalunga*, and *K. pelamis*

tissue of both tuna species; it is noted that *K. pelamis* had significantly ($p<0.05$) higher levels of As and Cu than *T. albacares.* Though there was a high variability of results, the sequence of elemental concentrations was Zn>Cu>As>Hg>Pb>Cd. As in other studies [9, 23], essential elements (Cu and Zn) were more concentrated than toxic metals. The relevance of metals and radionuclides in the marine environment lies in the relation of their concentration, distribution, and chemical forms to the health of humans and to the protection of the marine environment [24]. In the case of ^{210}Po, concentrations in marine biota are higher than those in terrestrial species [12]. Regarding metals, they are not biodegradable, and therefore, accumulation of some elements is enhanced in higher levels of trophic chains [23].

Correlations of metal concentrations in muscle tissue with weight of specimens were significant ($p<0.05$) in the case of *T. albacares*: Cd ($r=0.94$) and Pb ($r=0.96$) were positively correlated, whereas Cu ($r=0.98$) was negatively correlated. It has been widely recognized that tunas are fish with high metabolic activity. In the case of juvenile stages (as is the case of *T. albacares* in this study), such metabolic activity is even more accelerated [25]. Lower Cu concentrations in bigger specimens might result from the dilution effect that fish growth exerts on elemental levels [9]. Regarding toxic elements like Cd and Pb, their levels increased along with the weight of individuals of *T. albacares*; this issue is not completely clear nowadays; some studies have revealed that the balance between metal accumulation and excretion results in a net accumulation of the metal [9], but other results have suggested that Cd and Pb are not related to fish size [23] or are negatively correlated [26]. Since diet constitutes the main pathway of metal entrance to aquatic organisms [27], it is probable that food preferences and availability are associated to correlations of metal concentrations in muscle tissue with weight of tuna. In this context, it has been found that, in several cetacean species, the feeding behavior influences the type of correlations between metal levels and specimen size since younger organisms eat more cephalopods and crustaceans than older ones [28]. In the case of ^{210}Po, the concentrations in marine species have been related with the trophic levels, with lower concentrations in organisms occupying top positions (e.g., tuna) perhaps because of the combined feeding habits of tuna that feeds on small fish and zooplankton [12].

In order to have a general view of ^{210}Po activity and elemental concentrations in the current study, results were compared with published studies elsewhere (Table 2); available information was more abundant in the yellowfin tuna *T. albacares* than in the skipjack tuna *K. pelamis*. It is worth mentioning that more data are available in other tuna species, but when inspecting metal levels in organisms, it is important to make intraspecific comparisons [29], i.e., among organisms of the same species. Additionally, since location and fish size account for differences of elemental concentrations, it is important to consider such information in order to make a more objective comparison. In relation to

Table 3 Daily mineral intake (percentage) of Cu and Zn provided by 100 g wet weight of the edible portion of *T. albacares* and *K. pelamis* from the Eastern Pacific Ocean

Species	*n*	Cu mean (min–max)	Zn (males) mean (min–max)	Zn (females) mean (min–max)
T. albacares	14	5.1 (3.4–8.1)	3.6 (1.6–9.3)	4.5 (2.0–11.7)
K. pelamis	6	8.7 (6.6–11.2)	7.5 (3.8–10.8)	9.4 (4.8–13.5)

n number of specimens

Table 4 Percentage weekly intake (PWI) of As, Cd, Hg, and Pb and ^{210}Po doses (microsieverts per year) through consumption of *T. albacares* and *K. pelamis* from the Eastern Pacific

Species	*n*	As mean (min–max)	Cd mean (min–max)	Hg mean (min–max)	Pb mean (min–max)	^{210}Po mean (min–max)
T. albacares	14	0.925	0.09	0.855	0.105	
		(0.66–1.23)	(0.01–0.86)	(0.30–1.16)	(0.01–0.4)	(0.96–2.16)
K. pelamis	6	1.523	0.031	1.138	0.055	
		(1.43–1.59)	(0.02–0.04)	(0.85–1.57)	(0.01–0.09)	(2.82–6.37)

n number of specimens

T. albacares, As and Hg concentrations were more elevated in specimens from the Atlantic Ocean, Cd and Zn in organisms from Reunion Island and Mozambique Channel, Cu in fish from the Eastern Pacific, and Pb in specimens from the current study. In the compared studies with *T. albacares*, trace element concentrations were within the same order of magnitude in the case of As and Cu, whereas Cd, Hg, Pb, and Zn levels varied by one order of magnitude. Concerning the skipjack tuna *K. pelamis*, Cd, Hg, and Zn were more concentrated in fish from Reunion Island; Cu in tuna from the Eastern Pacific; and Pb in organisms from Japan. In general, essential metals (Zn and Cu) were detected in more elevated concentrations than the rest of the elements. Elemental variations in compared studies with *K. pelamis* were within the same order of magnitude in the case of Hg; with respect to Cd, Cu, Pb, and Zn, values varied by one order of magnitude. With respect to ^{210}Po, levels in muscle of analyzed tuna species in the current study (0.45 and 1.76 Bq kg^{-1} wet weight for *T. albacares* and *K. pelamis*, respectively) were lower than reported activity (5 Bq kg^{-1} wet weight) in tuna (composite sample of *T. albacares*, *Thunnus obesus*, *Thunnus alalunga*, and *K. pelamis*) from the coasts of Portugal [30].

Essential elements such as Cu, Fe, Mn, and Zn are required by humans for diverse metabolic activities in the order of milligrams per day [18]. In this context, the percentage of DMI of Cu and Zn provided by muscle tissue of analyzed specimens was estimated (Table 3). Mean percentages of DMI of Cu and Zn were slightly higher in *K. pelamis* than in *T. albacares*. In all cases, DMI values were below 10 %. In comparison with a study with *T. albacares* and *K. pelamis* from the Eastern Pacific collected in 2008 [13], DMI values of Cu and Zn were comparable. In the case of toxic elements, PWI values of As, Cd, Hg, Pb and doses of ^{210}Po by consumption of muscle tissue in *T. albacares* and *K. pelamis* are presented in Table 4. Mean percentages of As and Hg were higher in *K. pelamis*, while Cd and Pb were more elevated in *T. albacares*; nevertheless, none of the PWI figures (<2 %) represent a potential harm to human health. However, it is necessary to generate more accurate information regarding patterns of consumption in selected strata of the Mexican population as well as the contribution of toxic elements from other food items.

In this study, the estimated doses of ^{210}Po from consumption of *T. albacares* varied from 0.96 to 2.16 μSv year^{-1} and from 2.82 to 6.37 μSv year^{-1} for *K. pelamis*. Though no data of ^{210}Po in other seafood consumed in Mexico are available, considering results in analyzed tuna, doses account for 13.5 to 89.7 % of the median individual annual dose (7.1 μSv) for the world population [31] due to the consumption of marine fish and shellfish. The annual dose per capita by tuna consumption varied according to the tuna species (*K. pelamis*>*T. albacares*).

Conclusions

It can be concluded that elemental concentrations in muscle tissue of both species followed the sequence Zn>Cu>As>Hg>Pb>Cd. The skipjack tuna *K. pelamis* had significantly ($p<0.05$) higher levels of As and Cu than the yellowfin tuna *T. albacares*; regarding Pb, concentrations were significantly ($p<0.05$) more elevated in *T. albacares*. With respect to the variations of elemental concentrations in muscle tissue of tuna with weight of organisms, correlations were significant ($p<0.05$) in *T. albacares* (positive for Cd and Pb; negative for Cu). A comparison of measured elements with studies elsewhere showed that As and Cu in *T. albacares* and Hg in *K. pelamis* were in the same order of magnitude; the rest of the elements varied by an order of magnitude. DMI of Cu and Zn were below 10 %, and PWI of toxic elements were below 2 %. Therefore, at the consumption rates of tuna in Mexican population, the potential health risk is low. The annual dose of ^{210}Po per capita varied according to the tuna species (*K. pelamis*>*T. albacares*). A typical tuna consumer in Mexico receives a dose that accounts for 13.5 to 89.7 % of the median dose (7.1 μSv) from consuming marine fish and shellfish for the world population.

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