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Determination and assessment of total mercury levels in local, frozen and canned fish in Lebanon

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Abstract

Fish is an important constituent of the Lebanese diet. However, very little attention in our area is given to bring awareness regarding the effect of the toxicity of mercury (Hg) mainly through fish consumption. This study aimed to report analytical data on total mercury levels in several fish species for the first time in thirty years and to also made individuals aware of the presence and danger from exposure to mercury through fish consumption. Fish samples were selected from local Lebanese markets and fisheries and included 94 samples of which were fresh, frozen, processed, and canned fish. All values were reported as microgram of mercury per gram of fish based on wet weight. The level of mercury ranged from 0.0190 to 0.5700 μg/g in fresh samples, 0.0059 to 0.0665 μg/g in frozen samples, and 0.0305 to 0.1190 μg/g in canned samples. The data clearly showed that higher levels of mercury were detected in local fresh fish as opposed to other types thus placing consumers at higher risk from mercury exposure. Moreover, the data revealed that Mallifa (yellowstripe barracuda/*Sphyraena chrysotaenia*), Sargous (white seabream/*Diplodus sargus*), Ghobbos (bogue/*Boops boops*), and shrimp (*Penaeus* sp.) were among the types containing the highest amounts of mercury. On the other hand, processed fish such as fish fillet, fish burger, small shrimp and crab are found to contain lower levels of mercury and are associated with lower exposure risks to mercury. Lebanese population should therefore, be aware to consume limited amounts of fresh local fish to minimize exposure to mercury.

Key words: total mercury; fish; Lebanon

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Introduction

Fish are an important part of a healthy diet because they are considered to be an excellent source of high value protein and essential nutrients. Fish are low in saturated fat and contain polyunsaturated fatty acids such as omega-3 where a well-balanced diet can contribute to reduced heart disease and promotes the proper growth and development in fetuses and children (Burger and Gochfeld, 2004; National Research Council, 2000; Hunter et al.,1988; Kimbrough 1991; Horn, 1992; Anderson and Wiener, 1995). During the last decade, the importance of omega-3 fatty acids in the diet has been recognized. Such acids are believed to play an important role in protecting against heart disease by various ways including preventing blood cells from clotting and adhering to artery walls or decreasing triglycerides and low density lipoproteins (LDL) (Connor, 2000; Sidhu, 2003) in addition to having anti-arrhythmic effects (De Caterina et al., 2003). Fish consumption has been associated with improved pregnancy outcomes, including fewer preterm and low-birth-weight deliveries (Olsen and Secher, 2002) attributed in part, at least, to *n*-3 fatty acids (Allen and Harris, 2001). Fish are also thought to reduce the risk of stroke caused by blood clots (Kris-Etherton et al., 2002), and to play a role in decreasing inflammation while benefiting people with autoimmune diseases (Simopoulos, 2002). In addition, fish are believed to have beneficial effects on brain and retina development in children (Connor, 2000; Sidhu, 2003; Broadhurst et al., 2002). Ironically, fish can accumulate a high level of mercury and for more than thirty years, studies have shown positive correlations between mercury levels in humans and fish consumption (Bjornberg et al., 2003). Mercury in the muscle tissue of fish can, represent an ecological and human health hazard to those ingesting the fish (Boening, 2000; Henny et al., 2002; Mergler et al., 2007) thus the risks of consuming fish can counterbalance the benefits. Accordingly, there is a need to create a balance

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between maximizing the benefits from fish consumption while at the same time, minimizing the risk of mercury intake. Information on the risks from fish consumption is even more important in a country like Lebanon, a place that has been disturbed by many political issues, where health studies and research are not given priority by risk assessors or public health officials.

Generally, mercury occurs in the environment as metallic, inorganic (mercuric salts) or organic mercury. In aquatic environments, inorganic mercury is converted into methylmercury (the most common form of organic mercury) by microorganisms present in sediment. This causes the methylmercury to accumulate in the aquatic food chain, including fish and shellfish (molluscs and crustacea). Methylmercury is the most hazardous form of mercury encountered in food, and fish is the main source of exposure to methylmercury for most individuals (National Research Council, 2000). Methylmercury is readily absorbed (> 95%) from the gut following ingestion and is rapidly distributed via blood to the tissues (National Research Council, 2000; ATSDR, 1999). Although mercury may not produce a great risk in adults due to larger body weight, it can however be very harmful for developing fetuses and young children (National research Council, 2000; Jacobson et al., 1989, 1990; Sparks and Shepherd, 1994; Jacobson and Jacobson, 1996; Schantz, 1996; JECFA, 2003). For this reason, the Joint Food and Agriculture Organization/World Health Organization (FAO/WHO) Expert Group on Food Additives (JECFA, 2003) has suggested two separate upper safe levels of dietary intake (known as the provisional tolerable weekly intake, or PTWI) for the purposes of risk assessment. One level considered to be protective of the general population (3.3 μg/(kg body weight·week)) and another lower level considered being protective of the fetus (1.6 μg/(kg body weight·week)). However, the joint efforts of the FAO/WHO (2006) have made some additional considerations relating to the provisional tolerable weekly intake (PTWI) for methylmercury and have set that value at 1.6 μg/(kg body weight·week).

Methylmercury interferes with the structural design of the developing brain, thus disrupting the microtubule assembly (Graff et al., 1993) and interfering with the temporal sequencing of cell adhesion molecules that guide neuronal migration and connections (Dey et al., 1999). Studies have shown an association between contaminant levels in fish, fish consumption by pregnant women, and deficits in neurobehavioral development in children (Jacobson and Jacobson, 1996; Weihe et al., 1996; Grandjean et al., 1998). Such findings led the Food and Drug Administration (FDA, 2001) to issue a consumption advisory whereby pregnant women and women of childbearing age should avoid consuming shark, swordfish, king mackerel, and tilefish (FDA, 2001).

Despite the wide range of available data on mercury in fish globally (Rahimi et al., 2010; Islam et al., 2010; Voegborlo and Akagi, 2005, Lam and Sia Su, 2009), there is only one report that evaluates the total mercury levels in canned and frozen imported fish into Lebanon (Aftim et al., 1981). The study was carried out 36 years ago and did not include fresh fish caught in the area; a category which is preferred over frozen fish or canned fish by most Lebanese.

The current study, therefore, is designed to examine the total mercury levels in local, frozen and canned fish species that are normally available all years round and are in higher demand by the Lebanese population. The major objective is to provide concerned individuals information about the presence of mercury in the fish consumed in the area with the hope that such findings will help them to create a balance on how to benefit from consuming fish while keeping the risk of acquiring mercury to its minimum.

1 Materials and methods

1.1 Fish samples

Local fresh fish samples (10 species, 58 samples) were purchased from local fisheries extending along the Northern Lebanese shore starting from El Abde, Deir Aamar, Tripoli, El Qalamoun, Anfe, Chekka, and Batroun (Fig. 1). The types of fresh local fish selected for this study are based on the available species that are normally caught along the Lebanese shore and they constitute the majority of the species the population prefers to consume. The names of the species are listed using their Arabic name followed by their English, and scientific name. The samples consisted of Zellek (parrot-fish/*Sparisoma cretense*), Mallifa (yellowstripe barracuda/*Sphyraena chrysotaenia*), Sargous (white seabream/*Diplodus sargus*), Ghobbos (bogue/*Boops boops*), Ajaj (gilt-head seabream/*Sparus aurata*), Bezri (sand smelt/*Atherina boyeri*), Abou Shawki (marbeled spinefoot/*Siganus luridus*), Kraydiss (shrimp/*Penaeus* sp.), Sardine (european pilchard/*Sardina pilchardus*), and Sultan Brahim (yellow goatfish/*Upeneus moluccensis*). The frozen fish (27 samples) whether locally processed or imported consist of fish fingers, fish burgers, fish fillet (Lou'ouz (merou, *Serranidae*), Aarmout (merlin, *Saurida undosquamis*), and Cod (*Gadus morhua*)), shrimp (small, medium, large), crab sticks, breaded crab claw, calamari rings, and mussels. The frozen samples are pur-

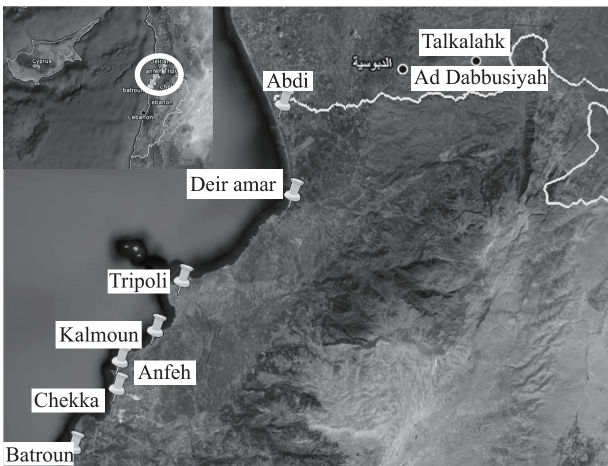

Fig. 1 A map of North Lebanon indicating the fresh fish sampling locations.

chased from well known supermarkets namely Spinneys (1 and 2, Tripoli), Bou Khalil (Tripoli), and Khoury shopping center (Batroun). The canned fish (9 samples) are brand names that can be found all years round and included mostly sardine and tuna. Table 1 shows a summary of all sample types and locations of purchase.

1.2 Sample preparation

For each type of fish collected, an amount of muscle tissue is removed from three or four individual fish, pooled together, homogenized, weighed, and dried in an oven at 80°C until a constant weight was reached. The moisture content of fish tissues is determined according to weight loss before and after drying. For canned fish (tuna or sardine), contents are drained as described in literature (Burger and Gochfeld, 2004), few grams of tissue are taken from separate cans of the same brand followed by pooling, homogenization, weighing, and drying in an oven as previously described. Sample preparation, digestion, and analysis are carried out in separate locations to minimize cross contamination between the three stages. All laboratory equipments and containers are soaked in 10% HNO_3 solution overnight prior to each application and rinsed three times with distilled de-ionized water.

Using the dried samples prepared, 0.5 g from each sample as well as from Certified Reference Material (CRM), DORM-3 (NRC, Canada) are accurately weighed and placed in microwave digestion vessels (Ethos-1 microwave oven, Thermo Scientific, Italy). Ten milliliter concentrated 65% HNO_3 are added to each vessel and left open in a fume extraction hood for at least 30 min to allow gases to escape. The vessels are then sealed and placed into the microwave oven and samples are digested using a three-step program; step 1: room temperature to 200°C, step 2: hold 200°C for 30 min (pressure $\leqslant$ 10 bars), step 3: bring to room temperature. Digested samples are transferred to 100 mL volumetric flasks prior to subsequent treatment. Potassium permanganate solution (6%, m/V) of 60 mL are added to each flask and left for 2 hr to ensure that all the mercury in the sample,

Table 1 Total mercury in fresh, frozen, and canned fish samples*

Type of fish	Location of purchase	Conc. (μg Hg/g)	CV	Type of fish	Location of purchase	Conc. (μg Hg/g)	CV	Type of fish (Brand name)	Location of purchase	Conc. (μg Hg/g)	CV
Fresh fish				**Fresh fish**				**Frozen fish**			
Zellek	El Qalamoun	0.0190	2.4	Bezri	El Qalamoun	0.0385	0.8	Fish finger (vici)	Khoury shopping center	0.0233	0.9
	Chekka	0.0240	4.1		Chekka	0.0459	0.7	Fish finger (alaska pollock)	Khoury shopping center	0.0634	1.8
	Batroun	0.0690	4.0		Batroun	0.0678	2.5	Shrimp (captain fisher)	Khoury shopping center	0.0180	1.7
	Tripoli	0.0778	0.5		Tripoli	0.0819	2.3	Shrimp (captain fisher)	Bou khalil	0.0214	1.6
	Anfeh	0.1463	0.4		Anfeh	0.1662	2.3	Shrimp (aqua blue)	Khoury shopping center	0.0423	2.0
	Deir Aamar	0.1700	0.8		Deir Aamar	0.1983	2.4	Shrimp (siblou)	Bou khalil	0.0531	1.4
	El Abde	0.2242	0.9		El Abde	0.2115	2.1	Fish burger (aqua blue)	Bou khalil	0.0119	0.4
Mallifa	Batroun	0.1601	2.0	Abou Shawki	El Qalamoun	0.0459	2.4	Fish burger (aqua blue)	Khoury shopping center	0.0121	0.2
	Tripoli	0.2099	0.2		Batroun	0.0571	1.9	Fresh burger (americana)	Spinneys 1	0.0221	0.3
	El Qalamoun	0.2254	0.7		Chekka	0.0648	3.2	Fish burger (siblou)	Bou khalil	0.0502	1.4
	Chekka	0.2483	0.5		Tripoli	0.0758	1.9	Fish filet merou, Lou'ouz (frumer)	Spinneys 2	0.0059	2.4
	Anfeh	0.3061	1.0		Anfeh	0.1582	1.6	Fish fillet (americana)	Khoury shopping center	0.0126	0.0
	Deir Aamar	0.3645	1.0		Deir Aamar	0.1610	1.7	Fish fillet (aqua blue)	Bou khalil	0.0151	0.2
	El Abde	0.4445	1.0		El Abde	0.1833	1.6	Fish filet merlen, Aarmout (frumer)	Spinneys 2	0.0177	0.8
Sargous	Chekka	0.1205	2.7	Shrimp	Anfeh	0.3586	0.7	Fish fillet (tanmia)	Bou khalil	0.0196	0.8
	Batroun	0.1262	0.0		Deir Aamar	0.5011	0.8	Fish fillet (tanmia)	Khoury shopping center	0.0202	0.5
	El Qalamoun	0.1263	2.3		El Abde	0.5697	6.5	Fish tilapia HGT (captain fischer)	Spinneys 1	0.0227	2.4
	Tripoli	0.1497	0.4	Sardine	El Qalamoun	0.1543	0.7	Fish filet Cod (frumer)	Spinneys 2	0.0299	0.1
	Anfeh	0.2386	1.7		Chekka	0.1739	0.8	Fish fillet (siblou)	Bou khalil	0.0313	1.5
	Deir Aamar	0.2850	1.5	Fresh shrimp, small	Spinneys 2	0.0198	2.7	Sultan Brahim, yellow (frumer)	Spinneys 2	0.0654	2.2
	El Abde	0.3408	1.6	Fresh shrimp, medium	Spinneys 2	0.0241	2.1	Crab claw breaded (golden meal)	Spinneys 1	0.0255	1.6
Ghobbos	Batroun	0.1042	0.0	Fresh shrimp, large	Spinneys 2	0.0547	2.1	Crab sticks (captain fisher)	Spinneys 1	0.0382	0.7
	Tripoli	0.1298	0.7	Sultan Brahim, fresh	Spinneys 2	0.0786	1.2	Crab sticks (surimi)	Spinneys 1	0.0665	2.8
	El Qalamoun	0.1901	1.7					Calamari rings breaded (siblou)	Spinneys 1	0.0346	4.6
	Chekka	0.2010	1.2					Calamari rings in crispy batter (emborg)	Spinneys 1	0.0397	0.6
	Anfeh	0.3101	0.9					Calamari rings (golden meal)	Spinneys 1	0.0429	0.8
	Deir Aamar	0.3502	0.8					Mussels (siblou)	Spinneys 2	0.0179	1.2
	El Abde	0.3831	0.9					**Canned fish**			
Ajaj	El Qalamoun	0.0611	0.9					Sardine (josiane)	Khoury shopping center	0.0305	1.5
	Chekka	0.0719	5.9					Sardine (deli)	Khoury shopping center	0.0487	1.6
	Batroun	0.1359	0.7					Sardine (maxims)	Khoury shopping center	0.0612	5.2
	Tripoli	0.1474	0.8					Tuna (maxims)	Khoury shopping center	0.0737	0.3
	Anfeh	0.1942	2.1					Tuna (sun bell)	Khoury shopping center	0.0916	1.0
	El Abde	0.2025	2.3					Tuna (white spring)	Khoury shopping center	0.1022	1.5
	Deir Aamar	0.2346	2.6					Tuna (golden plate)	Khoury shopping center	0.1038	0.8
								Tuna (star bell)	Khoury shopping center	0.1189	2.1
								Smoked salmon (spinneys)	Spinneys 1	0.0454	1.2

* Data are grouped according to concentrations where corresponding levels of mercury are listed from the lowest to the highest for each type, concentrations are expressed in μg Hg/g of fish based on wet weight.

including methylmercury, were reduced to the divalent state of mercury. This is followed by the addition of 15 mL of 20% (m/V) solution of hydroxylamine chloride to remove the excess potassium permanganate. Care must be taken during this addition, as this produces an exothermic reaction where the flask may become hot. Upon cooling, distilled de-ionized water is added to bring the volume of each flask up to 100 mL. To obtain matrix-matched standards ranging from 0 to 100 μg Hg/L, a 1000 μg Hg/L stock solution is used. Each standard is subjected to the same procedure as that of the samples and the reference material. The complete and detailed protocol is described in Food Safety Series-Accurate analysis of low levels of mercury in fish by vapor generation AA (www.joytech.com.tw/uploads/ass/11279788445215.pdf, accessed on November 2, 2009). Mercury is analyzed by using Thermo Scientific Graphite Furnace Atomic Absorption Spectrometer equipped with a vapor generation accessory, VP100 (M-series Atomic Absorption Spectrometry, Thermo Scientific, USA), capable of reaching detection limits of 0.07 μg/L in solution.

1.3 Quality control

For quality control, a necessary factor of any laboratory quality assurance program, all specimens are run in batches that included digestion blanks, matrix-matched standard calibration curve, spiked specimens as well as DORM-3 CRM as well as the determination of limit of detection of the method. All samples are analyzed in triplicates. Digestion blanks are used to check the cleaning process of all glassware throughout the procedure as well as for the digestion protocol. Further quality control includes software programming of periodic analysis of sample blanks and standards from the curve to test for any instrumental variations during the analysis.

2 Results and discussion

2.1 Mercury levels

Total mercury concentrations of all samples studied are shown in Table 1. For each type, the data is sorted according to concentration ranging from lowest to highest. Since the study focuses on the amount of mercury the population is exposed to, the data is interpreted by looking at which type of fish contains the highest concentration of mercury versus the area of purchase. First, the recoveries for CRM and spikes ranged between 90%–115% and 88%–107% respectively. The quality control samples (blanks, standards) were within specifications and passed the QC checks set by the software. The coefficient of variation on replicate samples ranged from 0 to 6.5%. This demonstrates the excellent stability of both the spectrometer and the VP100 accessory. The calibration curves show excellent linearity up to 100 μg/L with R^2 values ranging from 0.9976 to 0.9996, thus demonstrating the superb performance of the instrument over a wide concentration range. Furthermore, the limit of detection for the method used is found to be

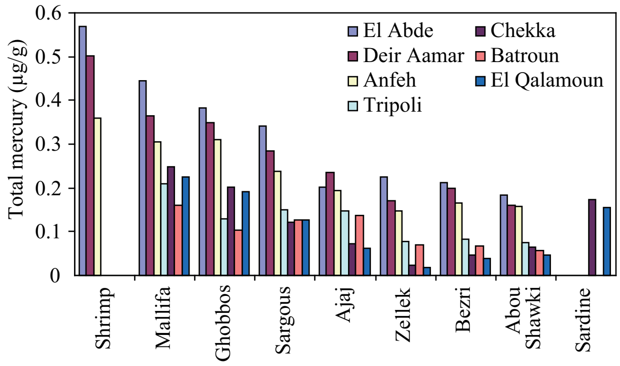

Fig. 2 Total mercury concentrations in fresh fish samples and their locations of purchase.

0.0680 μg/L in solution and is obtained by running 20 measurements of the blank solution followed by multiplying the standard deviation by three. This is equivalent to 0.0140 μg Hg/g in the original fish sample (assuming a sample mass of 0.5 g used for analysis).

2.2 Type of fish versus mercury levels

Based on type versus concentration of mercury (Fig. 2), the data show that shrimp contained the highest concentration of 0.5697 μg Hg/g fish, followed by Mallifa (0.4445 μg Hg/g), Ghobbos (0.3831 μg Hg/g), Sargous (0.3408 μg Hg/g), Ajaj (0.2346 μg Hg/g), Zellek (0.2242 μg Hg/g), Bezri (0.2115 μg Hg/g), Abou Shawki (0.1833 μg Hg/g), and lastly Sardine (0.1739 μg Hg/g). On average, if a 70-kg person consumes only 196 g of shrimp containing 0.5697 μg Hg/g, the PTWI would be already reached. Likewise, if 252 g of Mallifa is consumed, the tolerable level would be reached. In light of this data, Lebanese individuals (based on a 70-kg person) should limit their weekly intake of each type listed above to 196, 252, 292, 329, 477, 500, 530, 611, and 644 g, respectively.

2.3 Location of fresh fish versus mercury levels

Based on location versus concentration comparison (Fig. 2), the data show that the concentration of mercury was consistently higher in the El Abde, followed by Deir Aamar, then Anfeh region. The other four locations do not seem to show the same pattern and concentrations are varying randomly. This suggests that the ambient environment from which the fish caught along the shore of El Abde, Deir Aamar and Anfeh (and sold therein) is more polluted with mercury. The area of El Abde and Deir Aamar contain one of the largest coal power plants that, with no doubt, release significant amounts of mercury. Similarly is the case of Anfeh where cement producing factories highly affect the environment (air, land and sea).

2.4 Frozen and canned fish

Regarding the frozen fish (Table 1), the highest concentration of mercury is found in crab sticks (Surimi) to be 0.0665 μg Hg/g, followed by yellow Sultan Brahim (frumer) 0.0654 μg Hg/g, then fish fingers (Alaska Pollok) with a concentration of 0.0634 μg Hg/g. The data reveals that fresh fish contain around nine times more the concentration of mercury as opposed to the frozen ones when comparing the highest concentrations amongst the two. More interestingly, if we, for example, compare the highest concentration of fresh shrimp (El Abde, 0.5679 μg Hg/g) with that of the highest concentration of frozen shrimp (Siblou, 0.0531 μg Hg/g), the data show that the fresh ones contain 10.7 times mercury more than the frozen ones. This suggests that mercury exposure can be minimized by consuming frozen fish as opposed to fresh ones while at the same time raises the question to whether this has to do with where the fish is caught or is it due to food processing? The data also shows that an average person can consume around ten times more frozen fish as opposed to fresh ones before reaching the set PTWI.

Similarly, the canned type (Table 1) do not show elevated levels of mercury where Tuna (star bell) with a mercury concentration of 0.1189 μg Hg/g. in this case, a 70 kg person would need to consume 942 g of it to reach the PTWI.

In comparison with Aftim (1981), the total mercury levels in canned tuna and frozen fish imported into Lebanon were found to have mean values of 0.3 and 0.16 μg Hg/g respectively, which are 3 and 5 times higher than mean values obtained in our study. However, Aftim's results were obtained in the year 1974–1975, this gives a gap of 36 years in which many factors can change and thus preventing clear conclusions to be drawn. In comparison to recent studies elsewhere (Rahimi et al., 2010; Islam et al., 2010; Voegborlo and Akagi, 2005, Lam and Sia Su, 2009), mean values of total mercury were similar to those obtained in this study.

The results encourage that further studies should be implemented using sediments, water, air and fish samples through a regular monitoring program. Studies have suggested that mercury concentrations in fish have correlated to mercury levels in their ambient environment where sediments seem to be the major source for Hg accumulated in fish (Zhou and Wong, 2000).

In addition, this study reveals that there is a need to investigate the effect of different ways of processing fish (frozen, canned) on lowering the concentration of mercury in a similar way that has been carried by Burger (2003). In other words, the data for frozen and canned fish versus the fresh fish cannot be correlated since there is no information whether the processed fish are caught or imported from elsewhere. There are no studies that compare the levels of mercury in one type of fresh fish versus a frozen sample taken from the same (fresh) fish. This may shed light on the effect, if any, of fish processing on lowering the tissue mercury levels.

3 Conclusions

The study demonstrates that some of the areas along the Lebanese shore contain certain fish types with high mercury accumulation. Moreover, the data suggest that individuals should consume less of the local fish since they contain higher mercury concentrations in comparison to the frozen or canned ones. Based on PTWI values mentioned earlier and based on the data presented, small children and pregnant women should control their weekly intake of the types of fish in the fresh category since they are affected most. Adults, as well, should limit their intake of some of the high mercury containing types of fish. Awareness programs, advisories, care in general for public health and increasing the public basic knowledge regarding mercury contamination are essential to assist the individuals to reach to a balance between the benefits and the risks of fish consumption.

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