QATAR UNIVERSITY

COLLEGE OF ARTS AND SCIENCES

MERCURY AND METHYL MERCURY IN FISH: CONTAMINATION LEVELS AND HEALTH RISKS

BY

MAETHA M. AL-SULAITI

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COMMITTEE PAGE

The members of the Committee approve the Thesis of
Maetha AL-Sulaiti defended on 11/05/2022.

Dr. Lama Soubra
Thesis/Dissertation Supervisor

Prof. Mohammad Al-Ghouti
Thesis Co-Supervisor

Dr. Gouda Ramadan
Committee Member

Prof. Mohamed Najib Daly Yahia
Committee Member

Approved:

Ahmed Elzatahry, Dean, College of Arts and Sciences

ABSTRACT

AL-SULAITI, MAETHA, M., Masters : June : 2022, Environmental Sciences
Title: Mercury and Methyl Mercury in Fish: Contamination Levels and Health Risks
Supervisor of Thesis: Dr. Lama Soubra and Prof. Mohammad Al-Ghouti.

This study aims to assess the risks that mercury and methyl mercury would be posing on the health of fish consumers. The fish consumption patterns of Qatar residents aged 18 years and above were obtained using a fish frequency questionnaire. The Mercury contamination levels of the most consumed fish species were determined using a validated ICP-MS method. Total Mercury average concentration were 0.077 mg/kg ww and ranged between 0.001 mg/kg ww in Safi and 0.443 mg/kg ww in Hamour. PCA analysis was done for the contamination and the exposure. Results demonstrated that contamination levels are primarily affected by protein-lipid content in predatory species. Exposure to Mercury and Methyl mercury was determined via the deterministic approach, using both aggregated and disaggregated fish consumption data and simple distribution. Two scenarios were used to determine methyl mercury level from measured mercury level (MeHg100% and MeHg based on values reported in the literature). Hamour, Chanad, and canned tuna contributed significantly to the mercury exposure. The aggregated method revealed that the high fish consumption was the main source of the risk exposure. The median, 75^{th}, and 95^{th} percentile using the Hazard Quotient index (HQ) compared to the TWI and PTWI for all cohorts. Exposure to mercury from fish using aggregated method poses a risk on the health of Qatari women of the child-bearing age, and for all high fish consumers (P95).

DEDICATION

This thesis is dedicated to my mother for all the love and support. This is especially dedicated to the memory of my grandmother, she always supported women's education even though she did not read or write.

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

ABBREVIATION TABLE

Name	Abbreviation
Body weight	bw
Consumption	CNP
Dry weight	dw
Estimated daily intake	EDI
Estimated weekly intake	EWI
European Commission	EU Commission
European Food Safety Authority	EFSA
Food and Agriculture Organization	FAO
Gaseous elemental mercury	GEM
Hazard Quotient	HQ
Inorganic Mercury	IHg
Inorganic gaseous oxidized mercury	GOM
Mercury	Hg
Methyl Mercury	MeHg
Particulate Hg	p-Hg
Principal Component Analysis	PCA
provisional tolerable weekly intake	PTWI
Total Mercury	TotHg
Tolerable daily intake	TDI
Tolerable weekly intake	TWI
United States Environmental Protection Agency	EPA
Wet weight	ww
World Health Organization	WHO

CHAPTER 1: INTRODUCTION

1.1. Introduction

Metallic elements are naturally occurring in the soil and earth crust. Those elements are recycled by the biogeochemical cycles (Saleh, 2020). Some of those metals are essential for the human body, for example, iron (Fe), manganese (Mn), and zinc (Zn). Other, are non-essential or trace metals, which are considered harmful if they exceeded the safe limits, for example, mercury (Hg) and lead (Pb) (Rahmani, 2018). Fish constitutes the major source for human exposure to Hg and namely MeHg, which creates a serious challenge to balance between the toxicity of these and the healthy protein source (Clarkson, 2020). Fishing in the Arabian Gulf was the major food source before oil discovery in the 1930s since it is not hard to obtain and will not be affected by the arid climate like other meat sources (Cunningham, 2019). Fish is one of the most important protein sources around the world (Rahmani, 2018). In addition, fish is rich in omega 3, docosahexaenoic acid, linolenic acids, unsaturated fatty acids, micro & macronutrients, and different vitamins (Sofoulaki, 2019). Fish consumption shows protecting ability from non-communicable disease (NCD) particularly cardiovascular diseases, rheumatoid arthritis, and will maintain a normal neuronal development in children (Clarkson, 2020). Fish can be bought not only fresh but also, canned, frozen, smoked, dried, and salted (Rahmani, 2018). Fish tissues can contain different toxic heavy metals like Pb, and Hg; which accumulate through the food chain (Cunningham, 2019). Mercury is a major heavy metal contaminant in the aquatic environment and has serious neurotoxins to the human body due to fish consumption (Donadt, 2021; Melnyk, 2021). Mercury concentration in fish is highly influenced by different parameters, such as trophic level which influence the feeding habit, size, age, location (spatial and water depth), species, sex, water temperature, and salinity,

and surrounding industrial and petroleum activities (Elsayed, 2020; Milatou, 2020). Bioaccumulation and biomagnification are important factors that will influence the Hg concentration in fish. Bioaccumulation is the increase through continuous feeding, and biomagnification is the increase through the food chain (Gentès, 2021), see figure 1.

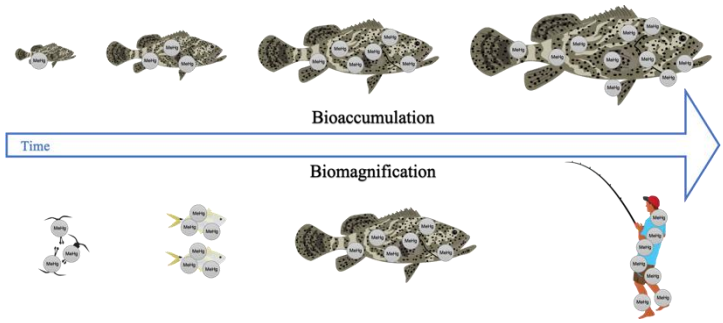


Figure 1. Bioaccumulation and Biomagnification

The high levels of mercury (Hg) in fish was always been a major concern around the world (Anual, 2018). In 2017, mercury is considered one of the main ten chemicals that is of concern for public health (UN, 2019). The United Nations (UN) introduced the Minamata Convention on Mercury in 2013. This convention was established to face the anthropogenic emissions of mercury. The convention takes into consideration the life cycle of mercury and waste management. The convention aims to regulate mercury compound emissions and the use of Hg in any form in the industry. The convention allowed the parties to design implementation plans within the regulation. The parties should report Hg sources, anthropogenic emissions (atmosphere) and releases (soil and water) of mercury, mercury amalgamation from artisanal and small-scale gold mining, and the use of Hg compound in the industry to the conference of the parties. Moreover,

the convention addresses the importance of research, development, and monitoring of mercury emissions and biotic media including fish (O'Connor, 2019; UN, 2019).

A limited number of studies were done to study the Hg level in sediments and marine species in the Arabian Gulf, with its physiologically arid environment, dusty, hot climate, high salinity, and poor water circulation that result in long water residence time (Cunningham, 2019). Moreover, the wars that existed in the region in the last two decades would have had an impact on the Hg levels. Since petroleum and oil industry are the main economic source in the Gulf region, Qatar has the main petroleum and oil plant in the south-east and north-east, in addition, the gulf region desalination and energy source is the seawater and for this reason, the plants are also on the cost (Al-Ansari, 2017). This contributed to increasing the mercury level in the semi-enclosed Gulf region (Cunningham, 2019).

The toxicity of mercury and its most toxic form methyl mercury (MeHg) are very well known (Dietz, 2021; Kimáková, 2018). In addition, MeHg having the molecular formula (CH_3HgX) in fish has the ability to bioaccumulate and transfer through the various trophic level of the food chain (Al-Ansari, 2017).

Humans are exposed to mercury and MeHg through the consumption of contaminated food (Al-Ansari, 2017). To avoid health risk issues as a result of the exposure to Hg from fish products the World Health Organization and the Food and Agriculture Organization (WHO/FAO) developed a research-based intake guideline (FAO/WHO, 2007). Those guidelines are based on the estimated intake from the contaminant per body weight over a lifetime without resulting in any risk in human health (Stelljes, 2008). The guidelines also indicated the acceptable daily or weekly intakes (Alva, 2020). These values are based on current contamination and exposure levels and based on careful toxicological considerations. Moreover, to identify the

quantify the current risk a health risk assessment should be conducted (Stelljes, 2008). Since Hg is considered to be non-carcinogenic, the risk assessment is generally determined by calculating the hazard quotient (Popovic, 2018). In Qatar, there are limited publications on the concentrations of mercury in fish and seafood (Al-Ansari, 2017; Elsayed, 2020). Previous studies focused on various levels of mercury in limited marine fish species at different trophic levels that were collected from selected sites. Moreover, according to our knowledge, no previous risk assessment for mercury and methyl mercury from fish consumption was conducted. Moreover, risk assessment studies are needed in Qatar and the Middle East region not to only assess current risks from exposure to contaminants, but also to draw legislative inferences (Saleh, 2019).

1.2. Aim and Objective

The primary aim of this study is to assess the risks that mercury from fish products would be posing on the health of the Qatari residents. Exposure assessment using assessment methods was conducted/ to assess the exposure to total mercury from fish consumption. The study aims to determine the concentration of total Mercury from the most common fish species from the Qatari market using a fish frequency questionnaire to collect fish consumption patterns.

The principal component analysis was used to investigate the trends of Mercury levels. Finally, the effect of pollution on Mercury levels in fish was analyzed.

CHAPTER 2: LITERATURE REVIEW

2.1. Heavy Metals

Heavy metals are elements that exist everywhere in the environment, persistent, stable, and non-biodegradable (Rahmani, 2018). Heavy metals can be classified into essential and non-essential. The essential heavy metals are important for the human body in small quantities, otherwise, they become toxic for the biochemical functions. On the other hand, non-essential heavy metals like Mercury (Hg) are not needed for the human body and cause adverse effects on health (Xu, 2021).

2.2. Mercury

Mercury (Hg) is a major trace element and non-radioactive pollutant in the environment. Similar to most heavy metals Hg is highly persistent which results in high toxicity levels (Álvarez-Fernández, 2020). Like any element on earth, Hg will be transported through the earth's spheres: atmosphere (air), geosphere (soil), hydrosphere (water), and biosphere (biotic) (Clarkson, 2020). However, to understand the Hg cycle we need to review the source-receptor relationships taking into consideration the change in the climate and emissions (Zhang, 2021). Hg has a different oxidation state and will bind to different elements and organic groups forming different species with different characteristics (Abass, 2018).

2.2.1. Mercury Categorization

Mercury can be categorized in three different ways; the first way is by its oxidation state: Hg is categorized through three oxidation states into elemental or metallic Hg^0, mercurous Hg(I)/ Hg^{1+}, and mercuric Hg(II)/ Hg^{2+}. The first oxidation state is observed only in the atmosphere due to its high volatility, and in highly polluted soil. The second oxidation state is unstable; this will make Hg difficult to be observed in any environmental compartment. The third oxidation state is produced mainly after

the dry/wet deposition from the atmosphere to form one of two types salts and minerals by ionic bond or organo-Hg by a covalent bond (Elsayed, 2020; O'Connor, 2019).

The second way to categorize mercury is based on its chemical form: elemental (natural gas), organic and inorganic forms. Methyl mercury (MeHg) is the most common organic form and the most toxic one (Kimáková, 2018). The term methylated Hg is consisting of MeHg and dimethylmercury (DMHg), in some cases, MeHg is used to represent the methylated Hg in general not only monomethyl mercury (Wang, 2020). The final way to categorize mercury is based on its physical characteristics: volatile (elemental and dialkyl mercury), insoluble (mercury sulfide HgS and mercury selenide HgSe), and soluble (monomethyl mercury halides, dialkyl mercury, and ionic conditions) (Saleh, 2020).

2.2.2. Methylation and Demethylation

The methylation processes are still considered to be ambiguous processes. The main locations are sediments, water, and some microorganisms on the macrophyte roots (Achá, 2011). The methylation and demethylation in incubation studies were found to follow first-order reactions, the rate of reaction will only depend on the concentration of IHg or MeHg and all the other surrounding conditions will not have a significant influence (Wang, 2020). MeHg is a form of Hg with a persistent ability, low elimination rate, and high absorption levels in the bloodstream (up to 95%) (Anual, 2018; Elsayed, 2020). This form of Hg makes up 95% of Hg in marine species (Al-Ansari, 2017). There are two main pathways for methylation and demethylation, the photo and bacterial (Luo, 2020). Bacterial methylation or bio-methylation of Hg to produce MeHg from the methylation/biomethylation process of inorganic Hg by soil and water heterogeneous anaerobic bacteria (Du, 2019; Elsayed, 2020; Luo, 2020; Thomas, 2020). The bacterial groups associated with methylation are iron-reducing

bacteria (FeRB or IRB), methanogens and fermentation bacteria, mainly sulfur-reducing bacteria (SRB), and sometimes methanogens, and acetogenic microorganisms (Luo, 2020; Wang, 2021). SRB methylation's ability to convert organic matter (OM) and Hg to MeHg are highly related to the sulfur biogeochemical cycle. Sulfur-rich sediments, sulfate (SO_4) and sulfides concentration, and other forms of sulfur, will create a strong ligand bond with Hg. The sulfur cycle will activate the methylation processes; however, the level of methylation is determined by the sulfur species. In general, it was found that the increase in SO_4 concentration will enhance the methylation (J. Wang, 2021). It was found that the methylation process accelerates in acidic pH levels, this was explained by the fact that Hg uptake into the fish tissues by the bacteria is more efficient at lower pH levels (Kelly, 2003; Thomas, 2020). Moreover, the negativity of dissolved organic carbon (DOC) in low pH levels will be weaker, this will limit the creation of a complex bond with Hg which will allow more methylation processes to take place (Kelly, 2003). But the main two factors are the availability of IHg and the microbial community (Hassan, 2019). Deacidification in water was found not only to decrease the Hg level but also the sulfate (SO_4^{2-}) concentration which will influence the methylation and will reduce the assimilation of Hg into fish cells, this can increase the Hg^0 in water (Kelly, 2003). In (Ziarati, 2017) study on Carcharhinus dussumie fish, the total Hg was 0.79 µg/g and MeHg was 0.78 µg/g, this means that MeHg makes around 99.76% of the total Hg. Photo-methylation occurs by solar irradiation on the surface of the water. However, the photo-methylation rate is slow and the intensity of light will not influence the concentration of MeHg in the surface water (Luo, 2020). Demethylation can occur by microorganisms, photodecomposition, or abiotic processes (Leopold, 2010; K. Wang, 2020). In some cases, demethylation can be processed by the same group of bacteria

that do methylation, the SRB (Achá, 2011). Photo-demethylation is the process of releasing the Hg from the methyl group by UV radiation and elemental Hg will emit back through the system. The greenhouse (GHG) effect will prevent and slow the demethylation processes (Jordan, 2019). Photo-demethylation in the marine system is highly influential and has a faster rate than photo-methylation. In 1972 and 1975 the first experimental study for Hg photochemical methylation was conducted. In 1972 they tested mercury chloride solution (HgCl) with methanol (CH_3OH), acetic acid (CH_3COOH), propionic acid ($CH_3CH_2CO_2H$), ethanol (C_2H_5OH) under 253.7 nm wavelength for 20 h. In 1975 they tested mercury acetate ($C_4H_6O_4Hg$) solution with solid sulfur, mercury oxide (HgO), and mercury sulfide (HgS). The results showed that the photosensitizers are HgO, mercury sulfate, and ($HgSO_4$). Additionally, when thiol (R-SH) group and CH_3COOH are available, thiol will work as photosensitizers and the reaction will be an intramolecular decarboxylation reaction (Luo, 2020). On the other hand, mercuric acetate solution in an acidic medium in the dark produced MeHg and when the solution was exposed to light the reaction slowed and photolysis of MeHg occur. Additionally, the radicle CH_3 group formed from acetate photolysis reacted with the dissolved oxygen (DO), not in methylation (Gårdfeldt, 2003).

2.3. Sources and Pollution of Mercury on Earth

Mercury exists on earth from natural and anthropogenic sources. Natural sources are soil and earth crust (earth crust off-gassing process into water), and they are highly affected by the geological location, the earthquakes activities, volcanic eruption, and can be as a result of natural seeps. Those factors can influence the concentration of Hg in raw petroleum products (Elsayed, 2020; Kimáková, 2018; Mao, 2020; Saleh, 2020). In soil part of the Hg is from natural sources like volcanoes, geothermal springs, and forest fire, however, anthropogenic source contributes a total of $86x10^6$ kg

(O'Connor, 2019). In natural gas, Hg is considered to be trace metal except if the soil's geographic location is rich in Hg (Hassan, 2019). Anthropogenic sources are highly related to industrial pollution. Mining and smelting, artisanal small-scale gold mining (ASM), coal burning, oil refining, Chloralkali process (mainly inorganic Hg), and cement production (O'Connor, 2019).

Heavy metals contamination in fish is a result of consuming toxic wastes which discharged into the water (Ziarati, 2017). According to (WHO, 2016), Hg from small-scale gold mining can bioaccumulate in predatory fish that feed on non-predatory fish. A Chlor-alkali plant on the Iranian coast results in increasing the Hg level in flounder fish (O'Connor, 2019). In Qatar, fish from the industrial city of Ras Laffan had a higher level of total Hg compared to Al-Khor, this can be as a result of high Hg salts (mercuric/ Hg(II)) loading (Al-Ansari, 2017). In the food industry, the canning process of tuna fish can introduce not only Hg but different heavy metals to the fish tissues (Rahmani, 2018). In the Arabian Gulf, the main source for Hg is related to oil fields like oil terminals, oil spills and natural seeps, and sewage discharge. The shore of Qatar has four coastal petrochemical plants (Elsayed, 2020). Hg accumulation in the soil is estimated to be around 25×10^{7}–1×10^{9} kg globally. In soil part of the Hg is from natural sources like volcanoes, geothermal springs, and forest fire, however, anthropogenic source contributes a total of $86x10^{6}$ kg (O'Connor, 2019). During petroleum refining processing and wastewater of discharge, Hg can leak out into soil and water, to result in soil and water contamination, however, this can be controlled and limited by using removal techniques (Saleh, 2020). Global warming will have a definite direct and indirect impact on the water chemistry (temperature & pH) which will influence the fish size, productivity, Hg level, and methylation rates in the fish (Thomas, 2020).

Carnivorous species fish can reflect the increase in Hg through the food chain (Al-majed, 2000). In general, the Hg level in the Arabian Gulf fish is considered to be below the maximum allowable levels (MALs) with only 10% above it. Most of the fish that were within the 10% are from two Chlor-alkali plants in Iran and Kuwait, both of those plants use to be discharging Hg rich waste in the water for a long period of time, the magnitude is estimated to be around 31 t in Iran and 21 t in Kuwait (Cunningham, 2019).

In Japan, in Minamata city, a chemical factory's waste discharged into the seawater for years resulted in Hg bioaccumulation and contaminated seawater. This has led to food poisoning of the fishermen and their families. Therefore, this was considered the first case of food poisoning caused by Hg bioaccumulation between the 1950s and 1960s (Clarkson, 2020).

2.3.1. Wastewater Discharged

The level of Hg in the wastewater (WW) discharged into the water bodies is a main global concern. According to the National Association of Clean Water Agencies around 36% of the Hg in WW is from dental clinics because of the use of Hg in the dental amalgam. In 2006 the EPA created a roadmap to characterize Hg sources, collection, and provided mitigation methods (Gbondo-Tugbawa, 2010). WW and contaminated liquids discharged into water bodies can have an adverse effect on animals and humans. The discharge of 100 Mg of Hg into rivers and fields in China from an organic chemical factory between 1971 -1997 resulted in underweight livestock, reduced the annual yield of the grain production by 30- 40%, and increased the cancer cases. And the pollution from explosives in chemical companies manufactured that make pesticides in Switzerland in 1980 reached France, Germany, and the Netherlands through the Rhine river (Zhao, 2021). Even though

modern wastewater treatment plants WWTP can remove up to 90% of totHg and 70% of MeHg from influent, the municipal wastewater will still have a MeHg concentration that is higher than the normal MeHg level in water bodies. This can be because of the availability of OM and ionic Hg in the effluent which will facilitate the methylation process. In the oxidation ditch process in China, the MeHg was uptake by the microorganisms but the totalHg will end up in the sludge. Moreover, the process and steps used for the municipal WWTP will have different Hg removal present. In General, the fate of Hg in WWTP still needs further studying (Gao, 2020). (Gao, 2020) stated that the primary sedimentation will not significantly reduce the Hg concentration and that the main factor for high totHg removal including MeHg is pH and temperature. The most famous Hg pollution case is in Minamata Bay in Japan when Chisso company discharged waste (CCDW) between 1932- 1961. Even though the totHg concentration reduced in time the bay still needed restoration, the restoration project started 30 years after 1961 by the Kumamoto prefecture from 1991- 2008, however, 2– 4% of the totHg from the CCDW is still present in the sediment (Akito, 2014; Matsuyama, 2019). Sediment samples from the bay were taken in 2012 resulting in a weighted average of 2.28 mg/kg (dry) from different sediment depths (0- 32.5 cm), the highest average was for sediment surface 2.96 mg/kg (Akito, 2014). Sediment samples from different sediment depths (0- 23 cm) in Yatsushiro sea in 2017 had a weighted average of 0.46 mg/kg (dry), and the highest average was for sediment surface 2.77 mg/kg, this study showed that the totHg from the CCDW was distributed out of the bay and still available in sediment (Matsuyama, 2019). The fact that the sediment still contains some Hg from the CCDW shows the resilience of Hg and the long-term danger.

Eutrophication

Eutrophication can alter the geochemistry of water to increases the autotroph species resulting in an increase in the MeHg concentration by increasing the methylation process, although Hg biomagnification will reduce (Razavi, 2015). Eutrophication occurs with algae bloom when the main growth factors the need high nutrient level is increased and with sunlight. Eutrophication will increase the sediment OM and hypoxic/ anoxia conditions created by the algae and lower pH levels (Ji, 2020; Jordan, 2019; Razavi, 2015). Phytoplankton production will increase, zooplankton will feed on the phytoplankton, however, zooplankton will not be able to limit the excess phytoplankton production (Jordan, 2019). Moreover, Phytoplankton produces OM which will be consumed by heterotrophic bacteria like microbial methylators (Ji, 2020). In addition, the algae will bioaccumulation MeHg, and after they die the MeHg will be released back into the water or biomagnification through the food chain. In (Ji, 2020), injecting the eutrophic waters with nanobubbles O^2 reduced MeHg concentration by 76% in water and 56% in sediment. However, MeHg absorption by algae can be used under control as the MeHg mitigation method (Quiroga-Flores, 2021). However, the dynamics between eutrophication and Hg still need further research (Hung, 2020), see figure 2.

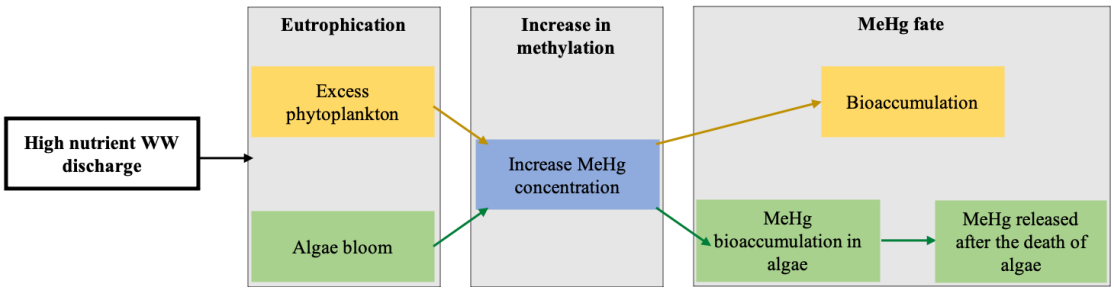


Figure 2. Two scenarios for the effect of eutrophication

Antibiotic discharged

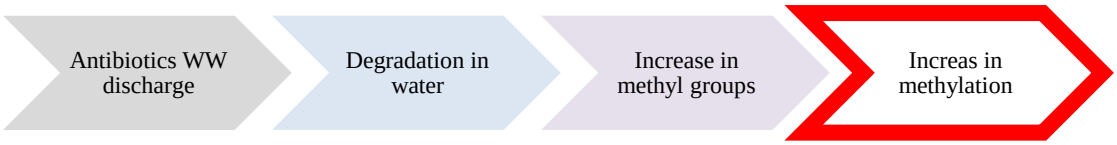


Figure 3. Antibiotics and MeHg formation (Liang, 2018)

Antibiotic is one of the top consumed pharmaceutical medicine globally, for example, two of the main used antibiotics are tetracyclines (TC) and oxytetracyclines (OC) (Liang, 2021). Antibiotics will decompose in sediment and methyl groups will be free in the water, this will make it easier for the methylation process to happen. The totHg will decrease in sediment polluted with antibiotics; this can be explained by the formation of the complex compound, however, with time the MeHg concentration will increase. This shows that bio-methylation might not be the main path for methylation. However, the effect of antibiotic WW on the methylation process is still lightly studied (Liang, 2018). See equation in figure 4 for the MeHg formation from antibiotics.

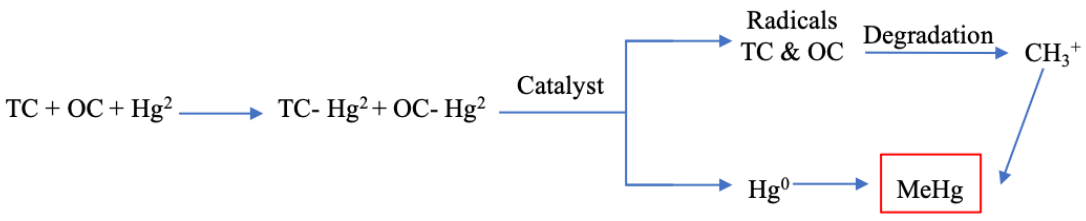


Figure 4. The assumed Antibiotics degradation to form MeHg by (Liang, 2018)

2.3.2. Air Pollution

To understand the transportation of air pollutants we need to establish a geospatial distribution for the pollutant from the source to the receptor in most cases, they are a point source. Geospatial distribution requires the exact location of the source using geographic coordinates and accurate emission values to each source or using the national emission estimates (Steenhuisen, 2015). The main pollutant sources are mainly related to industry and power generation, around 74% of atmospheric Hg comes from ASM, stationary combustion of coal, and non-ferrous metals production (AMAP/UN Environment, 2019; Steenhuisen, 2015). The ASM sources are difficult to create a geospatial distribution estimation for because of their large number and in many cases, they are transient over time. We also must take into consideration how close a source to the region is needed (Steenhuisen, 2015). The Middle East region (the Asian part) is accountable for 2.4% (40700– 93800 kg) of the global Hg emissions, the industrial sector contribute by more than 55% (AMAP/UN Environment, 2019), see figure 5. Qatar Hg emission from industries that use chloralkali process like cement production, medical and municipal waste, and the lost emitter is natural gas production ranging between 10^3 to $200x10^3$ ng/m^3 (Hassan, 2019).

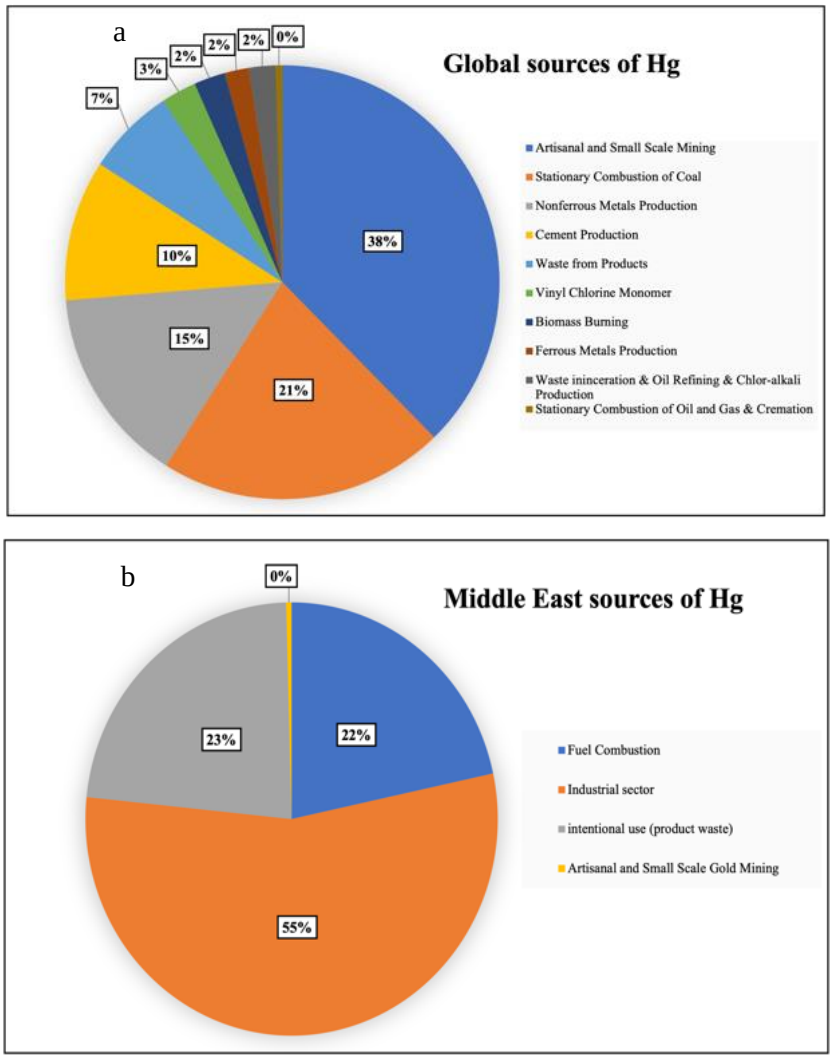


Figure 5. a) Global Sources of Hg & b) Middle East sources of Hg (AMAP/UN Environment, 2019)

2.4. Fate and Transport of Hg in the Environment

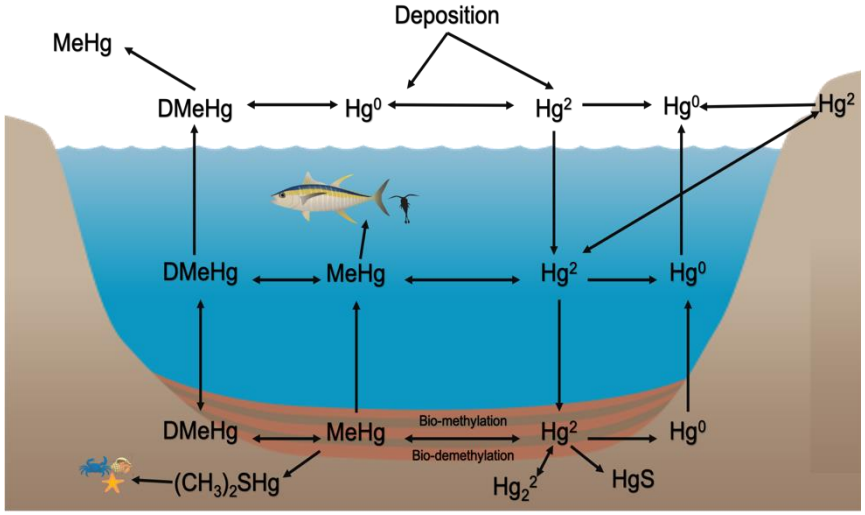


Figure 6. Hg biogeochemical cycle edited from (Leopold, 2010; Robles, 2014)

2.4.1 In the Atmosphere

In air elemental Hg^0 makes up to 95% of the mercury species and has a lifetime range from months up to one year, this will give it time to travel and to deposit from (dry/wet) far from the emission source (Zhang, 2021). There are three types of atmospheric elemental Hg which are named the total atmospheric mercury (TAM) (Yuan, 2021). The first one is the gaseous element mercury (GEM), which has the lowest deposition rate due to its low water solubility (Timonen, 2013). The second one is gaseous oxidized mercury (GOM) or reactive gaseous mercury (RGM), and the third type is particulate mercury (HGP) or particulate bound mercury (PBM) (Yuan, 2021). The PBM is an airborne particulate from the absorption of GEM or RGM (Sun, 2021). The RGM and PBM make only 10% of the TAM, however, they have a shorter life that ranges between hours to days or weeks and has a higher dry and wet deposition rate (Sun, 2021). The highly volatile Hg species like GEM and dimethylmercury are able to

go through long-range transport (Morosini, 2021). The mercuric mercury Hg^{2+} has a short lifetime in the atmosphere which will results in wet deposition into the water bodies and then will go through the methylation process to form MeHg (Clarkson, 2020). The highest Hg^{2+} wet deposition is around the equatorial from the influence of the low-pressure system and the trade winds along the line. The upper troposphere contribution makes 60% and in some areas by 70% of the despot Hg^{2+}. However, the lower troposphere is the highest contributor in the high latitude areas. The highest dry deposition will be in the subtropical anticyclones areas like the Middle East (mainly the Asian side), in high altitudes (e.g., the Himalayas), and around Antarctica. The upper troposphere is the main contributor by 79–82% except in the northern and southern poles, whereas the lower troposphere will be the main contributor in the water (Shah, 2017), see figure 7. The wet deposition for Hg^0 has a similar trend to Hg^{+2}. The total Hg^{+2} deposition is also similar to the total Hg^0deposition, however, in some regions, the Hg^0 is approximately higher in concentration. For example, the total deposition in the regions around the equator in Africa, Hg^{+2} is less than 15 µg/m^2y compared to 13-60 µg/m^2y for Hg^0, and in the poles (AMAP/UN Environment, 2019) see figure 8. In figure 9 we can see the comparison between their global totHg estimation the AMAP/UN estimations for 2019 show a significant increase compared to 2013 estimations, however, Kawai estimations show a significant decrease compared to the AMAP/UN (2013 & 2019) estimations (AMAP/UN Environment, 2013, 2019; Kawai, 2020).

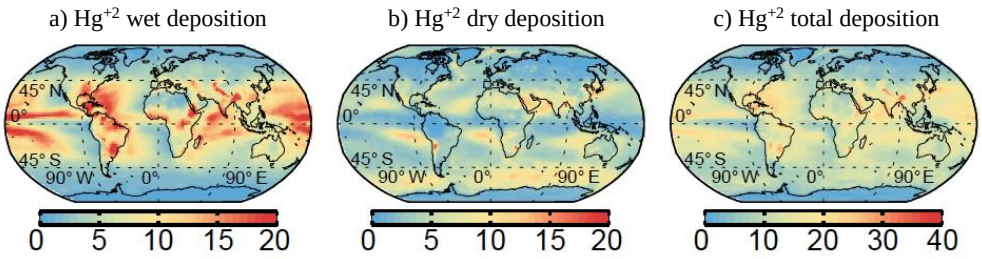


Figure 7. The fluxes of Hg^{+2} (a) wet deposition, (b) dry deposition, and (c) total (wet & dry) deposition in ($\mu g/m^2y$) (Shah, 2017)

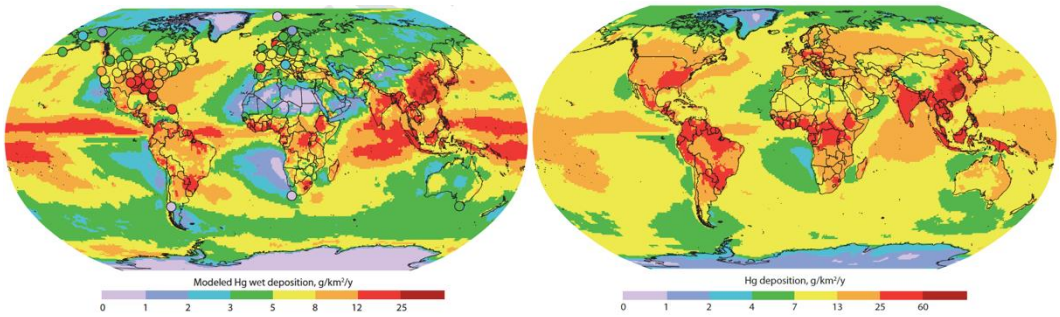


Figure 8. The fluxes of Hg^{+0}, left is wet deposition & right is total (wet & dry) deposition in ($g/km^2/y$) (AMAP/UN Environment, 2019)

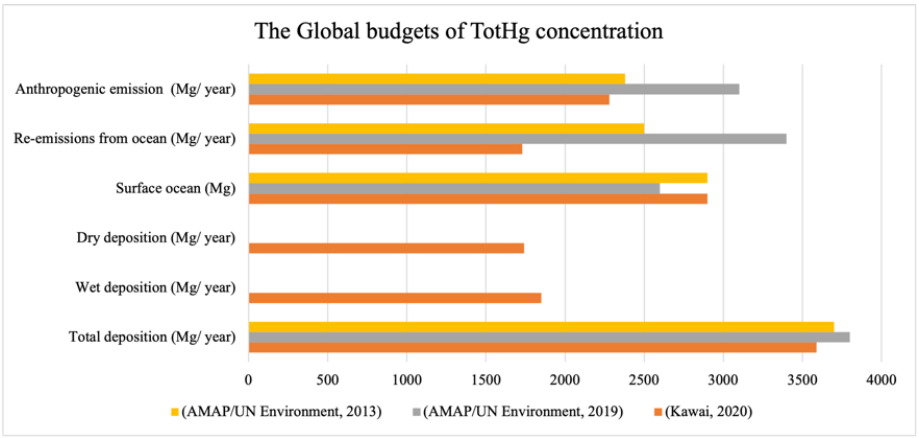


Figure 9. The Global budgets of TotHg concentration related to oceans (AMAP/UN Environment, 2013, 2019; Kawai, 2020)

2.4.2. In the Soil and Sediment

Soil is a natural Hg source, reservoir, and is considered to be a Hg sink; however, Hg contaminates the soil from the atmosphere or through the anthropogenic addition for example during the gold extraction process (Gyamfi, 2021; Y. Liu, 2022; Morosini, 2021; Song, 2021). The atmospheric pathway occurs in two ways, direct Hg deposition into the soil surface or when plants absorb the Hg and then recycled it into the soil through litterfall (Y. Liu, 2022). Mercury can also transport from the soil to the atmosphere through volatilization or to the hydrosphere to groundwater through infiltration. The IHg in the soil can be in different species one of the most stable species is Hg^{+2} sulfide or cinnabar HgS, however, Hg is transported through the food web and bioaccumulated as MeHg. Therefore, in order to understand the fate of Hg in the soil, we need to measure Hg using sequential extraction procedures (Morosini, 2021). In coastal areas, the sediment is considered to be the main source for MeHg not the Hg methylation in water. it is considered as Hg sink, however, it is the main location for Hg methylation and MeHg in the ocean (Mao, 2020; Whalin, 2007). The water-sediment interaction makes predicting the concentration of MeHg and totHg very difficult because of the sediment characteristics, molecular diffusion, the surrounding conditions, and the main one is advective transport (Mao, 2020). The mangrove ecosystem is considered to be a carbon sink system; this means that the organic matter (OM) will be high; this will enhance the bioaccumulation of Hg in the sediment. Moreover, mangrove sediment is low in pH, anoxic surrounding, adequate amount of sulfate and SRB, all those characteristics will create the perfect environment for Hg methylation. The litterfall from the mangrove tree was the main source for the OM, the OM from litterfall, and during anaerobic conditions, the MeHg concentration

increased (Duan, 2021). See table 1 for sediment totHg concentration from different locations.

Table 1. TotHg Concentration in Sediment From Different Locations in mg/kg (dry weight)

Concentration (mg/kg dry weight)	Location	Reference
0.09055	Alaska, USA, Atqasuk lakes	(Burke, 2020)
0.06279	Alaska, USA, Reindeer Camp lakes	
0.00108	Alaska, USA, Atqasuk lakes	
0.00012	Alaska, USA, Reindeer Camp lakes	
0.0239- 0.179	Qatar from 8 locations	(Kreish, 1999)
0.0007- 0.0167	Qatar from 5 locations	
0.0006- 0.0022	UAE	(De Mora, 2004)
<0.0001- 0.0112	Oman	
0.0025- 0.2202	Bahrain	
0.008- 0.034	Qatar 13 location	(Hassan, 2019)
0.02- 9.29	Parangipettai coastal region, India	(Satheeswaran, 2019)
0.46	Yatsushiro Sea, Japan	(Matsuyama, 2019)
0.1	Amakusa Sea, Japan	
3.24	Bandar Abbas, Persian Gulf, Iran	(Elsagh, 2021)
0.14- 44.0	China from 4 different locations	(Song, 2021)

2.4.3 In Hydrosphere

(Zhang, 2014) used ocean tracer model OFFTRAC to estimate the Hg distribution in the ocean for the present time. The high atmospheric wet deposition rate increased the Hg concentration in the oceanic mixed layer in the coastal area and around the midlatitude. The anthropogenic input contributed by 80- 100% to the overall Hg input, and it is concentrated in the middle and northern hemispheres, however, lower

around the poles. The overall Hg concentration increased with the increase in depth at around 1 km. Although, methylated Hg is considered to be the highest subsurface waters and part of the net OM (Wang, 2020). The organic particles deposition and the low oxygen levels increased the Hg concentration in the Gulf of Guinea, the Arabian Sea to south Asia, and the highest (~ 0.401 ng/L) spreading in the American side of the Pacific Ocean between 60°N and 30°S. The anthropogenic input is the highest in the North Atlantic Ocean and between 30°S and 60°S (Zhang, 2014), see figure 10. Rivers and estuarine are considered to be two of the main totHg sources for the aquatic system by contribution range between 50 to 80% compared to less than 10% from terrestrial (Whalin, 2007).

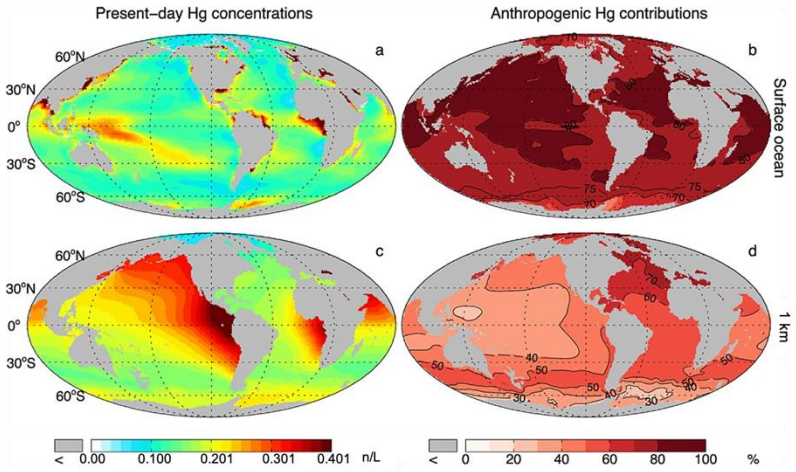


Figure 10. Hg concentrations and anthropogenic contribution spatial distribution from (Y. Zhang, 2014)

2.5. Health Impact on Humans

2.5.1. Pharmacokinetics

The chemical form will influence the absorption and distribution in the human body. Hg enter the human body through different pathways like respiration, orally, and through the skin (Kimáková, 2018). See table 2 for absorption levels. Elemental Hg is highly and very rapidly absorbed through respiration (32- 160 ng/d), as a result of the high solubility in the blood lipids, this results in 95% of this form is in the red blood cells (EPA, 1997; WHO, 2017). The elemental Hg oral pathway is mainly from dental amalgam (Abass, 2018). Elemental Hg distributes through the blood and can penetrate the brain. Elemental Hg can accumulate and oxidize in the brain to mercuric forms (EPA, 1997). Elemental Hg will be metabolized by oxidation which converts it to mercuric ions, however, this process can be slow (Abass, 2018; EPA, 1997). Elemental Hg will be discharged through bodily fluid extraction, feces, and exhaled air (EPA, 1997). Organic (MeHg) main absorption pathway is orally from food sources mainly fish. MeHg is the dominant form of Hg in the red blood cells and will be distributed to the brain by binding to the thiol group which works as a ligand to form MeHg-L-cysteine/ S-(Methylmercury)-L-Cysteine ($C_4H_9HgNO_2S$) complex which has the ability to pass through the blood-brain barrier, see figure 11 (Abass, 2018; National Center for Biotechnology Information, 2021). Opposite to the other form of Hg, MeHg is stable and rarely metabolized through demethylation to other forms (Abass, 2018). MeHg has an excretion half-life that ranged between 32 to 164 days, main route of elimination is through demethylation and excretion in the feces. Besides, MeHg can be secreted into the breast milk exposing thus newborns and infants (Abass, 2018; EPA, 1997). Inorganic Hg's main absorption pathway is through respiration. It was found that exposure to mercuric chloride for 4 hours a week (1 h per day) results in daily absorption between 37- 44 μg /kg. Opposite to the elemental and organic Hg, the

inorganic Hg will not penetrate the brain. Inorganic Hg can be reduced back to elemental Hg by mitochondrial proteins, NADPH and NADH. Since inorganic Hg has the lowest absorption ability around 85% of it will be eliminated by urine, feces, and a small amount by saliva, sweat, and breast milk (EPA, 1997).

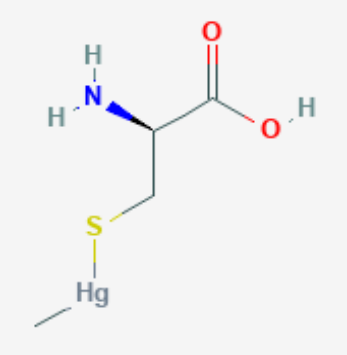


Figure 11. S-(Methylmercury)-L-Cysteine chemical structure (National Center for Biotechnology Information, 2021).

Table 2. Mercury Forms and Absorption Routes and Levels in the Human Body (H: High, M: Moderate, and L: Low) (EPA, 1997; Kimáková, 2018)

Exposure route	Elemental	Organic (MeHg)	Inorganic
Respiration	H for vapor (~85%)	H	L- M (40%)
Oral	Very L for liquid (0.01%)	H (95%)	L- M but high in infant (~15%)
Skin	M for vapor (3%)	L- M (~5%)	L- M (~3%)

2.5.2. Health Effects

Exposure of fetuses, infants, and young children to low levels of Hg can have a negative effect on child development, learning abilities, and behaviors (Kimáková, 2018). In adults, exposure to mercury at levels exceeding recommended ones can cause a decline in mental ability dementia, and dysarthria (Rahmani, 2018). MeHg is highly toxic compared to the other forms of Hg (Kimáková, 2018). MeHg is a neurotoxic compound with high penetration ability through plasma membranes, blood-brain barrier, and the placenta, and can also damage the cardiovascular system (Anual, 2018). Moreover, they can affect the Central nervous system (CNS) and cause loss of speech, hearing, vision, memory, and walking (Al-Ansari, 2017; Rahmani, 2018). See table 3 for the effect of Hg species on human health.

Table 3. Mercury Species Effect on Human Health (EPA, 1997, 2001b; WHO, 2000)

	Elemental Hg	Inorganic Hg	MeHg
Central nervous system (CNS)	Neurological dysfunction: erethism, memory loss, insomnia, nerve sensing, and motoring ability	Nerve pain, decreasing in the cerebellar and the brain weight, irregular arm movements, and dysphagia	Neurological dysfunction and poor development in children and fetus Acrodynia, seizures, losing sight and hearing, language disorders, memory loss, and paresthesia, and numbness in extremities and perioral area
Urinary system	Renal dysfunction	Renal transient proteinuria and failure	-
Digestive system	Gastrointestinal dysfunction,	Nausea, abdominal cramps, diarrhea, and corrosive to the gastrointestinal tract	-
Genotoxicity	-	Chromosomal aberrations and disorders	Chromosome breakage in lymphocytes
Others	Chest pain, dyspnea, and reduced the pulmonary function	Corrosive to skin and eyes	Cardiovascular system: elevation in blood pressure Immunotoxin

2.6. Impacts on Marine Species

Since MeHg has a low elimination rate, the bioaccumulation in the fish tissues, muscle, and liver is fast (da Silva, 2020; Elsayed, 2020). In many cases, the high levels of Hg can cause death to the aquatic organisms (Kimáková, 2018). Hg toxicity can increase the risk of fish organism oxidative damage which can be recognized by testing DNA damage and lipid peroxidation in muscles and liver (da Silva, 2020). Seabirds and marine mammals that feed on the contaminated fish will be at risk of Hg toxicity (Dietz, 2021). The contaminated fish will reach the toxic level as a result of Hg bioaccumulation, which results in decay in a fish population (Al-Ansari, 2017). This

would result in a decrease in the number of marine species in general and fish communities particularly and the biodiversity (Cunningham, 2019). In addition, MeHg will bind to metallothionein protein in the liver and increase hepatic methylation, this will result in increasing the MeHg in the muscle (Elsayed, 2020; Ferreira da Silva, 2020). Similar to the human body, the fish body will accumulate Hg in the tissue by binding to the thiol group of proteins (Kljaković-Gašpić, 2021).

2.7. Fish and Mercury

2.7.1. Mercury Concentration in Fish

MeHg has the highest bioaccumulation ability in the tissues compared to other forms of Hg (Elsayed, 2020). Hg exists as MeHg between 70 to 100% in fish. The concentration of total-Hg and MeHg in the Arabian gulf ranged between 0.012- 0.970 ppm (w/w) and 0.03- 0.248 ppm (w/w) respectively. Some studies only measured the totHg, therefore the highest ppm for the MeHg is low compared to total-Hg (Elsayed, 2020). The concentration of heavy metals and metalloids in fish is highly associated with the increase of the size, length of the fish, and it was found that secondary carnivores have higher levels compared to fish from lower trophic levels (Anual, 2018). Length and weight of the fish have a positive correlation with the MeHg concentration and this can be related to the increase in the fish life span and size (Al-Ansari, 2017; De Mora, 2004). In some fish species, the warmer temperature will enhance the feeding rate and others will not, for the species where the feeding enhances the metal bioaccumulation will increase, although warmer temperature might increase the growth rate which will dilute the content of the metal (Jordan, 2019; Walberg, 2011). Hg concentration is also related to the type of food the fish consume and the composition of the water. The functional proteins in the muscle of the fish have the highest level of Hg (Kimáková, 2018). Hg level in the fish parts will be by decreasing order in the liver, muscle, and finally gonad and other parts. This is a result of the liver's high metabolic

activity compared to the tissues and the other parts which will increase the metals accumulation ability (Elsayed, 2020). Moreover female fish have higher Hg levels compared to males (Al-Ansari, 2017). However, if the concentration of Hg in the muscles is 0.5 ppm and below, the liver's high detoxification ability will maintain a lower Hg level than the muscles (Elsayed, 2020). The Hg level in fish during the fall season is higher than in the other seasons as a result of the thermocline effect in the summer, which brings the Hg from the ocean ground sediment into the upper layers (da Silva, 2020). However, in Qatar study showed that there are no seasonal variations (Al-Ansari, 2017).

2.7.2. Fish Species and Mercury Bioaccumulation

Mercury bioaccumulates in the aquatic species' tissues through the ingestion of contaminated soil and its concentration increases through the trophic chain (Clarkson, 2020). Fish are considered to be at the top of the trophic levels in the marine ecosystem (Xu, 2021). The trophic chain starting from the bottom of the food chain to the top is as follows: heterotrophic (zooplankton & benthic invertebrates), herbivorous, and carnivore (predatory fish). Therefore, predatory fish contain higher levels of Hg (da Silva, 2020) since they are placed at the top level of the trophic chain. The carnivore fish sharp nose shark (*Rhizoprionodon oligolinx*) had the highest Hg concentration 1.287 ppm compared to 0.0068 ppm for the Badah (*Gerres oyena*) which is considered an omnivore fish (Elsayed, 2020), see figure 12. If the low or mid trophic level species have high Hg concentration this will result in an increase in the Hg level in the upper trophic level species (like in tuna fish). In addition, it was reported that the deeper the water column of the ocean, the higher the Hg level in the fish species, for example, benthic species have higher Hg levels than pelagic species (sardines & mackerels) (Al-Ansari, 2017; da Silva, 2020). However, a study showed that

mesopelagic fish had lower Hg levels compared to epipelagic fish (Al-majed, 2000). This was explained by the trophic level of the fish which will reflect on the feeding sources for the fish in each layer. Since the mesopelagic fish in this study are planktivorous (*Hilsha ilisha*) and the epipelagic are carnivorous, the Hg level will be influenced by the trophic level not the depth of the water (De Mora, 2004). Some carnivorous (*Lethrinus nebulosus*) can feed on echinoderms and crustaceans instead of small fish, which would lead to lower Hg levels compared to carnivorous (*Epinephelus coioides*) that feed strictly on small fish (Al-Ansari, 2017). Tables 4 and 5 present Hg and MeHg concentrations in different fish species from the Arabian Gulf. On the other hand, anchovy species *(Stolephorus indicus & Engraulis encrasicolus*) are low-trophic level fish that feed mainly on zooplankton, prawn, shrimp, and amphipods (Alizada, 2020). They are also prey for carnivorous species, and pelagic and demersal fish, and are consumed by human in different forms (Karsli, 2021). Another example for low-trophic level fish is sardine (*Sardina pilchardus*) a pelagic fish that feed on phytoplankton and zooplankton, sardine can feed in two ways filter-feeding and particulate feeding, depending on the food source available (da Silva, 2020; Sofoulaki, 2019). Hg concentration in the parts of anchovy from UAE ranged between 0.05- 0.18 mg/kg (w/w) (Alizada, 2020). Hg concentration in the muscles of sardine from Portuguese ranged between 0.0016 mg/kg - 0.0006 mg/kg (w/w) (da Silva, 2020).

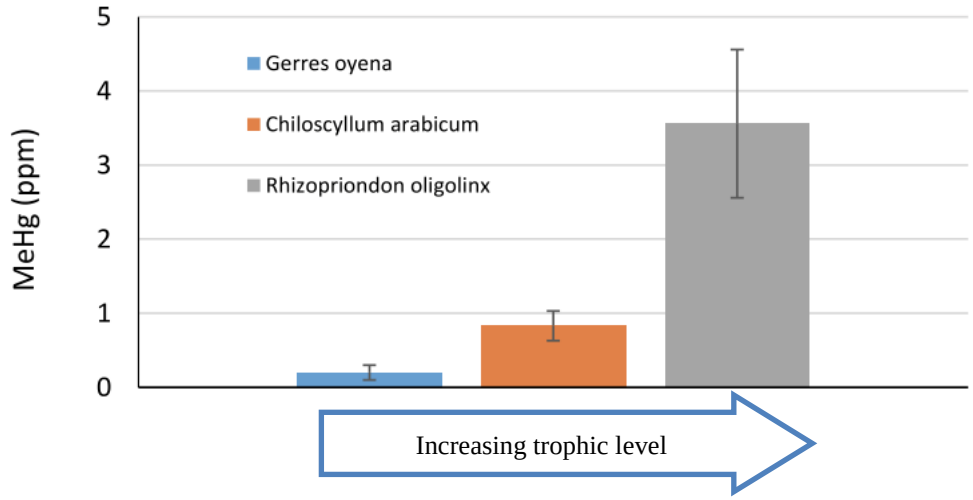


Figure 12. MeHg concentration and trophic level effect (Elsayed, 2020)

Table 4. Fish Species in the Arabian Gulf and Their Hg Concentration

Country	Fish	Part- dry or wet	Hg concentration (mg/kg (w/w))	Reference
Qatar	Sha'ri (*Lethrinus nebulosus*)	Liver - wet	0.773	(Al-Ansari, 2017)
UAE		Liver- dry	1.02	
Oman		Muscle- dry	0.522	
Qatar	Hamour (*Epinephelus coioides*)	Liver- dry	1.28	(De Mora, 2004)
Bahrain		Liver- dry	2.1	
UAE		Liver- dry	4.65	
Oman		Liver- dry	1.3	
Kuwait		Muscle- dry	0.01-3.92	(Al-majed, 2000)
UAE	Anchovy (*Stolephorus indicus*)	Tissues- wet	0.04- 0.18	(Alizada, 2020)

Table 5. Fish Species in the Arabian Gulf and MeHg Concentration

Country	Fish	Part- dry or wet	MeHg concentration (mg/kg (w/w))	Reference
Qatar	Sha'ri (*Lethrinus nebulosus*)	Liver - wet	0.771	(Al-Ansari, 2017)
Kuwait	Hamour (*Epinephelus coioides)*	Muscle- dry	0.001-3.27	(Al-majed, 2000)
Qatar	Badah *(Gerres oyena)*	Tissues- wet	0.0283	(Elsayed, 2020)
	(*Chiloscyllium arabicum*)	Tissues- wet	0.1662	
	or carpet shark (*Rhizoprionodon oligolinx*)	Tissues- wet	0.7942	
	or sharp nose shark			

2.7.3. Fish Preservation

Food preservation is an important process in the food industry since it makes storing food for a long time possible and preserves the nutritional content (Vafaei, 2018). Tuna is a predatory fish (top food chain) as a result that they have a high ability to bioaccumulate heavy metals in the tissue (Afonso, 2015). Canned tuna is one of the most consumed canned fish around the world (Mol, 2011). Tuna and salmon make up around 9.2% of the fish captured and produced around the world (Nong, 2021). Commercial handling and processing like canning increase the Hg level in tissues too (Mol, 2011). Exposing the fish to high temperatures like in cooking or the canning process will increase the Hg concentration, this can be explained by some chemical reaction between Hg species and sulfhydryl groups forming complexes compound, the loss of water during cooking (reduction in humidity), and mercury/mass ratio will increase because of the minerals loss and mass reduction (F. D. N. Costa,

2016). Freezing is an important method to preserve high-protein food sources and will not affect Carbon (C) and Nitrogen (N) concentrations (Ghazwan, 2016). Frozen fish from the Arabic/ Persian Gulf had an average of 0.79 μg/g Hg concentration, this is above the FAO/WHO non-predatory fish limit (Ziarati, 2017). A study done on frozen fish to see the effect of freezing on the Hg concentration, the different fish spices were frozen at –20°C temperature from 2002 to 2006 (around >600 days) the study concludes that there is no significant difference in the Hg concentration (Peterson, 2007). Since many parameters can increase the heavy metal concentration in fish, people avoid the high risk of heavy metal concentration by eating different fish species in smaller quantities (Islam, 2010).

2.8. Mercury Measurement Methods

2.8.1. Hair as Bioindicator

Using hair as a biomarker for Hg is by taking the advantage of the hair natural growing process by 1 cm/ month. This method can show the time period of Hg exposure (Xie, 2021). Another advantage to this method is the easiness of sample collection and storage. MeHg will accumulate during the hair formation from the blood. After the hair formation the MeHg will be preserved and an endogenous character in the hair (Wang, 2021).

2.8.2. Fish as Bioindicator

In general fish uptake heavy metals in two ways, direct through water and feeding and indirect through permeable membranes (Alizada, 2020). Metal's ability to interrupt the vital metabolic processes in the fish tissues makes fish a good bioindicator (Cunningham, 2019). The best way to study heavy metals pollution is using fish as a bioindicator for two main reasons bioaccumulation and biomagnification (Abolghait, 2015). The first factor is that the metals in general have strong persistent characteristics hence have the ability to bioaccumulate in the fish

tissues (Cunningham, 2019). The second factor is biomagnification when metals that were consumed by lower trophic level species will bioaccumulate in the tissues, and after that mid trophic level species will consume them and increase the concentration of the metal in their tissues, and the process will go until it reaches the human (Rahmani, 2018).

2.8.3. Mercury Stable Isotope

The isotope techniques, chemical tracers, or isotopic measurements is a method used to study the biogeochemical Hg cycle, sediment deposition rate into the water, and to study the Hg sources and transformations in rice (Jain, 2003; J. Liu, 2021; Tsui, 2020). Hg has seven stable isotopes and can be divided into five different types (Q. Huang, 2016; Tsui, 2020). Radioactive isotopes are ^{203}Hg for small marine species Hg uptake and efflux (Tsui, 2020). Spiking of highly enriched stable isotopes ^{199}Hg, ^{200}Hg, ^{201}Hg, or ^{202}Hg into the study area, to study the methylation and demethylation rates in soil and the Hg biogeochemical cycling in watersheds (J. Liu, 2021; Tsui, 2020). Natural abundance isotopes ratios ^{198}Hg ^{199}Hg, ^{200}Hg, ^{201}Hg, ^{202}Hg or ^{204}Hg, for MeHg photo-demethylation in water, and food chain in marine species and humans, moreover, to investigate the dry and wet deposition of Hg contribution. The last two types will use the isotopes of other elements, the light isotopes ^{13}C, ^{15}N, or ^{34}S, for food web complexities and trophic level estimation for MeHg biomagnification, and finally, the natural abundance stable ^{13}C isotope to study Carbone source for the MeHg (Q. Huang, 2016; Tsui, 2020).

2.8.4. Instrumental Used for Measurement

There are many methods to determine the total Hg and MeHg levels in foods. The method used in the fish will depend on the type of instrument used (Perelonia, 2021). Measuring the weight of the wet and/or dry tissues first, and if needed length

and age is required. The first steps consist of slicing and homogenizing the tested parts and then freezing at a very low temperature (-20 ^{o}C and below) until the time of analysis (Anual, 2018). Comparison between Some of the instruments used for total Hg and MeHg analysis in Table 6.

Table 6. Instrument Used for Total Hg and MeHg Analysis

Instrument	Advantages	Disadvantages	References
Inductively Coupled Plasma Mass Spectrometry (ICP-MS)	Multi-element detection High accuracy High sensitivity	Spectroscopic and non-spectroscopic interferences Pre-treatment required	(Anual, 2018; Perelonia, 2021)
Cold Vapor Atomic Absorption Spectrometry (CVAAS)	The prepared aqueous sample is easy to prepare and handle Low detection limits (0.2-10 µg Hg/L) Good for elemental Hg	High contamination risks High sample waste Pre-treatment required	(Brandão, 2005; EPA, 1994; Fernández, 2015)
Flame atomic absorption spectroscopy (FAAS)	High sensitivity High selectivity Easiness of use Short duration time Small volume Low cost	No direct detection Require preliminary separation	(Cui, 2010; Pourjavid, 2016)
Brooks Rand MERX System with Hg Speciation GC & Pyrolysis	Ultra-Low detection limits Easiness of use Trusted EPA Methods 1630 & 1631 Fast and stable	-	(Al-Ansari, 2017; Brooks Rand Labs, n.d.)

In a study conducted by Al Ansari et al., as a result of limited resources, they used a different set of samples between 2011 and 2012 to study total Hg and MeHg, this is because degradation of MeHg was noticed in samples age more than 6 months (Al-Ansari, 2017). It is important to take into consideration that the wet weight will

give a higher Hg level compared to the dry weight (Al-Ansari, 2017). Wet weight can provide more accurate Hg values since any preserving processes will affect the tissues (Crane, 2016). For example, in the liver, the Hg concentration for wet and dry weight were 0.600 and 0.018 mg/kg (w/w) respectively (Al-Ansari, 2017). To convert between wet to dry for the needed concentration:

$$wet \text{ weight } = (mass\ ratio) x\ dry \text{ weight}$$

The mass ratio is indicated by calculating the moisture percent, weighing the fish before and after drying tissue, and calculating the percentage (Al-Ansari, 2017; Burger, 2006).

Another way to calculate the Hg concentration for wet weight C_w (Milatou, 2020):

$$C_w = C_d \frac{M_d}{M_w}$$

C_d: concentration for wet weight, M_d: dry weight, and M_w: wet weight

2.9. Fish Consumption and Mercury Limits in Fish

2.9.1. Fish Consumption

MeHg bioaccumulation can be a result of consistent contaminated fish consumption (Elsayed, 2020). In many countries around the world, fish is considered one of the main protein dietary sources (Anual, 2018). Global fish consumption increased rapidly, from 9.9 kg in the 1960s to 20.5 kg per capita in 2017 (Cunningham, 2019). One of the commonly consumed fish species in Qatar and the Gulf region is sha'ri (Lethrinus nebulosus) with a 16.2% capturing rate (Al-Ansari, 2017). Table 7 presents fish consumption around the world.

Table 7. Examples for Fish Consumption Around the World

Country	Fish consumption	Average weight (kg)	Reference
International	20.3 kg per capita /year	-	
International (without China)	16 kg per capita /year	-	(FAO, 2020)
Asia	24.1 kg per capita /year	-	
Malaysia	160 g per capita /day	64	(Anual, 2018)
Qatar	10- 20 kg per capita /year 90% (52% of Qatari)	-	(FAO, 2020; Sana, 2020)
Brazil	10 kg per capita /year 175 per capita / week	60	(Alva, 2020; Ferreira da Silva, 2020)
KSA	Saudis:150 g /week Expatriates: 397 g/week	-	(Cunningham, 2019)
Mexico	250 g/ week or 35.71g/ day	Men 70 & women 60	(Murillo-Cisneros, 2021)
Greece	20 kg per capita /year 68.68 g/day	70	(Milatou, 2020; Sofoulaki, 2019)
Iran	20.3 kg per capita /year 21 – 147 g/person/day (frozen fish)	60	(Mansouri, 2021) (Ziarati, 2017)
Colombia	8- 10 kg per capita /year	-	(Alcala-Orozco, 2021)
USA	18.7 g per capita /day	-	(Sunderland, 2018)

2.9.2. Mercury Guideline Levels

To avoid Hg toxicity, it is recommended that fish consumption should be within the guideline levels (GLs) for non-predatory and predatory fish (FAO/WHO, 2007). According to a study, the consumption of sha'ri fish should be limited to three meals a week to avoid exceeding the FAO/WHO GLs (Al-Ansari, 2017). Tolerable weekly intake (TWI) is the amount of contaminate in a specific amount of food or water per unit of body weight, which a person can ingest weekly without having the risk to

develop adverse health effects (Anual, 2018). The term tolerable intake can also be used to address the daily intake (TDI) (WHO, 2017). The provisional tolerable weekly intake (PTWI) expresses the maximum safe long-term exposure intake from the contaminate (EFSA, 2012). The PTWI was set at 1.6 ug/kg bw for methyl mercury. Exceeding PTWI level is highly possible in nations where fish consumption and or contamination levels is/are high. When pregnant women exceed the PTWI level the Hg can influence the fetus's brain development (Anual, 2018).

$$\text{PTWI} = \frac{mean\ Hg\left(\frac{\mu g}{g} wet\ weight\right) x\ weekly\ fish\ consumption(g)}{body\ weight\ (kg)}$$

Governments can assess whether the fish contamination level is acceptable or not using the permissible limits or not. The EPA MeHg permissible limit in fish is 0.3 mg/kg and the EU Commission is 1 mg/kg of wet weight (EPA, 2001c; EU Commission, 2006).

The Environmental Health Criteria 101 Methylmercury estimated the intake for a different form of Hg in the general population. MeHg was 2.41 µg/day with 2.4 µg/day from fish. The inorganic Hg intake from fish was only 0.6 µg/day and 0 µg/day for elemental (WHO, 1990). See table 8 for the totHg and MeHg guidelines limits list.

Table 8. TotHg and MeHg Guideline limits (GLS) Summary

GLS	Concentration	Reference
Max IHg in drinking water	6 µg/L	(WHO, 2017)
Hg in Hair[1]	USEPA= 1.0 µg/g JECFA= 2.3 µg/g	(B. Wang, 2021)
Criterion level in fish (Hg)	0.3 ppm (wet wt)	(EPA, 2001b)
Predatory fish (MeHg)	1.0 ppm (wet wt)	(EU Commission, 2006; FAO/WHO, 2007)
Non-predatory fish (MeHg)	0.5 ppm (wet weight) or 2.5 ppm (dry wt)	(Al-Ansari, 2017; FAO/WHO, 2007)
PTWI (MeHg)	1.6 µg/kg	(FAO/WHO, 2007)
TWI (MeHg)	1.3 µg/kg	(EFSA, 2012)
PTWI (IHg)	4 µg/kg	(FAO/WHO, 2007)
TWI (IHg)	4 µg/kg	(EFSA, 2012)
TDI (IHg)	2 µg/kg	(WHO, 2017)

2.10. Risk Assessment

Risk assessment is a method used to qualitatively and/or quantitatively determine the health risks for a specific substance and time period. It helps understand the possible risks related to exposure under specific conditions (EPA, 2007; Stelljes, 2008). EPA/WHO method is used to assess human health risks by measuring and estimating the level and probability of health effects to occur when humans are exposed to substances. This method consists of four steps in figure 13.

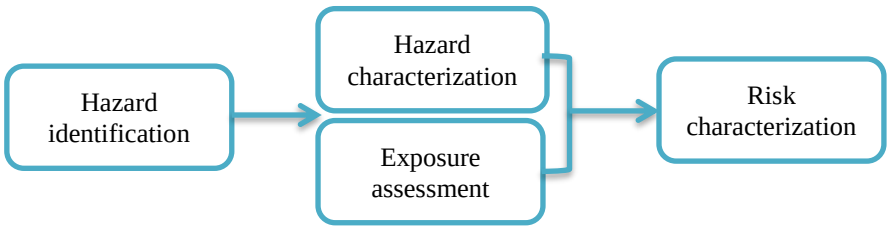


Figure 13. EPA/WHO method for risk assessment (Zolfaghari, 2018)

[1] Calculated using the reference dose from (USEPA) and PTWI from (FAO/WHO, 2007)

2.10.1. Hazard Identification

Hazard identification consists of identifying the health effects by collecting and evaluating toxicity data (EPA, 2014; Stelljes, 2008). This information can be collected from clinical data and studies, laboratory animal studies, and toxicokinetic studies (EPA, 2014).

2.10.2. Hazard Characterization

Hazard characterization consists of dose-response assessment which determines the associations between exposure doses and toxic effects. There are two estimation methods, one for carcinogenic compounds and the other for non-carcinogenic compounds. Hazard characterization yields the reference dose (RfD), reference concentration (RfC), TDI/ TWI, or acceptable daily intake (Locey, 2005; Stelljes, 2008). This is done by selecting the NOEL (the no observed adverse effect level) or LOAEL (the lowest observed adverse effect level) and dividing it by modifying and uncertainties factors for non-carcinogenic compounds (Stelljes, 2008; WHO, 2017). The PTWI and TWI for MeHg were measured by the FAO/WHO as seen in table 8.

$$\text{RfD } \left(\frac{\boldsymbol{\mu g}}{\boldsymbol{kg * w}}\right) = \frac{NOAEL \text{ or } LOAEL}{UF \times MF} = \text{PTWI or TWI}$$

The NOAEL is the highest measured contaminate level that does not have an adverse effect on health and LOAEL is the lowest dose that gives a toxic effect. UF: uncertainty factor, and MF: modifying factor (Renwick, 1993).

2.10.3. Exposure Assessment

Exposure to a contaminant is necessary for toxicity to occur. Exposure assessment is a process to quantify the amount to which humans are exposed to a certain contaminant (Filter, 2021). The assessment is done by identifying the receptor, exposure pathway, and contaminant dose in the environment (Stelljes, 2008). It consists

of multiplying the amount of intake/uptake by the contamination levels and then summing up data from all sources (EPA, 2014).

- The EDI or/and the estimated weekly intake (EWI) can be used and both represent the total Hg intake (Ferreira da Silva, 2020; Sofoulaki, 2019).

$$\text{EWI } \left(\frac{\boldsymbol{\mu g}}{\boldsymbol{kg * w}}\right) = \frac{CNP \times C}{bwt}$$

C: concentration of contamination, CNP: consumption per week, and bwt: body weight

Exposure assessment can be measured using mathematical models the deterministic and probabilistic models. The deterministic model outcome is determined by the relation between specific variables without randomness. This method can provide an understanding of how each variable affects each other's. The probabilistic model outcome is the product of random parameters that will result in its probabilities and have the ability to seize the uncertainties for each parameter (Bruce, 2007; Zamora-Arellano, 2017).

2.10.4. Risk Characterization

Risk characterization consists of estimating the risk level of a contaminant by integrating data obtained in the previous steps. In other words, it is the likelihood of adverse health effects occurrence based on the exposure level (Bleam, 2012). There are two methods to estimate the risk depending on the nature of the contaminant: non-carcinogenic, and carcinogenic. Since Hg is considered non-carcinogenic the hazard quotient equation (HQ) is used to estimate the risk level (Bleam, 2012; Rahmani, 2018). HQ is a risk measurement method for oral contamination intake. The HQ equation is the result of the division of the estimated daily intake EDI or weekly intake EWI over TDI or TWI, or RfD (Murillo-Cisneros, 2021). When HQ> 1, health risks are present

from current exposure; and if HQ< 1, no health risks are present from current exposure (Acosta-Lizárraga, 2020).

- The HQ result is used to assess the health risks using the below equation (Murillo-Cisneros, 2021)

$$HQ=\frac{EDI}{TDI}\ or\ \frac{EWI}{TWI}$$

The mercury permissible limits for the types of fish and the PTWI and CL are only for regulatory actions and cannot be used to assess the Hg/ MeHg actual risk level for a specific population in a specific context (Cunningham, 2019). For this purpose, The risk assessment should be based on data from consumption (such as favored consumed fish species and rate of consumption), contamination (the concentration of the Hg/ MeHg in the tissues), and the average consumer bodyweight from one side, and the reference dose (Rfd) or TWI, from the other side (Cunningham, 2019; Dietz, 2021). See figure 14 for quantitative risk assessment required data.

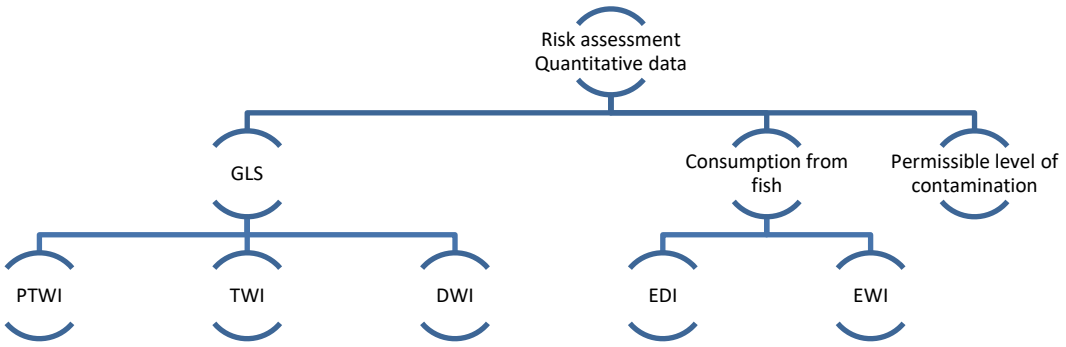


Figure 14. risk assessment measured data requirement

CHAPTER 3: MATERIALS AND METHODOLOGY

3.1. Material

3.1.1. Chemicals and Reagents

Seven fish species samples were collected for total Mercury (totHg) analysis. Deionized water was used for reagents, standard, and washing. Nitric acid solution (HNO_3) 1% were prepared from 65%- 70% HNO_3 from Fluka Analytical and were used as stock solution and blank. Mercury stock solution was prepared from 1000 ppm Hg and gold (Au) 50 ppm from PerkinElmer. The Au was used for the stabilization of the calibration curve standards. Hydrochloric acid solution from Scharlau (1.5 M) and Ethanol from Oxford lab were used for washing.

3.1.2. Apparatus and Instrumentation

The collected samples were dried in Kendro Laboratory Products Heraeus UT 20 oven at 80ºC, ground using a food processor and coffee grinder, and then stored in plastic zip bags. Standard, intermediate, and spiking were prepared using disposable polypropylene tubes (15 mL and 50 mL), and automatic micropipette 20 -200 µL and 100-1000µl. Acid digestion using Milestone ultraWAVE microwave digestion system was used.

The samples analysis was conducted using Perkin Elmer Inductively Coupled Plasma Mass Spectrometry (ICP-MS) NexION 350, equipped with an elemental scientific autosampler, and Syngistix software. ICP-MS Perkin Elmer temperature reaches 10,000 K, uses Helium as a carrier gas, and measure Hg^{208} isotope. Operating parameters were as the following: RF power (1600 W), plasma gas flow (18 L/min), auxiliary gas flow (1.2 L/min), nebulizer gas flow (0.97 L/min), carrier gas flow (3 ml/min), torch injector internal diameter (1.2 mm), injection volume (2 µL), running time (1 min), and Interface (ion focus) and Ni (1 mm sampler: 0.4 mm skimmer).

3.2. Methodology

3.2.1. Fish Consumption

A fish frequency questionnaire was designed to collect the fish-eating patterns for the resident of Qatar. A convenience sampling approach using the online google forms platform was used. The questionnaire was approved by the Institutional Review Board (IRB) (IRB 1807049-1). The questionnaire targeted adults aged 18 and above and had an open time frame to collect the required number of participants based on the sample size calculation. The number of needed participants was 600 and calculated based on a confidence level of 95% and a confidence interval of 4 for the 2,500,000 population. The questionnaire consisted of three sections. The first section was an introduction to the questionnaire and the study objectives. The second section included the general demographic information including age, gender, in addition to participant's body weight and height, pregnancy and breastfeeding status (for female participants), in addition to reporting whether the participant is following a high protein diet and the places from where they usually purchase the fish. The third section consisted of the fish frequency questionnaire that aimed at collecting semi-quantitative data on the participant's fish-eating patterns of fish species (number of portions and frequency/week) based on a reference period of the previous year. Nine species were included in the fish frequency questionnaire, together with their portion sizes and pictures to help participants recognize the fish species and identify the portion size that is usually eaten. Besides, the participant was provided with the option to report the consumption of any other species and/or portion outside the listed ones. The fish included in the questionnaire were selected based on the most available and sold ones in the Qatari market these were: Hamour (*Epinephelus coioides*), Safi (*Siganus rivulatus*), Chanad (*Scomberomorus commerson*), Sha'ri (*Lethrinus nebulosus*), Tuna (local: *Euthynnus affinis*), Salmon, Sea bass, Sardine, and Anchovy. The species chosen

were based on fish market observation and investigation, and the local species consumption were confirmed by previous study where Hamour, Safi, Chanad, and Sha'ri were the most consumed local species (Sana, 2020). The common name for each species was used in the questionnaire. Moreover, the participant had to report for each species whether it is purchased as fresh, frozen, canned, and/or dried. Finally, participants had to state their origin for species that exist as both locally captured or imported. The types for each fish were as follows: for Hamour, Safi, Chanad, Sha'ri, and Sea bass, fresh and frozen; for Tuna and Salmon, fresh, frozen, and canned; and for Sardine and Anchovy, fresh, frozen, canned, and dried. The intake rate was identical for all fish species and was expressed in terms of the number of portions per week as the following: 0, less than one per week, one per week, 2 per week, 3 per week, 4 portions per week, 5 per week, 6 per week, once a day, 2 per day and other. When the less than one per week option was selected, participants had to report the amount consumed based on the reference portion size. The fresh and frozen consumption portion was assigned as 200 g/w and the canned fish portion was based on the average drained tissue. Canned Salmon was 150 g/w, and canned tuna was 120 g/w.

3.2.2. Samples Collection and Preparation

The fish species that were selected for analysis were determined from the fish frequency questionnaire responses using a cut-off value for sample representativeness of 93% and fish that were consumed by less than 5% of the participants were not included. Based on this cut-off value seven fish species were selected and included. The species were Hamour, Safi, Chanad, Sha'ri, Tuna, Salmon, and Sea bass. For each selected fish species, a minimum of three samples were collected from each location and from where participants reported buying their fish. The samples were collected on different days based on their availability in the market between November 2021 and

January 2022. Whole fish samples for each species were bought with similar sizes and weights. The fish samples included fresh Hamour, Safi, Chanaad, Sha'ri, and Tuna locally caught and fresh imported Hamour, Salmon, Tuna, and Sea bass. Local Hamour and Tuna samples bought were among the medium size range, on the other hand, the imported Hamour, Tuna, and were only available in large sizes and bought as fillet on a plate. Salmon samples were bought as fillets on a plate and Sea bass samples were bought as whole fish. In addition, canned Tuna and Salmon samples from different brands were purchased from the local supermarkets in Qatar. Two Safi fish were mixed together with similar weight from each fish to represent one fish portion sample. This method was only done to Safi because of the small fish size that can make one portion from two fish. The original weight was measured in the purchasing location and Fish dissection was done in the location of purchasing to follow consumers' normal fish purchasing method. At the time of sample collection, only one frozen sample was purchased for the only available brand of Salmon. All bought samples were transported on the same day of their purchase to the lab in an ice compartment. In the Laboratory the samples were stored in the fridge at 5 ^{o}C till their preparation time. If the samples were not prepared on the same day, the samples were stored at -20^{o}C until preparation time. Edible muscle tissues and skin were collected in the lab using a steel knife and plastic cutting board to avoid contamination during sample preparation. Samples tissues were cut by knife into small pieces and were weighed using a balance from each of the fish samples and then dried in an aluminum tray in an oven at 80 ^{o}C until no change in weight was observed. The imported Hamour samples were slightly oilier than the local samples and needed two days for drying. The local and imported Tuna were red meat Tuna. The Sea bass and Salmon were significantly oily in texture compared to the other species and were kept in the oven for two days. The dry weight was determined and

ground using a DeLonghi coffee grinder. Finally, the analytical samples were collected in plastic zip bags and stored in a -20 ^{o}C deep freezer until analysis time. Tools were cleaned between each sample with 1.5 M HCl, ethanol, and DI water.

The moisture ratio was measured using the following formula:

- Moisture ratio= dry weight/ wet weight

Canned samples were purchased from supermarkets only for Tuna and Salmon. Three cans with the same lot number were mixed to represent one sample. Canned Tuna brands were chosen randomly from different brands and preservative mediums like brine, sunflower oil, olive oil, and canola oil. Canned Salmon was one of the only three available brands in the market. The canned samples were stored at room temperature until preparation time. Meat content was weighed after liquid drainage to calculate the average drained weight. The samples followed the same preparation method as described above for fresh and frozen fish samples.

3.3. Sample Analysis

3.3.1. Method Validation

The method validation of the inductivity inductively coupled plasma mass spectrometer (ICP-MS) was tested on milk powder and fish as a representative matrix for validation study, satisfactory recoveries at different concentration levels with recovery ranging between 95.073% - 115.32 %, and relative standard deviation (RSD%) lower than 20 %. The trueness of the method was validated by analyzing certified reference materials (CRM) with an accurate result. The limit of quantitation (LOQ) was 10 µg/L. The method showed to be linear from the LOQ 10 µg/L up to 500 µg/L with a correlation coefficient ≥ 0.99 and limit of detection 1 µg/L. For repeatability and precision, the measurement of expanded uncertainty expressed as relative standard deviation was 28.2 % at 95 % confidence level and coverage factor of k = 2. Proficiency tests and CRM samples were additionally applied to improve

confidence in the measurement results. Full in-house validation of the method intended for routine heavy metals analysis to support regulatory enforcement was carried out. The CRM were Mil powder, Soya flour, canned crab meat, and canned fish from Fapas - Proficiency Testing listed in table 9. The validation was done by the Ministry of Health- Central Food Laboratories.

3.3.2. Quality Control

The analysis procedures method was performed according to the Ministry Of Health standard operating procedures (SOPs) QMS code CTS-18 and using 1% HNO_3 as blank.

Two intermediates were prepared. Intermediate 1 (IM1) 10 ppm Hg in 50 mL tube and intermediate 2 (IM2) were prepared from IM1 in 10 ml tube using 1% HNO_3. Calibration curves were conducted by preparing six standards (ST) from IM2 with 1% HNO_3 and 100 µL of 50 ppm gold for stabilization. The internal standard (Bi 209) was measured with a range of 82-120% required for the method to run. Spiked samples were run between a maximum of 15 samples. A spiked sample was prepared by adding 50 µL IM2. Continuing Calibration Verification (CCV) by injecting a mid- range calibration standard to ensure the validity of the initial calibration of the instrument, ST3 was used as CCV. For the analysis repeatability, the following were measured between every 15 samples and must pass: blank, spiked samples, and CCV, see table 9.

Table 9. Calibration Curve Prepared Standards Concentrations, CCV Measured Concentration and Recovery, Passed Spiked Samples Concentration, CRM Used for Proficiency Tests With Recoveries.

Standards (ST)	IM2 (µL)	[ST] (ppb)
ST1	25	0.5
ST2	50	1
ST3	100	2
ST4	250	5
ST5	500	10
ST6	1000	20
	[Hg] (µg/L)	Recovery%
CCV	2.027	103.8
	2.086	114.8
	2.207	116.8
	1.983	104.0
	1.922	105.3
	1.908	108.7
	2.090	102.6
	2.087	105.4
	1.996	107.0
Spiked	[Hg] (µg/L)	Recovery%
Spike Sha'ri	569.203	107.3
	544.069	110.6
Spiked local Hamour	556.482	102.5
	536.963	106.7
Spiked fresh local Tuna	206.422	106.5
Spiked Chanad	412.883	104.2
	380.961	114.6
CRM	[Hg] (µg/L)	Recovery%
CRM 151-Mil powder	101	92.76
CRM 7204- Soya flour	491	84.13
CRM 7248- Soya flour	292	101.47
CRM 7279- Canned crab meat	106	88.89
CRM 7271- Canned fish	290	91.46

3.3.3. Analytical Procedure

Acid digestion was done using ultra WAVE microwave digestion system. In the Teflon digestion tubes samples were measured to the nearest 0.2500 g and 4 mL concentrated HNO_3. In the digestion Teflon vessel, 5 mL concentrated HNO_3 was added, and 130

mL deionized water. The samples were digested for 30 minutes under 110 bar at 60 ºC - 220 ºC, then cooled to room temperature for 30 minutes. After digestion, the samples were diluted in 50 mL tubes with deionized water to a total of 25 ml. With each digestion process, two spiked samples were prepared, see figure 15. The scheme in figure 16 presents the samples collection, preparation, and analysis.

Figure 15. digestion vessel on the left and digestion tubes on the right

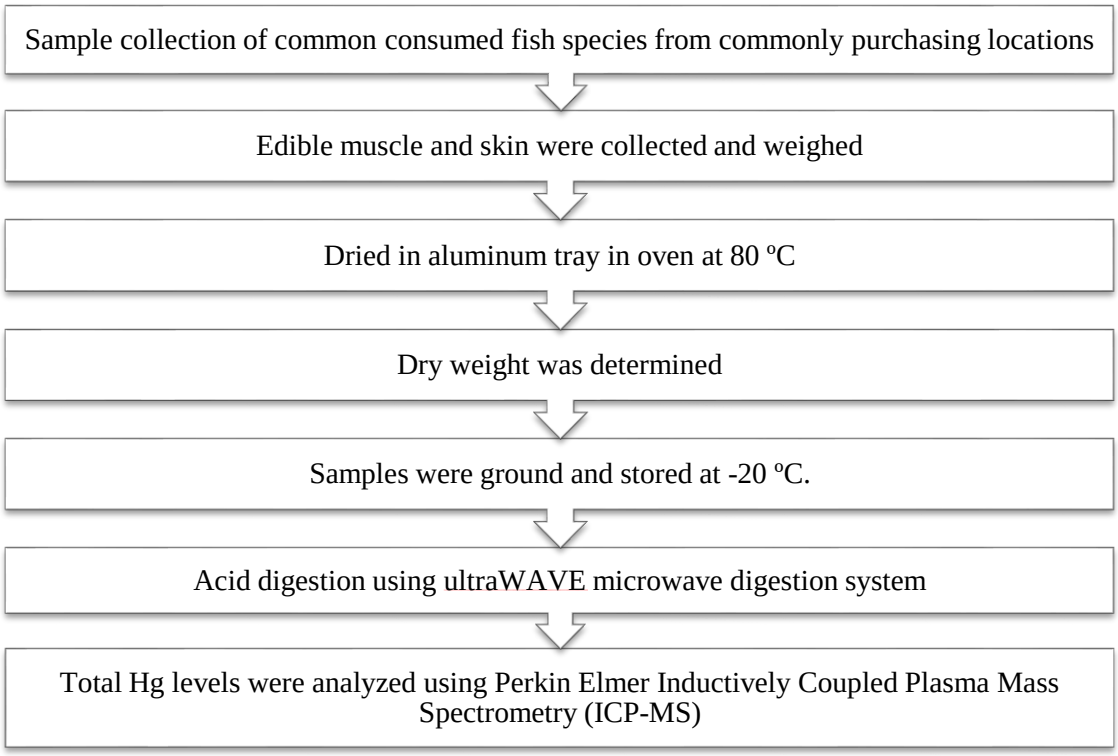


Figure 16. Samples Collection, Preparation, and Analysis.

3.4. Risk Assessment

3.4.1. Fish Consumption Levels

The data collected by the fish frequency questionnaire were entered into an excel file. Questionnaires that had missing/incomplete data regarding fish consumption were discarded. A random double check of entered data was performed to make sure that the data were entered correctly.

The percentage of participants following a specific high protein diet, pregnant and breastfeeding was determined. The participants were then grouped into the following cohorts: general population, Qatari population, and non-Qatari population. Cohorts were grouped into subgroups based on gender (females, males) and/or age (18-29 and 30 and above). The cutoff value of 30 years for age was based on the EFSA study on dietary reference values for energy (EFSA, 2017). In addition, according to EPA age grouping for exposure studies can be related to the change in the behavioral

and physical characteristics, since adults have minimum change the grouping can based on data collected (EPA, 2005). The two age groups were suitable for the number of responses received. Besides, in the female cohort a supplementary group based on the childbearing age of 18-40 years old was added. This upper age value was determined based on local observations since it is very rare to conceive after this age.

The fish frequency questionnaire was analyzed to determine the fish consumption for each participant and for each of the fish species in g per week. In case of a missing weight for a participant, the average for the same gender was used. Then the sum of all consumed fish species for each participant was determined.

The median, P75, and P90 of aggregated and disaggregated fish consumption were determined for the three cohorts.

3.4.2. Exposure Assessment

Exposure assessment consisted of multiplying the data collected on fish consumption from the fish frequency questionnaire and the contamination levels obtained from the analysis. Average contamination levels were used for each fish species and for the whole fish species.

Exposure assessment was done using two deterministic approach methods and simple distribution. Firstly, the disaggregated method (distribution of exposure). The disaggregated method was done by measuring the exposure (µg/w) for each individual respond using totHg contamination level (µg/g) for each fish. And secondly using the aggregated method used the average of Hg content of the analyzed samples, and this method was used in the risk characterization step. In the aggregated each response was analyzed separately to calculate the estimated weekly intake (EWI). Disaggregated fish samples contamination and consumption were used to calculate the exposure to Hg for each fish individually, this method is used to calculate an accurate value of

contamination. TotHg analysis results were multiplied by the weekly consumption to calculate the exposure per week for each fish type individually. The summation of exposure was divided by the body weight for each consumer to calculate EWI.

$$\text{EWI} = \frac{CNP \times C}{bwt}$$

C: concentration of contamination, CNP: consumption per week, and bwt: body weight

Since the analytical method was limited to totHg, we adopted two scenarios to assess the exposure to Methyl Mercury (MeHg). According to the FAO/WHO the percentage of MeHg (MeHg%) can range between 70 and 100%. The level will depend on species, size, age, and feeding habits. The EFSA indicated that 80% of FAO/WHO had MeHg% of 80% and higher. Two scenarios were used for the estimation of the EWI for MeHg. The first scenario was the conservative approach (MeHg 100%) by assuming that all totHg in the seven species is MeHg. The second scenario (MeHg ART) based on the MeHg% for each fish species from previous research as seen in table 10. The median, average, and 75^{th} and 90^{th} percentiles of EWI were compared to the (EFSA, 2012) TWI 1.3 µg/kg and (FAO/WHO, 2007) PTWI 1.6 µg/kg.

Table 10. MeHg% in the Seven Species From Previous Studies

Species	MeHg%	References
Hamour	97.3	(Freije, 2009)
Sha'ri	94.1	(Freije, 2009)
Chanad	97.6	(Freije, 2009)
Safi	94.5	(Burger, 2014b)
Tuna (Mackerel tuna)	93.0	(Ahmad, 2021)
Tuna (Yellowfin)	96.3	(Nicklisch, 2017)
Canned Tuna	90.5	(de Paiva, 2017; Dezfouli, 2018)
Salmon (fresh and canned)	80.5	(Afonso, 2015; Sarvan, 2021)
Sea Bass	81.1	(Maulvault, 2016)

Margin of exposure (MOE) is a metric method used to assess the level of safety of an exposure based on consumption patterns and contamination levels. MOE is calculated as the ratio between the reference dose (TWI) of the contaminant and the observed exposure to the contaminant. When MOE ≤ 1, this represents an exposure above the safe limits. On the opposite, when MOE >1, this represents an exposure within safe limits (EPA, 2014; WHO/FAO, 2009). This method was applied using exposure obtained from distribution of exposure method (disaggregated samples).

$$\text{MOE} = \frac{TWI}{EWI}$$

3.4.3. Risk Characterization

The risk characterization for non-carcinogenic risk using the aggregated method was done for the median, 75th, and 95th percentile using the Hazard Quotient index (HQ) compared to the TWI and PTWI for all participants, females, males, and two age groups (18-29 and above 30). For female only age 18-40 as child-bearing age. If the ratio HQ < 1, no risk, and if HQ ≥1 high risk.

$$\text{HQ} = \frac{EWI}{TWI \text{ or } PTWI}$$

3.5. Statistical Analysis

General Linear Model (ANOVA) was used for statistical analysis followed by Fisher comparisons test using the Minitab 20. TotHg concentrations in fish samples were statistically analyzed to compare the average Hg concentrations between the different 7 fresh and canned samples. TotHg concentrations in canned Tuna were analyzed based on medium, brand, and country of origin. General Linear Model and Fisher comparisons test were also used to determine if there was a significant association for the consumption per body weight a week and EWI between genders and age groups among the three population groups.

3.6. Principal Component Analysis (PCA) Analysis of Total Mercury

Multivariate analysis is a data set that contains several quantitative variables and aims to reduce the data variation without the loss of any information. PCA is a multivariate analysis method used for exploratory data by creating predictive models that identify the most influential variable to reduce the data dimensions (Ashfaq, 2019). The redundancy in the totHg concentration was reduced by using multivariate analysis (PCA). This is done by clustering the samples into groups based on the same variation characteristics and differences between them, according to the sample species and totHg concentration. PCA was used to investigate the influences of the main fish content (lipid, protein, and moisture) on the increase and decrease of totHg in different fish species. Moreover, PCA was used to investigate the influences of EWI and consumption per body weight for each fish species and type on MeHg concentration.

CHAPTER 4: RESULTS AND DISCUSSION

4.1. Questionnaire Analysis

4.1.1. Population Descriptive Statistics

A total of 619 responses were received, 600 were used in this study since they provide complete responses. A total of 35 nationalities participated in the questionnaire. The Qatari population contribution 56%, Jordanian 7%, Palestinian 6%, Egyptian 5%, and 26% from 35 different nationalities. Female gender represented was 71.1% and male gender represented was 28.9% of the total sample of participants. Participants' age ranged between was 18- 70 for females and 18-71 for males. The average body weight of the participants was 71.7± 17.1 kg for females and 89.2± 21.2 kg for males. The body mass index (BMI) was within the obese range i.e. BMI>30 for 31.9% of the participants. High protein diets were consumed by 19.6% of the total population, with 25.9% from the male population and 17.1% from the female population. Among female participants 4.5% were pregnant and 4.2% were breastfeeding, (Table 11). The main places from where participants reported buying their fish were as follows: Supermarket 53.3%, fish market 25.9%, supermarket & fish market 9.3%, fishermen 3.8%, and the remaining 7.5% from different sources like self-fishing, online, and more than one sources. Noting that some of those who reported buying from self-fishing, online, or any other source bought also from supermarket and fish market (Figure 17).

Table 11. The Characteristics of the Participants

Characteristics	All	Female	Male
Total	600	426	174
Qatari population	336	251	86
Non- Qatari population	264	176	88
Average age ± SD	35.8 ± 10.9	35.0 ± 10.5	37.7± 11.6
Average body weight (kg) ± SD	76.8 ± 20	71.7 ± 17.1	89.4 ± 21.2
Average height (cm) ± SD	164.9± 10	160.3± 6.2	175.7 ± 8.8
Average BMI± SD	28 ± 6.5	28 ± 6.3	29 ± 6.9
High diet	118	73	45
Pregnant	19	19	-
Breastfeeding	18	18	-

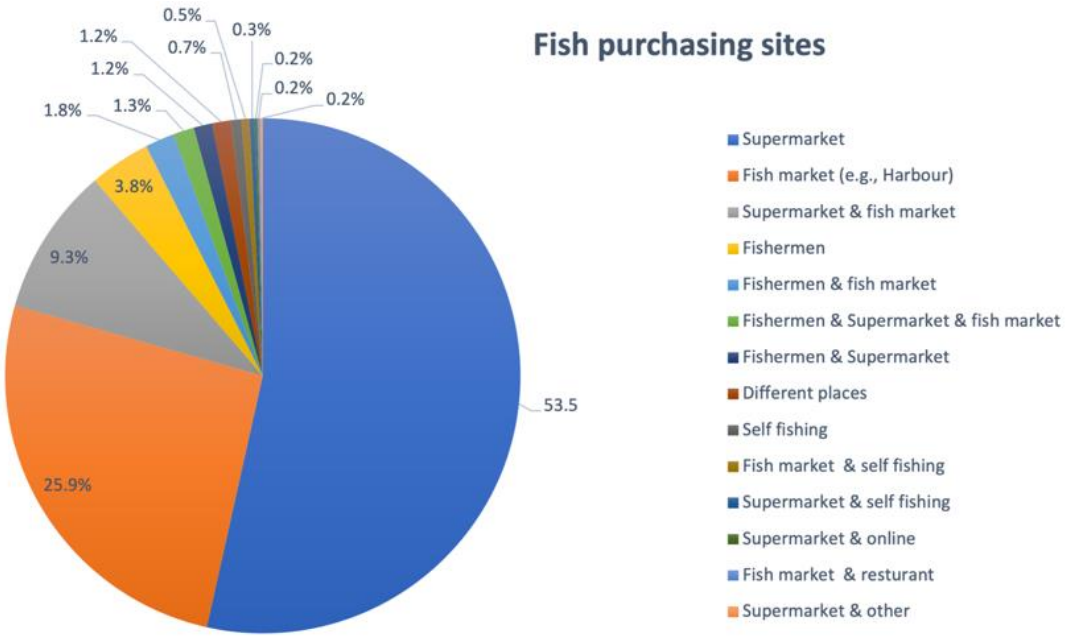


Figure 17. The percentage of the fish purchasing sites

4.1.2. Sampling

Based on the questionnaire results and using a cut-off value of 93%, seven fish species were included in the sampling plan. These were Hamour, Safi, Chanad, Sha'ri, Tuna, Salmon, and Sea bass. The samples were purchased from the five main supermarkets available in Doha, and one fish marketplace. The supermarkets were visited on different days to collect the samples. A total of 65 fish samples for the seven specie as presented in table 12 below were purchased. The canned fish portion was based on the average drained tissue. Canned Salmon was 150 g, and canned tuna was 120 g. The canned Tuna samples of four leading brands were purchased from the local supermarkets in Qatar. The mediums were as the following: 4 in brine, 7 in sunflower oil, 1 canola oil, 3 olive oil, and 1 extra olive oil. The canned Salmon samples were from two brands The mediums were as the following: 2 in brine and 1 in vegetable oil.

Table 12. Characteristics of Fish Samples Purchased From Different Locations in Qatar

Type	Common name - Scientific name	Trophic level	Local or imported	Wiled or farmed	Whole or fillet	Average fish or fillet weight (g)	Number of samples	Locations
Fresh	Hamour- *Epinephelus coioides*	Carnivores	Local	Wiled	Whole	1405.0	3	Fish market
							3	Supermarket A
			Imported	Unknown	Fillet	374.0	3	Supermarket B
	Safi- *Siganus rivulatus*	Herbivores	Local	Wiled	Whole	216.7	3	Fish market
							3	Supermarket A
	Chanaad- *Scomberomorus commerson*	Carnivores	Local	Wiled	Whole	1450.2	3	Fish market
							3	Supermarket A
	Sha'ri- *Lethrinus nebulosus*	Carnivores (non-predatory)	Local	Wiled	Whole	775.0	3	Supermarket B
							3	Supermarket C
	Tuna (local: *Euthynnus affinis)*	Carnivores	Local	Wiled	Whole	1443.3	3	Supermarket B
			Imported	Unknown	Fillet	164.7	3	Supermarket D
	Salmon- unknown	Carnivores	Imported	Farmed	Fillet	327.8	3	Supermarket A
							3	Supermarket D
	Sea bass- *Dicentrarchus labrax*	Carnivores	Imported	Farmed	Whole	507.9	3	Fish market
							3	Supermarket D
Frozen	Salmon- *Salmo salar*	Carnivores	Imported	Unknown	Fillet	498.3	1	Supermarket B
Canned	Tuna-different species	Carnivores-	Imported	Unknown	-	-	16	Supermarket A & B
	Salmon- unknown	Carnivores-	Imported	2 wiled & 1 unknown	-	-	3	Supermarket B & E
						Total	65	

4.1.3. Fish Consumption

The main fish species that were reported to be consumed by the participants are shown in Figure 18. The most consumed species were Hamour 17%, followed by Safi 14%, Chanad 14%, Sha'ri 11%, Tuna 15%, Salmon 12%, and Sea bass 10%. Anchovy and Sardines were reported to be consumed by less than 5% of the participants (Figure 18).

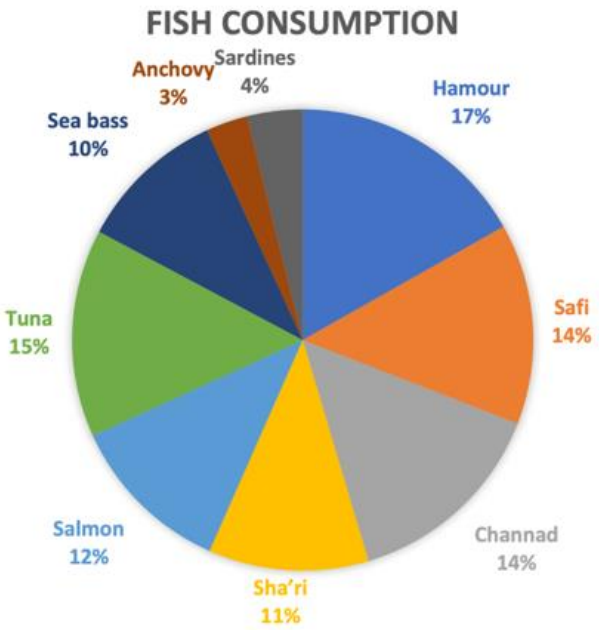


Figure 18. Fish consumption percentage

The average consumption for the general population was calculated as 1102± 1024 g/w, the Qatari population consumption average was 1185.63± 1005 g/w, and the non-Qatari were 1095±1042 g/w. This consumption yielded an average of fish consumption per capita per year of 61.82 kg per capita /year, which is significantly higher than the range 10- 20 kg per capita /year reported by the FAO (FAO, 2020; Sana, 2020). This difference may be explained by the difference in the method used to

estimate fish consumption. In fact, FAO estimates the consumption based on balance sheets and uses the whole population which might include fish consumers and non-consumers leading to a more flattened consumption.

Fish were mostly bought as fresh for all species (93%-98.5%) except for Tuna which was mostly bought as canned (93%). The only frozen fish found at the time of the sample collection was Salmon, this can indicate that the frozen fish is usually self-freezing, not store-bought frozen fish. The findings of this study agreed with (Sana, 2020) that the population preferred fresh over frozen fish and local over imported fish. For this reason, the frozen samples were summed with the fresh samples to represent consumption in grams per week.

The average weekly consumption for the species consumed in more than 5% of the participants is presented in Table 13. Among all species, Hamour had the highest average weekly consumption with 179.0± 187 g/w and Sea bass had the lowest 111.9± 182 g/w. When considering the different population groups, Safi had the highest consumption average in the Qatari population 232.9± 205 g/w, whereas Salmon had the highest consumption average in the non-Qatari population 183.0± 196 g/w.

Table 13. Fish Consumption per Week (CNP) in g/week ± SD and the Percentage of Consumers (C%) for the Selected Fish Species and the Three Population Groups.

Species	General population		Qatari population		Non-Qatari population	
	Fish CNP	C% (n= 600)	Fish CNP (g/w)	C% (n= 336)	Fish CNP	C% (n= 264)
Hamour	179.0± 187	83.0	198.4± 190	86.3	154.2± 181	78.8
Safi	178.7± 213	69.3	232.9± 205	87.5	109.8± 174	46.2
Chanad	174.3± 209	71.3	200.8± 204	82.4	140.5± 178	57.2
Sha'ri	127.0± 277	55.2	127.9± 341	52.1	125.9± 169	59.1
Tuna	158.1± 262	71.8	142.8± 271	68.5	178.1± 186	76.1
Salmon	173.0± 288	56.8	165.1± 288	54.2	183.0± 196	60.2
Sea bass	111.9± 182	51.3	118.1± 187	54.2	104.1± 190	47.7

The average fish consumption in grams per body weight a week is shown in table 14. The highest fish consumption was 16.72 g/kg w between age group 18-29 among the general population and it was the highest between 16.46 g/kg w females in the childbearing age (18-40). Moreover, fish consumption in females within childbearing age had the highest 75th percentiles value of 56.49 g/kg w. These high consumption rates observed in the females of the childbearing age can significantly contribute to the in utero exposure of the fetus to the contaminants contained in the fish, namely methyl mercury, which is a well-known neurotoxic agent (EFSA, 2012).

The average fish consumption in grams per body weight a week was the highest in age 18-29 and the highest in general population 16.72 g/ kg w and females 16.47 g/kg w. For childbearing age (18-40) the average consumption was 16.46 g/kg w and the highest 75th percentiles 56.49 g/kg w. This high consumption in females within the childbearing age can impose risk in case of pregnancy, especially since embryo and fetus are at higher risk for neurotoxicity (EFSA, 2012).

Table 14. The Average, Median, and 75th and 95th Percentile for Fish Consumption in Grams per Body Weight a Week (g/ bwt/ w) for the General Population

	Group	Average	Median	P75	P95
Female and male	All ages	15.14	11.28	17.98	42.31
	Age 18-29	15.85	10.93	17.67	55.58
	30 & above	14.82	11.38	18.27	36.24
Female	All ages	16.26	12.19	19.33	46.45
	Age 18-29	16.47	11.2	22.44	54.22
	30 & above	16.16	12.66	18.94	44.69
	Age18-40	16.46	11.67	18.94	56.49
Male	All ages	12.39	9.33	15.29	32.48
	Age 18-29	14.29	13.00	17.96	29.39
	30 & above	11.66	7.92	33.92	13.12

4.2. Samples Analysis

4.2.1. Total Hg (totHg) Contamination Levels

Table 15, and figures 19 & 20 present the total mercury concentration in the fish samples. TotHg concentrations in the tissues of the 65 samples were determined by ICP-MS, after acid digestion in an ultraWAVE microwave digestion system, as it was described in Section 3.3.3. TotHg was detected in all fish samples. TotHg concentrations ranged between 0.001 mg/kg ww in Safi and 0.443 mg/kg ww in imported Hamour and corresponding to 0.0041-2.2892 mg/kg dw, respectively. Statistically significant differences in average levels of totHg between the 7 fish species was observed. Results were considered statistically significant at $p<0.05$. The Fisher comparisons test was used to determine if there was a significant association between imported Hamour, local Hamour, Safi, Chanad, Sha'ri, local Tuna, imported Tuna, canned Tuna, Salmon, canned Salmon, and Sea bass. There was significant different in totHg concentration between imported Hamour, local Hamour, Safi,

Chanad, canned Tuna, and Salmon. The totHg concentration in local Tuna wasn't significantly different from Sha'ri and canned Salmon. There wasn't significant different in totHg concentration between local Tuna or between imported Tuna, canned Salmon, and Sea bass.

Imported Hamour had the highest average totHg concentration with a value of 0.4064± 0.04 mg/kg ww, while the lowest values were measured in Safi with 0.0012± 0.0002 mg/kg ww. Subsequently, arranging from highest to the lowest totHg concentrations, ranked as follows: imported Hamour > local Hamour > Tuna canned > Chanad > Sha'ri > local Tuna > Salmon canned > Sea bass >Salmon > Safi. When the average totHg concentrations were compared to the EPA Criterion level in fish for totHg of 0.3 mg/kg ww only imported Hamour exceeded the level (EPA, 2001a).

Table 15. Average totHg Concentration in Fish Species in (mg/Kg ww) ± SD

Fish	Dry	Wet	Moisture ratio range
Hamour (local)	0.670 ± 0.25	0.158^{b} ± 0.06	0.23-0.25
Hamour (imported)	2.109 ± 0.18	0.406^{a} ± 0.04	0.19
Safi	0.005 ± 0.001	0.001^{e} ± 0.0002	0.22-0.24
Chanad	0.332 ± 0.14	0.091^{c} ± 0.04	0.27-0.28
Sha'ri	0.278 ± 0.06	$0.063^{c,d}$ ± 0.01	0.19-0.26
Tuna (local)	0.132 ± 0.05	$0.040^{c,d,e}$ ± 0.01	0.29-0.33
Tuna (imported)	0.085 ± 0.004	$0.018^{d,e}$ ± 0.002	0.20-0.22
Tuna (canned)	0.273 ± 0.2	0.091^{c} ± 0.07	0.25-0.41
Salmon	0.024 ± 0.01	0.008^{e} ± 0.004	0.31-0.38
Salmon (canned)	0.070 ± 0.04	$0.023^{d,e}$ ± 0.01	0.31-0.36
Sea bass	0.055 ± 0.02	$0.021^{d,e}$ ± 0.007	0.35-0.47

Means that do not share a letter are significantly different.

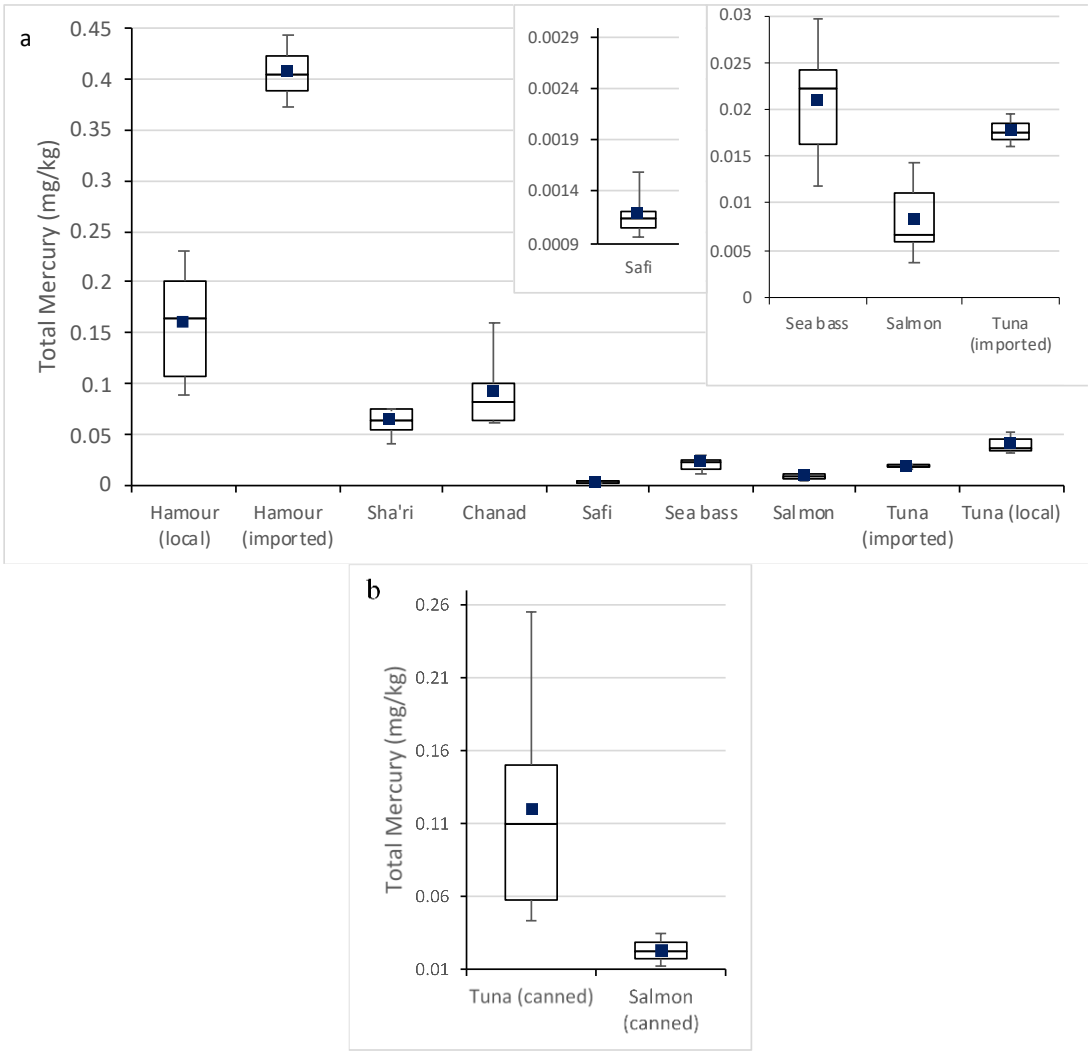


Figure 19. Box-and-whisker plot totHg concentrations (mg/kg ww) for seven fish species a) the concentration of totHg in the fresh samples; and b) the concentration totHg in the canned samples. The two interquartile boxes represent the Q1 & Q3 separated by the median, the blue squares near the middle of the box are the average values, the whisker represents the error bars, and the black circles are outliers

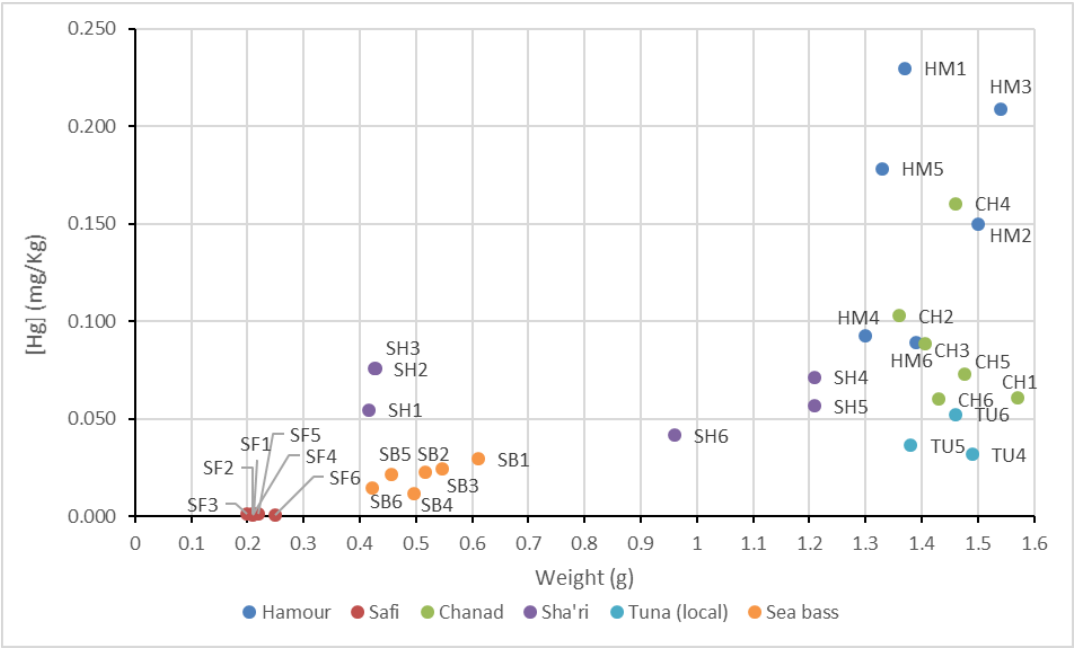


Figure 20. The concentration (mg/kg ww) of totHg in local Hamour, Safi, Chaanad, Sha'ri, local Tuna, and Sea bass as a function of the wet weight of the fish in g.

Hamour is significantly the highest consumed fish with the highest consumption level 184.2 g/w and the highest totHg concentration ranging between 0.089- 0.443 mg/kg ww. The totHg concentration in imported Hamour samples was significantly higher than the local, 0.372- 0.443 mg/kg ww and 0.089- 0.230 mg/kg ww, respectively. TotHg concentration ranges in Chanad, Sha'ri, and Sea bass, 0.060 - 0.161 mg/kg ww, 0.042- 0.076 mg/kg ww, and 0.012- 0.03 mg/kg ww, respectively. Hamour and Sha'ri are carnivores and demersal species, however, Hamour is predatory species that feed on small fish and crustaceans. In the other hand, Sha'ri is non- predatory species that feed on mollusks, echinoderms, and crustaceans (De Mora, 2004). Lower totHg concentration levels were found in Hamour samples in (Freije, 2009) in comparison to the local and imported Hamour samples from our study 0.110 mg/kg ww with a maximum value of 0.137 mg/kg ww. The totHg from Hamour samples from the Iranian market contained higher Hg level during winter 0.489 mg/kg ww and lower Hg level

during summer 0.317 mg/kg ww (Saei-Dehkordi, 2010). The MeHg% were 97.3% of the totHg with an average of 0.107 mg/kg ww and a maximum value of 0.126 mg/kg ww. Hamour samples from the South China Sea also had a lower totHg level of 0.056 mg/kg ww (Chen, 2018). In compare to our totHg 0.6701 in mg/kg for dry weight for local Hamour samples, similarly to higher concentrations were found by (De Mora, 2004) in samples from Qatar, Bahrain, UAE, and Oman were 0.97- 1.04 mg/kg dw, 0.67 -0.82 mg/kg dw, 1.62 -2.35 mg/kg dw, and 0.517-0.522 mg/kg dw, respectively. (De Mora, 2004) for Sha'ri samples from Qatar were close to our average in dry weight 0.343 mg/kg dw, in contrary UAE samples had a higher totHg range between 0.45-0.51 mg/kg dw. (Al-Ansari, 2017) totHg levels in Sha'ri were significantly higher ranging between 0.181- 0.508 mg/kg ww. The MeHg% in Sha'ri from (Freije, 2009) was 94.1%, however, the totHg concentration was lower than our average and ranged between 0.030–0.043 mg/kg ww.

Chanad is carnivore predatory species that feed in the epipelagic zone on small fish (Freije, 2009). Higher levels of Hg were found in Bahrain 0.126 mg/kg ww ranging between 0.117 – 0.137 mg/kg ww (Freije, 2009), in Iran 0.307 mg/kg ww (Saei-Dehkordi, 2010), in Kuwait 0.370 mg/kg ww (Laird, 2017), in Malaysia 0.061- 0.132 mg/kg ww (Anual, 2018), and in USA 0.446 mg/kg ww (Li, 2022). The MeHg% in (Freije, 2009) study was 97.6% with an average of 0.123 mg/kg ww. The MeHg% in (Laird, 2017) study was 62.1% with an average of 0.23 mg/kg ww. The MeHg% in (Ahmad, 2021) was 92.9% with an average 0.0838 mg/kg ww.

Safi had significantly the highest consumption level in the Qatari population 233.57 g/w. Contrary to Hamour, Safi had the lowest totHg concentration with small variation ranging between 0.0009- 0.0012 mg/kg ww. The trophic level can play an important role in the Hg concentration. Safi is a non- predatory herbivore that feeds

on algae and sea grasses which make this species at a lower trophic level. Those characteristics explain the significant low totHg concentration (Oksuz, 2010; Soykan, 2020). This species is part of the Siganus Genus, Siganus canaliculatus is species with similar characteristics to Safi became a target for farmed fish development in China because of its high nutritional value and significant low Hg level (X. Wang, 2020). Similarly to our results (Hakami, 2016) reported that totHg contents were 0.02 mg/kg ww. Lower levels of Hg were found in (Burger, 2014b) study with totHg and MeHg average concentration 0.002 mg/kg ww and 0.0019 mg/kg ww and ranging between ND - 0.004 mg/kg ww and 0.0012 − 0.0035 mg/kg ww, respectively.

Fresh Tuna samples totHg concentrations were significantly higher in local Tuna ranging between 0.032- 0.052 mg/kg ww, compared to 0.015-0.02 mg/kg ww in imported samples. However, the concentrations from our study are significantly low compared to different Tuna species from different studies. The local Tuna or mackerel tuna (*Euthynnus affinis*) is migratory species that migrate from the warm Indo-Pacific region. It is an epipelagic and apex predatory species that feed on small fish, mollusks, and crustaceans (Taghavi Motlagh, 2010; Vigneshwaran, 2018). Significantly higher totHg range was in Mackerel tuna from Malaysia ranging between 0.084- 0.132 mg/kg ww for small size sample (length 36–69 cm) (Anual, 2018). The imported Tuna species is unknown red meat species that can include different species like yellowfin tuna (*Thunnus albacares*), skipjack tuna (*Katsuwonus pelamis*), and a (*Euthynnus affinis*). The average totHg and MeHg concentrations in yellowfin tuna from South Africa ranged between 0.45 - 1.52 mg/kg ww and 0.23 - 1.24 mg/kg ww, respectively. The significantly high Hg level in this study can be related to the weight of the samples ranging between 25- 80 kg (Bosch, 2016). Yellowfin tuna from Sri Lanka with a weight between 25.5–91.6 kg had totHg concentration between ND-1.6 mg/kg ww, 5 samples

out of 65 were below the detection limit of 0.07 mg/kg ww. Yellowfin tuna from the North Pacific Ocean had the highest totHg concentration 0.602 mg/kg ww and the lowest was from the Northwest Pacific Ocean 0.064 mg/kg ww. Yellowfin tuna from the Indian Ocean was 0.245 mg/kg ww with a MeHg percentage was 96.3%. The weight of the fish samples in this study ranged between 10.1-186 kg, the fish did not correlate with the weight and length but with capturing site (Nicklisch, 2017). The study stated that there is a significant positive correlation between totHg concentration and weight (Jinadasa, 2019). In skipjack tuna, the average totHg concentration in the white muscle was 0.115 mg/kg ww compared to the dark muscle 0.124 mg/kg ww (Vieira, 2017). Lower levels of totHg were found in (Torres, 2016) with an average of 0.04 mg/kg ww. The totHg level in Skipjack tuna is mostly lower than other Tuna species because it mainly feeds on invertebrates (Ormaza-González, 2020). TotHg concentration in skipjack tuna and mackerel tuna from the red sea were 0.318 and 0.169 mg/kg ww, respectively (Al-Najjar, 2019). Tuna samples from Indonesia, Japan, and the Marshall Islands contained 0.3174, 0.182, and 0.0597 mg/kg ww, respectively (Nong, 2021) (Milatou, 2020) compared Hg level in different Tuna species from the Mediterranean Sea, Atlantic Ocean, Pacific Ocean, and the Indian Ocean. It was found that the wild bluefin Tuna from the Mediterranean Sea had a lower Hg level compared to open Oceans. Since Tuna is a group of migrated species the geographic nursery and migration can be significant to Hg levels.

(Vieira, 2017) studied the effect of the canning process on Skipjack tuna the average totHg concentration in the white muscle and dark muscle and the totHg increased by 16.5 and 25%, respectively. However, no canned sample from our study exceeded the EPA Criterion level. The canned Tuna samples had a wide range totHg concentrations ranging between 0.009 – 0.255 mg/kg ww. Our study on canned Tuna

was done on 16 samples from 5 country of origin, 6 brands, and 4 preservative mediums (brine, sunflower oil, olive oil, and canola oil). According to the FDA, canned white Tuna is mainly albacore Tuna that usually grow to large size and light Tuna is skipjack, a mix of Tuna species, or mainly small size Tuna species. This means that light Tuna is the safer choice (FDA, 2021). However, our study showed no significant difference in totHg concentration between albacore, skipjack, or light Tuna ($p > 0.05$). Also, there was no significant difference in totHg concentration between the brands or the main 3 medium ($p > 0.05$), see table 16. In this study, similar non-significant differences between brine, sunflower oil, and olive oil medium or the brands that were obtained from canned Tuna samples from Spain. The highest and the lowest totHg concentration were preserved in olive oil (González-Estecha, 2013). TotHg concentration Tuna preserved in vegetable oil from Poland 0.0369 mg/kg ww (Kowalska, 2020). The average totHg concentration from 5 different brands preserved in oil and water showed no significant differences with 0.169 and 0.173 mg/kg ww, respectively (de Paiva, 2017). The only significant differences were between the species ($p=0.004$) skipjack, white Tuna, and light Tuna had totHg concentration ranging between 0.299 - 0.322 mg/kg ww, 0.225- 0.965 mg/kg ww, and 0.03- 1.176 mg/kg ww, respectively (González-Estecha, 2013). The only significant difference in totHg concentration from our study in canned Tuna samples from our study was observed between country A and the other 4 countries ($p < 0.05$). The Fisher comparisons test showed significant different in totHg concentration between country of origin "A" and the other countries companied. According to the Thai Fish Inspection and Quality Control Division released in 2011, the maximum Hg level in the imported canned Tuna fish to KSA should not exceed 0.5 mg/kg (Fish Inspection and Quality Control Division, 2011). The average totHg concentration in canned albacares and tonggol (mixed) preserved in oil

(not significant) from 2 different brands from Iran ranged between 0.024- 0.0394 mg/kg ww (Mansouri, 2021). Culture bluefin tuna that were feed small fish that contain low totHg levels, raised in a narrow cage, and in Hg controlled water showed no increase in Hg concentration per body weight even with the increase in body weight. This is because the rate of Hg extraction from the body through faces was faster than the accumulation rate and this can be an explanation for the low Hg levels in the imported and canned Tuna samples (Nakao, 2007). The totHg and MeHg analysis from canned Tuna in Brazil estimated that MeHg% ranged between 82- 99% (de Paiva, 2017).

Table 16. TotHg Concentration in (mg/Kg ww) From Different Canned Tuna Samples From Different Brand, Country of Origin, and Preservative Medium

Country of origin	**Brand**	**Medium** Species totHg concentration							
		sunflower oil		**Brine**		**olive oil**		**Canola oil**	
A	**1**	Tonggol 0.069	Light meat 0.009	Tonggol 0.071	Light meat 0.033				
	2	Albacore 0.060	Light meat 0.040			Albacore 0.015			
		Skipjack 0.059							
B	**3**	Skipjack 0.129				Skipjack 0.123			
C	**4**	Light meat 0.110		Light meat 0.206					
D	**5**			*Light meat 0.072		Light meat 0.083	**Light meat 0.126		
E	**6**							no species identification 0.255	

*From country A
**Extra olive oil

TotHg concentration in fresh Salmon ranged between 0.004- 0.014 mg/kg ww. In general, Salmon is considered to contain low Hg levels and is stated among the best choices category by the EPA and FDA (USDA, 2021). Higher levels of totHg were found in fresh samples from the USA market from Alaska and unknown sources in Salmon samples without skin were 4 times greater in than Salmon with skin, 0.157 mg/kg ww and 0.0437 mg/kg ww, respectively (Li, 2022). TotHg concentrations in Salmon samples from Denmark and Chile were 0.0287, and 0.004 mg/kg ww, respectively (Nong, 2021). TotHg concentration in Norwegian Salmon were 0.006 mg/kg ww (Nong, 2021) and 0.0098 mg/kg ww (Panichev, 2015). TotHg concentration in Norwegian Salmon 0.0098 mg/kg ww was significantly lower than the Canadian Salmon 0.109 mg/kg ww (Panichev, 2015). TotHg in fresh farmed Salmon were significantly lower than in wild Salmon from Spain ($P < 0.05$), 0.011 mg/kg ww and 0.048 mg/kg ww, respectively. This is as a result of the controlled growing conditions for the farmed fish (Fernández-Bautista, 2022). MeHg in Salmon samples from Portugal made 80% of totHg, the totHg and MeHg concentration was 0.028 mg/kg ww and 0.023 mg/kg ww, respectively (S. Costa, 2015).

TotHg concentration in canned Salmon samples from our study ranged between 0.012- 0.035 mg/kg ww. Our canned Salmon samples were from 2 brands and 2 different countries. Two samples from brand 1 were wild pacific Salmon preserved in brine and had a concentration of totHg for pink Salmon 0.022 mg/kg ww and red Salmon 0.035 mg/kg ww. The sample from brand 2 was preserved in vegetable oil without species identification had a concentration of totHg 0.012 mg/kg ww. The difference in totHg concentration is related to the Salmon feeding habits. Pink Salmon feeds mainly on zooplankton and red Salmon or sockeye feeds on zooplankton, fish eggs, and small fish (Qin, 2016). Similarly, (Panichev, 2015) reported that totHg

concentration of canned pink Salmon was 0.048 mg/kg ww. Contrary to our results, the totHg concentration in canned red Salmon was slightly lower than in canned pink Salmon, 0.0328 and 0.0361 mg/kg ww, respectively (Ikem, 2005).

Sea bass is a demersal predatory freshwater fish that feeds on shrimps, mollusks, and small fish (Maulvault, 2016). In this study, the totHg concentration in Sea bass in dry weight ranged between 0.025- 0.077 mg/kg dw. Similarly, (Mieiro, 2011) reported that the range on totHg concentration from Portugal 0.04-0.46 mg/kg dw. Sea bass samples from Greece had a higher totHg concentration. The samples were obtained from different aquaculture sites with an average range between 0.034- 0.105 mg/kg ww (Renieri, 2019).In this study, a similar totHg concentration of 0.025 mg/kg ww from Turkey was observed (Bat, 2022).

Differences in Hg levels within the same species from different regions can be associated with different levels of Hg contamination from the fishing sites. The differences between the data obtained from our study and the previous studies are mainly due to different pollution levels, and other biotic factors like age and gender (da Silva, 2020; Elsayed, 2020).

4.2.2. Methyl Mercury (MeHg) Contamination Levels

The concentration of MeHg from the two scenarios are presented in table 17. The average MeHg concentration form scenario 1 was 0.077± 0.09 mg/kg ww and the average MeHg concentration form scenario 2 was 0.072± 0.09 mg/kg ww.

Table 17. Average MeHg Concentration in Fish Species in (mg/Kg ww) for the Two Applied Scenarios

Fish	MeHg 100%	MeHg ART
Hamour (local)	0.158	0.154
Hamour (imported)	0.406	0.395
Safi	0.001	0.001
Chanad	0.091	0.089
Sha'ri	0.063	0.059
Tuna (local)	0.040	0.038
Tuna (imported)	0.018	0.017
Tuna (canned)	0.091	0.083
Salmon	0.008	0.007
Salmon (canned)	0.023	0.019
Sea bass	0.021	0.017

The majority of commercially available fish species in Qatar are considered to have low Hg levels. Only the imported Hamour exceeded the EPA criterion level and all the samples were below the FAO/WHO and EU Commission guideline for MeHg concentration in predatory and non- predatory fish species even if the MeHg% were 100% of totHg (EU Commission, 2006; FAO/WHO, 2007). Our findings explain that the most consumed fish species in Qatar might have a low risk of MeHg. However, the exposure in our study was evaluated with caution because the contamination levels can have divergent results from time to time based on the fish age, body-size parameter, and capturing location and season.

4.3. Exposure Assessment to Methyl Mercury

The EWI can be calculated by different calculation method. (Alva, 2020) presented the calculation for individual respond or for group of people sharing similar characteristics. In our study the estimated weekly intake EWI (median, average, and 75^{th} and 90^{th} percentile) for general population were calculated in two calculation

methods distribution of exposure and aggregated method. The distribution of exposure was calculated by measuring the exposure (µg/w) for each induvial respond using totHg contamination level (µg/g) for each fish:

$$\mathrm{EWI}(\frac{\mu g}{kg}) = \sum_{n=0}^{n} \left(\frac{\mathrm{CNP} \times \mathrm{C}}{bwt}\right)_n$$

The aggregated method by using the average totHg contamination level as a constant with the change of consumption per body weight.

$$\mathrm{EWI}(\frac{\mu g}{kg}) = \left(\sum_{n=0}^{n} \left(\frac{\mathrm{CNP}}{bwt}\right)_n\right) \times \overline{C}$$

CNP: fish consumption (g/w), body weight: bwt, and C: MeHg contamination level for each fish & $\overline{C}$: average MeHg contamination level in (µg/g)

The average consumption per body weight were calculated for each respond individually. The aggregated method takes account to the variability that exists in fish consumption patterns and to provide data on the exposure levels distribution within the studied population. The aggregated method resulted in slightly higher average EWI values for the three population groups. The percentage change was 32%, 21%, and 19%, for general population, Qatari, and non-Qatari, respectively. This indicate that the risk is from high fish consumption not from high level of contamination.

The estimated weekly intake (EWI) of MeHg for the three population groups from Scenario 1 is presented in table 18 for the distribution of exposure and table 19 for the aggregated method. The distribution of exposure method highest 95th and 75th percentiles were 4.05 µg/kg w and 1.57 µg/kg w for the non-Qatari females aged 18-29, respectively. The highest median and average were 0.82 µg/kg w and 1.10 µg/kg w for Qatari females aged 18-29, respectively. The aggregated method highest 95th and

75th percentiles were 4.55 µg/kg w for Qatari females at childbearing age and 1.67 µg/kg w for Qatari females aged 30 & above, respectively. The highest median and average were 1.08 µg/kg w for Qatari males aged 18-29 and 1.33 µg/kg w for Qatari females at childbearing age, respectively.

The estimated weekly intake (EWI) of MeHg for the three population groups from Scenario 2 is presented in table 20 for the distribution of exposure and table 21 for the aggregated method. The distribution of exposure method highest 95th and 75th percentiles were 3.92 µg/kg w and 1.51 µg/kg w for the non-Qatari females aged 18-29, respectively. The highest median was 0.78 µg/kg w among Qatari females and males aged 18-29 and the highest average was 1.05 µg/kg for Qatari females aged 18-29. The aggregated method highest 95th and 75th percentiles were 4.29 µg/kg w for Qatari females at childbearing age and 1.57 µg/kg w for Qatari females aged 30 & above, respectively. The highest median and average were 1.02 µg/kg w for Qatari males aged 18-29 and 1.25 µg/kg w for Qatari females at childbearing age.

The percentage of Females (23.94%) exceeding the TWI limit was higher than males (14.94%) and it was the highest among the females in the Qatari population (25.5%) . The number and percentage of females exceeding the TWI limit were not significant that are pregnant (5- 1.17%) or breastfeeding (4 – 0.94%). The number and percentage of respondents for people on a high protein diet and who exceeded the TWI limit were higher in Qatari males (9.3%), see table 22.

Table 18. Scenario 1 EWIs Using the Distribution of Exposure Method in µg/kg w

Groups		General population				Qatari				Non- Qatari			
		P75	P95	Median	Average	P75	P95	Median	Average	P75	P95	Median	Average
Females & males	All ages	1.20	2.81	0.73	0.97	1.26	3.06	0.79	1.02	1.05	2.50	0.63	0.90
	Age 18-29	1.20	3.12	0.74	1.00	1.23	3.12	0.81	1.06	1.29	2.81	0.66	0.99
	30 & above	1.22	2.59	0.72	0.95	1.28	3.08	0.76	1.01	1.00	2.42	0.62	0.86
Female	All ages	1.27	3.03	0.75	1.02	1.32	3.24	0.81	1.08	1.14	2.81	0.68	0.95
	Age 18-29	1.38	3.17	0.76	1.07	1.31	3.22	0.82	1.10	1.57	4.05	0.65	1.04
	30 & above	1.25	2.97	0.74	1.00	1.33	3.41	0.80	1.07	1.07	2.33	0.70	0.90
	Age 18-40	1.25	3.19	0.75	1.04	1.31	3.47	0.80	1.09	1.15	2.81	0.67	0.98
Male	All ages	0.94	2.12	0.60	0.83	0.98	2.09	0.76	0.88	0.93	2.36	0.57	0.79
	Age 18-29	1.12	2.02	0.81	0.91	1.11	3.19	0.81	0.97	1.16	1.97	0.77	0.81
	30 & above	0.91	2.38	0.57	0.80	0.92	2.18	0.57	0.83	0.89	2.49	0.53	0.78

Table 19. Scenario 1 EWIs Using the Aggregated Method in µg/kg w

Groups		General population				Qatari				Non- Qatari			
		P75	P95	Median	Average	P75	P95	Median	Average	P75	P95	Median	Average
Females & males	All ages	1.38	3.24	0.86	1.16	1.45	3.27	0.97	1.23	1.20	3.29	0.75	1.07
	Age 18-29	1.35	4.26	0.84	1.22	1.48	3.94	1.00	1.26	1.33	3.85	0.71	1.17
	30 & above	1.40	2.78	0.87	1.14	1.45	3.23	0.95	1.22	1.15	2.74	0.76	1.03
Female	All ages	1.48	3.56	0.93	1.25	1.64	3.91	1.03	1.30	1.28	3.60	0.78	1.17
	Age 18-29	1.57	4.16	0.86	1.26	1.60	4.34	0.97	1.29	1.25	4.12	0.64	1.23
	30 & above	1.45	3.43	0.97	1.24	1.67	3.86	1.07	1.30	1.33	3.32	0.86	1.14
	Age 18-40	1.45	4.33	0.89	1.26	1.57	4.55	0.99	1.33	1.22	3.89	0.71	1.18
Male	All ages	1.17	2.49	0.72	0.95	1.32	2.81	0.81	1.03	0.99	2.45	0.64	0.87
	Age 18-29	1.38	2.25	1.00	1.10	1.37	3.21	1.08	1.17	1.38	2.43	0.79	0.99
	30 & above	1.01	2.60	0.61	0.89	1.22	3.00	0.63	0.96	0.94	2.53	0.60	0.84

Table 20. Scenario 2 EWIs Using the Distribution of Exposure Method in µg/kg w

Groups		General population				Qatari				Non- Qatari			
		P75	P95	Median	Average	P75	P95	Median	Average	P75	P95	Median	Average
Females & males	All ages	1.14	2.70	0.69	0.92	1.21	2.96	0.75	0.98	0.99	2.40	0.60	0.85
	Age 18-29	1.14	3.00	0.71	0.95	1.18	3.00	0.78	1.02	1.24	2.70	0.61	0.94
	30 & above	1.15	2.51	0.69	0.91	1.22	2.93	0.72	0.96	0.95	2.30	0.58	0.81
Female	All ages	1.21	2.92	0.72	0.98	1.27	3.08	0.77	1.03	1.09	2.70	0.64	0.90
	Age 18-29	1.31	3.03	0.73	1.02	1.25	3.07	0.78	1.05	1.51	3.92	0.61	0.99
	30 & above	1.19	2.83	0.70	0.95	1.27	3.24	0.77	1.02	1.02	2.23	0.66	0.86
	Age 18-40	1.20	3.05	0.72	0.99	1.25	3.27	0.76	1.03	1.09	2.70	0.64	0.93
Male	All ages	0.89	2.03	0.57	0.79	0.93	2.00	0.73	0.84	0.88	2.25	0.54	0.75
	Age 18-29	1.07	1.95	0.77	0.87	1.06	3.07	0.78	0.93	0.93	1.73	0.56	0.66
	30 & above	0.87	2.27	0.54	0.76	0.88	2.08	0.56	0.79	0.86	2.38	0.52	0.74

Table 21. Scenario 2 EWIs Using the Aggregated Method in µg/kg w

Groups		General population				Qatari				Non- Qatari			
		P75	P95	Median	Average	P75	P95	Median	Average	P75	P95	Median	Average
Females & males	All ages	1.30	3.05	0.81	1.09	1.37	3.08	0.91	1.16	1.13	3.09	0.71	1.01
	Age 18-29	1.28	4.01	0.79	1.14	1.40	3.71	0.94	1.18	1.25	3.63	0.67	1.11
	30 & above	1.32	2.62	0.82	1.07	1.37	3.04	0.89	1.15	1.08	2.58	0.72	0.97
Female	All ages	1.40	3.35	0.88	1.17	1.54	3.68	0.97	1.22	1.21	3.39	0.73	1.10
	Age 18-29	1.48	3.91	0.81	1.19	1.51	4.09	0.91	1.21	1.17	3.88	0.60	1.16
	30 & above	1.37	3.23	0.91	1.17	1.57	3.64	1.00	1.23	1.25	3.13	0.81	1.07
	Age 18-40	1.37	4.08	0.84	1.19	1.48	4.29	0.93	1.25	1.15	3.66	0.67	1.11
Male	All ages	1.10	2.34	0.67	0.89	1.24	2.65	0.76	0.97	0.94	2.30	0.60	0.82
	Age 18-29	1.30	2.12	0.94	1.03	1.29	3.02	1.02	1.10	1.30	2.29	0.75	0.93
	30 & above	0.95	2.45	0.57	0.84	1.14	2.83	0.59	0.91	0.89	2.38	0.57	0.79

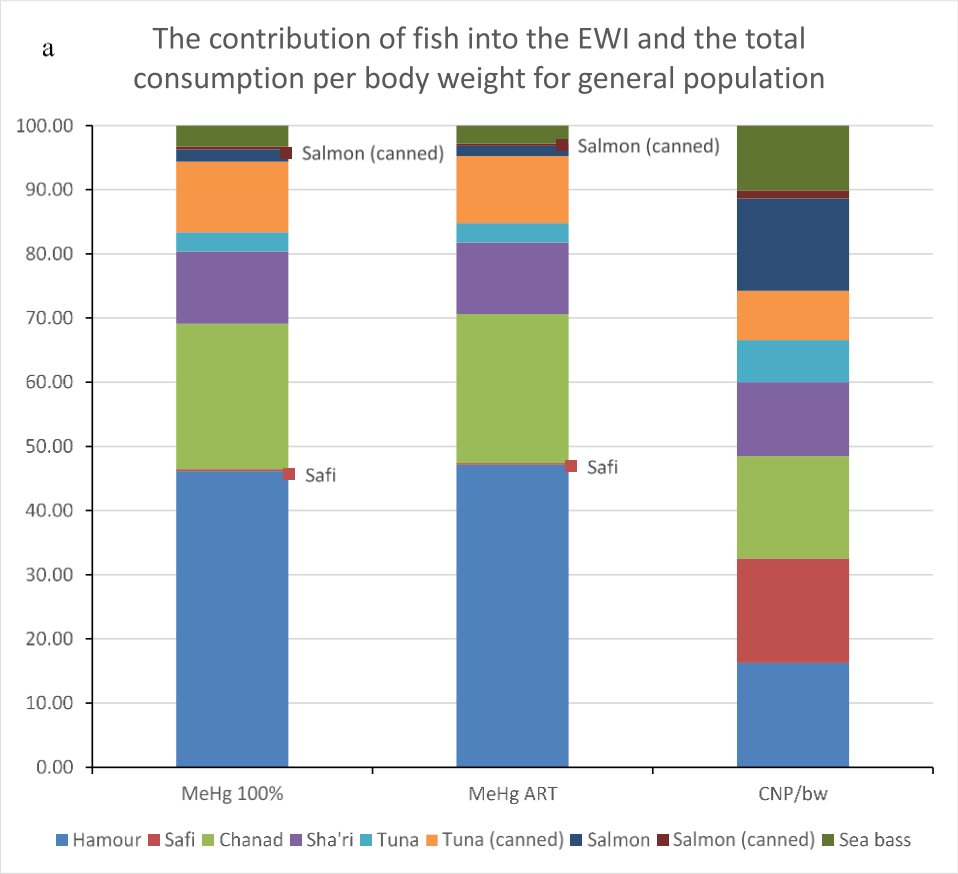
Table 22. The Percentage of Respondents in Which the EWI Exceeded TWI and the Percentage of the Population in High Protein Diet Exceeding TWI for Distribution of Exposure Method

Population	Gender	MeHg 100%	MeHg ART	On high protein diet
General population	All (n=600)	21.33	20.33	5
	Female (n=426)	23.94	22.54	4.23
	Male (n=174)	14.94	14.94	7.9
Qatari	All (n=336)	23.21	22.02	5.9
	Female (n=251)	25.50	23.90	4.78
	Male (n=86)	16.28	16.28	9.3
Non-Qatari	All (n=264)	18.94	18.18	3.8
	Female (n=176)	21.59	20.45	3.41
	Male (n=88)	13.64	13.64	4.55

There was significant variation ($p<0.05$) in consumption per body weight a week (CNP/kg) and EWI (distribution of exposure) in between genders for general population. The Fisher comparisons test confirmed the significant association between genders. The grouping information in Fisher comparisons for CNP/kg and EWI showed significant variations between females and males aged 30 & above, and between females aged 18-29 and males aged 30 & above. There were no significant variations in CNP/kg ($p>0.05$) for Qatari population, however, the grouping information in Fisher comparisons for CNP/kg for showed significant variations between females and males aged 30 & above.

Between one individual to another the fish choice and consumption will vary considerably and it is highly influenced by the culture, price, and availability in the market (Ikem, 2005). Therefore, the contribution of the different consumed fish species to the exposure of totHg would be variable among the population. The contribution of the studied fish to the exposure to totHg presented in figure 21 was found to be mainly influenced by the Hg contamination level of the fish. This was mainly confirmed for Safi and canned Salmon. Safi is among the highest consumed fish species whereas canned Salmon is the lowest consumed source. However, because of its low Hg level, the contribution of Safi to the exposure to totHg was lower than canned salmon. Similarly, fresh Salmon a had lower consumption rate than fresh Tuna. However, the higher Hg concentration in fresh Tuna made the contribution to the totHg exposure significantly higher. This observation shed light that the exposure to totHg is mainly driven by the consumption rather than the contamination level. Therefore, the higher the consumption of a species with high contamination levels, the higher the exposure to totHg is. This was confirmed for Hamour and Chanad which both contributed to 70%, 73%, and 65.7%, to the general population, Qatari, and non-Qatari, respectively.

Similarly, to our results Hamour was the highest consumed species in Kuwait and the main exposure to Hg contributor by 58% (Laird, 2017). It was also the highest consumable species in KSA among the Saudi and the expats, 72% and 60%, respectively (Burger, 2014a).



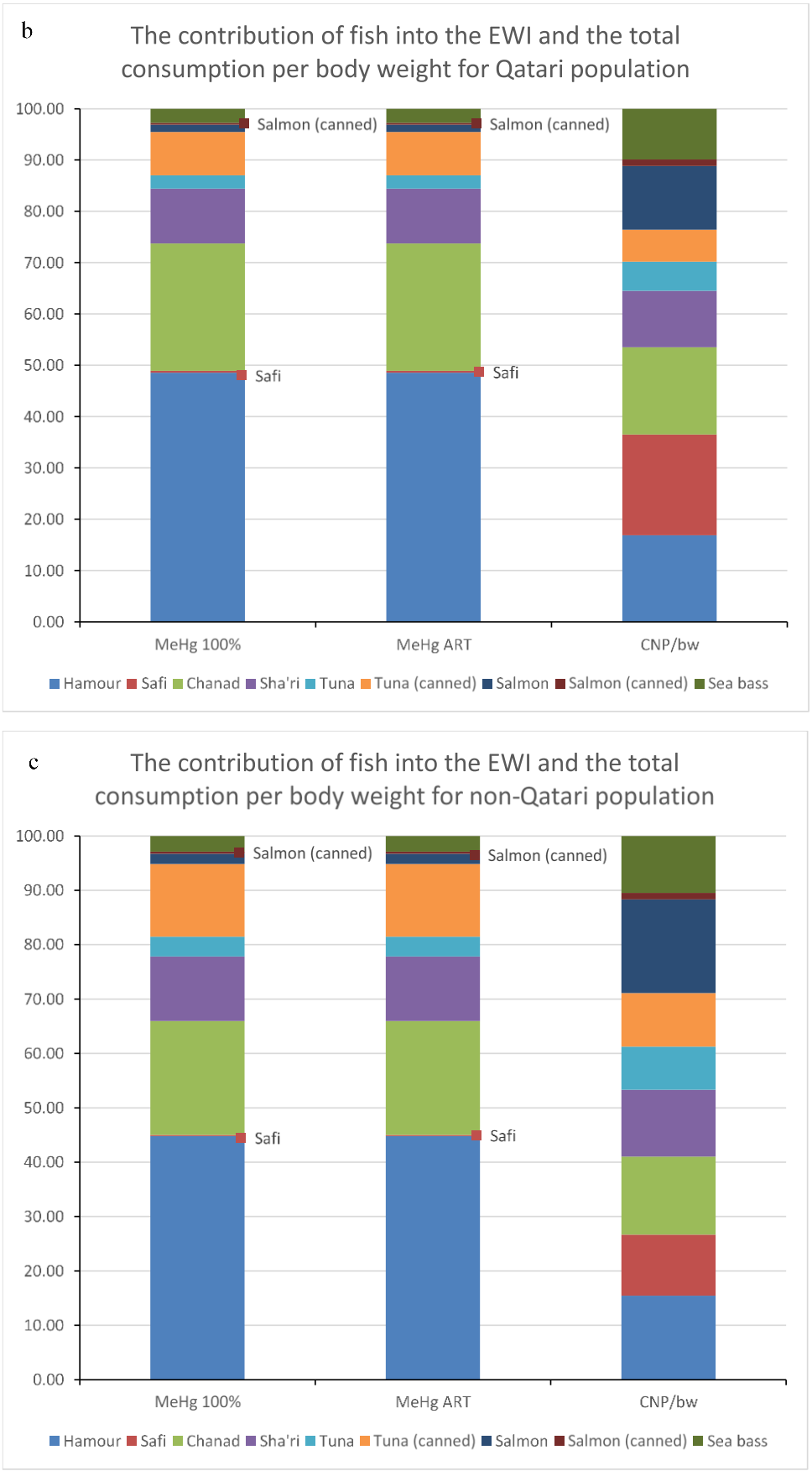


Figure 21. Fish contribution to EWI (µg/kg) for the two implemented scenarios and the consumption (g/kg w): a) general population, b) Qatari; and c) non-Qatari

Widely applied, TWI represents MeHg concentration that can be ingested over a lifetime without adverse health risks. Among the studied fish samples from scenario 1 as seen in table 23, the highest values calculated were EWI 0.49 μg/kg and TWI% 37.69% for Hamour among Qatari population. The highest total TWI% 78.82% among the Qatari population. Among the studied fish samples from scenario 2 as seen in table 24, the highest values calculated were EWI 0.47 μg/kg and TWI% 36.46% for Hamour among Qatari population. The highest total TWI% 75.09% among the Qatari population. Safi were always the lowest in EWI average and TWI%. The results were below the limits for MeHg intake recommended by EFSA.

Table 23. EWI Average of MeHg From Scenario 1 and TWI% of in the Studied Fish

MeHg 100%	General population		Qatari population		Non- Qatari population	
	Average	TWI%	Average	TWI%	Average	TWI%
Hamour	0.45	34.37	0.49	37.67	0.39	30.18
Safi	0.003	0.22	0.004	0.29	0.002	0.14
Chanad	0.22	16.88	0.25	19.10	0.18	14.05
Sha'ri	0.11	8.41	0.11	8.52	0.11	8.29
Tuna	0.03	2.22	0.03	2.02	0.03	2.47
Tuna (canned)	0.11	8.19	0.09	7.04	0.13	9.66
Salmon	0.02	1.41	0.02	1.29	0.02	1.56
Salmon (canned)	0.004	0.33	0.005	0.36	0.004	0.30
Sea bass	0.03	2.46	0.03	2.55	0.03	2.35
Total	0.97	74.50	1.02	78.82	0.90	68.99

Table 24. EWI Average of MeHg From Scenario 2 and TWI% of in the Studied Fish

MeHg ART	General population		Qatari population		Non- Qatari population	
	Average	TWI%	Average	TWI%	Average	TWI%
Hamour	0.42	32.49	0.47	36.46	0.38	29.37
Safi	0.003	0.21	0.003	0.27	0.002	0.13
Chanad	0.21	16.47	0.24	18.64	0.18	13.71
Sha'ri	0.10	7.92	0.10	8.01	0.10	7.80
Tuna	0.03	2.14	0.03	1.95	0.03	2.38
Tuna (canned)	0.10	7.41	0.08	6.37	0.11	8.74
Salmon	0.01	1.13	0.01	1.03	0.02	1.25
Salmon (canned)	0.003	0.27	0.004	0.29	0.003	0.24
Sea bass	0.03	2.00	0.03	2.07	0.02	1.90
Total	0.91	70.03	0.98	75.09	0.85	65.52

The margin of exposure (MOEs) representing above safe limits from both scenarios is presented in tables 25 and table 26. All MOEs for median and average consumers in the three-population group were below safe limits. All HQs for 95th percentile consumers in the three-population group were above safe limits.

Table 25. Margin of Exposure (MOE) for Scenario's 1 Average and 75th Percentile When MOE≤ 1

Groups		General population		Qatari population		Non- Qatari population	
		Average	P75	Average	P75	Average	P75
Female and male	All ages	-	-	-	-	-	-
	Age 18-29	-	-	-	-	-	-
	30 & above	-	-	-	-	-	-
Female	All ages	-	-	-	0.98	-	-
	Age 18-29	-	0.94	-	0.99	-	0.83
	30 & above	-	-	-	0.98	-	-
	Age 18-40	-	-	-	0.99	-	-
Male	All ages	-	-	-	-	-	-
	Age 18-29	-	-	-	-	-	-
	30 & above	-	-	-	-	-	-

Table 26. Margin of Exposure (MOE) for Scenario's 2 Average and 75th Percentile When MOE≤ 1

Groups		General population		Qatari population		Non- Qatari population	
		Average	P75	Average	P75	Average	P75
Female and male	All ages	-	-	-	-	-	-
	Age 18-29	-	-	-	-	-	-
	30 & above	-	-	-	-	-	-
Female	All ages	-	-	-	-	-	-
	Age 18-29	-	0.99	-	-	-	0.86
	30 & above	-	-	-	-	-	-
	Age 18-40	-	-	-	-	-	-
Male	All ages	-	-	-	-	-	-
	Age 18-29	-	-	-	-	-	-

The permissible levels of Hg in fish were implemented to calculate EWI and then compared to the TWI 1.3 µg/kg and PTWI 1.6 µg/kg limits to investigate if the exposure limits are adequate to consumption rate in Qatar as seen in figure 22 (EU Commission,

2006; FAO/WHO, 2007). The implementation of the (FAO/WHO, 2007) MeHg limits in predatory and non- predatory levels, 1.0 ppm ww and 0.5 ppm ww, respectively, showed alarming results. Hamour, Chanad, Salmon, and Sea bass consumers exceeded the PTWI limit for the three population groups. Safi consumers exceeded the TWI limit for the Qatari population and canned Tuna consumers exceeded the TWI limit for the non-Qatari population. The FAO/WHO reported in 2016 that the limits that were adopted in 1991 did not take into consideration the variation in fish consumption patterns across the populations (FAO/WHO, 2016). For this reason, the PTWI was reduced from the 3.3 µg/kg to 1.6 µg/kg. On the other hand, the results from implementing the EPA permissible Hg limits 0.3 ppm ww results showed that the PTWI or TWI were not exceeded for the consumers of the different fish species (EPA, 2001b). However, the sum of consumption for all species significantly exceeded the PTWI and TWI. In 2012 the European Food Safety Authority (EFSA) reduced the safe MeHg from the FAO/ WHO 2003 limit 1.6 µg/ kg bw to 1.3 µg/ kg bw. The reason for this reduction is because the EFSA think that the studies regarding the benefit of the omega 3 fatty acid had effected the FAO/ WHO limit (González-Estecha, 2013). For this reason, we will take conservative approach by using TWI limit for risk assessment.

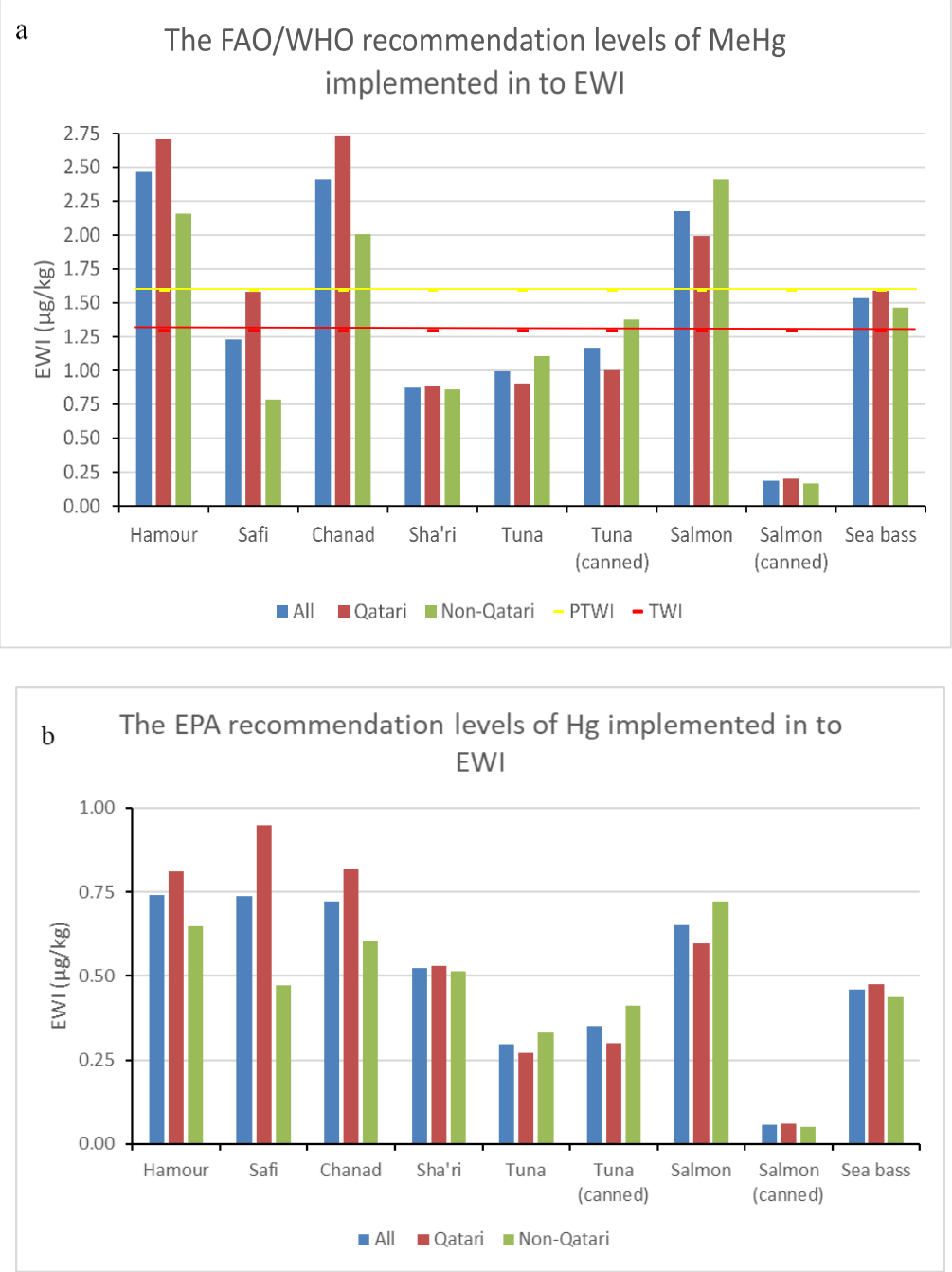


Figure 22. Implementing the recommendation levels to calculate EWI: a) implementing the (FAO/WHO, 2007) for MeHg in fish; and b) implementing the (EPA, 2001b) Hg criterion level in fish.

4.4. Risk assessment

The HQ resulting from applying the first scenario where it was assumed that 100% of totHg is MeHg, is presented in tables 27 and figure 23. All Hazard Quotients

(HQs) for median consumers in the three-population group were below the value of 1. All HQs for 95th percentile consumers in the three-population group were above the value of 1. The 95th percentile HQs ranged between 1.73 for males aged 18-29 and 3.33 for females in childbearing age among the general population, 2.16 for males from all ages and 3.5 for females in childbearing age among Qatari population, and 1.87 for males aged 18-29 and 3.17 for females aged 18-29 among non-Qatari population. As for the 75th percentile the HQ values ranged between 0.94 in males' Qatari population from 30 & above and 1.28 in Qatari females aged 30 & above. The HQs for the average consumers in the general population and non-Qatari were all below the value of 1. In contrast, Qatari females all ages, 30 & above, and childbearing age (18-40) were above the limit value, 1, 1, and 1.02, respectively. Although, the average HQs for the female's general population (0.97 and 0.97) and non-Qatari age (0.91 and 0.95) 18-29 and childbearing age were significantly close to value of 1, respectively.

Table 27. The Estimated Hazard Quotient (HQ) for Scenario's 1 Average and 75th Percentile When HQ≥ 1

Groups		General population		Qatari population		Non- Qatari population	
		Average	P75	Average	P75	Average	P75
Female and male	All ages	-	1.06	-	1.12	-	-
	Age 18-29	-	1.04	-	1.14	-	1.03
	30 & above	-	1.08	-	1.12	-	-
Female	All ages	-	1.14	1.00	1.26	-	-
	Age 18-29	-	1.21	-	1.23	-	-
	30 & above	-	1.12	1.00	1.28	-	1.02
	Age 18-40	-	1.12	1.02	1.21	-	-
Male	All ages	-	-	-	1.01	-	-
	Age 18-29	-	1.06	-	1.06	-	1.06
	30 & above	-	-	-	-	-	-

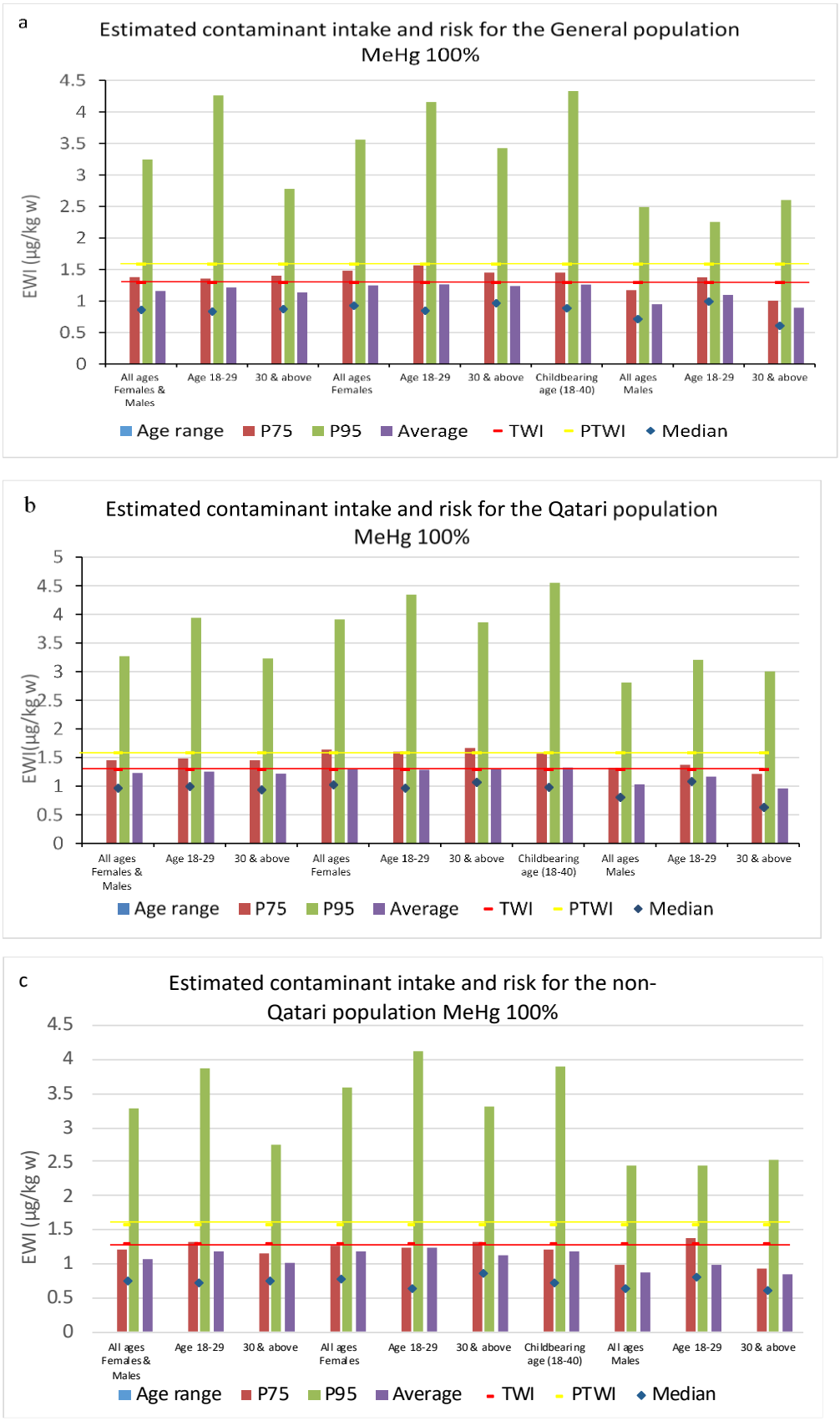


Figure 23. Scenario's 1, 75th and 95th percentile, median, and average EWI compared to PTWI and TWI: a) general population, b) Qatari; and c) non-Qatari

The HQ resulting from applying the second scenario where it the MeHg% from each fish species based on previous studies, are presented in tables 28 and figure 24. All Hazard Quotients (HQs) for median consumers in the three-population group were below the value of 1. All HQs for 95th percentile consumers in the three-population group were above the value of 1. The 95th percentile HQs ranged between 1.63 for males aged 18-29 and 3.14 for females in childbearing age among the general population, 2.03 for males from all ages and 3.3 for females in childbearing age among the Qatari population, and 1.76 for males aged 18-29 and 2.98 for females aged 18-29 among the non-Qatari population. As for the 75th percentile the HQ values ranged between 0.73 in the general population 30 & above and and 1.21 in Qatari females aged 30 & above. The HQs for the average for the three-population groups were all below the value of 1. However, the exposure in some cases was close to the safety margins especially for Qatari females and mainly the childbearing age.

Table 28. The Estimated Hazard Quotient (HQ) for Scenario's 2 Average and 75th Percentile When HQ≥ 1

Groups		General population		Qatari population		Non- Qatari population	
		Average	P75	Average	P75	Average	P75
Female and male	All ages	-	1.00	-	1.05	-	-
	Age 18-29	-	-	-	1.08	-	-
	30 & above	-	-	-	1.05	-	-
Female	All ages	-	1.07	-	1.19	-	-
	Age 18-29	-	1.14	-	1.16	-	-
	30 & above	-	1.05	-	1.21	-	-
	Age 18-40	-	1.05	-	1.14	-	-
Male	All ages	-	-	-	-	-	-
	Age 18-29	-	1.00	-	-	-	1.00
	30 & above	-	-	-	-	-	-

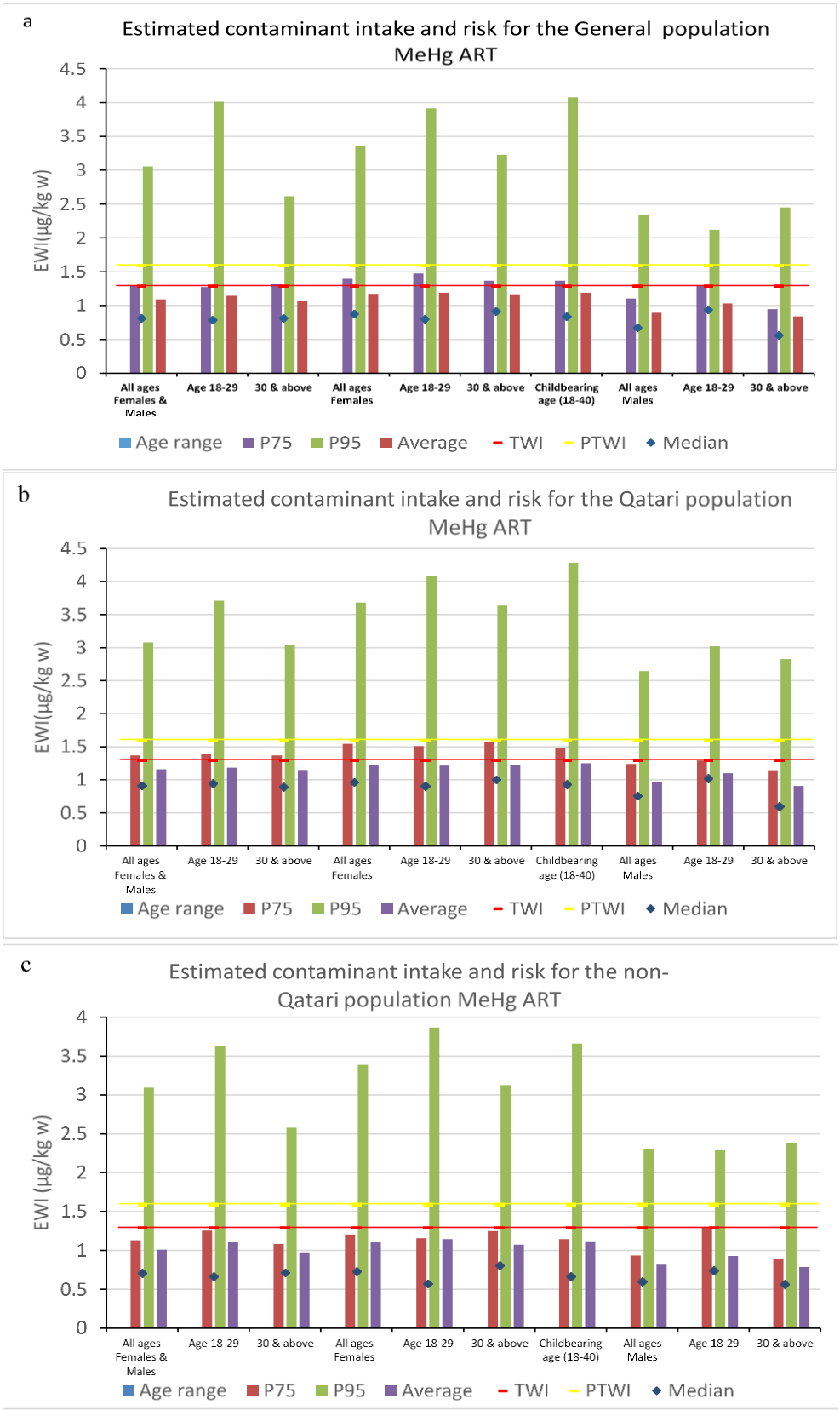


Figure 24. Scenario's 2, 75th and 95th percentile, median, and average EWI compared to PTWI and TWI: a) general population, b) Qatari; and b) non-Qatari

Males aged 18-29 have a higher fish consumption rate than the average all ages group for the three-population groups. This was observed in the 75th percentile HQs values. The highest consumed fish for Qatari males were from Hamour, Safi, and Chanad with a 50.9% contribution to the EWI and HQ. The highest consumed fish for non-Qatari males were from Hamour, Chanad, and Salmon with a 38.94% contribution to the EWI and HQ. The largest contribution to HQs in the Qatari females was from the high consumption of Hamour and Safi. The average consumption for Hamour and Safi is 198 g/w and 232.9 g/w, female consumption was 194 g/w and 227.1 g/w, respectively. The consumption per body weight in Qatari females was the highest for age groups 18-29 and 18- 40. This is similar to the results in Kuwait for females of childbearing age and Hamour contribution to the estimated daily intake (EDI) was 68%. The 75th and 95th percentile were significantly higher than our results and accounted for 0.43 and 0.88 µg/kg d corresponding to 3.01 µg/kg w and 6.16 µg/kg w, respectively. The average EDI was 0.32 µg/kg d corresponding to 2.24 µg/kg d. The risk was measured based on the PTWI and the average HQ was 1.4 (Laird, 2017). Additionally, some of the most consumed fish are from species containing the highest Hg levels. This indicates that even following the distribution of exposure methods the same groups will be at risk or close to risk value.

The maximum amount of Fish (MAF) consumption per week or the maximum allowable consumption rate that can results in HQ value 1 and above can be calculated using the following equation (Ferreira da Silva, 2020; Zolfaghari, 2018):

$$\text{MAF } (\boldsymbol{g}) = \frac{TWI \text{ or } PTWI \times bwt}{C}$$

C: concentration of contamination, bwt: body weight, and w: week

In table 29, the MAF consumption was measured based on the average weight of each group. The MAF revealed the influence of the body on the EWIs and HQs.

Males in general have the ability to consume more fish compared to females as a result of the higher body mass. The HQs for three fish species (grouped by size) from Baja California, Mexico with a consumption rate of 250 g/w with an average body weight of 70 kg for males and 60 kg for females. Females were at risk for more species compared to males. The highest HQ for females was 2.77 and for males was 2.37 (Murillo-Cisneros, 2021).

Table 29. MAF Value in Grams for scenario 1 for All Population Groups and Ages

Gender	Age	General population		Qatari		Non- Qatari	
		TWI	PTWI	TWI	PTWI	TWI	PTWI
Female and male	All ages	1297.0	1596.4	1303.7	1604.5	1288.6	1585.9
	Age 18-29	1183.3	1456.4	1195.8	1471.8	1168.8	1438.6
	30 & above	1348.9	1660.2	1353.4	1665.7	1342.6	1652.5
Female	All ages	1210.5	1489.8	1228.1	1511.5	1185.4	1459.0
	Age 18-29	1097.5	1350.8	1107.4	1362.9	1085.2	1335.6
	30 & above	1265.2	1557.2	1281.8	1577.7	1237.2	1522.7
	Age 18-40	1177.8	1449.6	1187.0	1461.0	1165.6	1434.5
Male	All ages	1509.0	1857.2	1523.5	1875.0	1494.1	1838.8
	Age 18-29	1435.3	1766.6	1431.1	1761.4	1442.1	1774.9
	30 & above	1537.0	1891.7	1570.4	1932.8	1509.5	1857.9

The high fish consumption among females of childbearing age can have an increased risk of toxicity increase of pregnancy and during. Study on mother's hair and blood after childbirth from Croatia. Hair as mentioned in section 2.8.1 in bioindicator for MeHg consumption. The study revealed a strong positive correlation between seafood consumption and MeHg concentration in the mother's hair and blood

(Sekovanić, 2020). (Sulimanec Grgec, 2020) compared the contribution of the EWI to TWI and the HQs for two and four meals per week for 12 fish species in females with an average body weight of 65 kg. Four species were above value 1 for two meals per weeks and five species were above value 1 for four meals per weeks. The influence of high consumption of highly Hg contaminated fish species in Portugal was observed in the blood of pregnant women. The number of fish meal consumption reaches to 8 meals per week. The study finding revealed that the Hg level in the blood of 30% of those women exceeded the maximum safe level of the Health Organization/United Nations Environment Programme. The finding was higher than other nations where fish consumption is the main food source like in Hawaii (USA), South Korean, and Japan (Caetano, 2019).

The HQs from fish consumption in Malaysia were determined based on the following groups: age, gender, and ethnicity. The highest fish consumption was in age 60 and above, males, and Malays. Only 60 and above were at risk with HQ value 1.0489 and the Malays population had an HQ value of 0.969. Childbearing females had a lower HQ value of 0.817 in comparison to our both scenarios (Ahmad, 2021).

(Saei-Dehkordi, 2010) investigated the risk fish impose on the Iranian population with average body weight of 70 kg for fish caught from the Arabian Gulf (Persian Gulf) through two seasons winter and summer. The study calculated the HQ using the JECFA 1972 PTWI limit 5 µg/ kg w (FAO/WHO, 2010). There was no risk and after calculating the HQ using TWI no risk was observed. However, the study used a constant consumption rate of 147 g/w for average body weight of 70 kg. The Hg in the fish samples was significantly higher than our finding but the study presented an increase in the Hg level during winter which increased the EWI. Since our study was done on samples bought during the fall season, we can indicate a small increase in the

Hg level during the winter and subsequently increasing EWI and HQs above the value of 1. Another study done in Iran for different fish species from wetland and the Caspian Sea showed variation in the HQs ranging between 0.009- 1.12 for fish consumption 29.23 g/d equivalent to 204.61 g/w. The fish that resulted in the highest HQ value had a higher daily consumption rate than the MAF (daily) and that resulted in a high HQ value (Zolfaghari, 2018).

The HQs for median, 75th percentile, and 95th percentile for different fish species consumed at a rate of 332 g/w by the Greek population with an average weight of 70 kg, were all significantly below the value of 1 (Renieri, 2019). The HQs for the weekly consumption of Sea bass were 0.13, 0.228, and 0.36 for median, 75th percentile, and 95th percentile, respectively.

The daily fish consumption for one of the highest fish consumption areas on the Amazon, Brazil during two seasons a November (flood season) 525 g and May (dry season) 519 g. The EDI was measured for each fish species induvial. Similarly, to our results females consumed more fish and had higher EDI levels. The highest EDI value was during the Flood season 4.55 µg/kg d equivalent to 31.85 µg/kg w and the HQ value will be significantly higher than 1. The lowest EDI was among men during the dry season 0.17 µg/kg d equivalent to 1.19 µg/kg w and was the only value below the TWI level (Ferreira da Silva, 2020).

The difference in the risk imposed from the consumption of Nephrops and fish species in Norway using two scenarios (low and high consumption) was observed. The consumption of Nephrops showed no risk. However, for consumers that consumed Nephrops, and fish or fish only within the high consumption scenario, the intake exceeded the TWI limit. Consumers that consumed fish only with the highest MeHg concentration had an average weekly intake of 130 µg for an average 80 kg body

weight. This corresponds to EWI equivalent to 1.6 µg/kg w and HQ equivalent to 1.25 (Wiech, 2021).

The risk from the consumption of 15 fish that belongs to the elasmobranch species in Italy for three consumption levels was estimated. The consumption levels were 497.0, 276.1, and 140.0 g/w, and resulted in EWI of 5.20, 2.84, and 1.46, respectively. The highest two consumption levels had EWI that exceeded the PTWI limit and had HQ value above 1. On the other hand, the lowest consumption level exceeded the TWI and will have an HQ value above 1 (Storelli, 2022).

4.5. Principal Component Analysis (PCA)

4.5.1. Analysis of Mercury

Three PCA was created to investigate what influences the increase and decrease of totHg in different fish species. PCA1 biplots (figure 25-a), taken together F1 and F2 captured 100% of the variability of the data; F1 explained approximately 72.01% of the variation in the data and had a strong positive correlation with the moisture ratio and strong negative correlation with totHg concentration. The F2 captured approximately 27.99% of the variation in the data and had a positive correlation with totHg concentration and moisture ratio. The increase in moisture ratio indicates a relationship with the water and lipid content. PCA 2 biplots (figure 25-b), was used to investigate the negative correlation between totHg concentration and moisture ratio by comparing the average totHg for the fresh species to the lipid, protein, and moisture content from previous studies as listed in table 30. The variance in PCA 2 was between F1, F2, F3, and F4 amounted to 52.06%, 37.41%, 10.29, and 0.23%, respectively, F1 and F2 are chosen because they have higher variance in the data. PCA3 biplots (figure 25-c), taken together F1 and F2 captured 84.08% of the variability of the data; the variance for F1, F2, F3, F4, and F5 amounted to 50.68%, 33.4 %, 14.24%, 1.53%, and 0.156%,

respectively. PCA3 biplots (figure 25-c), were done to reduce the redundancy of the same variables used for PCA2 with the addition of the fish weight.

PCA1 paralleled the fish species clustering together and organized in sequence significantly through the F1 axis. Based on PCA1 Salmon, local Tuna, and Sea bass had low totHg concentration and high moisture ratio, and a strong positive correlation with F1 and F2. Safi, Sha'ri, and imported Tuna had low totHg concentration and low moisture ratio and correlated negatively with F2. On the other hand, Hamour and Chanad samples with the high totHg concentration and lower moisture ratio had a significant negative correlation with F1. Chanad samples hand different correlations between them and the F1 and F2 axis.

Based on PCA2, through F1 and F2 Hg concentration had a positive correlation with moisture this indicates that increasing the water content will always increase totHg concentration. F1 explained the strong negative correlation the lipid has with totHg concentration and moisture. However, through F2 there was a weak positive correlation. This indicates that increasing the lipid content can reduce totHg concentration. Conversely, protein had a similar scenario. F2 explained the strong negative correlation protein has with Hg concentration and moisture. On the other hand, totHg had a weak positive correlation with protein in F1. This indicates that there are other factors that can reflect the correlation between the Hg level and the lipid and protein content. TotHg in Hamour was mainly influenced by the high moisture content. Salmon and Sea bass were mainly influenced by the high lipid content. Safi, Sha'ri, and Tuna were mainly influenced by the high protein content and the moisture. Chanad was the only species that showed weak influence with the three contents.

The positive correlation between Hg level and protein and the negative correlation between Hg level and lipid can be explained by the ligand complex bond

between Hg and metalloproteins mainly amino acids containing the thiol group. Around 30.2- 37.6% of the totHg in Tuna and Salmon tissues was bonded to beta-actin protein (Nong, 2021). Hg has a high binding affinity in soluble proteins and binds as MeHg mainly to the myofibrillar protein (Ando, 2008). In addition, MeHg was the main species binding to the protein and had a higher binding affinity with amino acids containing thiol group and nitrogen-containing compounds than Selenium. This can explain the positive correlation between protein and moisture (Charette, 2021; Nong, 2021). The percentage of soluble protein bond to Hg in Tuna is 50% compared to 35.4% in Salmon, the difference is because Salmon have higher lipid content than Tuna (Nong, 2021). However, this was noticeable in the condition of high lipid accumulation, which creates a dilution effect to the protein content, and with that, the binding site will be reduced (Balshaw, 2008; Nakao, 2007). Safi has low-fat content and high protein content but low totHg level because of the lower trophic level and feeding source. According to other articles, Sha'ri and the local Tuna species (*Euthynnus affinis*) have lower fat content, however, the totHg level was low (Musaiger, 2008; Saoud, 2007). Yellowfin tuna from different capturing sea bodies had different Hg levels. MeHg level had no correlation with lipid content and the highest MeHg were in the sample with the lowest lipid content samples from the North Pacific Ocean. MeHg samples from this study correlated with the location of capturing (Nicklisch, 2017). Sha'ri's food source can explain the low level of totHg and fish gender can also influence the Hg level, female Sha'ri samples from (Al-Ansari, 2017) study showed significantly higher totHg and MeHg levels than in males.

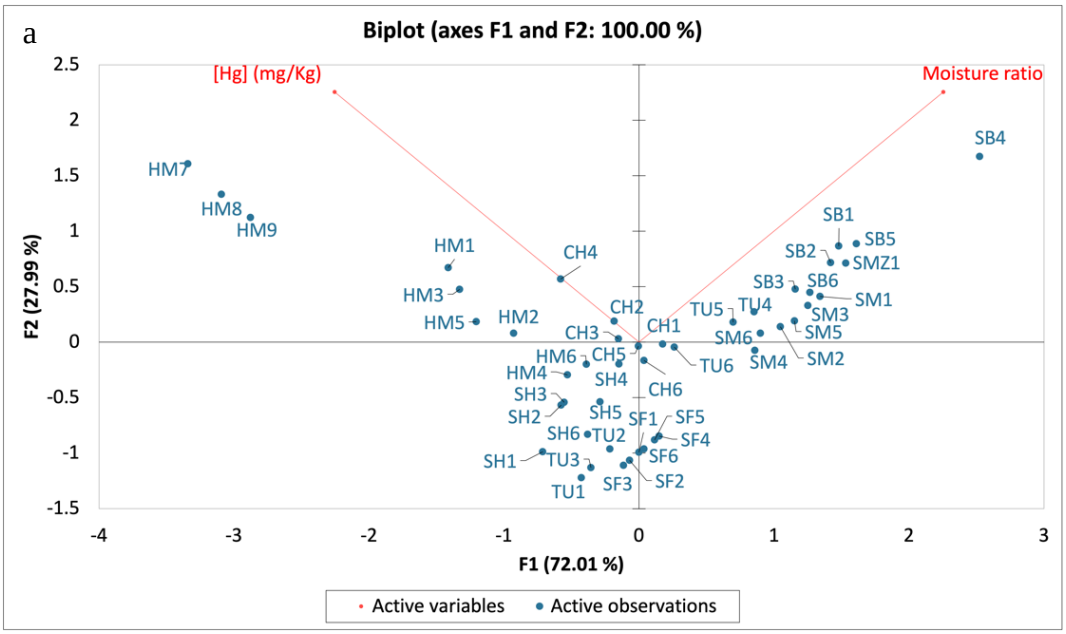
The low totHg level in Tuna can be related to low contamination level or to the age and size of the fish. The totHg level is also influenced by the metabolism, chemical contaminants from water and atmosphere, geographic region of the fish habitat, niche,

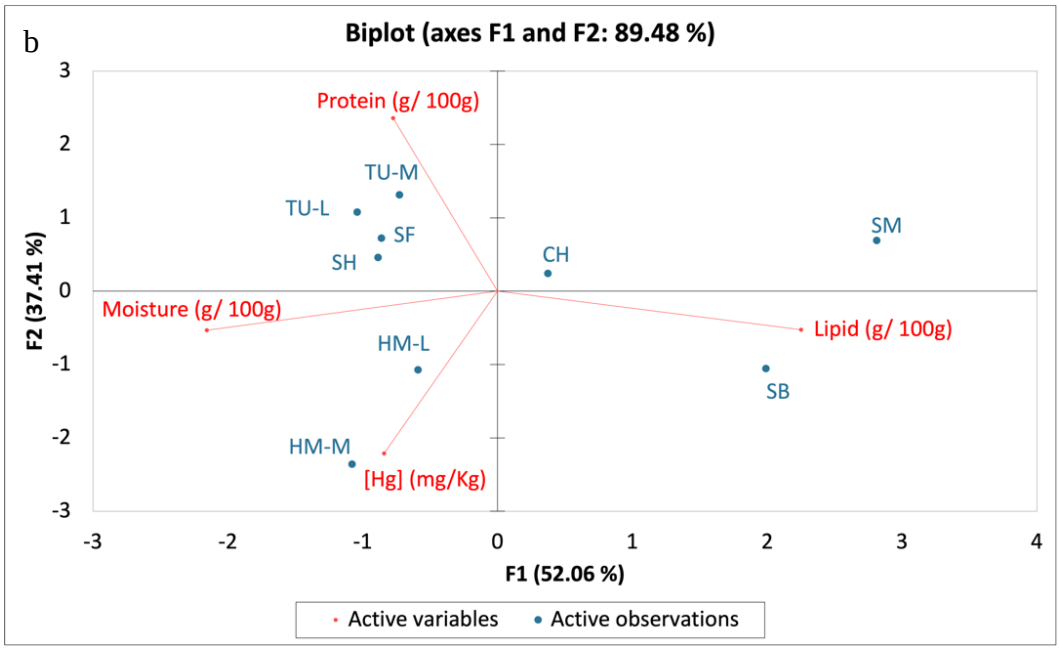
and harvesting (Li, 2022). Since the imported Hamour and Tuna were bought as fillets the depth of the cut can be also a factor that affected the Hg level. In Striped Bass fish the tissues around the lateral line contained fewer MeHg levels. Besides MeHg levels in tissues decreases with the decrease in the depth of the tissues. However, this was not the same for Northern Pike fish which shows no diverse distribution of Hg through the muscular system. This indicated that each species would have different Hg distribution. It was found that fish species with a wider variation range between % N (protein) and %lipids will have a variation in the distribution of Hg through the muscular system (Charette, 2021). Demersal species like Hamour can be associated with the level of Hg in the sediment. Hg in sediment from the northern coastline of Qatar that had a higher Hg concentration in comparison to the coastline from Doha to the southern region (Hassan, 2019). Moreover, sunlight has the ability to degrade 80% of the MeHg on surface water but in deep water, the MeHg will have a longer lifetime (Olsvik, 2021).

Based on PCA3, the correlation between totHg concentration protein and lipid were the same, however, the moisture had a weak correlation with totHg. The weight had a strong and significant positive correlation with totHg concentration through F1 and F2. There was a strong negative correlation between weight and lipid through F1. The concentration of Hg will increase with the increase in age and body-size parameter of the fish as a result of bioaccumulation regardless of the moisture, protein, and lipid content (Al-Ansari, 2017; Bosch, 2016). Since lipid always correlates negatively with the totHg level with F1, the increase in fish weight from protein and moisture will yield more increase in Hg level than if the weight increase was from lipid.

When Tuna samples were studied for the change in totHg concentration through canning processes it was found that there was an increase in totHg concentration,

protein, however, there is a small increase in lipid content and moisture content (Rasmussen, 2007). Other studies compared to (Rasmussen, 2007) study showed an increase in totHg concentration and protein content, a significant increase in lipid content, and a significant decrease in the moisture content. MeHg is a lipophilic compound, this means that the increase in lipid content can enhance the MeHg cell membrane penetration (Pawlaczyk, 2020). After that the MeHg will accumulate in the protein by forming a strong stable bond with the sulfhydryl groups Hg–S bond (Kutscher, 2012). This makes the fish muscles the primary MeHg reservoir, however, (Kutscher, 2012) study in Tuna fish indicated that the Hg-S is highly influenced by the protein molecular weight and that MeHg is mostly bonded with high-molecular protein and inorganic Hg bond with low- molecular protein (Balshaw, 2007; Kutscher, 2012). Those finding can indicate when fish that contain high lipid content is exposed to high Hg contamination levels the mobility of MeHg to the binding site on the protein will be enhanced. Therefore, the low Hg level in Sea bass compared to Hamour can be a result of the difference in the Hg contamination level.


a
Biplot (axes F1 and F2: 100.00 %)
[Hg] (mg/Kg)
Moisture ratio
HM7
HM8
HM9
HM1
HM3
HM5
HM2
CH4
CH2
CH3
CH1
CH5
CH6
HM6
HM4
SH4
SH3
SH2
SH5
SH6
SH1
TU2
TU3
TU1
SF3
SF1
SF5
SF4
SF6
SF2
TU5
TU4
TU6
SM6
SM4
SM5
SM3
SM2
SM1
SB1
SB5
SB2
SMZ1
SB3
SB6
SB4
F2 (27.99 %)
F1 (72.01 %)
Active variables
Active observations


b
Biplot (axes F1 and F2: 89.48 %)
Protein (g/ 100g)
TU-M
TU-L
SF
SH
CH
SM
Moisture (g/ 100g)
Lipid (g/ 100g)
HM-L
SB
HM-M
[Hg] (mg/Kg)
F2 (37.41 %)
F1 (52.06 %)
Active variables
Active observations

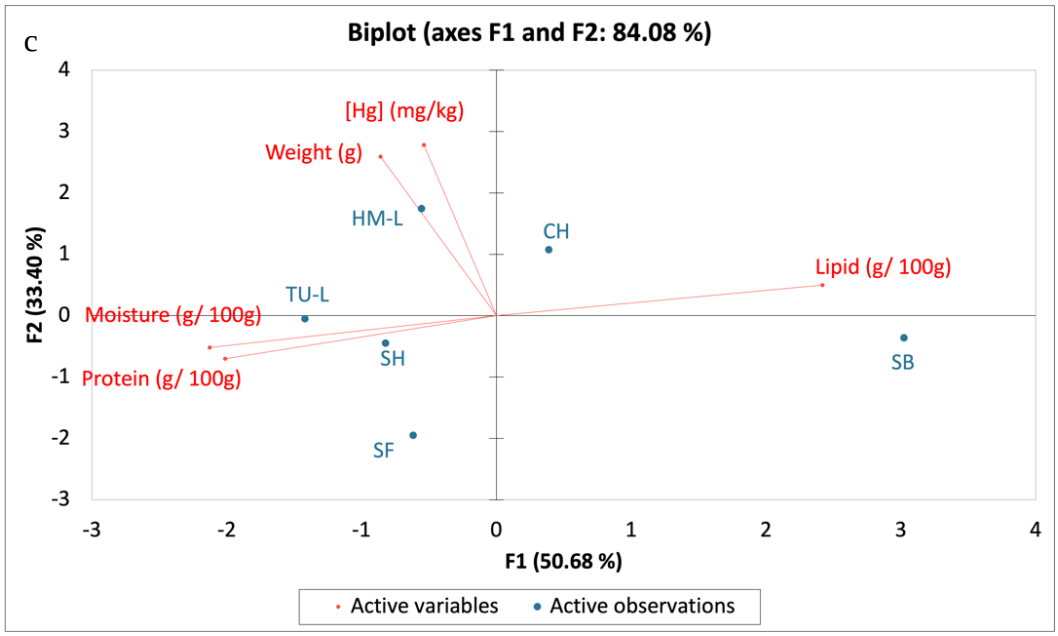


Figure 25. Principal component analysis (PCA) of totH. a) PCA1: totHg distribution in fresh fish samples and moisture ratio; b) PCA 2: totHg distribution in fresh fish samples and the fish lipid, protein, and moisture content, and c) PCA3: totHg distribution in whole fresh fish samples, fish average weight, and the fish lipid, protein, and moisture content. Abbreviations: Hamour (HM), Safi (SF), Chanad (CH), Sha'ri (SH), Tuna (TU), Salmon (SM), Sea bass (SB), local (L), and imported (M).

Table 30. Proximate Analysis (Lipid, Protein, and Moisture Content) Used for PCA.

Code	[Hg] (mg/Kg)	Lipid (g/ 100g)	Protein (g/ 100g)	Moisture (g/ 100g)	References
Hamour (imported)	0.158	4.58	14.05	76.45	(Momenzadeh, 2017)
Hamour (local)	0.406				
Safi	0.001	1.96	19.71	76.75	(Patrick Saoud, 2008)
Chanad	0.091	9.3	19.5	70.4	(Musaiger, 2008)
Sha'ri	0.063	1.1	19.7	75.2	(Musaiger, 2008)
Tuna (local)	0.040	0.93	22.73	75.38	(P., 2017)
Tuna (imported)*	0.018	1.52	23.18	73.28	(Biji, 2018)
Salmon	0.008	19.9	19.1	59.2	(S. Costa, 2015)
Sea bass	0.021	18	10.3	68.83	(Khrystenk, 2015)

Yellowfin tuna*

4.5.2. Analysis for Exposure to Methyl Mercury

The PCA was done for the EWI results from the distribution of exposure method for the Qatari and non-Qatari population to investigate the influence of MeHg level, EWI and CNP/bwt in the consumed fish sources. PCA1 biplots (figure 26-a) represent the Qatari population, taken together F1 and F2 captured 99.14% of the variability of the data; F1 explained approximately 72.58% of the variation in the data and had a positive correlation with the CNP/bw and strong negative correlation with MeHg concentration and EWI. The F2 captured approximately 26.57% of the variation in the data and had a strong positive correlation with the CNP/bw. Hamour and Chanad had a strong positive correlation with EWI and MeHg through F1. However, Hamour is

more influenced by MeHg concentration than CNP/bw. Safi, on the other hand, correlated negatively with the three variables, however, through F2 Safi correlated positively with CNP/bw. PCA2 biplots (figure 26-b) represent the non-Qatari population. Taken together F1 and F2 captured 99.39% of the variability of the data; F1 explained approximately 73.67% of the variation in the data and had a positive correlation with the CNP/bw and a strong negative correlation with MeHg concentration and EWI. The F2 captured approximately 25.72% of the variation in the data and had a strong positive correlation with the CNP/bw. Canned Tuna had a positive correlation with the CNP/bw, MeHg concentration, and EWI through F1. The non-Qatari population has higher canned Tuna consumption rate. Additionally, Hamour, Chanad, and canned Tuna have the highest Hg level. The PCA confirms the results from section 4.3 for the fish species/ types and the contribution to EWI. In a study done in Malaysia, it was observed that for the higher fish consumers the demersal fish species were always the highest contributor to the EWI (Ahmad, 2021).

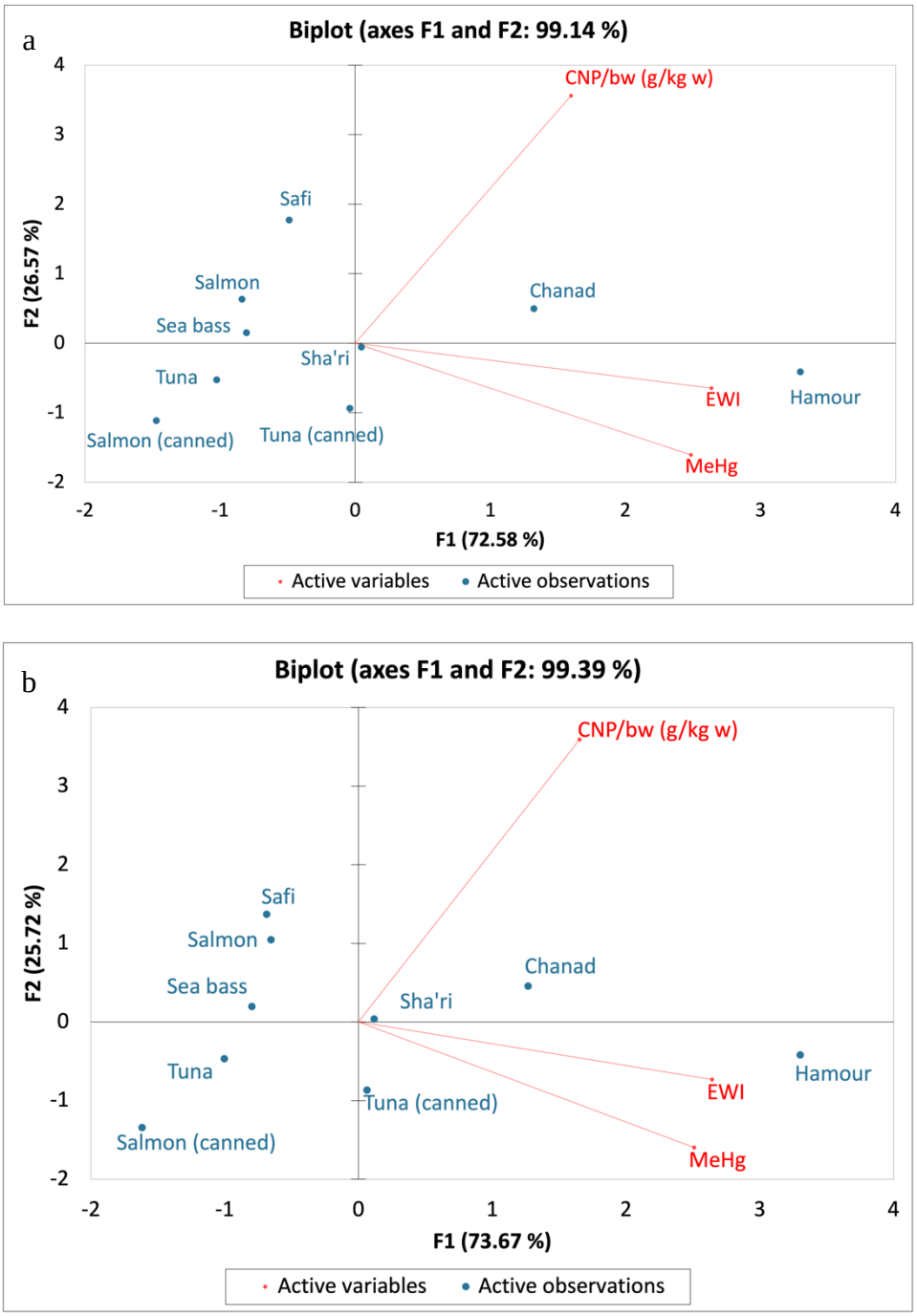


Figure 26. Principal component analysis (PCA). a) PCA1: Qatari population and; b) PCA 2: non-Qatari population.

4.6. Pollution Effect on Mercury Levels in Fish

The marine boundary layer (MBL) and surface ocean will shorten the lifetime of the inorganic gaseous oxidized mercury (GOM) and particulate Hg (p-Hg) as a result of the air-sea exchange. At the same time, the air-sea exchange can also be a source for re-emitting Hg back into the atmosphere (Soerensen, 2010). The finding of the totHg concentration from our study showed low Hg levels that did not exceed the EPA criterion level of Hg in fish with the exception of imported Hamour samples. Sha'ri samples from our study and previous studies had wide variation. Even though Sha'ri is a demersal and non-predatory species the tot-Hg concentration was higher than Sea bass which is a demersal and predatory species. If we neglected the lipid-protein-Hg relation the differences in concentration can be related to the level of Hg in the sediment or the possibility of the Sea bass being farmed in a controlled environment (Nakao, 2007). However, even for farmed species caution is still required since it was discovered that farming location can be influenced by water salinity and temperature, feeding source, and mainly the pollution level (Di Bella, 2021).

The level of Hg in local fish samples can be related to factors other than sex, size, age, or lipid, and protein content. Those factors can be trophic level, water depth, migration, or the pollution level in the fish niches and habitat. The imported and canned samples can be related to the same factors in addition to the canning process for canned fish and the geographic location. Naturally, predatory fish contain higher levels of Hg since they are at the top level of the trophic (da Silva, 2020). The geographic location and the water column depth or the thermocline depth can influence the level of Hg in water which will reflect on the marine species. In some regions, the surface water can contain a higher Hg level in case of exposure to aeolian dust. However, open oceans will be less exposed to Hg pollutants and for this reason, fish that grow in the open

water can contain lower contamination levels than fish that grow near the coastal areas (Ormaza-González, 2020). The level of Hg will increase with the increase in the depth of the water column as a result of the change in the microbiological and chemical conditions. Fish that ingested contaminated sediment will have a rapid increase in Hg concentration and will introduce more Hg into the food chain (Clarkson, 2020). A study from the Portuguese coastal area showed seasonal variation as a result of the thermocline effect in the summer, where Hg from the ocean ground sediment will be mixed with the upper layers, although, these phenomena was not observed in the Qatari water (Al-Ansari, 2017; da Silva, 2020).

The level of Hg in sediment is highly important to the food chain and for demersal species like Hamour and Sha'ri. Different studies compared the concentration of Hg in demersal species to other species like pelagic or benthopelagic species. The demersal predatory species always contain higher Hg levels as a result of living on the bottom of seas and feeding on other fish species. For example, the demersal species had totHg concentration of 0.312 mg/kg ww compared to 0.288 mg/kg in pelagic species (Saei-Dehkordi, 2010).

The totHg concentration from Qatari sediments samples from different locations showed a significant decrease from 1999 to 2019 (De Mora, 2004; Hassan, 2019; Kreish, 1999). The totHg level from sediments from 8 locations in 1994 ranged between 0.0239- 0.179 mg/kg dw, the highest concentrations were from the industrial coastal area of Umm-Saeed followed by the samples from the Doha coastal area. However, the concentrations from Doha were significantly lower than samples from a 1994 study where the totHg concentration ranged between 0.19-1.75 mg/kg (Kreish, 1999). Sediment samples from 5 locations in 2004 ranged between 0.0007- 0.0167 mg/kg this range is lower than the 1999 range (De Mora, 2004). However, sediment

samples from13 locations in 2019 had a slightly higher range of 0.008- 0.034 mg/kg. The concentration of Hg was highly influenced by the current movement and the highest Hg levels were around the northeastern of Qatar. Additionally, the Hg levels near Doha coastal area were considered moderately low regardless of the high exposure to anthropogenic emissions (Hassan, 2019).

The local and imported Hamour contained a higher level of Hg compared to Hamour from China, on the other hand, Chanad contained a lower level of Hg compared to other studies. Additionally, local Tuna contained lower Hg levels compared to the non-predatory Sha'ri. Since Chanad and Tuna are pelagic species, this can indicate that the influence on Hg level is from the sediment is the main contributor to the Hg level. Since MeHg is mainly the product of methylation, the seabed is a good methylation environment since it is a totHg sink and habitat for sulfur reducing bacteria (SRB). According to the 1985 National Research Council, the oil pollution in the Arabian Gulf is 4.7 % of the total global oil pollution. This will influence the Hg level in sediment which was observed on the benthic species (Elsagh, 2021; Sarasiab, 2014).

The geographic location can influence the level of Hg significantly. The fjord of Sørfjorden in Norway is influenced by the input from two rivers in which one of which is influenced by the present of hydroelectric power plants. The water from the power plant is from the hypolimnion layer which provides ideal methylation conditions. The other river is considered as an Hg source as a result of atmospheric deposition. Additionally, the rivers were influenced by run-off that contain Hg. The Hg in the run-off is a residue of zinc plant that was closed in the 1980s, however, the Hg is still available in the water and sediment. Those pollution sources influence the downstream species by increasing the Hg concentration in plankton and fish (Azad, 2019).

Wetland ecosystems like mangroves can be a source of Hg. Mangroves have a high ability to tolerate heavy metals accumulation without facing the risk of losing their biomass. Mangrove forest is considered to be a nursery, habitat, and niche to wide range of species (Rezaei, 2021). Mangrove can accumulate Hg; however, it will limit the mobility and bioavailability. The system had the ability to produce MeHg and the biodiversity in this system creates concern especially when there is anthropogenic addition into the system (Duan, 2021). The mangrove system is unique since it has plants, seawater, and sediment direct interaction, it also combines the photochemical reactions with the microbial processes (S. Huang, 2020). In mangrove forest under anaerobic conditions a decomposition of sediment compounds will occur, and Hg will be released. Moreover, mangrove is rich in organic matter (OM) and with the availability of the methylation microorganisms MeHg will be produced from inorganic Hg (Duan, 2021; S. Huang, 2020). Moreover, Phytoplankton can produce OM which will be consumed by heterotrophic bacteria like microbial methylators (Ji, 2020). See figure 27 for the interaction between litterfall, bacteria, and phytoplankton to produce MeHg.

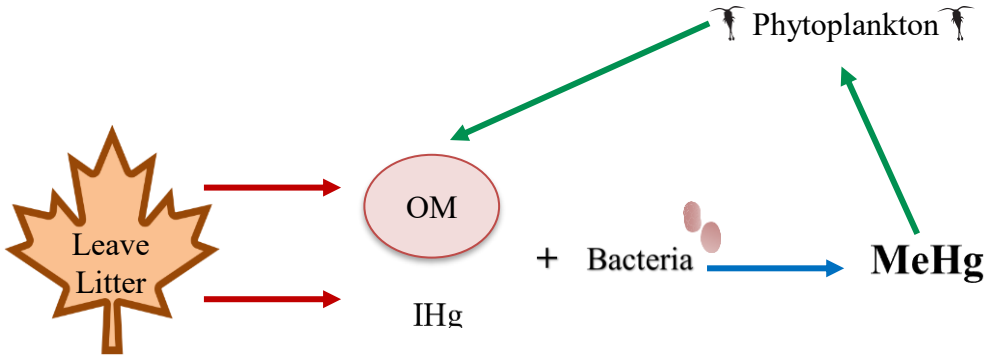


Figure 27. The biotic factors interact with inorganic Hg (IHg) and organic matter (OM) to produce MeHg. (Red arrow: anaerobic decomposition product, blue line: reaction, & green line: consuming & producing) (Duan, 2021; Ji, 2020)

CHAPTER 5: CONCLUSION

This study is the first study done in in the State of Qatar to assess the health risks of Mercury and Methyl Mercury as a result of fish consumption among fish consumers. The fish frequency questionnaire was used to collect the most consumed fish species and forms, consumption data, and from various demographics. Total Mercury contamination levels of the highly consumed fish species was determined using a validated ICP-MS method. Exposure assessment was done using deterministic approach methods and simple distribution, and risk assessment was done using the HQ. The aggregated method was used for the risk characterization. PCA analysis was used to analyze the influence that some of the nutrient compositions (lipid, protein, and moisture content) have on Mercury accumulation in the fish tissues.

According to the results, fresh, and predatory species including Hamour, Safi, Chanad, and Salmon are the main consumed fish species by Qatari and non-Qatari. The only highly consumed non-fresh form was canned Tuna. Average consumption of all fish species was 1102 g/w. The high consumption rate can indicate either high fish consumption in Qatar or the response was mainly from the highest consumers. Mercury levels on the 65 analyzed samples were all below the permissible level of 0.3 ppm except for imported Hamour. Total Mercury levels ranged between 0.001 mg/kg ww in Safi and 0.443 mg/kg ww in Hamour, both species were among the highest consumed fish in Qatar. The concentration of total Mercury in canned Tuna had significant variation ranging between 0.009 mg/kg ww and 0.255 mg/kg ww, the only significant difference was between the samples from country A and the other 4 countries.

The distribution of the exposure method showed that the level of Mercury in fish is significantly influencing the contribution to the total EWI. Although Safi was the most consumed fish species for the Qatari population and Salmon for the non-Qatari

population, the contribution to the total EWI was negligible because of their low Mercury contamination level. The aggregated method revealed that the EWI is highly influenced by the high fish consumption rate. Moreover, the implementation of the (FAO/WHO, 2007) permissible limits of 0.5 ppm (non-predatory) and 1 ppm (predatory) for Methyl Mercury and using actual consumption rates showed alarming results for the general population, with a high contribution from Hamour, Chanad, and Salmon. Additionally, for the Qatari population Safi was found to be the main contributor to the exposure, whereas canned Tuna for the non-Qatari population.

Two scenarios of Methyl Mercury were used for the risk characterization in this study: the conservative approach (scenario 1) assuming that 100% of total Mercury was Methyl Mercury and the second approach (scenario 2) where the Methyl Mercury% was derived from total Mercury based on previous published data for each included fish species. According to the risk characterization analysis conducted in this study, Mercury exposure from scenario 1 is not likely to results in adverse health effects in the non-Qatari population but the risk was observed between Qatari females aged 30 & above and childbearing age. However, the consumer from the 75th percentile Qatari females and non-Qatari males aged 18-29 were at risk. In addition, based on the scenario 2, Mercury exposure from fish is not likely to results in adverse health effects in the general population.

Based on the PCA analysis result predatory fish species that contain higher lipid content will mostly contain lower Mercury levels and can be a safer alternative. However, the lipophilic Methyl Mercury characteristics can facilitate the mobility to protein binding site in polluted locations. Moreover, non-predatory and non-demersal species regardless to the protein lipid content is also a safer alternative. The PCA

analysis confirmed the influence of the Mercury level on the consumed fish and that EWI is mostly influenced and controlled by the high Mercury level.

During the implementation of the study, strengths and gaps were observed that may need to be considered in future studies. Using questionnaire data collection method provided good sample size and can provide real-life data for a year and lifetime consumption, however, the consumption over a year can contain over or underestimation. The aggregated method for the risk assessment is the recommended method and can provide accurate results. The main three gaps in this study are not including children, the lack of real Methyl Mercury analysis, and the influence of different fish weight on Mercury level and the exposure. Moreover, the results might be over estimating fish consumption and therefore the exposure to Mercury, because we used online surveys, convenience sampling, and a fixed portion size for all fresh samples. Despite its limitations, this study shed the light on the necessity to establish Methyl Mercury permissible levels in fish (predatory and non- predatory) based on real consumption data obtained from nationwide surveys. Besides, since females at child baring age 18-40 at increased risk of Hg toxicity as a result from there fish consumption patterns, therefore recommendations from health authorize targeting this category is needed. However, pilot-scale implementation followed by the collection of hair or blood samples as human biomonitoring to determine the real Methyl Mercury exposure level is needed to support and validate our current results. The result of such a study will provide an accurate evaluation of the impact of fish consumption in Qatar. Additionally, the analysis of Hg in sediment and water from capturing site, and the content of (lipid, protein, and moisture) in the captured fish can provide an understanding for the correlation between them.

CHAPTER 6: REFERENCES

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APPENDIX

Appendix1: Sample Preparation

1) Fresh fish samples as whole fish and pre-cut in plate. Left: Salmon and right Sha'ri

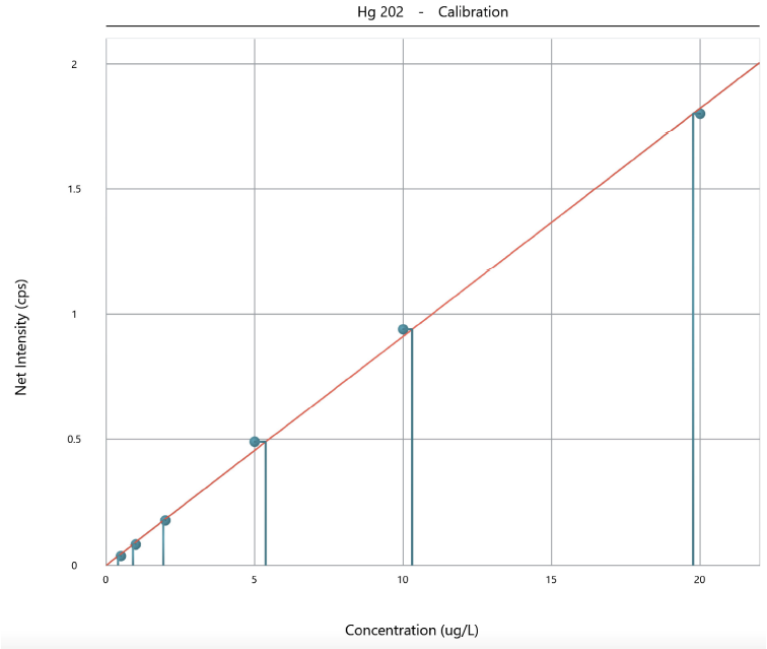
2) Sample cutting using knife and cutting board

3) Samples in the aluminum foil tray and dried samples grinding

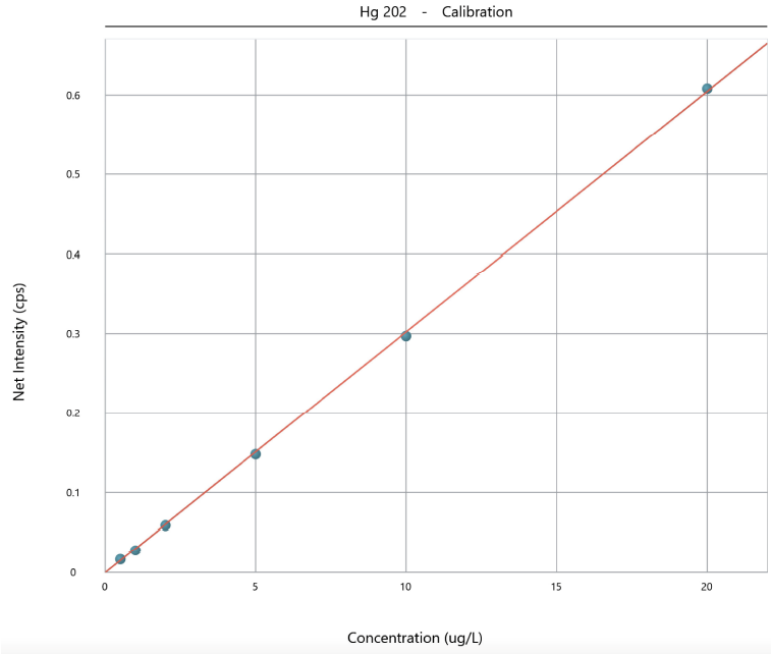
4) Acid digestion using Milestone ultraWAVE microwave digestion system

Appendix 2: Calibration Curve

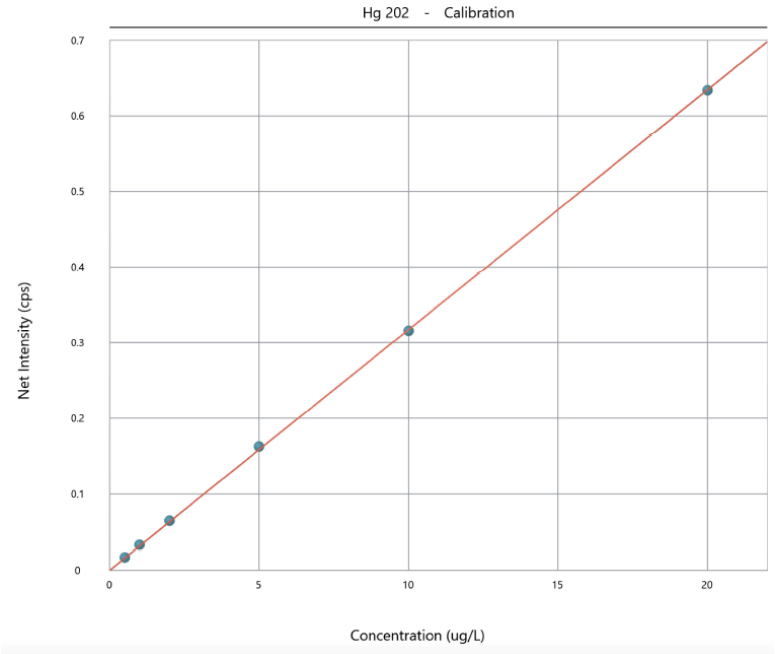
1) Calibration curve of Standard



2) Calibration curve of Standard



3) Calibration curve of Standard



Appendix 3: ANOVA Tables and Fisher Comparisons Test

ANOVA table 1: Total Hg concentration in mg/kg and different fish samples

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Fish	10	0.48033	0.048033	27.35	0.000
Error	54	0.09483	0.001756		
Total	64	0.57516			

Fisher Comparisons 1: Total Hg concentration in mg/kg and different fish samples. Hamour imported (HMM), Hamour local (HML), Tuna canned (TUC), Chanad (CH), Shari (SH), Tuna local (TUL), Salmon canned (SMC), Sea bass (SB), Tuna imported (TUM), Salmon (SM), and Safi (SF).

Fish	N	Mean	Grouping		
HMM	3	0.406384	A		
HML	6	0.158209	B		
TUC	16	0.091171		C	
CH	6	0.091005		C	
SH	6	0.062686		C	D
TUL	3	0.040378		C	D E
SMC	3	0.023071			D E
SB	6	0.020853			D E
TUM	3	0.017691			D E
SM	7	0.008384			E
SF	6	0.001176			E

Means that do not share a letter are significantly different.

ANOVA table 2: Total Hg concentration in mg/kg in different canned Tuna and country of origin as factor

Source	DF	Adj SS	Adj MS	F-Value	P-Value
country	1	0.03935	0.039351	20.59	0.000
Error	14	0.02675	0.001911		
Total	15	0.06610			

Fisher Comparisons 2: Total Hg concentration in mg/kg in different canned Tuna and country of origin as factor

country	N	Mean	Grouping	
b	7	0.147404	A	
a	9	0.047434		B

Means that do not share a letter are significantly different.

ANOVA table 3: The consumption per body weight for general population

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Age	1	195	194.8	0.93	0.335
Gender	1	1178	1178.3	5.63	0.018
Age*Gender	1	116	116.3	0.56	0.456
Error	596	124825	209.4		
Total	599	126898			

Fisher Comparisons 2: The consumption per body weight for general population Gender as factor

Gender	N	Mean	Grouping	
F	426	16.3168	A	
M	174	12.9236		B

Means that do not share a letter are significantly different.

Fisher Comparisons 3: The consumption per body weight for general population interaction between Age & gender

Age*Gender	N	Mean	Grouping	
18-29 F	139	16.4737	A	
30 & above F	287	16.1600	A	
18-29 M	49	14.1465	A	B
30 & above M	125	11.7008		B

Means that do not share a letter are significantly different.

ANOVA table 4: The EWI for general population

Source	DF	Adj SS	Adj MS	F-Value	P-Value
Age	1	0.745	0.74487	0.91	0.341
Gender	1	3.454	3.45444	4.21	0.041
Age*Gender	1	0.020	0.02001	0.02	0.876
Error	596	489.603	0.82148		
Total	599	495.029			

Fisher Comparisons 4: EWI for general population Gender as factor

Gender	N	Mean	Grouping	
F	426	1.03683	A	
M	174	0.85310		B

Means that do not share a letter are significantly different.

Fisher Comparisons 5: EWI for general population the interaction between Age & gender

Age*Gender	N	Mean	Grouping	
18-29 F	139	1.07250	A	
30 & above F	287	1.00116	A	
18-29 M	49	0.90275	A	B
30 & above M	125	0.80345		B

Means that do not share a letter are significantly different.

Fisher Comparisons 6: The consumption per body weight for Qatari population. The interaction between Age & gender

Age*Gender	N	Mean	Grouping	
30 & above F	173	17.0035	A	
18-29 F	77	16.8087	A	B
18-29 M	29	15.2140	A	B
30 & above M	57	12.5657		B

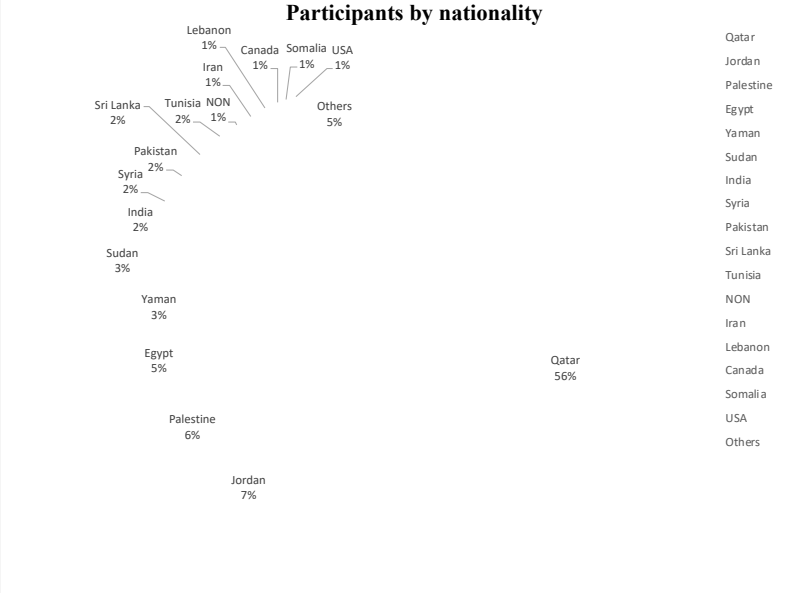
Means that do not share a letter are significantly different.

Appendix 4: Additional Tables and Figure

1) The percentage of consumption type for each fish

Species	Number of consumers	Fresh%	Frozen%	Canned%
Hamour	500	97.6	31.6	-
Safi	419	97.4	31.5	-
Chanad	431	97.7	30.9	-
Sha'ri	334	98.5	28.4	-
Tuna	434	33.9	14.5	93.3
Salmon	344	93.0	28.2	18.3
Sea bass	309	97.4	28.2	-

2) Participant nationalities. Others are a total of 27 participant from 18 different nationalities



3) Mercury concentration in the 65 samples in mg/kg ww.

Hamour local (HM-L), Hamour imported (HM-M), Safi (SF), Shari (SH), Chanad (CH), Tuna local (TU-L), Tuna imported (TU-M), Tuna canned (TU-C), Salmon (SM), Salmon frozen (SM-Z), Salmon canned (SM-C), and Sea bass (SB).

Code	TotHg/ MeHg100%	MeHg ART
HM-L1	0.230	0.224
HM-L2	0.150	0.146
HM-L3	0.209	0.203
HM-L4	0.093	0.090
HM-L5	0.178	0.174
HM-L6	0.089	0.087
HM-M7	0.443	0.431
HM-M8	0.404	0.393
HM-M9	0.372	0.362
SF1	0.002	0.002
SF2	0.0012	0.001
SF3	0.0012	0.001
SF4	0.0010	0.001
SF5	0.0011	0.001
SF6	0.0009	0.001
CH1	0.061	0.059
CH2	0.103	0.100
CH3	0.089	0.087
CH4	0.161	0.157
CH5	0.073	0.071
CH6	0.060	0.059
SH1	0.055	0.051
SH2	0.076	0.071
SH3	0.076	0.072
SH4	0.071	0.067
SH5	0.057	0.053
SH6	0.042	0.039
TU-L1	0.032	0.030
TU-L2	0.037	0.034
TU-L3	0.052	0.049
TU-M4	0.016	0.015
TU-M5	0.020	0.019
TU-M6	0.017	0.017
TU-C1	0.069	0.062
TU-C2	0.071	0.065
TU-C3	0.033	0.030
TU-C4	0.009	0.008
TU-C5	0.255	0.231
TU-C6	0.129	0.116
TU-C7	0.123	0.111
TU-C8	0.060	0.054
TU-C9	0.015	0.013
TU-C10	0.040	0.036
TU-C11	0.059	0.053
TU-C12	0.110	0.100
TU-C13	0.206	0.186
TU-C14	0.072	0.065

TU-C15	0.083	0.075
TU-C16	0.126	0.114
SM1	0.006	0.005
SM2	0.008	0.006
SM3	0.007	0.005
SM4	0.006	0.005
SM5	0.004	0.003
SM6	0.014	0.012
SM-Z1	0.014	0.011
SM-C1	0.022	0.018
SM-C2	0.035	0.028
SM-C3	0.012	0.010
SB1	0.030	0.024
SB2	0.023	0.018
SB3	0.025	0.020
SB4	0.012	0.010
SB5	0.021	0.017
SB6	0.014	0.012