Review

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Heavy metals in marine fish meat and consumer health: a review

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Abstract

The numerous health benefits provided by fish consumption may be compromised by the presence of toxic metals and metalloids such as lead, cadmium, arsenic and mercury, which can have harmful effects on the human body if consumed in toxic quantities. The monitoring of metal concentrations in fish meat is therefore important to ensure compliance with food safety regulations and consequent consumer protection. The toxicity of these metals may be dependent on their chemical forms, which requires metal speciation processes for direct measurement of toxic metal species or the identification of prediction models in order to determine toxic metal forms from measured total metal concentrations. This review addresses various shortcomings in current knowledge and research on the accumulation of metal contaminants in commercially consumed marine fish globally and particularly in South Africa, affecting both the fishing industry as well as fish consumers.

Keywords: heavy metals; fish muscle; consumer health; maximum allowable limits; PTWI; metal speciation

INTRODUCTION

Many populations globally depend on fish as part of their daily diet (FAO, http://www.fao.org/fishery/sofia/en) as fish and seafood are healthy components of human nutrition providing many essential nutrients such as high-value proteins, various vitamins and minerals and polyunsaturated omega-3 fatty acids. In some communities, fish can be a primary food source that contributes substantially to food security (FAO, http://www.fao.org/fishery/sofia/en). Fish and other marine organisms are, however, not independent of the environment in which they live. Both essential and harmful minerals and metals present in the environment can be absorbed into living organisms from the surrounding water, sediment and diet.[1] Even though fish and seafood carry numerous health benefits, contaminants in this food group can also pose a significant threat to the health of consumers. Of the various environmental contaminants, metals and metalloids (which will be discussed hereafter in combination as 'metals') are amongst the most commonly accumulated toxins in fish and seafood which can lead to health defects when consumed in amounts exceeding safe consumption levels.[2,3]

Metal contaminants are naturally present in the environment but can be increased through industrial activity and pollution.[4] The concentrations and uptake of these metals in marine organisms are subject to environmental and species-specific biological factors as well as the chemical and physical state of the metals.[4–6] Canli and Atli[6] have shown that different fish species accumulate metals at different rates and to different levels; that different metals accumulate differently within the same fish species; and also that one specific metal is accumulated at different levels in different tissues within one fish. Therefore it is imperative to consider these factors when determining the consumer safety of fish with regards to metal content.[4,6]

It is important to note that not all metals are hazardous and toxic to fish and humans. They form part of a larger group of elements, some of which are essential to human health.[7] These can therefore be classified as essential, non-essential or toxic. Essential elements which play a specific role in body metabolism include iron (Fe), copper (Cu), zinc (Zn) and selenium (Se). Non-essential elements are elements that have no known specific function in the body, but are also not considered toxic in any significant amount and, lastly, toxic elements such as chromium (Cr), nickel (Ni), cadmium (Cd), mercury (Hg) and lead (Pb) are generally related to pollution and can have harmful effects on living organisms when exceeding certain concentrations. Some elements (e.g. Se) are essential in small quantities or up to certain concentrations above which they can have toxic effects. Schroeder and Darrow[8] have also grouped metals into categories according to toxicity levels as follows: those that easily attain toxic levels [Pb, Ni, antimony (Sb), beryllium (Be), Cd and Hg] and those that can become toxic at extreme levels [barium (Ba), arsenic (As), germanium (Ge) and tungsten (W)]. Several other metals are known to be inert and are considered non-toxic. Regulatory limits and main sources of

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essential, non-essential and toxic metals present in commonly consumed marine organisms are summarised in Table 1.

As individual metals have different degrees of toxicity, maximum allowable limits (MALs) and provisional tolerable weekly intake (PTWI) for metals in foodstuffs are determined specific to each metal for the protection of the consumer.[15] The MALs are specific to food products and provides a limit above which consumers are likely to be exposed to harmful contaminant levels, whereas PTWI represents 'permissible human weekly exposure to metal contaminants unavoidably associated with the consumption of otherwise wholesome and nutritious foods'.[16] These limits can also be species-specific as metal accumulation is affected by different development and metabolic rates of different organisms. Individual countries or governing bodies can have specific MALs that differ from the general regulations as fish consumption patterns of specific population groups are taken into consideration (Table 1).

Although numerous foodstuffs may contain metal contaminants above regulatory limits, marine fish tend to have some of the highest levels where metals such as As, Cd, Hg and Pb predominate.[2,3] Due to frequent high concentrations of these four metals in marine fish and their potential harmful effects to consumers, they will be the metals of focus in the current review.

TOXIC METALS

Arsenic

Arsenic (As) is widely distributed in nature due to environmental sources[17,18] and anthropogenic pollution which is largely due to smelting activities, glass manufacturing, manufacture and use of arsenic pesticides, herbicides, fungicides and wood preservatives.[17,19,20] Arsenic has a complex chemistry and can be present in several organic (trivalent and pentavalent arsenic) and inorganic (elemental, trivalent and pentavalent arsenic) forms which vary in their degree of toxicity. Inorganic As is seen as the most toxic form as it is stable and soluble and therefore absorbed by the digestive tract, abdominal cavity and muscles in the human body,[18] whilst organic As does not accumulate in the human body due to rapid excretion.[17,18] Inorganic As is often found in high levels in drinking water whereas organic As is primarily found in fish and meat.[17,19] Seafood can contain several times the amount of As than other foods and is therefore the main source of dietary intake in humans.[2,21] Although high concentrations of As (up to 100 ppm) have been found in certain edible marine species (Table 2),[18,22–25] in most of these cases it is the total As concentrations that are measured instead of the toxic inorganic form (arsenite). Up to 90% of As in fish muscle is present in the non-toxic arsenobetain form.[17,45] Nonetheless, total As concentration is the current standard whereby regulatory limits are set at 3.0 mg kg^{-1} in fish and processed fish by the South African Department of Health.[9] Measuring individual As species will produce more accurate results in terms of the true As toxicity in seafood as a PTWI of 15 µg kg^{-1} body weight has been set for inorganic As.[3] Early symptoms of As exposure in humans include abdominal pain, vomiting, diarrhoea, muscle weakness and skin flushing whereas chronic As toxicity has led to skin defects and cancer.[8,18]

Cadmium

Cadmium (Cd) is a metal contaminant which is introduced into the environment through both natural processes (volcanic emissions and weathering of rocks) and anthropogenic activities such as the smelting of other metals, burning of fossil fuels, incineration of waste materials and the use of certain fertilisers.[32] Cadmium is most commonly found as inorganic compounds in the 2+ oxidation state and is mainly present as [$CdCl_2{}^0$] and [$CdCl^+$] complexes in seawater.[46] Cadmium can readily cross various biological membranes, and once inside living cells, has a high affinity to bind to ligands and form Cd complexes which can be more stable.[32] For example, in fish muscle most of the Cd present tends to bind to proteins.[32] Cadmium absorbed into the fish body is therefore eliminated at a very slow rate, causing bioaccumulation in the body. Cadmium can enter fish by passive diffusion across the gills or by entering the marine food chain at the plankton and microorganisms level and thereby entering fish through the diet.[4] As Cd is most readily taken up by aquatic organisms in its free form (Cd^{2+}), the high salinity in seawater which causes Cd to readily form complexes ([$CdCl_2{}^0$] and [$CdCl^+$]) seems to reduce this bioaccumulation.[6] Nonetheless, fish is still considered a major source of Cd,[19] which has frequently been found to exceed maximum allowable limits in a number of commonly consumed fish species (Table 2).

Cadmium is highly toxic to humans and has a long biological half-life preventing the reduction of the accumulated body burden.[4,32] Effects on human health include hypertension and cardiovascular function, neurological disorders, carcinogenic effects and skeletal weakness and defects.[8,17] Cadmium exposure in humans is predominantly through food ingestion[19] where fish, meat and fruit can contain 1–50 µg kg^{-1} Cd.[17] The European Commission has set a PTWI of 7 µg kg^{-1} body weight (BW) and MAL as seen in Table 1.[13] The Food and Agriculture Organization of the United Nations presents species-specific maximum limits for Cd in fish from 0.05 mg kg^{-1} fresh weight in fishery products to 1.0 mg kg^{-1} fresh weight in bivalves and cephalopods.[12] Within South Africa, the Department of Health's regulatory limit for Cd in fish and processed fish is 1.0 mg kg^{-1}.[9]

From a survey across 18 European Union (EU) Member States, Iceland, Australia and three commercial organisations, 4.8% ($n = 305$), 8.2% ($n = 102$) and 2.0% ($n = 7$) of all samples from three respective categories of fish species had Cd levels exceeding the maximum limits in fish muscle according to FAO and EU regulations.[32] Even though Cd is a common contaminant in edible fish meat, how and where (muscle, bone, gills and organs) Cd is accumulated in marine fishes is not homogenous[47] and therefore needs to be investigated in a wide variety of fish species in order to determine the true danger that Cd poses to the fish consumer.

Lead

Lead is one of the primary contaminants present in the environment[8,19] and naturally occurs in rocks, soils and in the hydrosphere.[48] However, Pb is also the most widely used metal and industrial Pb contributes a considerable quantity to that found in the natural environment.[49] Large amounts of lead tetraethyl can be completely converted to aerosols through the combustion of gasoline, subsequently contributing to atmospheric Pb.[50,51] The atmosphere, in turn, is the main source of Pb deposition in the marine environment, therefore acting as a Pb pathway from the terrestrial to the marine environment. Since it became evident that leaded petrol was the predominant source of atmospheric lead,[50] regulations were adopted on the allowable gasoline lead content.[51] This reduction in anthropogenic lead pollution was evident in a reduction in seawater lead concentrations[50] forming a direct link from terrestrial sources to effects in the marine environment. Once in the marine environment, Pb is easily absorbed

Table 1. Maximum allowable limits (MALs) with specifications for individual metals in fish by various regulatory bodies **mg kg^{-1}**

Metal	MAL	Regulatory body	Specifications
Essential but toxic in excess amounts			
Sn	50 mg kg^{-1}	DOH, 2004[9]	For all uncanned meat and meat products
Fe	–	–	–
Cu	30 mg kg^{-1}	FAO, 1983	
	20 mg kg^{-1}	UK, Spain[10]	–
	5 mg kg^{-1}	Turkey[10]	
Cr	0.1 mg kg^{-1}	Brazil Standard[11]	–
Zn	30 mg kg^{-1}	FAO, 1983	–
	50 mg kg^{-1}	Turkey[10]	
Se	0.3 mg kg^{-1}	–	–
Toxic			
As	3.0 mg kg^{-1} g	DOH, 2004[9]	Fish and processed fish meat
	2.0 mg kg^{-1}	Australia New Zealand Food Authority, 2011[10]	–
Sb	0.15 mg L^{-1}	DOH, 2004[9]	All liquid foodstuffs
Cd	0.05 mg kg^{-1} 0.1 mg kg$^{-1,a}$	FAO. Heavy Metals Regulations Legal Notice No 66/2003[12] Commission Regulation (EC) No 1881/2006[13]	[a] For the following species: bonito (*Sarda sarda*), wedge sole (*Dicologoglossa cuneata*), eel (*Anguilla Anguilla*), European anchovy (*Engraulis encrasicholus*), louvar/luvar (*Luvarus imperialis*), horse mackerel or scad (*Trachurus trachurus*), grey mullet (*Mugil labrosus labrosus*), common two-banded seabream (*Diplodus vulgaris*), European pilchard or sardine (*Sardina pilchardus*), mackerel (*Scomber* species), sardinops (*Sardinops* species), tuna (*Thunnus* species, *Euthynnus* species, *Katsuwonus pelamis*)
	1.0 mg kg^{-1}	DOH, 2004[9]	Fish and processed fish
	0.3 mg kg^{-1}	Commission Regulation (EC) No 1881/2006[13]	Muscle meat of swordfish
	0.2 mg kg$^{-1,b}$ 0.3 mg kg$^{-1,bb}$	Commission regulation (EC) No 629/2008[14]	[b] bullet tuna (*Auxis* species) [bb] anchovy (*Engraulis* species) swordfish (*Xiphias gladius*)
Hg	1 mg kg$^{-1,c,1}$ 0.5 mg kg$^{-1,ccc1}$	DOH, 2004[9]	[c] Predatory fish including swordfish [ccc] All other fish and processed fish [1] As methylmercury
		FAO. Heavy Metals Regulations Legal Notice No 66/2003[12] Commission regulation (EC) no 629/2008 [14, cc]	[c] Anglerfish (*Lophius*), Atlantic catfish (*Anarhichas lupus*), Bass (*Dicentrarchus labrak*), Blue ling (*Molva dipterygia*), Bonito (*Sarda* spp.), Eel (*Anquilla* spp.), Halibut (*Hippoglossus hippoglossus*), Little tuna (*Euthunnus* spp.), Marlin (*Makaira* spp.), Pike (*Esox lucius*), Plain bonito (*Orcynopsis unicolor*), Portuguese dogfish (*Centroscymnes coelolepis*), Rays (*Raja* spp.), Redfish (*Sebastes marinus, S. mentella, S. uiviparus*), Sail fish (*Istiophoms platypterus*), Scabbard fish (*Lepidopus caudatus, Aphanopus carbo*), Shark (all species), Snake mackerel or butterfish (*Lepidocybium flavobrunneum, Ruvettus pretiosus, Gempylus serpens*), Sturgeon (*Acipenser* spp.), Swordfish (*Xiphias gladius*), Tuna (*Thunnus* spp.). [cc] add: emperor, orange roughy, rosy soldierfish (*Hoplostethus species*), grenadier (*Coryphaenoides rupestris*), kingklip (*Genypterus capensis*), megrim (*Lepidorhombus species*), mullet (*Mullus species*), pink cusk eel (*Genypterus blacodes*), poor cod (*Tricopterus minutes*), seabream, pandora (*Pagellus* species). [ccc] edible parts of the fishery products

Table 1. Continued

Metal	MAL	Regulatory body	Specifications
Pb	0.5 mg kg^{-1}	DOH, 2004[9]	Fish and processed fish
	0.20 mg kg$^{-1,f}$ 0.4 mg kg$^{-1,ff}$	FAO. Heavy Metals Regulations Legal Notice No 66/2003[12]	[f] Edible parts of the fishery products [ff] Wedge sole (*Dicologoglossa cuneata*), Eel (*Anguilla anguilla*), Spotted seabass (*Dicentrarchus punctatus*), Horse mackerel or Scad (*Trachurus trachurus*), grey mullet (*Mugil labrosus labrosus*), Common two-banded seabream (*Diplodus vulgaris*), Grunt (*Pomadasys benneti*), European pilchard or sardine (*Sardina pilchardus*)
	0.3 mg kg$^{-1,d}$	Commission regulation (EC) No 1881/2006[13]	Muscle meat of fish

into the fish's bloodstream and accumulated in the body tissues, bones, gills, kidneys, liver and scales.[52] It can thus enter the human body through the diet and can accumulate, especially when seafood is consumed regularly.

The toxicity of Pb is dependent on its chemical form[4,17] where the organolead compounds are more toxic than the inorganic Pb form.[1] Lead is mostly found in its dissolved form in the ocean, of which a large proportion (50–70%) is organic compounds.[50] As was shown by a series of studies by Sánchez-Marín *et al.*,[53–55] the bioavailability of Pb in the environment as organic compounds can be significantly increased by the presence of dissolved organic matter (DOM). The more methyl or ethyl carbon groups linked to the Pb molecule, the higher its toxic effect.[1] The marine environment is therefore a significant source of toxic Pb exposure in fish and humans due to consumption (Table 2). In certain communities fish consumption is the main source of Pb exposure[56] where excess exposure can result in neurological problems, haematological effects, renal failure, hypertension and cancer.[1,17] A PTWI of 50 µg kg^{-1} BW was first set by the Expert Committee on Food Additives and Contaminants (JECFA), which was replaced in 1993 by a new PTWI of 25 µg kg^{-1} BW for all age groups.[57] At present, according to the South African Department of Health,[9] the MAL for Pb in fresh and processed fish is 0.5 mg kg^{-1} with a MAL of 0.3 mg kg^{-1} set by the European Commission (Table 1).[13]

Mercury

Mercury (Hg) is a metal that is liquid at ambient temperature and pressure and can be present in several different chemical forms and compounds in the environment. It is the metal that presents the most concern with regards to fish and seafood consumption and human health[43] and will thus be reviewed in more detail. Fish is considered the primary source of Hg in humans[3,58] and there are numerous reports (examples in Table 2) of high levels of Hg in fish muscle, exceeding the allowable maximum limits.

Sources

Mercury levels in the environment have increased markedly since the early 20th century due to both natural processes and human activity.[59] Natural Hg sources include forest fires and volcanic activity;[60] however, one- to two- thirds of the Hg present in the atmosphere and aquatic environment is from anthropogenic origin.[60,61] Mercury is used for the production of paint, electrical equipment, batteries and fungicides as well as in medicine, dentistry, wood pulping and the military sector.[61] In addition, mining contributes significantly to Hg water pollution whilst the burning of fossil fuels and the smelting of Pb, Cu and Zn ores are major sources of atmospheric Hg pollution.[61] Due to increasing awareness of Hg-related health hazards, the use of Hg in many industries and consequently atmospheric Hg pollution has diminished in recent years.[59] However, current environmental Hg levels are still 10 times higher than in pre-industrial times.[59]

Due to anthropogenic input from various activities, seawater, sediments and biota near cities, harbours and industrial areas tend to have higher Hg concentrations compared to rural locations.[62] A number of marine-based studies have corroborated such claims where black-mouthed dogfish, carp spp. and catfish, for example, had overall higher Hg concentration when sampled from industrialised and developed sites compared to those areas considered rural, less developed and/or clean.[63–65] Rivers also carry metal contaminants from inland industrial and agricultural sources towards the ocean, affecting marine fish in estuaries and near river mouths.[66]

Chemistry and accumulation of mercury species

Mercury consists and is present in the environment in several chemical forms, each displaying different characteristics (mobility and toxicity).[67] Elemental Hg (Hg^0) and mercuric ions (Hg^{2+}) are the predominant natural forms in the environment and generally do not accumulate in fish.[61] Although not directly accumulated, elemental Hg is easily vapourised and transported through the atmosphere, providing circulation of Hg from land sources to the oceans[61] where it can be converted into other more soluble chemical forms (inorganic and organic Hg). The toxicity of these Hg compounds is dependent on their chemical form, which affects their ability to be accumulated and excreted from the fish and human body.[68,69] Organic Hg compounds are considered toxic as they are more stable and are more readily accumulated in fish tissue and in the human body whereas inorganic Hg compounds are considered non-toxic as they are accumulated in fish tissue in much lower concentrations and have a high rate of excretion from the human body and is therefore not accumulated to quantities at which it becomes toxic and negatively affects the human body.[60,61] Inorganic Hg include compounds such as mercuric chloride ($HgCl_2$), mercurous chloride (Hg_2Cl_2), mercuric acetate ($HgC_4H_6O_4$) and mercuric sulfide (HgS) which is the most common form in nature, but is also insoluble.[70] Even though these inorganic forms are considered non-toxic and some of them insoluble, they can be methylated in the environment to form organic Hg compounds, such as methylmercury (MeHg),[71] which are considered toxic. This methylation occurs either by a photochemical reaction (photomethylation) or a process catalysed

Table 2. Summary of cases where fish and fish products contain Pb, Cd, As and Hg concentrations exceeding the respective maximum allowable limits (MALs) as per study region

Metal > MAL	Fish/fish products sampled	Metal concentration (mg kg^{-1})	Country/region
As	Flathead sole[22]	19.5 ± 1.01	Aleutian Islands
	Rock sole[22]	4.34 ± 0.70	Aleutian Islands
	Horse mackerel[23]	6.85 ± 6.22	Croatia
	Sardine[23]	8.08 ± 2.43	Croatia
	Hake[23]	10.03 ± 0.82	Croatia
	Hake[23]	23.30 ± 3.56	Croatia
	Red mullet[26]	59.91 ± 9.49	Italy
	European hake[26]	38.70 ± 7.69	Italy
	Blue whiting[26]	35.30 ± 2.82	Italy
	Atlantic mackerel[26]	30.76 ± 9.95	Italy
Cd	Canned tuna[27]	0.06	Jordan
	European conger eel[28]	0.11 ± 0.01	Italy
	Blackbellied angler[28]	0.09 ± 0.02	Italy
	Rosefish[28]	0.10 ± 0.02	Italy
	Brown ray[28]	0.08 ± 0.04	Italy
	Red mullet[28]	0.08 ± 0.04	Italy
	European pilchard[29]	0.045 ± 0.020	Sicily
	Red mullet[29]	0.084 ± 0.069	Sicily
	Red mullet[30]	0.053 ± 0.027	Italy
	Salted anchovies[31]	0.06 to 0.61	Italy
	Various species[32]	0.092 ± 0.267	EU Member States, Iceland and Australia
	Louvar[33]	0.08 ± 0.01	Spain
	Albacore[34]	0.05 ± 0.03	Mediterranean Sea
	Grey mullet[35]	0.10 to 0.40	Turkey
	Atlantic mackerel[36]	0.49 ± 0.01	Nigeria
	European pilchard[36]	0.19 ± 0.0001	Nigeria
	Blue whiting[26]	0.10 ± 0.06	Italy
	European hake[26]	0.05 ± 0.04	Italy
	Red mullet[26]	0.07 ± 0.05	Italy
Pb	Rudd[37]	4.31	Bulgaria
	Algae[38]	–	New Jersey, USA
	Salmon[39]	0.4	Lithuania
	European anchovy[29]	0.32 ± 0.22	Sicily
	Canned sardines[11]	2.15 ± 0.85	Brazil
	Rednose labeo[40]	0.8	South Africa
	Atlantic mackerel[36]	0.46 ± 0.02	Nigeria
Hg	Shortfin mako[41]	2.65 ± 1.16	New England
	Common thresher[41]	0.88 ± 0.71	New England
	Albacore tuna[41]	0.46 ± 0.14	New England
	Yellowfin tuna[41]	0.30 ± 0.09	New England
	Dolphinfish[41]	0.21 ± 0.17	New England
	European conger eel[28]	1.14 ± 0.46	Italy
	Rosefish[28]	1.04 ± 0.56	Italy
	Brown ray[28]	1.09 ± 0.39	Italy
	Blackbellied angler[28]	0.96 ± 0.32	Italy
	Red mullet[28]	0.43 ± 0.55	Italy
	Shortfin mako[42]	1.83 ± 0.17	New Jersey
	Atlantic bluefin tuna[42]	0.52 ± 0.03	New Jersey
	Striped bass[42]	0.39 ± 0.02	New Jersey
	Bluefish[42]	0.35 ± 0.02	New Jersey
	Swordfish[33]	0.93 ± 0.07	Spain
	Louvar[33]	0.99 ± 0.06	Spain
	Albacore[34]	1.56 ± 0.49	Mediterranean Sea
	Blue whiting[43]	–	Italy
	Atlantic horse mackerel[43]	–	Italy
	Bullet tuna[43]	–	Italy
	European hake[44]	–	Italy

Table 2. Continued

Metal > MAL	Fish/fish products sampled	Metal concentration (mg kg^{-1})	Country/region
	Spiny dogfish[43]	6.53 ± 2.19	Italy
	Small-spotted catshark[43]	–	Italy
	Thornback ray[43]	–	Italy
	Blackbellied angler[43]	–	Italy
	Sandy ray[43]	–	Italy
	Brown ray[43]	–	Italy
	Mediterranean starry ray[43]	–	Italy
	Silver scabbardfish[43]	–	Italy
	Dogtooth tuna[44]	0.38–4.40	Seychelles
	Bonito[44]	0.07–1.26	Seychelles
	Carangue balo[44]	0.03–1.51	Seychelles
	Becune[44]	0.26–1.58	Seychelles
	Kingfish[44]	0.06–1.46	Seychelles
	European hake[26]	0.59 ± 0.14	Italy

by microorganisms such as bacteria in the sediment[63] or in the gills or gut of fish themselves.[61] Sulfate-reducing bacteria have proven to be responsible for the bulk of Hg methylation in natural waters.[60] Other organic Hg forms include dimethylmercury (DMHg) and ethylmercury (EthHg). Dimethylmercury is unreactive because its carbon–metal bonds are stable in water and it is therefore not absorbed into the food chain, except if partial demethylation of DMHg occurs, in which case it can then be absorbed as MeHg complexes (usually CH_3HgCl and CH_3HgOH).[60] EthHg is also considered a toxic organic form of Hg, but is not significantly absorbed and accumulated in fish tissue.[72] Methylmercury is therefore the main chemical form absorbed into the food chain and also the most toxic. MeHg is passed easily across cell membranes as it is a stable organometallic compound, and has a high affinity for the sulfhydryl groups of amino acids[20,63] and is therefore easily absorbed into and bioaccumulated up the marine food chain.[17,73] The average proportion of MeHg to total Hg increases from approximately 10% in the water column to 15% in phytoplankton, 30% in zooplankton and 95% in fish flesh.[74] MeHg generally accounts for 75 to 100% of the total Hg present in most fish species.[75] In the current review the term 'mercury' (Hg) will refer to total Hg (tHg) which is the sum of the inorganic Hg (iHg), MeHg, EthHg and any other Hg forms present.

Due to the significant role of diet in Hg accumulation,[76,77] fish at higher trophic levels are more likely to be exposed to and accumulate higher levels of Hg than those at lower trophic levels.[62,78] This process of Hg accumulation up the food chain is referred to as bioaccumulation.[75,79] In addition, Hg can also be biomagnified within a single species with older/larger individuals having higher levels of accumulated Hg.[61,80] Methylmercury has a longer half-life than inorganic Hg resulting in a strong correlation between the percentage of total Hg present as MeHg and the total Hg levels,[81] therefore, the percentage of total Hg present as MeHg tends to approach 100% with increasing total Hg burden and fish size/age.[81]

Distribution

Bioavailable Hg is primarily found in the muscular tissue of fish, hence its risk to consumer health as this part is most frequently consumed.[82] More specifically, Hg is known to be associated with the protein fraction of the muscle as it binds to thiol group complexes.[68,82,83] The protein distribution (protein type and concentration) within a fish carcass and how it varies for example between white and dark muscle could therefore provide a link to the nature of Hg accumulation and distribution across the carcass.

Most large predatory fish such as tuna and shark have distinct muscle groups which are categorised as either white or dark muscle. These individual muscle groups have distinct functions (either fast, strong muscle movement or slow, continuous muscle movement) and distributions across the carcass.[84,85] The function and location of a muscle can affect the rate of development and composition of the muscle cells.[86] Therefore, as metals are stored in muscle cells,[87] these differences in muscle cell development and composition could in turn influence the rate and degree of metal accumulation within the muscle. Several studies have found variation of Hg accumulation within carcasses of tuna fish,[82,88,89] but reasons for Hg variation have not been clearly identified.[89] Some authors suggest that lipid concentrations might have a diluting effect on accumulated Hg;[83] however, the Hg and lipid content relationship was insignificant in cultured Bluefin tuna and wild Albacore tuna.[83,90] These studies measured total Hg concentrations and, as far as the authors are aware, no studies have been published on variation in the accumulation of individual Hg species and therefore variation in Hg toxicity across the fish carcass.

Accumulation and effects in the human body

Methylmercury is the main stable organic form of Hg that is taken up by the human body via seafood consumption. More than 95% of MeHg ingested is absorbed from the intestinal tract after consumption[91] and is then distributed to all tissues and target organs via the bloodstream. MeHg readily crosses the blood–brain barrier,[69,92] resulting in significant deposition (about 10% of the total Hg burden) in the brain region.[93] The accumulation of MeHg in the brain causes loss of cells in specific brain areas such as the cerebellum, visual cortex and other focal areas.[92] Other main target organs include the pituitary gland, liver and kidney.[17,69] Methylmercury readily crosses the placental barrier subsequently affecting the neurological development in developing foetuses.[69,92]

Symptoms of MeHg intoxication in humans include impaired vision and hearing, headaches, paraesthesia, movement difficulties and loss of coordination, fatigue, tremors and ataxia.[59]

Low-level exposure of MeHg can adversely affect the cardiovascular system whereas chronic Hg exposure impacts the pituitary gland and the liver and leads to a compromise of the immune system.[91] Children exposed to Hg prenatally often show delays in the development of their speech and motor functions.[59,94] The onset of these various symptoms can take up to a few months from the time of Hg exposure or ingestion. This is especially dangerous to pregnant women as doses of one-fifth the toxic dose to adults could have adverse effects on the developing nervous system of a foetus or child[59] and a high Hg intake can therefore affect the foetus before any signs of Hg poisoning are visible in the mother.

Rather than limiting fish intake, attention should be focused on determining which fish are safe for consumption and which should be avoided in regards to Hg levels as seafood is the main source of polyunsaturated fatty acids such as eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA),[95] which have major beneficial effects on human health and neurocognitive development (especially DHA).[96,97] It has been recommended that high-trophic-level predatory fish such as shark, swordfish, king mackerel, tilefish and albacore should be avoided or consumed in smaller quantities (FDA, www.cfsan.fda.gov). Advisory committees such as the United States Food and Drug Administration (FDA) have published recommendations for safe fish consumption (FDA, www.cfsan.fda.gov) and should be consulted when consuming seafood.

Disposal of Hg from the body is a slow process and occurs mainly via the faecal route. MeHg is secreted in the bile from where a fraction is reabsorbed in the gallbladder and gastrointestinal tract.[69] Some secretion may also occur across the intestinal membrane as intestinal flora in the gastrointestinal tract are capable of breaking the carbon–Hg bond, converting MeHg to iHg which is poorly absorbed and is then mostly excreted in the faeces.[69] Mercury intake should therefore be limited and monitored in order to prevent toxic build-up of Hg which occurs when the amount of Hg absorbed exceeds that being excreted.

The maximum tolerable weekly intake for Hg, as recommended by the Expert Committee on Food Additives and Contaminants (JECFA) under the joint Food and Agriculture Organization (FAO) and the World Health Organization (WHO), as part of the international safety guidelines, is 1.6 $\mu g\ kg^{-1}$ BW[13] which replaces the previous PTWI of 3.3 $\mu g\ kg^{-1}$ BW.[98] The first regulatory MALs for Hg in seafood were set as 0.5 $mg\ kg^{-1}$ fish meat, except for large predatory species, which were found to frequently exceed this limit and therefore only had to comply to a Hg MAL of 1 $mg\ kg^{-1}$ according to the European Commission.[13,59] However, these limits were set for Hg and not MeHg and it is known that the latter is more toxic to humans. The South African Department of Health now requires this limit of 1 $mg\ kg^{-1}$ fish meat as specifically for MeHg.[9]

Analysis of mercury

In order to monitor the compliance of commercial fish meat to Hg maximum limit regulations, accurate and efficient analytical methods are required. A number of methods that include atomic fluorescence spectrometry (AFS), various forms of atomic absorption spectrometry (AAS), inductively coupled plasma atomic emission spectrometry (ICP-AES) and inductively coupled plasma mass spectrometry (ICP-MS) have been developed for measuring total Hg in seafood and are currently widely used.[99] South African regulations require Hg analysis to be done by cold vapour atomic absorption spectrometry (CVAAS).[100] However, in order to accurately monitor levels of toxic Hg in fish meat,[101,102] metal speciation techniques should be used.

Metal speciation, is defined by Florence[103] as 'the determination of the concentrations of the individual physico-chemical forms of the element in a sample that together, constitute its total concentration' (p. 345). The analytical techniques used for Hg speciation are combinations of separation techniques such as gas or liquid chromatography and detection techniques such as CVAAS, inductively coupled plasma mass spectrometry (ICP-MS), flame atomic absorption spectrometry (FAAS), electron-capture detection (ECD), cold vapour atomic fluorescence spectrometry (CVAFS) or atomic emission spectrometry (AES).[104–106] These separation and detection techniques can be used in different combinations with the choice of speciation technique depending on the performance priorities and requirements, as each technique has its own strengths and advantages.[107]

For these aforementioned speciation analyses, all analytical steps need to be properly planned from sample preparation to species separation and detection in order to ensure that all the Hg species in the samples analysed remain in their original form and none are lost or changed along the process. Although the total Hg content of a sample is stable and cannot be reduced through losses during processing steps, individual Hg species can be interconverted between organic and inorganic forms affecting measurement results of Hg toxicity.[108] Little is known about the stability of Hg species in biological samples during sample storage, but fresh samples are usually deep frozen or lyophilised in darkness for storage before analysis.[107] Sample preparation/digestion is a critical step as the analyte needs to be fully extracted from the sample matrix, but without losses, contamination or changes in species.[106] Subsequent to metal extraction, the individual elemental species need to be separated as cleanly as possible prior to the detection process.[106] Current metal speciation techniques are costly and time consuming, which is not beneficial for routine monitoring in the industry as analytical results on Hg toxicity of fish samples should be obtained before entire batches of fish are distributed onto the market.[109]

The fish and seafood industry is still in need of a time and cost effective, accurate way of determining levels of toxic MeHg for a true measurement of food safety for human consumption. The ratio of the total Hg burden present as MeHg can vary with more than 30% within one species and average ratios vary from approximately 50–100%.[81] It is therefore clear that a fixed conversion factor will not provide accurate estimates of toxic Hg concentrations in health assessments. There are, however, significant correlations between the percentage of total Hg present as MeHg and the total Hg concentrations as well as between total Hg concentrations and fish size/age.[81] Further research should therefore further explore these relationships for the possibility of setting up a model for calculating toxic MeHg levels from total Hg measurements.

HEAVY METALS IN SEAFOOD

Hg accumulation in both marine and freshwater fish has been widely studied (a summary is given in Table 3) as a result of an increased focus on Hg poisoning and its toxic effects due to a number of large-scale human poisoning incidences.[59,126] These studies cover a wide variety of fish species and aquatic organisms, not all of which are commercially consumed, as some are merely used as biomonitors of environmental pollution.

Even though a large part of the current review has focused on Hg as a main contaminant, the possible hazardous effects of other metals should not be disregarded. The accumulation and effects of individual metals are not always independent of each other and

Table 3. A summary of studies on mercury and methylmercury in a variety of organs/muscles in fish from various continents or seas per trophic level

Trophic level	Continent/sea	Species	Marine/ freshwater	Mercury levels (ppm) (mean ± SD)					
				Liver	Gills	White meat	Red meat	Overall meat	MeHg
Apex predators	Africa	Smoothhound (*Mustelus mustelus*)[110]	Marine	–	–	–	–	0.9	–
	Atlantic Islands	Blue Shark (*Prionace glauca*)[111]	Marine	0.03–0.96	–	–	–	0.22–1.30	–
	Atlantic Islands	Blue Shark (*P. glauca*)[111]	Marine	0.15–2.20	–	–	–	0.68–2.50	–
	Atlantic Islands	Swordfish (*Xiphias gladius*)[111]	Marine	0.05–8.50	–	–	–	0.03–2.40	–
	Atlantic Islands	Swordfish (*X. gladius*)[111]	Marine	1.10–9.80	–	–	–	0.90–2.10	–
	Australia/NZ	Southern Bluefin Tuna (farmed) (*Thunnus maccoyii*)[82]	Marine	–	–	0.32 ± 0.03	–	–	–
	Australia/NZ	Dog Shark (*Deania calcea*)[112]	Marine	–	–	–	–	7.2 ± 2.3	–
	Australia/NZ	Dog Shark (*Centroscymnus crepidater*)[112]	Marine	–	–	–	–	4.3 ± 2.4	–
	Australia/NZ	Dog Shark (*Centroscymnus owstonii*)[112]	Marine	–	–	–	–	11.9 ± 1.1	–
	Australia/NZ	School Shark (*Galeorhinus australis*)[113]	Marine	–	–	–	–	0.9	–
	Australia/NZ	Gummy Shark (*Mustelus antarcticus*)[113]	Marine	–	–	–	–	0.37	–
	East Asia	Swordfish[114]	Marine	–	–	–	–	0.50 ± 0.01	0.41 ± 0.02
	East Asia	Dall's Porpoise[115]	Marine	–	–	–	1.26 ± 0.53	–	–
	East Asia	Baird's Beaked Whale[115]	Marine	–	–	–	1.64 ± 1.26	–	–
	East Asia	Pantropical Spotted Dolphin[115]	Marine	–	–	–	4.72 ± 0.39	–	–
	East Asia	Risso's Dolphin[113]	Marine	–	–	–	5.42 ± 4.68	–	–
	East Asia	Rough-toothed Dolphin[115]	Marine	–	–	–	6.00	–	–
	East Asia	Pilot Whale[115]	Marine	–	–	–	7.59 ± 6.12	–	–
	East Asia	Bottlenose Dolphin[115]	Marine	–	–	–	9.55 ± 6.01	–	–
	East Asia	Striped Dolphin[115]	Marine	–	–	–	15.0 ± 27.1	–	–
	East Asia	False Killer Whale[115]	Marine	–	–	–	46.9 ± 29.7	–	–
	East Asia	Tiger Shark (*Galeocerdo cuvier*)[116]	Marine	1.17 ± 3.14	–	–	–	0.78 ± 0.29	–
	East Asia	Silvertip Shark (*Carcharhinus albimarginatus*)[116]	Marine	0.70 ± 0.42	–	–	–	1.80 ± 0.45	–
	Mediterranean	Tuna[117]	Marine	–	–	–	–	0.48	–
	Mediterranean	Swordfish[117]	Marine	–	–	–	–	1.93	–
	Mediterranean (Adriatic Sea)	Blackmouth Dogfish (*Galeus melastomus*)[63]	Marine	–	–	–	–	2.66 ± 1.24	2.11 ± 0.96
	Mediterranean (Ionian Sea)	Blackmouth Dogfish (*G. melastomus*)[63]	Marine	–	–	–	–	0.82 ± 0.62	0.74 ± 0.52
	Mediterranean (Aegean Sea)	Blackmouth Dogfish (*G. melastomus*)[63]	Marine	–	–	–	–	2.14 ± 1.44	1.55 ± 1.23

Table 3. Continued

Trophic level	Continent/sea	Species	Marine/ freshwater	Mercury levels (ppm) (mean ± SD) Liver	Gills	White meat	Red meat	Overall meat	MeHg
	Mediterranean	Small Spotted Shark (*Scyliorhinus canicula*)[63]	Marine	–	–	–	–	1.49 ± 0.61	1.23 ± 0.49
	Mediterranean	Kitefin Shark (*Dalatias licha*)[63]	Marine	–	–	–	–	4.38 ± 1.07	3.81 ± 0.69
	Mediterranean	Gulper Shark (*Centrophorus granulosus*)[63]	Marine	–	–	–	–	9.66 ± 0.69	0.09 ± 0.83
	Mediterranean	Longnose Spurdog (*Squalus blainvillei*)[63]	Marine	–	–	–	–	4.53 ± 1.19	4.05 ± 1.29
	Mediterranean	Velvet Belly (*Etmopterus spinax*)[63]	Marine	–	–	–	–	0.63 ± 0.29	0.58 ± 0.26
	Mediterranean	Smoothhound (*M.mustelus*)[63]	Marine	–	–	–	–	0.31 ± 0.06	0.23 ± 0.05
	Mediterranean	Sharpnose Sevengill (*Heptranchias perlo*)[63]	Marine	–	–	–	–	1.20 ± 0.17	1.20 ± 0.17
	Mediterranean	Hammerhead (*Sphyrna zygaena*)[63]	Marine	–	–	–	–	18.29 ± 0.03	16.06 ± 0.04
	Pacific Islands	Yellowfin Tuna[118]	Marine	–	–	–	–	0.21 ± 0.11	–
	South America	Scalloped Hammerhead (*Sphyrna lewini*)[65]	Marine	–	–	–	–	4.84	–
	South America	Catfish (*Galeichthys peruvianus*)[65]	Marine	–	–	–	–	1.58	–
	South America	Blue Shark (*Prionace glauca*)[119]	Marine	–	–	–	–	0.05 ± 0.03	–
	South America	Mako Shortfin Shark (*Isurus oxyrinchus*)[119]	Marine	–	–	–	–	0.03 ± 0.02	–
	South America	Brazillian Sharpnose Shark (*Rhizoprionodon lalandii*)[120]	Marine	–	–	–	–	0.20 ± 0.16	–
	USA	Swordfish[58]	Marine	–	–	–	–	1.07	–
	USA	Shark[58]	Marine	–	–	–	–	0.96	–
	USA	Bull Shark (*Carcharhinus leucas*)[121]	Marine	–	–	–	–	0.77 ± 0.32	–
	USA	Blacktip Shark (*Carcharhinus limbatus*)[121]	Marine	–	–	–	–	0.77 ± 0.71	–
	USA	Atlantic Sharpnose Shark (*Rhizoprionodon terraenovae*)[121]	Marine	–	–	–	–	1.06 ± 0.63	–
	USA	Bonnethead Shark (*Sphyrna tiburo*)[121]	Marine	–	–	–	–	0.50 ± 0.36	–
	USA	Tuna (canned)[75]	Marine	–	–	0.41 ± 0.17	–	–	–
	New England	Shortfin mako shark[41]	Marine	–	–	–	–	2.65 ± 1.16	–
	New England	Thresher shark[41]	Marine	–	–	–	–	0.87 ± 0.71	–

Table 3. Continued

Trophic level	Continent/sea	Species	Marine/ freshwater	Mercury levels (ppm) (mean ± SD) Liver	Gills	White meat	Red meat	Overall meat	MeHg
	New England	Albacore tuna[41]	Marine	–	–	–	–	0.45 ± 0.14	–
	New England	Yellowfin tuna[41]	Marine	–	–	–	–	0.32 ± 0.09	–
	New England	Dolphinfish[41]	Marine	–	–	–	–	0.20 ± 0.17	–
Mid-trophic level species	Africa (u'Mgeni River)	Sharp Toothed Catfish (*Clarias gariepinus*)[66]	Freshwater	–	–	–	–	0.4	–
	Africa	Atlantic Mackerel[122]	Marine	–	–	–	–	0.116 ± 0.070	–
	Africa	Atlantic Bonito[122]	Marine	–	–	–	–	0.064 ± 0.180	–
	Africa	European Conger[122]	Marine	–	–	–	–	0.049 ± 0.002	–
	Africa (Inanda Dam)	Sharp Toothed Catfish (*Clarias gariepinus*)[66]	Freshwater	–	–	–	–	0.2	–
	Africa (Nagle Dam)	Sharp Toothed Catfish (*Clarias gariepinus*)[66]	Freshwater	–	–	–	–	0.14	–
	Africa (Orange River)	Sharptooth catfish[123]	Freshwater	–	–	–	–	0.73 ± 0.02	–
	Africa (Vaal River)	Sharptooth catfish[123]	Freshwater	–	–	–	–	0.05 ± 0.01	–
	South America (Guaymas Harbour)	Striped Mullet (*Mugil cephalus*)[65]	Marine	–	0.07 ± 0.00	–	–	–	–
	South America (Altata-Ensenadadel Pabellón)	Striped Mullet (*Mugil cephalus*)[65]	Marine	–	0.20 ± 0.08	–	–	–	–
	Australia/NZ	Orange Roughy (*Hoplostethus atlanticus*)[124]	Marine	–	–	–	–	0.5	–
	Mediteranean	Mackerel[117]	Marine	–	–	–	–	0.09	–
	Mediteranean	Salmon[117]		–	–	–	–	0.05	–
	Mediteranean	Hake[117]	Marine	–	–	–	–	0.19	–
	Mediteranean	Red Mullet[117]		–	–	–	–	0.23	–
	Mediteranean	Sole[117]	Marine	–	–	–	–	0.08	–
	Adriatic Sea	Red mullet[26]	Marine	–	–	–	–	0.48 ± 0.09	–
	Adriatic Sea	European hake[26]	Marine	–	–	–	–	0.59 ± 0.14	–
	Adriatic Sea	Blue whiting[26]	Marine	–	–	–	–	0.38 ± 0.10	–
	Adriatic Sea	Atlantic mackerel[26]	Marine	–	–	–	–	0.36 ± 0.09	–
	USA	Pollock[58]	Marine	–	–	–	–	0.15	–
	Canada	Chinook salmon[125]	Marine	–	–	–	–	0.088 ± 0.077	–
	Canada	Sockeye salmon[125]	Marine	–	–	–	–	0.077 ± 0.028	–
	Alaska	Black rockfish[22]	Marine	–	–	–	–	0.145 ± 0.018	–
	Alaska	Dolly Varden[22]	Marine	–	–	–	–	0.114 ± 0.013	–
	Alaska	Pacific Halibut[22]	Marine	–	–	–	–	0.148 ± 0.044	–
	Alaska	Great Sculpin[22]	Marine	–	–	–	–	0.294 ± 0.054	–
	Alaska	Pacific Cod[22]	Marine	–	–	–	–	0.173 ± 0.012	–

Table 3. Continued

Trophic level	Continent/sea	Species	Marine/ freshwater	Mercury levels (ppm) (mean ± SD)					
				Liver	Gills	White meat	Red meat	Overall meat	MeHg
	Alaska	Rock greenling[22]	Marine	–	–	–	–	0.099 ± 0.014	–
	Alaska	Yellow Irish Lord[22]	Marine	–	–	–	–	0.272 ± 0.029	–
	Alaska	Pink Salmon[22]	Marine	–	–	–	–	0.042 ± 0.005	–
	Alaska	Flathead Sole[22]	Marine	–	–	–	–	0.276 ± 0.012	–
	Alaska	Rock Sole[22]	Marine	–	–	–	–	0.095 ± 0.023	–
Lower-trophic level species	Africa	Cape Silverside (*Atherina breviceps*)*	Estuarine	–	–	–	–	0.5–5.3	–
	Africa	Cape Silverside (*Atherina breviceps*)*	Estuarine	–	–	–	–	0.3	–
	Africa	Barehead Goby (*Caffrogobius nudiceps*)*	Marine	–	–	–	–	0.3–1.0	–
	Africa	Barehead Goby (*Caffrogobius nudiceps*)*	Marine	–	–	–	–	0.4–0.9	–
	Africa	Common Carp (*Cyprinius carpio*)*	Freshwater	–	–	–	–	0.3–0.8	–
	Africa	Round Herring (*Gilchristella aestuaria*)*	Estuarine	–	–	–	–	0.2–0.8	–
	Africa	Round Herring (*Gilchristella aestuaria*)*	Estuarine	–	–	–	–	0.3–0.4	–
	Africa	Mullet (*Liza richardsonii*)*	Estuarine	–	–	–	–	0.3–0.8	–
	Africa	Mullet (*Liza richardsonii*)*	Estuarine	–	–	–	–	0.3–0.6	–
	Africa	Tilapia (*Oreochromis mossambicus*)*	Estuarine	–	–	–	–	0.3–0.5	–
	Africa	Tilapia (*Tilapia sparrmanii*)*	Estuarine	–	–	–	–	0.4–1.0	–
	Africa	Common Carp (*Cyprinius carpio*)[66]	Freshwater	–	–	–	–	0.35	–
	Africa	Common Carp (*Cyprinius carpio*)[66]	Freshwater	–	–	–	–	0.11	–
	Africa	Common Carp (*Cyprinius carpio*)[66]	Freshwater	–	–	–	–	0.37	–
	Africa	Pacific Thread Herring (*Opisthonema libertate*)[65]	Marine	–	–	–	–	–	–
	Africa	European Pilchard[122]	Marine	–	–	–	–	0.084 ± 0.080	–
	Africa	Rubberlip Grunt[122]	Marine	–	–	–	–	0.059 ± 0.020	–
	Africa	Atlantic Horse Mackerel[122]	Marine	–	–	–	–	0.034 ± 0.030	–
	Africa	Bogue[122]	Marine	–	–	–	–	0.194 ± 0.008	–
	Africa	*Trisopterus capelanus*[122]	Marine	–	–	–	–	0.097 ± 0.020	–
	Mediteranean	Sardine[117]	Marine	–	–	–	–	0.08	–
	Mediteranean	Anchovy[117]	Marine	–	–	–	–	0.08	–
	USA	Flatfish[58]	Marine	–	–	–	–	0.09	–

*Hutchings K, Clark BM, unpublished.

Table 4. Summary of African research done on metals in various marine and freshwater fish and organisms

Metals analysed	Samples studied	Country/region
Marine		
Zn, Cd, Cu, Pb, Mn, Ni, Co, Bi	Mussel: *Chorornytilus meridionalis*	Saldanha Bay, South Africa[132]
Pb, Cd, Cu, Zn	Phytoplankton, zooplankton, Shrimps	North-West Africa[133]
Cd Cu, Zn, Hg	round sardinella, chubmackerel, Atlantic horse mackerel, painted comber, golden grouper, Niger hind, West African goatfish	Mauritanian coast[134]
Al, Cr, Mn, Fe, Co, Cu, Zn, As, Cd, Pb	Shortnose spurdog, Smooth hound shark	Southeastern Coast of South Africa[4]
Al, Cr, Cu, Fe, Mn, Pb, Zn	Groovy mullet	Mhlathuze Estuary, South Africa[135]
Cu, Cd, Pb, Hg	Various marine fish	Egypt[136]
As, Cd, Hg	Manta Ray	Ghana[137]
Cd, Cu, Cr, Fe, Mn, Ni, Pb, Zn	Harder, estuarine round herring, Tilapia, silverside, crabs, polychaete worms, insect larvae	Diep River Estuary, South Africa[*]
Hg	basa, calamari, shrimp, mussels, sardines, salmon (fresh and tinned), sole, fishfingers, red snapper, monktail, silver, snoek, tinned tuna, butterfish, angelfish, yellowtail, kingklip, dorado, fresh tuna, rockcod	Supermarkets and seafood restaurants in Gauteng, South Africa[138]
Si, Ti, Al, Fe, Mn, Mg, Ca, Na, K, P, Cd, V, Cr, Ni, Cu, Zn, Sr, Zr, Ba, Pb, U	*Dentex* spp., *Galeodes decadactylus,* *Chloroscombrus chrysurus,* *Trichurus lepturus*, Mussel spp.	Togo[139]
Hg	Smooth hound shark	Langebaan, South Africa[110]
Fe, Pb, Ni, Cd, Zn, Cu	*Scomber scombrus* *Sardina pilcladus* *Jack mackerel* *Gadus macrocephalus*	South Western Nigeria[140]
As, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, Se, Zn	*Drapane africana*, *Cynoglossus senegalensis*, *Pomadasys peroteti*	Ghana[141]
Status of marine pollution research in SA[109]		
Freshwater (South Africa)		
Fe, Mn, Zn, Cu, Ni, Pb	Southern mouthbrooder	Transvaal, South Africa[142]
Fe, Zn, Pb, Ni, Cu, Cd, Mn	Tigerfish	Olifants River, South Africa[143]
Cr, Mn, Ni, Pb	Moggel	Witbank dam, South Africa[52]
Hg	Sharp toothed catfish, wide mouthed bass	KwaZulu-Natal, South Africa[66]
Al, Sb, As, Ba, B, Cd, Cr, Co, Cu, Fe, Pb, Mn, Ni, Se, Ag, Sr, Sn, V, Zn	Rednose laboe	Olifants River, South Africa[40]
Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Se, Ag, Pt, Au, Cd, Hg, Pb, U	Sharptooth catfish	Vaal River, South Africa[123]
Al, Sb, As, Ba, B, Cd, Cr, Co, Cu, Fe, Pb, Mn, Ni, Se, Ag, Sr, Sn, V, Zn	Sharptooth catfish	Olifants River, South Africa[144]

[*] Hutchings K and Clark BM, unpublished.

correlations between various heavy metals have previously been identified for numerous fish species.[10,127] These correlations can be positive where certain metals facilitate absorption of other metals or negative where certain metals dominate and therefore decrease the uptake of other metals and minerals. The presence of Hg, for example, has been found to decrease the uptake of Cu and Zn in certain organisms[4] while Se has been shown to have a detoxifying effect on organic Hg in the liver of certain fish.[111,127]

The effects of various external (marine environment) and internal (fish carcass parameters) factors can lead to variation in metal accumulation and inter-metal correlations within and among fish species, locations and seasons.[22] One such widely documented relationship is the size–age effect on metal concentration. In general, Hg concentrations increase as fish size/age increases (especially in predatory fish).[4,6,116,118,128] However, this trend is not apparent in all other metals.[34] Rather, several metals (Cr, Cu, Fe, Cd, Ni, As and Pb) have negative correlations with fish size/age in a number of fish species,[4,6,129] which may be due to higher metabolic rates of younger individuals.[6] Similarly, not all metals are bioaccumulated up the food chain as was previously described for Hg.[130] Arsenic, for example, is found in higher concentrations in lower trophic level fish species.[131] Continuous research on individual metals and how they relate to each other in various fish species and various locations is therefore fundamental in understanding the overall food safety levels of fish meat with regards to metal contaminants.

In Africa, where malnutrition is a major underlying cause of death, assurance of food safety of the continent's natural resources is of utmost importance. Fish meat is one of these natural food sources. The average contribution of fish protein to the total animal protein supply in Africa is 19.4% which is exceeded in many countries with high per capita fish consumption, especially in coastal West Africa (FAO, http://www.fao.org/fishery/sofia/en). Research on metal contaminants in marine fish around the African continent is, however, very limited (Table 4) and further research is needed to ensure that fish which is commonly known as a healthy food source, does not carry unknown hazards to consumer health.

South Africa's extensive coast line (close to 3000 km) and diverse ocean systems have facilitated the development of a major fishing industry where approximately 80% of fish is exported globally and the remainder is further processed and/or consumed locally. However, a number of factors (over-fishing, global climate change, habitat destruction and pollution) have and continue to pose problems for the national fishing industry. Hutchings and Clark (unpublished) identified a number of South African estuarine systems (Diep and Berg Estuaries) with sediment and biota trace metal levels exceeding the recommended safe levels for natural environments.[145] This is largely due to anthropogenic activity such as wastewater treatment works, storm water and industrial wastewater, which can in turn have a significant effect on the consumption safety of South African fishes. Few studies have been done on metals in South African freshwater fish from contaminated rivers and dams (Table 4). However, even though commercial fish are being monitored on an on-going basis,[100] the lack of reported information on heavy metals and especially Hg in South African marine fish is of great concern with regards to consumer health and industry economics as the fishing industry has no guidelines as to which fish or which areas to avoid to minimise catches of fish containing Hg levels above allowable limits.

RECOMMENDATIONS FOR FUTURE RESEARCH

Research on heavy metal concentrations in commonly consumed fish species is still needed, especially in Africa, yet such research is essential in order to understand true toxicity and eventual effects on the consumer. However, the majority of published studies to date predominantly focus on only the few most toxic metals (Hg, As, Cd, Pb) in fish meat; their concentrations and comparisons to various allowable limits. As has been stated earlier, there are numerous factors which can affect the levels of heavy metals detected in a fish and therefore to understand how and why this can vary is essential. Some studies have described mercury speciation and total metals present in toxic form (Hg and As) as obtained from fish tissue; however, fish monitoring programmes continue to only measure total As and Hg assuming that 100% of total Hg is present in its toxic MeHg form. This is largely due to unwanted extra costs and time which metal speciation techniques would add to routine monitoring.[109] Therefore, the identification of a toxicity predictive model could allow for a more accurate and time- and cost-effective method of monitoring true toxicity in fish samples.

Standardised sampling strategies are necessary to allow cross-study and species comparisons. To date, no standard protocol for sampling fish anatomical sections exists;[109] however, different muscles have different functions and can absorb and utilise nutrients and pollutants differently. Therefore, research on cross-carcass metal accumulation, especially between different muscle types (dark and white) of large predatory fish is recommended.

Heavy metal concentrations are species, location and trophic level dependent which can result in considerable variation making comparison and meaningful interpretation difficult. Therefore, more research is required to cover each of these aspects. Research on: (1) trophic level disparities can aid the understanding as to how metals accumulate within the food chain, while (2) spatial scale studies (between and within species) may provide links between environmental pollution and the effects on fish contamination and consequently food safety and consumer health.

Monitoring of metals in seafood is compulsory according to South African legislation;[100] however, the results are not publically available and therefore generally remain unpublished. In addition, very limited research has been published on the human effects of metal contamination through fish and seafood consumption on the African continent. This lack of knowledge and information transfer has led to large knowledge deficits for scientists, consumers and industry as a whole. It is therefore suggested that all data collected be made publically available within a predetermined time from collection.

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