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Review

Assessment of metal contamination in Arabian/Persian Gulf fish: A review

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ABSTRACT

Metal contamination in fish is a concern worldwide, including in the Arabian/Persian Gulf region. This review summarizes the findings from 55 papers about metal concentrations in Gulf fish. Metal concentrations in muscle tissue were screened against the most recent maximum allowable levels (MALs) for fish in international commerce. We identified metals, fish species, and locations where concentrations exceeded the MALs. For some metals, recent MALs have been set to lower concentrations as more toxicological data have become available. Mean fish tissue concentrations exceeded the MAL in 13% (arsenic), 76% (cadmium), 56% (lead), and 10% (mercury) of species means. We identified 13 fish species with the potential to serve as bioindicators of metal contamination for use in four Gulf habitats: pelagic, benthopelagic, demersal, and coral reefs. Recommendations are provided for a regional approach to improve consistency of sampling, data analysis and reporting of metal concentrations in Gulf fish.

1. Introduction

1.1. The marine environment of the Gulf

The marine environment of the semi-enclosed Gulf[2] is one of the most physiologically stressful environments (Price et al., 1993; Sheppard, 1993), exhibiting extreme fluctuations in water temperatures (> 20 °C seasonally; Riegl and Purkis, 2012; Sheppard, 1993) and elevated salinities (> 43 ‰ and between 70 and 80 o/oo in shallow isolated lagoons; Naser, 2013a, 2013b; Sheppard, 1993). The Gulf environment is also characterized by poor water circulation, resulting in a long residence time of water (between 3 and 5 years) and is impacted by multiple anthropogenic activities (Burt, 2014; Carpenter et al., 1997; Freije, 2015; Naser, 2013a, 2013b; Sheppard et al., 2010). Because of the long residence time, environmental contaminants, including metals discharged into Gulf waters, receive limited dilution, thus dissipating more slowly than would occur in a more open circulating coastal system (Freije and Awadh, 2009).

The marine environment is the final repository for most of the natural and anthropogenically generated metal-contaminated runoff from the Gulf landscape that makes its way through watersheds into rivers, and then into Gulf coastal waters or which is discharged directly from shoreline facilities into the Gulf. Countries bordering the Gulf are characterized as having arid to hyper-arid landscapes (Kalbus et al., 2011; Udden et al., 2007). On the northern and western Gulf shores, metal contaminants in surface runoff or industrial discharges from Iran and Iraq flow into rivers flowing into the Gulf, as well as into the Gulf directly. The largest freshwater discharge into the Gulf is from the Shatt Al-Arab River system, which drains the combined waters of the Tigris and Euphrates Rivers of Iraq and the Karun River of Iran (Fig. 1). Several other Iranian rivers, including the Helleh, Hendijan, and Mond, also empty into the Gulf along its northwestern shore. In contrast, along the southern Gulf shore, from Kuwait south to the United Arab Emirates (UAE), there are no rivers; thus, surface runoff and effluent discharges flow directly into the Gulf.

Gulf countries have experienced substantial population growth over the past several decades. This rapid population growth has increased both economic and industrial development (Sale et al., 2011), resulting in increasing discharges of metals into coastal systems, further adding to the already naturally stressed Gulf environment (Burt, 2014; Naser, 2013a, 2013b; Sheppard et al., 2010). This population growth has also necessitated the expansion of harbor facilities to accommodate more

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[2] The name of this waterbody is contentious; therefore, the term "Gulf" is used henceforth to avoid offending any parties.

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Fig. 1. Map of the Gulf showing countries, cities, major rivers, and islands.

trade, increases in the number of desalination plants to produce drinking water, and the construction of power plants to meet rising energy needs and wastewater treatment plants to meet industrial and municipal demands (Freije, 2015; Naser, 2013a, 2013b; Sheppard et al., 2010; Vaughan et al., 2019). Accidental oil spills (El-Shahawi and Al-Yousuf, 1998) and regional political conflicts such as the 1991 Gulf War (Fowler et al., 1993; Sheppard et al., 1992) have added to metal pollution in the Gulf. Seasonally occurring dust storms may also be a source of metal input into the Gulf (Behairy et al., 1985; Bu-Olayan and Subrahmanyam, 1996). Human activities and natural processes are sources to varying degrees for releases of metals into Gulf waters and subsequent bioaccumulation of metals through marine food chains into fish tissues at concentrations that may impact human health (El-Shahawi and Al-Yousuf, 1998; Freije, 2015; Naser, 2013a; Sheppard et al., 2010).

1.2. Fish as bioindicators of metal contamination

Heavy metals are persistent estuarine and marine contaminants that are found worldwide and can cause ecotoxicological, including human health, problems (Chiarelli and Roccheri, 2014; Järup, 2003). Some metals (e.g., Cu, Fe, Zn) are essential to maintaining healthy cell function, whereas others (e.g., Cd, Pb, Hg) are non-essential and highly toxic to cellular processes. Still other metals (e.g., As, Cr, Ni) are intermediate because they are essential to the functions of some organisms, but they produce toxic effects in others (Chiarelli and Roccheri, 2014).

In the marine environment, fish have been used extensively as bioindicators of metal pollution because they accumulate elevated concentrations of metals in their tissues (many times the concentrations detected in ambient seawater) (Authman et al., 2015; Burger, 2006; Burger and Gochfeld, 2001; Gerhardt, 2009; U.S. EPA, 2000b). Fish pass these metals up through the food chain to apex predatory fish and higher trophic–level organisms, including fish-eating sea birds, sea mammals, and human consumers (Biddinger and Gloss, 1984; Freije, 2015; Naser, 2013a; Sheppard et al., 2010; U.S. EPA, 2000b). These bioindicator species accumulate metal contaminants from their environment (seawater and their prey) over time. Therefore, their tissue metal concentrations can be used to provide an integrated view of environmentally available metals (Gerhardt, 2009) rather than an instantaneous snapshot of contaminant concentrations detected by analysis of seawater or sediment samples. As bioindicators, fish must also be able to accumulate metals, but not be damaged by these metals over a wide range of concentrations so that they can react to these environmental stressors (Gerhardt, 2009).

Fish are good bioindicators of the effects of metals because some metals disrupt vital metabolic processes that can affect reproduction, weaken the immune system, and cause pathological changes in tissues or cellular structure (Authman et al., 2015). High concentrations of metals in fish tissue can also cause ecological degradation (i.e., reduced abundance and biodiversity of the fish community) and health effects for fish-consuming wildlife and humans (Authman et al., 2015; Burger, 2006; Burger and Gochfeld, 2001). Fish is a source of protein and nutrients in the human diet; however, consuming metal-contaminated fish can cause adverse health effects, particularly for sensitive populations (e.g., pregnant and nursing women, infants and young children), as well as for recreational or subsistence fishers who typically consume large quantities of fish (U.S. EPA, 2000b).

Before the discovery of oil in the Gulf in the late 1930s, fishing was a major source of food and income for many Gulf coastal populations (Al-Jedah et al., 1999). Fishing is still the most important renewable resource of the Gulf (Carpenter et al., 1997; Grandcourt, 2008; Sale et al., 2011). The nutritional importance and popularity of fish consumption to local communities in the Gulf have been described in exposure assessments of various contaminants associated with fish consumption. These studies have been conducted in Bahrain (Freije and Awadh, 2009), Iran (Hosseini et al., 2015; Naji et al., 2016; Raissy and Ansari, 2014; Saei-Dehkordi et al., 2010; Sahebi et al., 2011), Kuwait (Bu-Olayan and Al-Yakoob, 1998; Khordagui and Al-Ajmi, 1991), Qatar (Al-Jedah and Robinson, 2001; Al-Jedah et al., 1999), Saudi Arabia (El-Bahr and Abdelghany, 2015; Krishnakumar et al., 2016), and the UAE (Davidson et al., 2012; Kosanovic et al., 2007).

1.3. Factors that affect bioaccumulation of metals in fish

Many factors, including both intrinsic species differences and extrinsic environmental conditions, affect the accumulation of metals in fish. Intrinsic species-dependent factors include trophic status and feeding strategy, which are linked to major constituents of the diet (e.g., algae, phytoplankton, invertebrates, fish) (Al-Majed and Preston, 2000; Pourang, 1995; Saei-Dehkordi et al., 2010; Tremain and Adams, 2012). In addition, intrinsic factors include the age of the fish (Al-Hashimi and Al-Zorba, 1991) and the gender and phase of sexual reproduction, which is dependent upon the season when fish are collected (Al-Hashimi and Al-Zorba, 1991; Al-Yousuf et al., 2000; Nejatkhah et al., 2014; Mortazavi and Sharifian, 2012; Pourang, 1995; Saei-Dehkordi et al., 2010). These factors also include the tissue type analyzed (Ahmad and Al-Ghais, 1997; Al-Najare et al., 2015; Al-Yakoob et al., 1994; Al-Yousuf and El-Shahawi, 1999; Al-Yousuf et al., 2000; Ashraf, 2005; El-Shahawi and Al-Yousuf, 1998; Khoshnood et al., 2010; Rahimi and Gheysari, 2015; Rahmanpour et al., 2014; Zamani et al., 2015) and body size (length and/or weight; Al-Yousuf et al., 2000; Barak and Mason, 1990; Mortazavi and Sharifian, 2012).

Extrinsic factors affecting metal bioaccumulation include the chemical form of the metal, the severity of the contamination, and presence of other contaminants in the marine environment. Extrinsic factors also include associated water quality conditions, such as salinity, temperature, dissolved oxygen concentrations, pH, total suspended solids, and the nature of the bottom sediments, which can serve as a reservoir for metal adsorption and resuspension (Al-Darwish et al., 2005; Barak and Mason, 1990; ButTayban and Preston, 2004; Saei-Dehkordi et al., 2010).

1.4. Purpose of this review

The purpose of this review was to examine studies of metal contamination in Gulf fish over a 26-year period. First, we identified and selected primary studies of metal contamination in edible muscle tissue of Gulf fish. Then, we compiled data regarding metal concentrations in fish from the selected papers and screened the mean species metal concentrations reported to determine exceedances of the maximum allowable limits (MALs) of metals in fish in international commerce that might pose a health risk to human consumers. We also assessed the spatial extent of contamination in Gulf waters by mapping the sites where fish tissue data were collected and later analyzed and discussed in the reviewed studies. Lastly, we identified important characteristics of bioindicators for consideration and recommended fish species that could serve as bioindicators for metal contamination Gulf-wide. To our knowledge, our paper is the first in-depth review of the literature that focuses on both the geographic extent of contamination and the range of concentrations of metals in muscle tissue of Gulf fish.

2. Methods

2.1. Literature search methodology

During this review, fish contamination studies conducted in the Gulf over a 26-year period were identified and evaluated. Publications were identified by searching records in English for the following terms: fish tissue AND (Arabian or Persian Gulf) AND (heavy metals OR methylmercury) for the period from January 1990 through December 2016. The following three databases were searched:

1. PubMed
2. Web of Science, which includes Science Citation Index Expanded, Social Sciences Citation Index, conference proceedings, and citation indices for science and for social science and humanities
3. Environmental Science and Pollution Management, which includes Aquatic Science and Fisheries Abstracts, Aquatic Pollution and Environmental Quality, Water Resources Abstracts, TOXLINE, Toxicology Abstracts, Environmental Abstracts, Pollution Abstracts, and Conference Papers Index.

Research papers obtained from this literature search were cross-checked against the reference lists provided in other recent review articles on metal contamination in Gulf fish (e.g., Freije, 2015; Naser, 2013a, 2013b). In addition, the Reference section of each paper was reviewed to maximize inclusion of all relevant literature. The electronic search focused on identifying peer-reviewed journal articles.

A Microsoft Access database was used to compile the fish concentration data from individual publications for use in our screening analyses. Publications were reviewed from all eight Gulf countries (i.e., Bahrain, Iran, Iraq, Kuwait, Oman, Qatar, Saudi Arabia, and the UAE). From each publication that met our selection criteria (see the Selection Criteria for Fish Tissue Studies section), we compiled data for each fish species by using the accepted scientific and common names, study type, and the metal concentrations (muscle tissue) in wet weight (WW) units for the mean ± standard deviation (SD) and maximum concentrations reported. Sampling locations were identified by name, country, and latitude and longitude coordinates.

2.2. Selection criteria for fish tissue studies

The major focus of this review was to identify primary research studies that contained information about metal concentrations in fish muscle tissue from the Gulf. Overall, 85 separate studies were identified, and the following six criteria were used to select 55 of these studies for evaluation during our review:

1. The reference must be a primary source published in English. Only data from primary sources were included. Secondary review articles were used exclusively to obtain background information and to locate additional primary sources.
2. Fish must be collected from estuarine or marine waters in the Gulf. Only fish from Gulf waters were included. Fish analyzed from fish market surveys were included when the market was located along the Gulf coast or when the authors of the papers indicated that only Gulf-harvested fish were analyzed. Fish from inland markets were not included because these outlets may sell seafood purchased from local vendors of Gulf fish; regional fish from the Arabian, Caspian, and Red Seas; and imported fish from other international sources.
3. Fish must be identified by their scientific genus and species names. Studies that did not identify fish to the species level or that used only common or local names were not included in our review. Precise identification of the fish species is of paramount importance because the ability to bioaccumulate metals can differ greatly, even among closely related species, based on their feeding strategies (Tremain and Adams, 2012). FishBase was used as the primary source for currently accepted scientific and common names (Froese and Pauly, 2017) because it was most often cited by the authors of the reviewed Gulf papers. For some species, the scientific name given in the original publication needed to be updated to the currently accepted nomenclature appearing in FishBase. Table 1 lists the species names reported in the reviewed studies that were updated to their currently accepted scientific and common names.

Table 1
Species names that were updated to reflect currently accepted nomenclature.[a]

Reported scientific name	Currently accepted scientific name	Currently accepted common name
Euryglossa orientalis	*Brachirus orientalis*	Oriental sole
Arius thalassinus	*Netuma thalassina*	Giant catfish
Liza subviridis	*Chelon subviridis*	Greenback mullet
Valamugil schelli	*Moolgarda seheli*	Bluespot mullet
Hilso ilisha	*Tenualosa ilisha*	Hilsa shad
Sparus sarba	*Rhabdosargus sarb*	Goldlined seabream
Acanthopagrus cuvieri	*Sparidentex hasta*	Sobaity seabream
Lutjanus coccineus	*Lutjanus gibbus*	Humpback red snapper
Otolithus argenteus	*Otolithus ruber*	Tigertooth croaker
Lethrinus kalioptrus	*Lethrinus erythracanthus*	Orange-spotted emperor
Epinephelus microdon	*Epinephelus polyphekadion*	Camouflage grouper
Epinephelus jayakari	*Epinephelus multinotatus*	White-blotched grouper
Epinephelus suillus	*Epinephelus coioides*	Orange-spotted grouper
Epinephelus tauvina	*Epinephelus coioides*	Orange-spotted grouper
Caranx kalla	*Alepes djedaba*	Shrimp scad
Caranx sem	*Caranx heberi*	Blacktip trevally
Mullodichthyes auriflamma	*Parupeneus forsskaii*	Red Sea goatfish
Barbus grypus	*Arabibarbus grypus*	Shabout

[a] FishBase was used as source of accepted fish nomenclature.

One example of the nomenclature issue involved the orange-spotted grouper (*Epinephelus coioides*), which is one of the most frequently consumed Gulf species. This species was originally identified as the greasy grouper (*E. tauvina)* by Fowler et al. (1993) and in several other early studies. However, according to Heemstra and Randall (1993), *E. tauvina* has been widely misidentified in the past and does not occur in the Gulf (Edwin Grandcourt, personal communication). Thus, all instances of *E. tauvina* in the research papers were revised to *E. coioides* before data were entered into our database. Another early synonym for this species, *E. suillus* was also revised to *E. coioides* based on information in FishBase. Additional information about the taxonomic status of the genus *Epinephelus* is discussed by Ketchum et al. (2016).

4. Fish tissue must be analyzed from raw uncooked samples.
   Studies that analyzed processed fish products (i.e., dried, cooked, or canned fish) were not included in the review. The only acceptable sample type was uncooked fish tissue that was fresh or previously frozen.
5. Tissues analyzed for metals must include a separate analysis of muscle tissue.
   Muscle tissue is most often used for contaminant analyses because it is the most commonly consumed part of the fish and is therefore most important for assessing human health risks from consuming metal-contaminated fish (U.S. EPA, 2000b). During our review, we found that muscle and several other tissue types were analyzed in some studies. The analysis of multiple tissue types within the same species provides a context for a comparison of which tissues are most involved in metal bioaccumulation. However, for this review, only metal concentrations in muscle were evaluated as being most relevant to human health and as the comparable tissue type to examine across all of the reviewed studies.
6. Analyses of individual fish or composite samples were acceptable.
   The papers that we reviewed reported either the mean metal concentrations from analyses of several individual fish or from several composite samples of fish from a single species. Both of these sample types were appropriate for inclusion in our review. In all cases, the mean metal concentrations presented in this review are for only one species.

2.3. Challenges encountered in analyzing the data

During our review, four major challenges in analyzing various aspects of the data were encountered which are discussed in detail below.

2.3.1. Spatial mapping of study locations

For each study, the location of the fish collection sites provided by the respective authors was mapped. If the latitude and longitude coordinates were provided, then these were used. If geographic coordinates were not provided, but a map or location description of the sampling site was provided (e.g., cities, island, other geographic reference points), then the description was entered in Google Earth to obtain approximate coordinates. Because there were various levels of precision associated with locational information among different studies and different methods were used, the sampling locations that we mapped should be considered as approximations.

Our main purpose in mapping the sampling sites was to visualize the locations where sampling has occurred in the Gulf over the 26-year period we reviewed. By mapping the sampling sites and linking locational information to metal concentrations that exceeded the MALs, potential hotspots of contamination, as well as information about unmonitored or under monitored areas where future monitoring might be appropriate, could be identified. For this review, “hotspots” were defined as contaminated areas where the metal concentrations in fish exceeded the MAL.

2.3.2. Presentation of metal concentrations on a wet weight (WW) basis

Tissue concentrations can be analyzed equally well on a WW or dry weight (DW) basis; however, data presented on a WW basis were preferred because the MALs used to screen the concentration data are in WW. If the authors of the papers reported concentrations on a DW basis, then the concentrations were converted to WW for comparison to the available MALs. Many authors, however, did not provide the WW concentration and did not provide the percent moisture of the tissues to facilitate this conversion. If the percent moisture was not provided, then we assumed that muscle tissue was 78% water, based on the median value reported for several Gulf species (Attar et al., 1992). Then, we used the conversion method in Eq. (1) to convert DW to WW concentrations (Lusk et al., 2005). Note: All data results discussed in this review are presented in WW concentrations only to facilitate comparison with the MALs.

$$\textit{Metal concentration}\ (WW) = \textit{metal concentration}\ (DW) \times [(1 - (\%\textit{moisture}) \div 100)] \quad (1)$$

Using Eq. (1) was not an ideal way to convert DW to WW, but it was necessitated because only a few authors (El-Bahr and Abdelghany, 2015; Attar et al., 1992; Monikh et al., 2012; Pourang et al., 2005) provided information about the percent moisture of fish tissue or a conversion factor that could be used to calculate the WW concentration. The authors of three papers (i.e., El-Bahr and Abdelghany, 2015; Mortazavi and Sharifian, 2011; Naji et al., 2016) handled this issue best by providing both the WW and DW concentrations for each metal and for each species analyzed.

2.3.3. Selection of MALs for metals in commercial fish

A list of international MALs for metals in fish in international commerce was compiled from a variety of sources. MALs are not available for many metals that were monitored in Gulf fish, and there is no standard source for current information about MALs as previously discussed by Burger and Gochfeld (2005). A summary of the MALs we compiled is presented in Table 2. Note: The values in bold type in Table 2 were selected as the MAL values for screening mean fish tissue concentrations. In general, we selected the most recently promulgated MALs from international regulatory organizations, when possible, rather than national standards. The MALs that we used were more stringent than many of the previous MALs promulgated by these same international organizations because as more information about the toxicity of a metal has become available, the MALs have been adjusted

Table 2
International MALs (mg/kg WW) for metal contaminants in fish and fish products (Note: the values in bold type were selected as the MAL screening values).

Metal	CFIA standards[a] (mg/kg)	FSANZ[b] (mg/kg)	EU[c] standards (mg/kg)	FDA[d] standards (mg/kg)	Median[e] (range) (mg/kg)	CAC[f] (mg/kg)
Arsenic (total)	**3.5**	2.0[g]	–	–	1.4 (0.1–5)	–
Cadmium	–	–	**0.05**[h]	–	0.35 (0.05–2)	–
Lead	0.5	0.5	**0.3**[i]	–	1.75 (0.5–6)	**0.3**
Mercury (see table notes [a], [b], [c], and [j])	**0.5**[a]	**0.5**[b]	**0.5**[j]	–	**0.5** (0.1–1)	–
Methylmercury (see table notes [d] and [f])	–	–	–	1.0[d]	–	**0.5**[f]

[a] CFIA (Canadian Food Inspection Agency) (2014). Note: For most fish, the MAL is 0.5 mg/kg of mercury (Hg). The MAL is 1.0 mg/kg of Hg for swordfish, shark, fresh or frozen tuna, escolar, orange roughy, and marlin.

[b] Federal Register of Legislation (2013). Note: For most fish, the MAL is 0.5 mg/kg of Hg. The MAL is 1.0 mg/kg of Hg for gemfish, billfish (including marlin), southern bluefin tuna, barramundi, ling, orange roughy, rays, and all shark species.

[c] Commission of the European Communities (2001).

[d] FDA (U.S. Food and Drug Administration, 2011. Note: The MAL is 1.0 mg/kg of methylmercury (CH_3Hg^+) for fish, shellfish, crustaceans, and other aquatic animals (fresh, frozen, or processed).

[e] Nauen (1983).

[f] CAC (Codex Alimentarius Commission) (2011). Note: For most fish, the MAL is 0.5 mg/kg for CH_3Hg^+. For predatory fish (i.e., shark, swordfish, tuna, and pike) in fresh or processed fish and fish products, the MAL is 1.0 mg/kg of CH_3Hg^+.

[g] Federal Register of Legislation (2013). Note: This MAL is for inorganic arsenic (As). This MAL was not used for screening because only one study of Gulf fish analyzed for inorganic As; all other studies analyzed muscle tissue only for total As.

[h] European Union Commission Regulation (2014). Note: The MAL is 0.10 mg/kg of cadmium (Cd) for mackerel (*Scomber* species), tuna (*Thunnus* species, *Katsuwonus pelamis*, *Euthynnus* species), and bichique (*Sicyopterus lagocephalus*). The MAL is 0.15 mg/kg of Cd for bullet tuna (*Auxis* species). The MAL is 0.25 mg/kg of Cd for anchovy (*Engraulis* species), swordfish (*Xiphias gladius*), and sardine (*Sardina pilchardus*).

[i] European Union Commission Regulation (2015).

[j] European Union Commission Regulation (2011). Note: The MAL is 1.0 mg/kg of Hg for anglerfish, Atlantic catfish, bonito, eel, emperor, grenadier, halibut, kingklip, marlin, megrim, mullet, orange roughy, pandora, pike, pink cuskeel, plain bonito, poor cod, Portuguese dogfish, rays, redfish, rosy soldierfish, sailfish, scabbard fish, seabream, shark (all species), snake mackerel, sturgeon species, swordfish, and tuna (*Thunnus* and *Euthynnus* species, *Katsuwomus pelamis*).

accordingly, generally to lower concentrations. In all cases, we used MALs that are most protective of human health.

Both an adequate amount of fish tissue concentration data and a MAL were available for the following four metals: arsenic (As), cadmium (Cd), lead (Pb), and mercury (Hg). All four of these metals were analyzed in studies conducted in each of the Gulf countries, but with several exceptions. Arsenic was not analyzed in fish from Qatar, and Hg was not analyzed in fish from the Gulf waters of Saudi Arabia. No studies conducted in Oman and Iraq were included because we could not determine whether the fish were harvested from the Gulf or because the studies were not available in English.

It should be noted that MALs were promulgated for both total As and inorganic As and for total Hg and methyl mercury (CH_3Hg+) (Table 2). During this review, we used the MAL for total As because all 13 studies analyzed for total As and only one study (Krishnakumar et al., 2016) also conducted speciation analysis for inorganic As. The U.S. Environmental Protection Agency (U.S. EPA, 2000b) recommends that if total As is analyzed that it is assumed that 10% of total As is inorganic As, the more toxic species to humans. This assumption, however, is highly controversial. Similarly for Hg, all studies analyzed for total Hg, and only one study also analyzed for CH_3Hg+ (Freije and Awadh, 2009). To be conservative when estimating potential health effects for humans, the U.S. EPA (2000b) recommended that if total Hg is analyzed, it should be assumed that 100% of the mercury detected is CH_3Hg+ —the most toxic form to humans. This assumption is supported by several studies that analyzed the percentage of CH_3Hg+ compared with total Hg in marine fish tissues (Amlund et al., 2007; Freije and Awadh, 2009; Kannan et al., 1998; Nostbakken et al., 2015; Wagemann et al., 1997).

2.3.4. Selection of potential bioindicator species

The Gulf's northern inshore area is characterized by deep water over a muddy bottom that extends offshore to the center of the Gulf (Carpenter et al., 1997). The northwestern portion of the Gulf contains the only estuarine areas around the Shatt Al-Arab delta and at the mouths of the Mond, Hilleh, and Hendijan Rivers. In contrast, the Gulf's southern inshore area is characterized by warm, high salinity waters over a shallow, primarily sandy bottom punctuated with coral reefs and seagrass beds that also extends offshore to the center of the Gulf (Carpenter et al., 1997). This diversity in underwater landscapes and water conditions creates different habitats with unique fish communities, making the selection of a small number of Gulf-wide bioindicator species difficult.

Our review focused on identifying fish species that are likely candidates to serve as bioindicators for metals in the Gulf. The use of bioindicators allows for comparisons of fish tissue concentrations among various sites over a wide geographic area. Various intrinsic species factors make it difficult to compare metal concentration results within a nation's jurisdiction or across jurisdictions unless the fish data are from the same species (U.S. EPA, 2000b). Because the spatial distribution and habitat requirements of each species vary, it is almost impossible to sample just a single species across all Gulf sites. One solution is to identify a limited number of bioindicator species that are distributed widely enough to allow their sampling at many different sites Gulf-wide (U.S. EPA, 2000b).

The U.S. EPA (2000b) recommends that three primary criteria be used when selecting bioindicator species for assessing human health effects. These criteria are that the species should (1) be commonly consumed in the area, (2) be widely distributed geographically, and (3) have the potential to accumulate high metal concentrations. The bioindicator should also be easy to identify taxonomically, relatively abundant, easy to capture, and large enough to provide adequate tissue mass for residue analysis (Authman et al., 2015; Gerhardt, 2009; Tremain and Adams, 2012; U.S. EPA, 2000b).

To select species that met these criteria a matrix table was constructed that helped assess various species characteristics (e.g., ability to bioaccumulate metals, habitat type, feeding strategy, trophic level, status of commercial fishery and inclusion in fish market surveys, and sustainability of the species' population in the Gulf). For each metal, the mean concentrations reported for each species were screened against the respective MAL, and then ranked from highest to lowest concentrations. Fish species with the highest mean concentrations (in the upper quartile of ranked mean values and that exceeded the MAL for that metal) were potential candidates. To ensure that a species had proven potential to accumulate metals, however, only species with tissue concentrations that exceeded the MALs for at least two metals were recommended as potential bioindicators.

Bioindicators should also include species that are representative of the major fish habitats and feeding strategies (U.S. EPA, 2000b). Habitat information was obtained to determine whether the species was pelagic, benthopelagic, demersal, or associated with coral reefs and to identify prey organisms prevalent in the diet. Saei-Dehkordi et al. (2010) used the same habitats that we selected except for coral reefs because the Iranian coast includes few areas of this habitat type (Vaughan et al., 2019). FishBase was used as the source of information about the habitat types, feeding strategies, and trophic status (Froese and Pauly, 2017). Trophic status was obtained by accessing each species file in FishBase, and then drilling down into “More information” under the “Ecology” category. The trophic status numeric value provided in FishBase was developed by using an estimation method derived from a number of food items using a randomized resampling routine (Froese and Pauly, 2017). The species that accumulated two or more metals were grouped by habitat designation and ranked from highest trophic status (5 to 4) to lowest (3 to 2). To identify species that were commonly consumed, information from both fish market surveys and species of commercial importance were identified. The commercial fisheries status for each species was obtained from FishBase (Froese and Pauly, 2017) and Carpenter et al. (1997). Commercial fishery categories in FishBase included highly commercial, commercial fishery, minor fishery, or no commercial interest. Lastly, the International Union for Conservation of Nature (IUCN, 2019) extinction rating of each potential bioindicator fish species was reviewed. The IUCN Red List categorizes the extinction ratings (from lowest to highest risk) as follows: “Not evaluated,” “Data deficient,” “Least concern,” “Near threatened,” “Vulnerable,” “Endangered,” “Critically endangered,” “Extinct in the wild,” and “Extinct” (IUCN, 2019). Based on this matrix of information about each species, we developed a list of potential bioindicators.

3. Results

3.1. Overview of the study findings

A total of 55 studies (out of 85 identified in the literature search) met the six criteria that were established for our review. Summary information about these 55 studies is presented in the Supplementary data, including each reference by author and year of publication, country where the study was conducted, number of metals analyzed, number of fish species analyzed, sample type (composite or individual sample), and number of fish per sample mean. The Supplementary data also provides general information about what types of data are provided in each study, including mean metal concentration ± SD, maximum metal concentration, and range in metal concentrations for the species. Out of the 55 reviewed studies, 34 (57%) were conducted in Iran; 6 studies each were conducted in Kuwait, Saudi Arabia, and the UAE; and 4 studies each were conducted in Bahrain and Qatar. In addition, two of the reviewed studies analyzed fish data from more than one Gulf nation.

The following number and types of studies were also identified: (1) 30 baseline monitoring studies (69.6%), (2) 5 targeted pollutant studies (5.5%), and (3) 20 fish market surveys (24.9%). The large number of baseline studies and small number of targeted studies were not surprising. Many baseline studies were initiated to survey areas that had not been previously monitored, and the targeted pollutant studies were conducted where previous studies had identified metal contamination. Fish market surveys included purchasing fish directly from artisanal fishermen at the docks or at local fish markets. This method was recommended by the Regional Organization for the Protection of the Marine Environment (ROPME, 1999) for human health studies. Presumably, researchers used this latter method of collecting fish to minimize costs as compared with the field sampling studies. Using fish market surveys also offered researchers the opportunity to visually select the species and size of fish they needed if they wanted to evaluate size class differences in metal concentrations either for individual or composite samples.

Overall, a total of 22 metals and trace elements were detected in fish across the Gulf studies reviewed. However, for many metals, very few studies with limited concentration data were available and/or no MALs were identified; therefore, these concentration data could not be screened to determine exceedances of the MALs. The mean, median, and minimum and maximum concentrations for each of these metals and trace elements are summarized in Table 3. Our review focused on an in-depth analysis of four toxic metals (As, Cd, Pb, Hg) because both adequate concentration data and a MAL were available. The largest number of studies were conducted for Pb (34), followed by Cd (32), Hg (24), and As (13). Similarly, data regarding the largest number of fish species means were reported for Pb (322), Cd (256), Hg (224), and As (146).

ROPME (1999) guidance recommends that for fish studies involving public health, fish should be collected from the point of sale to the public, typically from fish markets or collected from the main fishing areas. The fish species that were most commonly sampled (> 50 analyses) for metal contamination in fish market surveys are listed in Table 4. In addition, the species listed were compared to determine which species were consumed in the largest number of jurisdictions in the Gulf. The species that were most widely sampled included the orange-spotted grouper (six Gulf countries), the spangled emperor (five Gulf countries), and the narrow-barred Spanish mackerel and the white-spotted spinefoot (each from four Gulf countries).

3.2. Screening metal concentrations against MALs for commercial fish

The results of our screening of fish muscle tissue concentrations against the MALs are summarized in the remainder of this subsection for As, Cd, Pb, and Hg. To show the geographic extent of these exceedances, we mapped the sampling sites where metal concentrations were assessed. The tables listing the mean concentrations from the most contaminated species for each respective metal are also provided to characterize the magnitude of the contamination detected at these hotspots. For Cd and Pb, we found so many studies where tissue concentrations exceeded the MAL that we discuss results from additional studies not shown in the tables. For As and Hg, such a low percentage of the species means exceeded the MALs that the discussion was confined to the exceedances in those tables.

3.2.1. Arsenic

The spatial extent of As sampling and the study sites where fish means exceeded the MAL (hotspots) are shown in Fig. 2. Fish species samples with mean total As values exceeding the MAL were clustered along the Saudi Arabian coast from Ad Damman north to the Kuwaiti border (15 of 70 species means), in Kuwait City (1 of 11), in Bushehr City (1 of 1), in Bahrain at Fasht Al-Jarim reef (1 of 1), and off Dubai, UAE (3 of 9).

Out of the 146 fish species means evaluated, only 20 means (13%) exceeded the MAL (3.5 mg/kg) for total As. Total As mean concentrations across all species ranged from 0.046 to 32.3 mg/kg. The As mean and median of all species were 1.97 mg/kg and 0.4 mg/kg, respectively, and both values were below the MAL (Fig. 3). The highest As mean (32.3 mg/kg) and maximum (73.7 mg/kg) concentrations were found in the twobar seabream (*Acanthopagrus bifasciatus*) from Ad Damman, Saudi Arabia (Attar et al., 1992) and these concentrations exceeded the MAL by factors of almost 10 and 20, respectively.

The top 10 species whose mean total As concentration exceeded the MAL are presented in Table 5. In addition to the twobar sea bream, Attar et al. (1992) reported mean As concentrations for four other species from Saudi Arabia that exceeded the current MAL. The mean was 9.95 mg/kg for blackspotted rubberlip (*Plectorhinchus gaterinus)*,

Table 3
Summary of 22 metals and trace elements analyzed in Gulf fish tissue.[a,b]

Metal name (abbreviation)	Number of studies	Number of species samples	Mean of study means (mg/kg)	Maximum value (mg/kg)	Minimum value (mg/kg)	Median of study means (mg/kg)
Aluminum (Al)	2	5	2.1	7.52	0.3	1.5
Antimony (Sb)	2	5	0.31	3.4	0.08	0.2
Arsenic (As)	**13**	**146**	**1.34**	**73.7**	**0.003**	**0.355**
Beryllium (Be)	1	3	0.1	0.2	0.02	0.1
Cadmium (Cd)	**32**	**256**	**0.53**	**46**	**0.002**	**0.214**
Chromium (Cr)	5	61	5.8	147	4	6
Cobalt (Co)	9	106	0.36	18	0.05	0.28
Copper (Cu)	31	318	1.83	4.85	0.01	1.6
Iron (Fe)	12	115	8.07	11	1.37	5.39
Lead (Pb)	**34**	**322**	**0.65**	**33**	**0.06**	**0.45**
Magnesium (Mg)	2	15	279	441	147	275
Manganese (Mn)	10	38	0.14	1.09	0.03	0.1
Mercury (Hg)	**24**	**224**	**0.33**	**21.6**	**0.003**	**0.12**
Molybdenum (Mo)	1	3	2	4	0.4	2
Nickel (Ni)	21	175	1.34	100	0.02	0.61
Selenium (Se)	4	21	9.5	230	0.006	0.62
Silver (Ag)	2	18	0.029	0.205	0.002	ND[c]
Strontium (Sr)	1	2	3.26	8.66	0.74	ND[c]
Thallium (Tl)	1	3	0.12	0.3	0.02	0.1
Tin (Sn)	1	2	0.135	0.5	0.03	ND[c]
Vanadium (V)	7	77	0.40	15	0.91	0.11
Zinc (Zn)	25	225	6.16	64.1	0.009	5.5

[a] The four metals evaluated in this review are shown in bold type. For these metals, there were an adequate number of species samples and a MAL to screen the concentration data.
[b] All concentrations presented are in mg/kg WW.
[c] Median value was not determined.

3.83 mg/kg for cobia (*Rachycentron canadus*), 3.55 mg/kg for sharp tooth jobfish (*Pristipomoides typus*), and 5.34 mg/kg for golden trevally (*Gnathanodon speciosus*). More recently, Krishnakumar et al. (2016) reported that four species exceeded the MAL from coastal Saudi Arabia, with mean As concentrations ranging from 12.32 to 16.72 mg/kg (see Table 5). In addition to the orange spotted grouper from Bahrain, Fowler et al. (1993) also found As concentrations in Dubai (UAE) exceeded the current MAL. These As concentrations included 4.84 mg/kg for orange spotted grouper and 3.52 mg/kg for the spangled emperor (*Lethrinus nebulosus*). Rahimi and Gheysari (2015) reported elevated As concentrations (22.29 mg/kg) in the narrow-barred Spanish mackerel collected from a market in Boushehr City, Iran. Moore et al. (2015) reported muscle tissue of seven individual smooth tooth blacktip sharks from Kuwait, and all samples exceeded the MAL. The mean As concentration of the seven individual sharks ranging in age from juveniles to adults was 27.43 ± 9.96 mg/kg, with a maximum As concentration of 41 mg/kg.

3.2.2. Cadmium

The spatial extent of Cd sampling and the study sites where fish means exceeded the MAL (hotspots) are shown in Fig. 4. Fish species with mean Cd concentrations exceeding the MAL were identified along the Iranian coast near Bandar Abbas, including Qeshm Island (35 out of 40 species means), Bushehr City (3 of 3), Kharg Island (2 of 2), and in the area of the Shatt Al-Arab and the Musa Estuary (19 out of 24). In addition, mean Cd concentrations near Ad Damman, Saudi Arabia (55 of 55), near Doha (28 of 28) and Halul Island (30 of 30) in Qatar, and in Abu Dhabi and Dubai in the UAE (8 of 15) also exceeded the MAL (Fig. 4).

Out of the 256 fish species means evaluated, 185 means (72%) exceeded the MAL (0.05 mg/kg) for Cd. Six mean + SD values also exceeded the MAL (4%), and thus a total of 191 (76%) of the means exceeded the MAL, the largest percentage of exceedances of any of the four metals. The mean Cd concentrations of all species ranged from 0.00005 to 27.21 mg/kg. Across all species, the mean and median concentrations were 0.54 mg/kg and 0.23 mg/kg, respectively and both values exceeded the MAL (Fig. 3). The highest mean Cd concentration (27.21 mg/kg) was detected in the pink ear emperor (*Lethrinus lentjan*), and the highest maximum concentration (19.14 mg/kg) was reported in the narrow-barred Spanish mackerel. The highest mean and maximum Cd concentrations exceeded the MAL by a factor of 544 and 382, respectively.

Table 4
Gulf species most frequently analyzed for metals in fish market surveys.

Common name	Species name	Number of analyses conducted on this species[a]	Country
Orange-spotted grouper	*Epinephelus coioides*	211	Bahrain, Iran, Kuwait, Qatar, Saudi Arabia, and the UAE
Spangled emperor	*Lethrinus nebulosus*	192	Bahrain, Iran, Qatar, Saudi Arabia, and the UAE
Narrow-barred Spanish mackerel	*Scomberomorus commerson*	110	Bahrain, Iran, Qatar, and Saudi Arabia
White-spotted spinefoot	*Siganus canaliculatus*	100	Bahrain, Kuwait, Qatar, and Saudi Arabia
Tigertooth croaker	*Otolithes ruber*	80	Iran and Kuwait
Diamond mullet	*Planiliza alata*	72	Bahrain, Kuwait, and Saudi Arabia
Haffara seabream	*Rhabdosargus haffara*	61	Bahrain and Saudi Arabia
Belanger's croaker	*Johnius belangerii*	61	Iran
Oriental sole	*Brachirus orientalis*	59	Iran
Pink ear emperor	*Lethrinus lentjan*	55	Qatar and the UAE
Silver pomfret	*Pampus argenteus*	51	Iran and Kuwait

[a] The number of times that muscle tissue from each species was analyzed for the various metals across all fish market surveys.

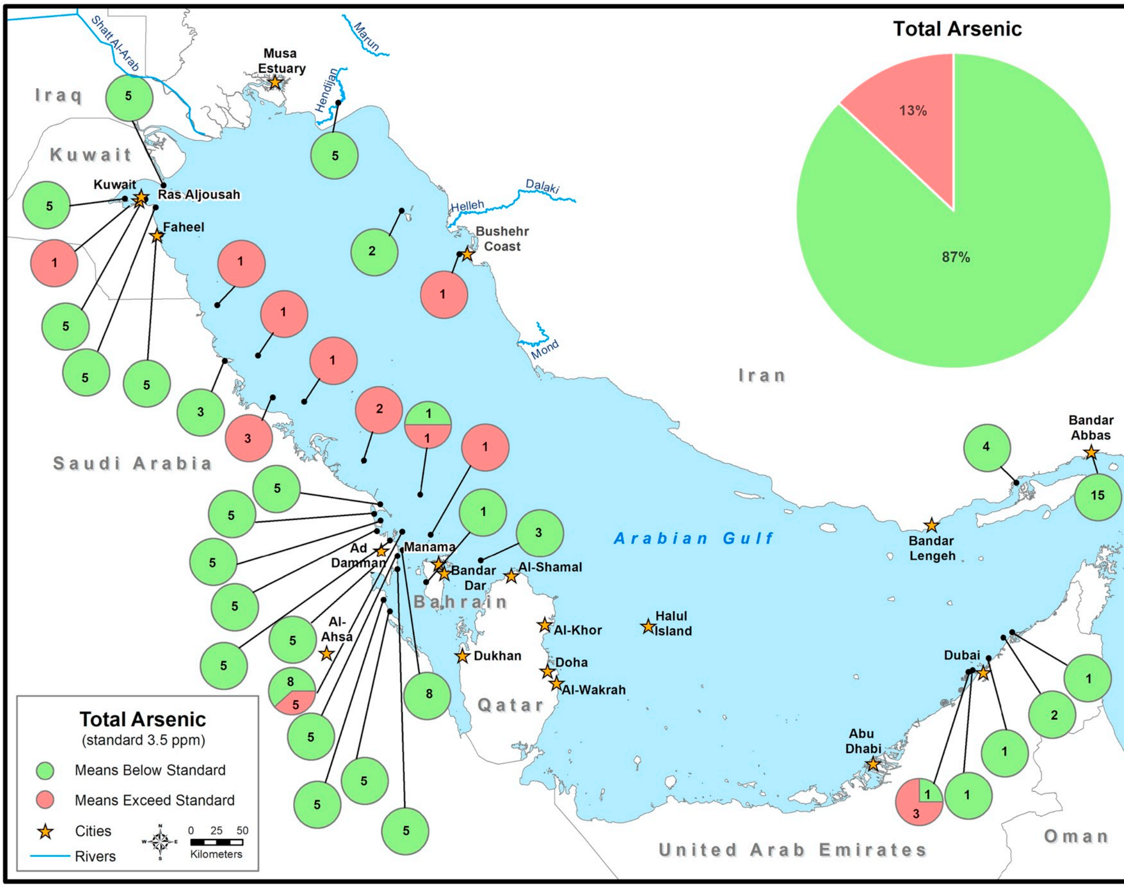

Fig. 2. Exceedances of the MAL of 3.5 mg/kg (ppm) for total arsenic in Gulf fish.

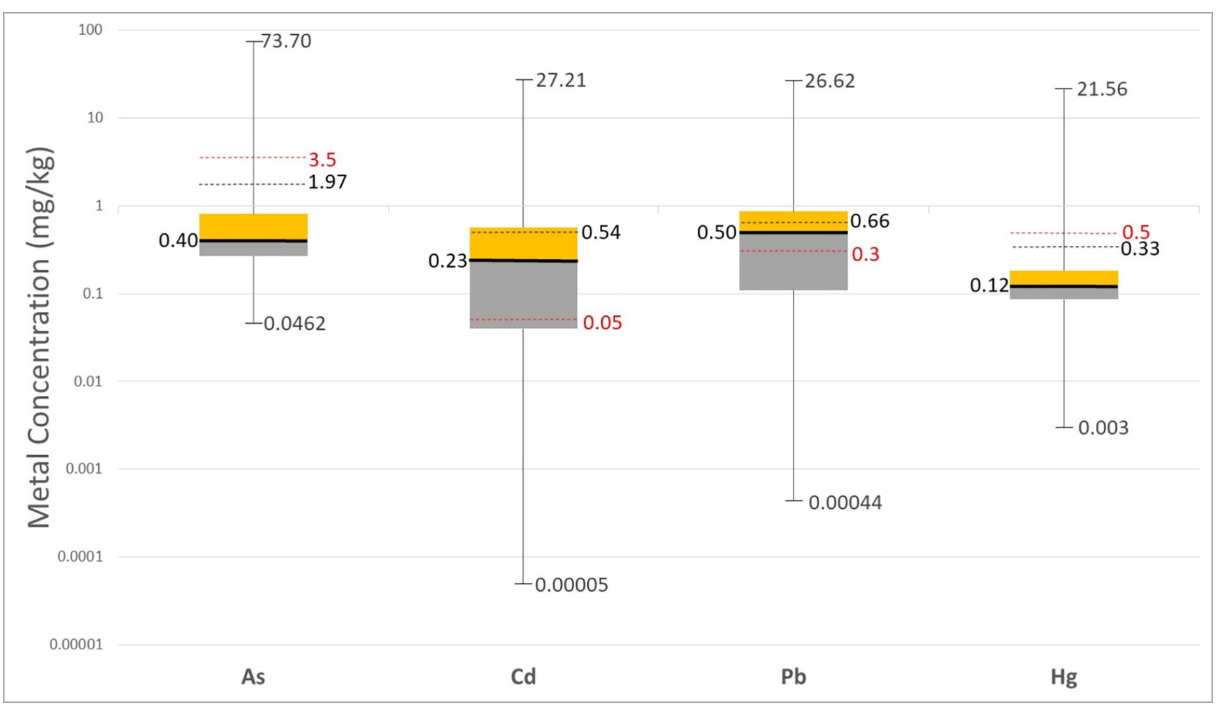

Fig. 3. Comparison of metal concentration data for As, Cd, Pb, and Hg to MAL standards.
Note: The red dotted lines denote the MAL concentration used for each of the respective metals. The black dotted line is the mean value, and the solid black line is the median value for all species. The maximum and minimum concentrations detected for each metal also are provided in the figure. All values are in wet weight (WW). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 5
Top 10 species means that exceeded the MAL for total arsenic (3.5 mg/kg WW).

Common name	Species name	Mean ± SD (mg/kg)[a]	Maximum (mg/kg)[a]	Country	Reference
Twobar seabream	*Acanthopagrus bifasciatus*	32.3	73.7	Saudi Arabia	Attar et al., 1992
Smooth tooth blacktip shark	*Carcharhinus leiodon*	27.43 ± 9.96	41.0	Kuwait	Moore et al., 2015
Narrow-barred Spanish mackerel	*Scomberomorus commerson*	22.29 ± 2.17	32.56	Iran	Rahimi and Gheysari, 2015
Giant catfish	*Netuma thalassina*	16.72	17.60	Saudi Arabia	Krishnakumar et al., 2016
Giant catfish	*Netuma thalassina*	15.84	NA	Saudi Arabia	Krishnakumar et al., 2016
Giant catfish	*Netuma thalassina*	14.30	15.62	Saudi Arabia	Krishnakumar et al., 2016
Pearly goatfish	*Parupeneus margaritatus*	12.54	14.52	Saudi Arabia	Krishnakumar et al., 2016
Twobar seabream	*Acanthopagrus bifasciatus*	12.32	20.24	Saudi Arabia	Krishnakumar et al., 2016
Yellow spotted trevally	*Carangoides fulvoguttatus*	12.32	12.54	Saudi Arabia	Krishnakumar et al., 2016
Orange-spotted grouper	*Epinephelus coioides*	11.66	NA	Bahrain	Fowler et al., 1993

NA = Maximum value was not reported for this species.
[a] All concentrations in this table are in mg/kg WW.

The top 10 species whose mean Cd concentration exceeded the MAL are presented in Table 6. One of the early papers, Kureishy (1993) found Cd concentrations ranging from 0.21 to 1.32 mg/kg in 13 fish species around Doha, Qatar, and ranging from 0.31 to 1.21 mg/kg in 16 fish species from Halul Island (an oil storage and transfer facility), offshore of Qatar. All species sampled by Kureishy (1993) exceeded the current MAL and seven species had Cd concentrations ≥1 mg/kg (Table 6). Zyadah and Taweel (2013) found that mean Cd concentrations were higher during winter in fish near Ad Damman along the Saudi Arabian coast; however, even summer concentrations exceeded the Cd MAL. Besides the Haffara seabream (0.99 mg/kg), the other three species analyzed by Zyadah and Taweel (2013) included the diamond mullet (*Planiliza alata)* (0.70 mg/kg), white-spotted spinefoot (*Siganus canaliculatus*) (0.61 mg/kg), and the narrow-barred Spanish mackerel (0.57 mg/kg). More recently, Rahimi and Gheysari (2015) reported the mean Cd concentration of 13.28 ± 1.55 mg/kg for Gulf-caught narrow-barred Spanish mackerel collected in Boushehr Province, Iran.

Because we identified 32 separate Cd studies and almost three quarters of species means exceeded the current MAL, we discuss results here from additional studies not shown in Table 6. Saei-Dehkordi and Fallah (2011) reported Cd concentrations in 8 out of 12 species of Gulf fish that exceeded the MAL (0.05 mg/kg), including the pelagic narrow-barred Spanish mackerel (0.078 ± 0.048 mg/kg), pickhandle barracuda (*Sphyaena jello*) (0.053 ± 0.022 mg/kg), longtail tuna (*Thunnus tonggol*) (0.106 ± 0.047 mg/kg), and the demersal orange-spotted grouper (0.076 ± 0.023 mg/kg), bartail flathead (*Platycephalus indicus*) (0.111 ± 0.078 mg/kg), Indian halibut (*Psettodes erumei*) (0.097 ± 0.015 mg/kg), and yellowfin seabream (*Acanthopagrus latus*) (0.072 ± 0.021 mg/kg) in a fish market survey in Bandar Abbas, Iran.

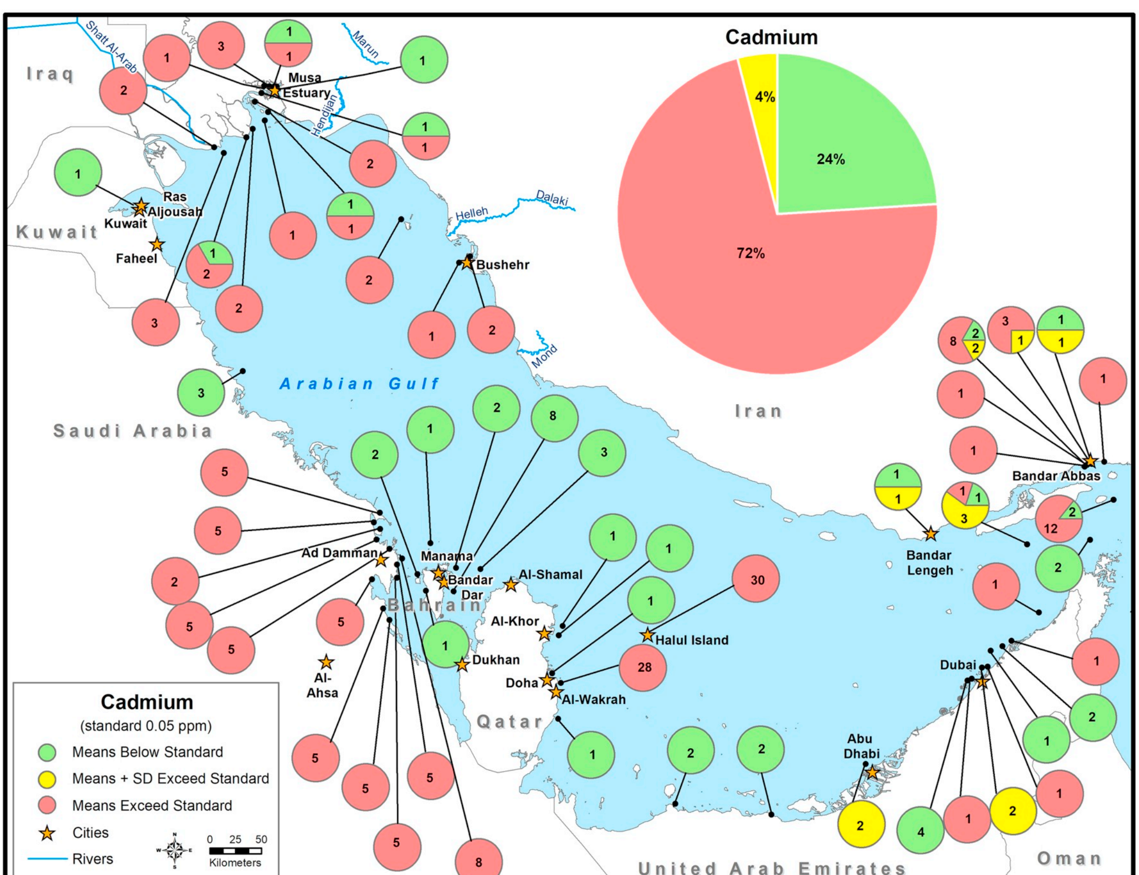

Fig. 4. Exceedances of the MAL of 0.05 mg/kg (ppm) for cadmium in Gulf fish.

Table 6
Top 10 species means that exceeded the MAL for cadmium (0.05 mg/kg WW).

Common name	Species name	Mean ± SD (mg/kg)[a]	Maximum (mg/kg)[a]	Country	Reference
Pink ear emperor	*Lethrinus lentjan*	27.21	NA	Qatar	Kureishy, 1993
Narrow-barred Spanish mackerel	*Scomberomorus commerson*	13.28 ± 1.55	19.14	Iran	Rahimi and Gheysari, 2015
Delagoa threadfin bream	*Nemipterus bipunctatus*	1.31	NA	Qatar	Kureishy, 1993
Delagoa threadfin bream	*Nemipterus bipunctatus*	1.27	NA	Qatar	Kureishy, 1993
Largetooth flounder	*Pseudorhombus arsius*	1.25	NA	Qatar	Kureishy, 1993
Blackfin scad	*Alepes melanoptera*	1.21	NA	Qatar	Kureishy, 1993
Deep flounder	*Pseudorhombus elevatus*	1.08	NA	Qatar	Kureishy, 1993
Malabar trevally	*Carangoides malabaricus*	1.01	NA	Qatar	Kureishy, 1993
Largetooth flounder	*Pseudorhombus arsius*	1.00	NA	Qatar	Kureishy, 1993
Haffara seabream	*Rhabdosargus haffara*	0.99 ± 0.28	NA	Saudi Arabia	Zyadah and Taweel, 2013

NA = Maximum value was not reported for this species.
[a] All concentrations in this table are in mg/kg WW.

These authors noted that there were differences in the international regulations regarding permissible limits of Cd in fish ranging from 0.05 to 1.0 mg/kg, but they used the most stringent MAL available (0.05 mg/kg) as did we to identify exceedances.

More recently, Naji et al. (2016) reported mean Cd concentrations in Kluzinger's mullet (0.09 mg/kg) and Indian mackerel (*Rastrelliger kanagurta*) (0.07 mg/kg), both exceeded the MAL we used to screen their study data collected in Bandar Abbas, Iran. Naji et al. (2016) used a MAL of 1 mg/kg (European Commission, 2006), although this organization issued a more recent MAL (0.05 mg/kg) in 2015 (European Union Commission Regulation, 2015). Naji et al. (2016) reported that children were at greater risk of harmful health effects from consuming Cd contaminated fish than adults. Niri et al. (2015) also reported that muscle tissue concentrations of Cd in the popular tigertooth croaker (*Otolithes ruber)* (0.12 mg/kg) from the Musa Estuary area were more than two times higher than the MAL, and these concentrations could pose a threat to fish consumers in the region.

3.2.3. Lead

The spatial extent of Pb sampling and the study sites where fish means exceeded the MAL (hotspots) are shown in Fig. 5. Fish species with mean Pb concentrations exceeding the MAL (0.3 mg/kg) were concentrated on the Iranian coast near Bandar Abbas, including Qeshm Island (28 out of 38 species means), Bushehr City (4 of 4), Kharg Island (1 out of 2) and in the area of Shatt Al-Arab and the Musa Estuary (17 out of 27). In addition, mean Pb concentrations near Ad Damman, Saudi Arabia (53 of 53), and near Doha (28 of 29) and Halul Island in Qatar (30 of 30) exceeded the MAL. No exceedances of the Pb MAL were reported for any fish samples from the UAE.

Out of the 322 fish species means evaluated, 181 means (56%) and 6 additional mean + SD values (2%) or 58% exceeded the MAL. Mean Pb concentrations across all species ranged from 0.00044 to 20.17 mg/kg, and the mean and median concentrations were 0.66 mg/kg and 0.50 mg/kg, respectively. Both the mean and median Pb concentrations exceeded the MAL (Fig. 3). Both the highest mean (20.17 mg/kg) Pb concentration and the maximum (26.62 mg/kg) concentration were found in the narrow-barred Spanish mackerel an apex predator and these Pb concentrations exceeded the MAL by a factor of 67 and 89, respectively.

The top 10 species whose mean values exceeded the MAL are presented in Table 7. Rahimi and Gheysari (2015) reported a mean Pb concentration in narrow-barred Spanish mackerel of 20.17 mg/kg (range of 13.86 to 26.62 mg/kg) in a fish market survey in Boushehr City, Iran. All of the fish analyzed were in exceedance of the Pb MAL (0.3 mg/kg). Bastami et al. (2015) reported mean Pb concentrations of 3.12 mg/kg, 1.28 mg/kg and 0.71 mg/kg in Klunzinger's mullet from three adjacent sites near Bandar Abbas, Iran that all exceeded the MAL. The three collection sites were in the vicinity of industrial and municipal waste discharges. Gholami et al. (2016) reported the mean (6.52 mg/kg), maximum (13.42 mg/kg), and minimum (1.00 mg/kg) Pb concentrations in the demersal Indian halibut and the mean (0.71 mg/kg), maximum (1.02 mg/kg) and minimum (0.31 mg/kg) Pb concentrations in the spangled emperor collected from Qeshm Island, Iran. These authors reported that measured concentrations in the fish studied were less than the standard levels proposed by the WHO and FAO however their selected regulatory standards (0.5–1.5 mg/kg and 0.5 mg/kg, respectively) were established in the mid-1980's and early 1990s. All of the fish samples analyzed for both species exceeded the current MAL for Pb. Kureishy (1993) reported Pb concentrations in fish from two sites in Qatar. Lead concentrations off Doha were highest in the pink ear emperor (3.61 mg/kg) and three largetooth flounder (see Table 7) and ranged from 0.58 to 3.61 mg/kg in 13 other fish species. Lead concentrations off Halul Island, a major petroleum storage and offloading facility were highest for the deep flounder (*Pseudorhombus elevates*) (1.72 mg/kg) and ranged from 0.52 to 1.72 mg/kg Pb for 15 other finfish species. All of these Pb concentrations exceeded the MAL. Bu-Olayan and Subrahmanyam (1996) analyzed 12 finfish species from Kuwait harbor and 22 finfish species from the coastal area just south of Kuwait City at Al-Ahmadi. Surprisingly, these authors did not find Pb exceedances in fish from Kuwait City (Bay area), but three species along the south coast exceeded the Pb MAL. These included the obtuse barracuda (Table 7) and short-nosed tripod (*Triacanthus biaculeatus*) (1.17 mg/kg) and the bluespot mullet (*Moolgarda seheli*) (0.62 mg/kg). These authors attributed elevated Pb concentrations to vehicle exhaust releases from leaded gasoline that still was in use during their study.

Because we identified 34 studies of Pb and more than half of the species means were in exceedance of the MAL, we discuss results from additional studies in the Gulf that reported lead concentrations in exceedance of the MAL. Saei-Dehkordi and Fallah (2011) reported tissue concentrations of Pb in several species of Gulf fish, including the demersal Indian halibut (0.310 ± 0.119 mg/kg) and Arabian yellowfin seabream (*Acanthopagrus arabicus*) (0.471 ± 0.109 mg/kg) in an Iranian fish market survey in Bandar Abbas. These concentrations exceeded the MAL (0.3 mg/kg) that we employed during our review. However, Saei-Dehkordi and Fallah (2011) noted that there were substantial differences in the international regulations regarding permissible limits of Pb in fish that ranged from 0.2 to 0.5 mg/kg WW. Saei-Dehkordi and Fallah (2011) selected a less stringent MAL (0.5 mg/kg) to screen their data so only the Arabian yellowfin seabream (0.534 ± 0.102 mg/kg) exceeded the MAL used. However, using the current MAL, the orange spotted grouper (0.367 ± 0.090 mg/kg), and Indian halibut (0.397 ± 0.049 mg/kg) were also in exceedance. Niri et al. (2015) also reported that muscle tissue concentrations of Pb in the tigertooth croaker (0.34 mg/kg) were higher than the more stringent MAL (0.2 mg/kg) that they used as well as the MAL (0.3 mg/kg) we used. These authors suggested that Pb could pose a threat to fish consumers in the Musa Estuary region of Iran. In another Iranian health study by Naji et al. (2016), the authors reported mean Pb concentrations in longtail tuna (1.21 mg/kg), Kluzinger's mullet (0.94 mg/kg), Indian mackerel (*Rastrelliger kanagurta*) (0.50 mg/kg), and pickhandle

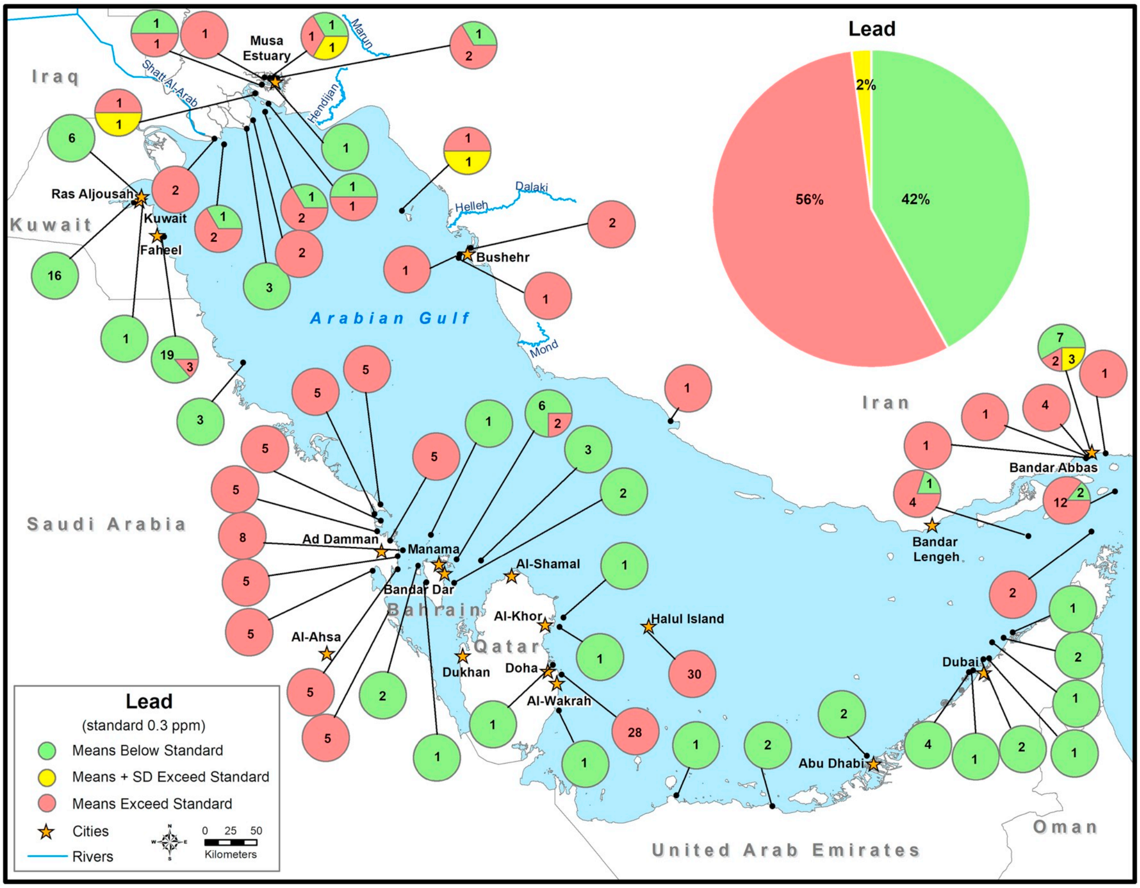

Fig. 5. Exceedances of the MAL of 0.3 mg/kg (ppm) for lead in Gulf fish.

barracuda (1.30 mg/kg). These species all exceeded the MAL (0.3 mg/kg) that we used however, these authors used an older MAL (1.50 mg/kg) (European Commission, 2006) and therefore reported no exceedances. Despite using the less stringent MAL, Naji et al. (2016) recommended continued monitoring for Pb in Gulf fish to protect public health.

3.2.4. Mercury

The spatial extent of Hg sampling and the study sites where fish means exceeded the MAL (hotspots) are shown in Fig. 6. Fish species with mean Hg concentrations exceeding the MAL were found primarily on the Iranian coast near Bandar Abbas including Qeshm Island (6 of 22 species means), Bushehr City (1 of 1), and in the area of Shatt Al-Arab and the Musa Estuary (11 of 25). In addition, mean Hg concentrations in Kuwait (1 of 1), near Manama, Bahrain (2 of 15), and Al Mirfa in the UAE (1 of 1) were in exceedance of the MAL. No exceedances for Hg were reported for any samples from Doha or Halul Island, Qatar.

Out of the 224 fish species means evaluated, 13 means (6%) and 10 additional mean + SD values (4%) for a total of 10% of the species means exceeded the Hg MAL (0.5 mg/kg), the lowest percentage of the four metals. Mean Hg concentrations across all species ranged from 0.003 to 18.3 mg/kg. The mean and median Hg concentrations of all species were 0.33 mg/kg and 0.12 mg/kg, respectively and these values were both lower than the MAL (Fig. 3). The highest mean Hg concentration (18.3 mg/kg) was detected in a demersal species, the Oriental sole (*Brachirus orientalis*) and exceeded the MAL by a factor of 36. The maximum Hg concentration (21.56 mg/kg) was detected in the narrow-barred Spanish mackerel and exceeded the MAL by a factor of 44.

The 12 highest mean Hg concentrations were detected in species

Table 7
Top 10 species means that exceeded the MAL for lead (0.3 mg/kg WW).

Common name	Species name	Mean ± SD (mg/kg)[a]	Maximum (mg/kg)[a]	Country	Reference
Narrow-barred Spanish mackerel	*Scomberomorus commerson*	20.17 ± 2.02	26.62	Iran	Rahimi and Gheysari, 2015
Indian halibut	*Psettodes erumei*	6.52 ± 0.81	13.42	Iran	Gholami et al., 2016
Pink ear emperor	*Lethrinus lentjan*	3.61	NA	Qatar	Kureishy, 1993
Obtuse barracuda	*Sphyraena obtusata*	3.21 ± 0.53	NA	Kuwait	Bu-Olayan and Subrahmanyam, 1996
Klunzinger's mullet	*Liza klunzingeri*	3.12 ± 0.32	NA	Iran	Bastami et al., 2015
Largetooth flounder	*Pseudorhombus arsius*	2.47	NA	Kuwait	Kureishy, 1993
Largetooth flounder	*Pseudorhombus arsius*	2.10	NA	Kuwait	Kureishy, 1993
Largetooth flounder	*Pseudorhombus arsius*	1.94	NA	Kuwait	Kureishy, 1993
Spangled emperor	*Lethrinus nebulosus*	1.92	NA	Kuwait	Kureishy, 1993
Malabar trevally	*Carangoides malabaricus*	1.91	NA	Kuwait	Kureishy, 1993

NA = Maximum value was not reported for this species.
[a] All concentrations in this table are in mg/kg WW.

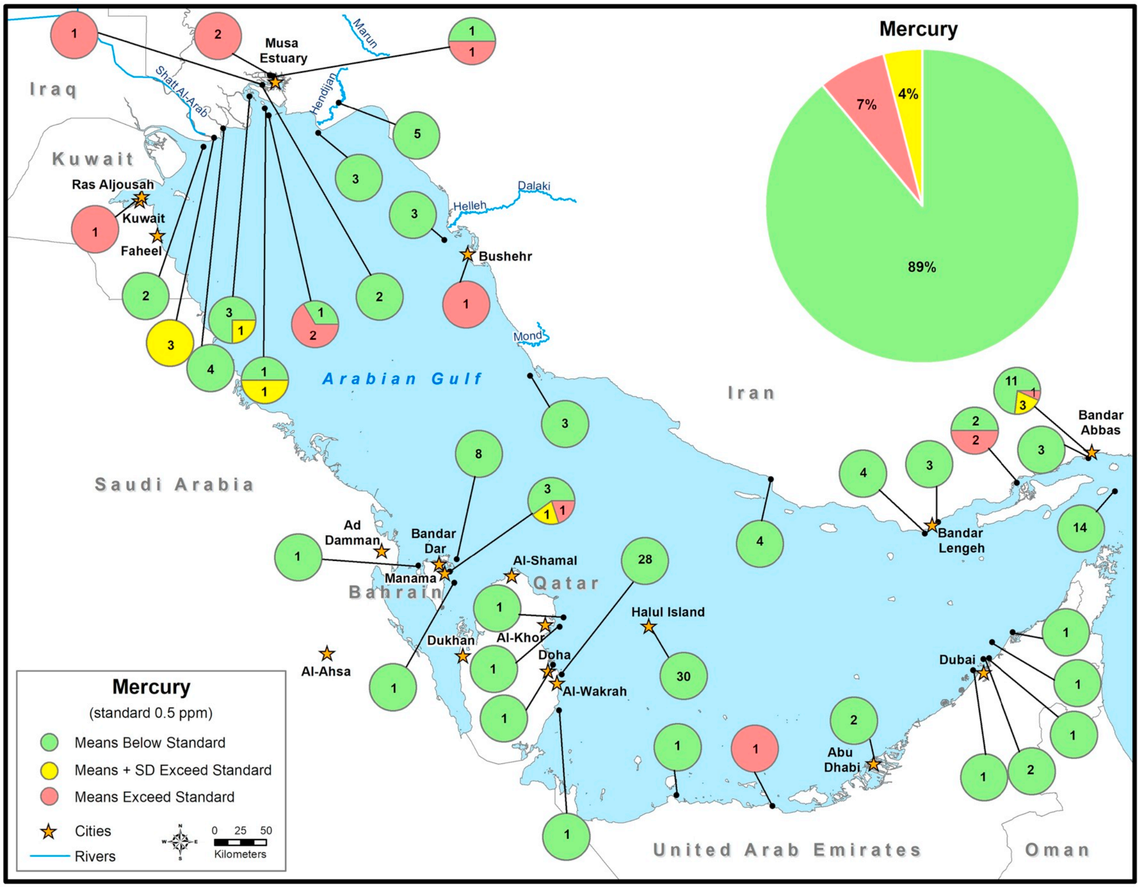

Fig. 6. Exceedances of the MAL of 0.5 mg/kg (ppm) for total mercury in Gulf fish.

identified in Table 8. Haghighat et al. (2011) studied Hg concentrations in the Oriental sole in the Musa Estuary (Iran) a highly contaminated estuarine system. The highest mean Hg concentration (18.3 mg/kg) was detected at the most polluted site (closest to a chlor-alkali plant), while Hg concentrations decreased (0.46 mg/kg) as the distance from the plant increased and reached the lowest concentration (0.11 mg/kg) at the most distant site. Fard et al. (2015) reported Hg in Belanger's croaker at a mean and maximum concentration of 3.15 mg/kg and 17.19 mg/kg, respectively also from the Musa estuary. Hosseini et al. (2016) also found Belanger's croaker and Roho labeo in exceedance of the MAL from the Khuzestan coast of Iran. Some of these concentrations are clearly in exceedance of the MAL by a factor of > 30 and pose a risk to human health. Rahimi and Gheysari (2015) reported a mean Hg concentration in narrow-barred Spanish mackerel of 10.78 mg/kg (range of 4.18 to 21.56 mg/kg) in a fish market survey in Boushehr City, Iran. All of the fish analyzed in this study were in exceedance of the Hg MAL (0.5 mg/kg). Saei-Dehkordi et al. (2010) reported 0.548 mg Hg/kg in the longtail tuna. While the concentration exceeded the MAL for most fish, tuna and other predatory species have a Hg MAL of 1 mg/kg (see Table 2) so this species concentration did not exceed the legal regulatory limit, but even so would pose potential health risks to pregnant and nursing women and young children at this concentration. Mercury concentrations in two species collected in the Hara biosphere reserve near Qeshm Island, Iran also contained concentrations exceeding the MAL; bartailed flathead and silver sillago contained Hg concentrations of 0.81 mg/kg and 0.69 mg/kg, respectively. The Hara

Table 8
Top 12 species means that exceeded the MAL for mercury (0.5 mg/kg WW).

Common name	Species name	Mean ± SD (mg/kg)[a]	Maximum (mg/kg)[a]	Country	Reference
Oriental sole	*Brachirus orientalis*	18.3	NA	Iran	Haghighat et al., 2011
Narrow-barred Spanish mackerel	*Scomberomorus commerson*	10.78 ± 1.05	21.56	Iran	Rahimi and Gheysari, 2015
Smooth tooth blacktip shark	*Carcharhinus leiodon*	4.37 ± 3.31	9.5	Kuwait	Moore et al., 2015
Belanger's croaker	*Johnius belangerii*	3.15 ± 1.98	17.19	Iran	Fard et al., 2015
Oriental sole	*Brachirus orientalis*	1.6	NA	Iran	Haghighat et al., 2011
Oriental sole	*Brachirus orientalis*	1.0	NA	Iran	Haghighat et al., 2011
Bartail flathead	*Platycephalus indicus*	0.81 ± 0.04	2.65	Iran	Mohammadnabizadeh et al., 2013
Silver sillago	*Sillago sihama*	0.69 ± 0.05	2.8	Iran	Mohammadnabizadeh et al., 2013
Belanger's croaker	*Johnius belangerii*	0.55 ± 0.08	NA	Iran	Hosseini et al., 2016
Roho labeo	*Labeo rohita*	0.54 ± 0.08	NA	Iran	Hosseini et al., 2016
Longtail tuna	*Thunnus tonggol*	0.53 ± 0.19	NA	Iran	Saei-Dehkordi et al., 2010
Orange-spotted grouper	*Epinephelus coioides*	0.52	NA	UAE	De Mora et al., 2004

NA = Maximum value was not reported for this species.
[a] All concentrations in this table are in mg/kg WW.

biosphere site is impacted by a cement plant, power generating facility, lead and zinc factory, paint manufacturing plants, and an aluminum smelting facility. In addition, petroleum products from commercial vessels and refinery water discharges are released to the area (Mohammadnabizadeh et al., 2013.

The mean Hg concentration (4.37 ± 3.31 mg/kg) for one endangered shark species, the smooth tooth blacktip shark, also exceeded the Hg MAL (Moore et al., 2015). The muscle tissue from all individual sharks (both juveniles and adults) that were analyzed in Kuwait exceeded the MAL (1.0 mg/kg) for total Hg specifically established for sharks and other apex predatory species (see Table 2). Mercury concentrations in the individual shark muscle samples ranged from 1.1 to 9.5 mg/kg (Moore et al., 2015). De Mora et al. (2004) found muscle tissue of the orange spotted grouper (0.517 mg/kg) at Al Mirfa in the UAE exceeded the Hg MAL.

3.3. Selection of potential bioindicator species for use in the Gulf

Several fish species were identified that met the three criteria recommended by the U.S. EPA (2000b) for the selection of bioindicator species for use in human health studies (Table 9). The U.S. EPA (2000b) also recommends that at a minimum, bioindicator species should be selected to include both a predator and a bottom-feeding species. Using fish species from at least two distinct ecological groups allows the bioindicator species to represent a wider spectrum of habitat types, feeding strategies, and intrinsic and extrinsic factors that determine differences in contaminant bioaccumulation potential. In the Gulf, four ecological habitats occur that should be considered when selecting representative bioindicator species: pelagic, benthopelagic, demersal, and coral reef–associated habitats.

Species listed in Table 9 were also evaluated as to the IUCN Red List rating (IUCN, 2019). Pelagic species, for example, include the narrow-barred Spanish mackerel, an apex predator of high commercial value and one of the species frequently reported in the fish market surveys (FMS). However, this species has an IUCN rating of "Vulnerable" in the Gulf—a less than ideal distinction for a bioindicator species. Similarly, the yellowfin tuna has an IUCN rating of "Near threatened" that is also suggestive that the species' population may not be sustainable. In contrast, the pink ear emperor, yellow spotted trevally, and Indo-Pacific king mackerel are all listed by the IUCN as species of "Least concern" with respect to their populations and could serve as pelagic bioindicator species. Data are deficient for assessing the IUCN Red List rating for the longtail tuna, which is a highly commercial species.

Benthopelagic species that feed on prey from bottom, mid, and surface waters include the tiger tooth croaker, silver pomfret, and diamond mullet, which are all species found in the fish market surveys and meet the requirement of being consumed by local populations. In addition, the black pomfret is a commercially valuable species, although it was not a dominant species in the fish market surveys we reviewed. Unfortunately, the IUCN Red List rating for the tigertooth croaker and silver pomfret is "Vulnerable" and only the black pomfret and diamond mullet populations are rated as "Least concern" within the benthopelagic species group.

Coral reef–associated fish include many commercially valuable species that are eaten locally and have an IUCN Red List rating of "Least concern." This group includes the Malabar trevally, golden trevally, spangled emperor, twobar seabream, and white-spotted spinefoot. The orange-spotted grouper, while also in this coral reef–associated group, is a species with an IUCN Red List rating of "Vulnerable" in the Gulf (IUCN, 2019) and so should not be considered as a bioindicator. Similarly, the Haffara seabream has an IUCN Red List rating of "Near threatened" and should also not be used as a bioindicator. Two other coral reef associated species for consideration include the pickhandle barracuda and blackspotted rubberlip, which are commercially valuable.

Finally, the demersal bottom-feeding species included Indian halibut, largetooth flounder, king soldier bream, deep flounder, Oriental sole, Belanger's croaker, largescale tonguesole, and Klunzinger's mullet. These species are commercially valuable with the Oriental sole and Belanger's croaker also being listed as species evaluated in fish market surveys (Table 4). All of the demersal species have an IUCN Red List rating of "Least concern" except for the Indian halibut where data are deficient and Klunzinger's mullet which has a "Vulnerable" rating and should not be selected as a demersal bioindicator species. The IUCN (2019) designated the smooth tooth blacktip shark as "Endangered" — the highest extinction rating designated for any Gulf fish on our potential bioindicator list. Fortunately, this species is not of any commercial interest in the Gulf region. For these reasons, the smooth tooth blacktip shark also should not be considered as a bioindicator, despite its demonstrated capacity for bioaccumulating both As and Hg.

Based on examination of the matrix table (Table 9) developed from our study results, we selected 13 species as having the best combination of characteristics to serve as bioindicator species in the Gulf for the 4 metals studied here (Table 10).

4. Discussion and recommendations

4.1. Arsenic

Inorganic As has long been recognized as a poison in humans that can cause adverse health effects including stomach and intestinal problems (ATSDR, 2007a). Diet is the largest source of As for humans, and the predominant source of As in the human diet is generally seafood (ATSDR, 2007a). However, bioconcentration factors (BCFs) for As species in fish under experimental conditions are relatively low BCF = 17 for inorganic As (As + 3), BCF = 6 for inorganic As (As + 5) and BCF = 9 for organo-arsenicals (U.S. EPA, 1985a). Most of the As in fish tissue is in the form of organic arsenobetaine, which has a much lower toxicity than inorganic forms of As (ATSDR, 2007a; U.S. EPA, 2000b). Limited information is available, however, about the toxicity of oral exposures to organic As compounds in humans (ATSDR, 2007a).

Krishnakumar et al. (2016), the only study that conducted an analysis for both total and inorganic As species in Gulf fish, found inorganic As concentrations from 0.002 mg/kg to 0.005 mg/kg, with inorganic As representing < 0.08% of the total As content. The maximum inorganic As concentration is a factor of 400 below the MAL for inorganic As (2 mg/kg) (Table 2). A study by Peshut et al. (2008) reported similar concentrations of inorganic As species in fish in American Samoa. In that study, inorganic As averaged 0.5% of total As (ranging from < 0.05% to 4.4%) for fish, with concentrations ranging from 0.0091 to 0.0258 mg/kg. Peshut et al. (2008) concluded that their findings supported two important points about As toxicity. First, the total As concentration in seafood is not a good benchmark of As toxicity to humans. Secondly, chemical speciation analysis is needed to accurately assess the human health risks of inorganic As in seafood.

Because methods for As speciation analysis were emerging in the late 1990's, the U.S. EPA, 2000b recommended inorganic As for analyses in fish tissue samples for determining human health risks or if total As was measured, then assume that 10% of the total As is the more toxic inorganic As. This conversion approach for estimating inorganic As from the total As concentration, however, has been questioned by several researchers and may overestimate (by a factor of 10 to ≥100) the true human health risks associated with inorganic As that have been reported in several fish studies (Greene and Crecelius, 2006; Krishnakumar et al., 2016; Peshut et al., 2008). A literature review by Lorenzana et al. (2009) of As species found in fish worldwide, reported that the percent of inorganic As in estuarine and marine fish generally does not exceed 7.3%. All four of the authors previously mentioned concur with the U.S. EPA (2000b) that human risk assessment decisions should be based on an analysis of inorganic As species, not the total As concentration. Similarly, because there appears to be no correlation

Table 9
Potential Gulf bioindicator species—Metals[a] bioaccumulated, Trophic status, Feeding strategy, Habitat designation, Commercial status, and IUCN Red List Rating.

Common name	Species name	As	Cd	Pb	Hg	Trophic status[b]	Feeding strategy[c]	Habitat designation[d]	Commercial status	IUCN Red List Rating[e]
Yellowfin tuna	***Thunnus albacares***		●	●		**4.48**	**Fish, squid, and crustaceans**	**Pelagic (migratory)**	**Highly commercial**	**Near threatened**
Narrow-barred Spanish mackerel	***Scomberomorus commerson***		●	●	●	**4.38**	**Small fish, squid, and shrimp**	**Pelagic**	**Highly commercial; FMS**	**Vulnerable**
Indo-Pacific king mackerel	*Scomberomorus guttatus*		●	●		4.28	Small fish, squid, and crustaceans	Pelagic	Highly commercial	Least concern
Yellow spotted trevally	*Carangoides fulvoguttatus*	●	●	●		4.02	Small fish and invertebrates	Pelagic, coral reef–associated	Commercial fishery	Least concern
Longtail tuna	*Thunnus tonggol*		●	●	●	4.01	Fish, cephalopods, and prawn	Pelagic	Highly commercial	Data deficient
Pink ear emperor	*Lethrinus lentjan*		●	●		3.87	Crustaceans, echinoderms, mollusks, worms, and fish	Pelagic, coral reef–associated	Highly commercial; FMS	Least concern
Tigertooth croaker	***Otolithes ruber***		●		●	**3.60**	**Fish, prawn, and other invertebrates**	**Benthopelagic, coral reef–associated**	**Commercial fishery; FMS**	**Vulnerable**
Silver pomfret	***Pampus argenteus***		●	●	●	**3.23**	**Zooplankton: salps and medusa ctenophores**	**Benthopelagic**	**Highly commercial; FMS**	**Vulnerable**
Black pomfret	*Parastromateus niger*		●	●		2.93	Zooplankton	Benthopelagic, coral reef–associated	Highly commercial	Least concern
Diamond mullet	*Planiliza alata*		●	●		2.33	Phyto and zooplankton, detritus, snails, benthic algae	Benthopelagic	Minor fishery; FMS	Least concern
Pickhandle barracuda	*Sphyraena jello*		●	●		4.29	Fish and squid	Coral reef–associated	Commercial fishery	Least concern
Blackspotted rubberlip	*Plectorhinchus gaterinus*	●	●	●		3.99	Benthic crustaceans and finfish	Coral reef–associated	Commercial fishery	Least concern
Orange-spotted grouper	***Epinephelus coioides***		●	●	●	**3.95**	**Small fish, shrimp, crabs**	**Coral reef–associated**	**Commercial fishery; FMS**	**Vulnerable**
Malabar trevally	*Carangoides malabaricus*		●	●		3.90	Crustaceans, squid, and fish	Coral reef–associated	Commercial fishery	Least concern
Golden trevally	*Gnathanodon speciosus*	●	●	●		3.84	Small fish and benthic invertebrates	Coral reef–associated	Minor fishery	Least concern
Spangled emperor	*Lethrinus nebulosus*		●	●		3.64	Fish, worms, echinoderms, molluscs, and crustaceans	Coral reef–associated	Highly commercial; FMS	Least concern
Haffara seabream	***Rhabdosargus haffara***		●	●		**3.50**	**Benthic invertebrates**	**Coral reef–associated**	**Commercial fishery; FMS**	**Near threatened**
Twobar seabream	*Acanthopagrus bifasciatus*	●	●	●		3.39	Benthic worms, molluscs, and crustaceans	Coral reef–associated	Minor commercial fishery	Least concern
White-spotted spinefoot	*Siganus canaliculatus*		●	●		2.76	Benthic algae and sea grasses	Coral reef–associated	Commercial fishery; FMS	Least concern
Smooth tooth blacktip shark	***Carcharhinus leiodon***	●			●	**4.0**	**NA**	**Demersal**	**No commercial interest**	**Endangered**
Indian halibut	*Psettodes erumei*		●	●	●	4.34	Fish and benthic invertebrates	Demersal	Commercial fishery	Data deficient
Large tooth flounder	*Pseudorhombus arsius*		●	●		3.84	Fish and benthic invertebrates	Demersal	Commercial fishery	Least concern
King soldier bream	*Argyrops spinifer*		●	●		3.71	Benthic invertebrates, mainly molluscs	Demersal	Commercial fishery	Least concern
Deep flounder	*Pseudorhombus elevatus*		●	●		3.50	Benthic animals	Demersal	Commercial fishery	Least concern
Oriental sole	*Brachirus orientalis*		●	●	●	3.50	Invertebrates, mainly crustaceans	Demersal	Commercial fishery; FMS	Least concern
Belanger's croaker	*Johnius belangerii*		●		●	3.27	Benthic invertebrates, mainly worms	Demersal	Minor commercial fishery; FMS	Least concern
Largescale tonguesole	*Cynoglossus arel*		●	●		3.26	Benthic invertebrates	Demersal	Commercial fishery	Least concern
Klunzinger's mullet	***Liza klunzingeri***		●	●	●	**2.63**	**Detritus, algae, small invertebrates, and fish eggs**	**Demersal**	**Commercial fishery**	**Vulnerable**

FMS = This fish species was one of the most frequently analyzed in the fish market surveys
NA = Information not provided in FishBase (Froese and Pauly, 2017).
Notes: Fish species that are shown in bold type have a higher IUCN extinction rating and therefore should not be used as bioindicators if their populations are threatened or are not sustainable.
[a] These species were found to be in the top quartile of species that accumulated each metal and exceeded the MAL for two or more of the four metals of concern.
[b] Trophic status: Derived from values in FishBase (Froese and Pauly, 2017) in which herbivores are designated as 2 to 3, carnivores are 3 to 4, and top-level apex predator species are 4 or greater. Note: Species are ranked by trophic status from highest to lowest score within each of the habitat designations (pelagic, benthopelagic, coral reef associated, and demersal).
[c] Feeding strategy: Derived from information in FishBase (Froese and Pauly, 2017) about prey preference.
[d] Habitat: Derived from information in FishBase that includes pelagic, benthopelagic, demersal, and coral reef–associated species (Froese and Pauly, 2017).
[e] The species extinction rating was based on the IUCN's Red List of Threatened Species version 2019–1 (https://www.iucnredlist.org/), which determines the overall trends in species extinction risk. Red List ratings range from low to high risk (i.e., “Not evaluated,” “Data deficient,” “Least concern,” “Near threatened,” “Vulnerable,” “Endangered,” “Critically endangered,” “Extinct in the wild,” and “Extinct”).

Table 10
Recommended bioindicator fish species for use in the Gulf.[a]

Pelagic	Benthopelagic	Coral reef associated	Demersal
Pink ear emperor *Lethrinus lentjan*	**Black pomfret** *Parastromateus niger*	**Spangled emperor** *Lethrinus nebulosus*	**Oriental sole** *Brachirus orientalis*
Indo-Pacific king mackerel *Scomberomorus guttatus*	**Diamond mullet** *Planiliza alata*	**White-spotted spinefoot** *Siganus canaliculatus*	**Large tooth flounder** *Pseudorhombus arsius*
Yellow spotted trevally *Carangoides fulvoguttatus*		**Malabar trevally** *Carangoides malabaricus*	**King soldier bream** *Argyrops spinifer*
		Twobar seabream *Acanthopagrus bifasciatus*	**Deep flounder** *Pseudorhombus elevatus*

[a] Species with an IUCN rating of Least concern are recommended as bioindicators for the Gulf.

between the proportion of total As and inorganic As in fish samples in their respective studies, Krishnakumar et al. (2016), Lorenzana et al. (2009), and Peshut et al. (2008) believe it is misleading to use a conversion approach for deriving inorganic As concentrations from total As concentrations. Collecting quantitative data on the concentration of total As, but not the more toxic inorganic As, also has led to inappropriate regulatory actions. Based on total As analysis, a fish consumption advisory was issued by the State of Delaware (USA) when, ultimately, subsequent speciation analysis for inorganic As showed that the advisory was unnecessary (Greene and Crecelius, 2006).

It is noteworthy that we did not identify any exceedances of the MAL for inorganic As species (2.0 mg/kg) from the one study (Krishnakumar et al., 2016), which analyzed for inorganic As, despite these authors detecting elevated concentrations of total As (maximum of 20.24 mg/kg) that exceeded the total As MAL. Additional chemical speciation studies are needed that analyze for inorganic As in additional fish species in the Gulf to confirm whether the exceedances for total As also reflect exceedances for the more toxic inorganic As. If speciation analysis is undertaken, then use of the MAL of 2.0 mg/kg for inorganic As (Federal Register of Legislation, 2013) is appropriate.

Future monitoring for As contamination in fish should focus on industrial facilities associated with smelting operations (Schmitt et al., 1999) and petroleum combustion sites such as power plants as crude oil contains from 0.0024 to 1.63 mg As/kg (Pacyna, 1987). Monitoring should also focus on manufacturing sites where As-containing products are or have been produced including ammunition, lead-acid storage batteries, gallium-arsenide semiconductors for telecommunication products and solar cells, bottle glass, medicinal animal feeds, phosphate fertilizers, pressure-treated lumber, and As-containing herbicides (ATSDR, 2007a; Carapella, 1992; Eisler, 1988a; U.S. EPA, 2006; Senesi et al., 1999).

4.2. Cadmium

The widespread toxic effects of Cd in humans were first reported in a population living in the Jinzu River Basin, a heavily industrialized area of Japan, from consumption of Cd-contaminated rice grown in contaminated river water and fish from the Jinzu River. The exposure to Cd resulted in softening or brittleness of the bones, and decreased bone density leading to fractures in the long bones in affected individuals, typically post-menopausal women with poor nutritional status (ATSDR, 2012; Nordberg et al., 1997; Shigematsu, 1984). The effects of Cd exposure on bones were due to kidney damage (Elinder, 1985; Nogawa et al. 1987 and 1990) that disrupted renal vitamin D metabolism leading to changes in calcium absorption and increased calcium excretion (ATSDR, 2012; Buchet et al., 1990).

In the current review, over three quarters of the fish species samples contained mean Cd concentrations that exceeded the MAL—the highest percentage of species means exceeding any of the four MALs. Because of this and the toxicity of this metal to humans, continued surveillance of Cd in Gulf fish would be prudent. Future Cd monitoring should focus on industrial facilities associated with combustion of fossil fuels such as power plants, waste incineration and disposal sites, manufacturing plants for phosphate fertilizers, and smelting operations associated with non-ferrous metals (Cu, Pb, and Zn) (ATSDR, 2012; U.S. EPA, 1985b). Cadmium is also used in production of consumer products including batteries, pigments, and coatings and as a stabilizer in plastics (ATSDR, 2012) and sites where these activities occur also should be monitored for Cd contamination.

4.3. Lead

Fish can bioaccumulate Pb to a small degree, but Pb is not biomagnified up the aquatic food chain to fish (ATSDR, 2007b). For example, the median BCF value of 42 for fish is much lower than for shellfish (BCF values of 536 for oysters and 2570 for mussels) (Eisler, 1988b). Consuming fish even with minimal Pb concentrations, however, is problematic for pregnant and nursing women because this metal can cause severe neurological and cognitive impairment, especially in developing fetuses, nursing infants, and young children (ATSDR, 2007b). Exposure to high concentrations of Pb can severely and irreversibly damage the brain, may cause stillbirths and miscarriages, and inhibit fetal development in pregnant women (ATSDR, 2007b; Eisler, 1988b). Although the U.S. EPA (2000b) did not provide a screening value for Pb for protection of recreationally or subsistence fish consumers, the agency strongly recommended that any Pb exposure from fish consumption should be minimized to the greatest extent possible.

In the current review, more than half of the fish species means exceeded the MAL for Pb (Fig. 5). Because of its prevalence in the fish studies we reviewed and due to its toxicity especially to pregnant and nursing women, infants, and young children, continued surveillance of Pb in Gulf fish is highly recommended. Future monitoring for Pb contamination in fish should target areas receiving discharges from industrial facilities involved in combustion of fossil fuels (power plants), waste incineration sites, and facilities that were or are currently involved in manufacturing or recycling of Pb-containing products (ATSDR, 2007b). Lead and its compounds are primarily used in storage batteries for motor vehicles, but Pb also has diverse uses in a variety of consumer products including ammunition, weights, and pigments for glazes, dyes, and paints (ATSDR, 2007b). Many former uses of Pb in gasoline, paint, and Pb-arsenate pesticides, have been strictly regulated or eliminated entirely because of the metal's toxicity and persistence in the environment (ATSDR, 2007b; Eisler, 1988b).

4.4. Mercury

Methylmercury accumulates up the food chain, so that fish at the top of the food chain will typically have the highest concentrations in their tissues (ATSDR, 1999; Burger and Gochfeld, 2005; U.S. EPA, 1997). The major source of CH_3Hg+ for humans is through the consumption of Hg-contaminated fish. During the 1959 Minamata disaster in Japan, Hg in effluents from an industrial facility contaminated fish and shellfish, and many of the first victims were fishermen and their families (ATSDR, 1999). Consumption of Hg-contaminated fish results in a range of neurological problems (ATSDR, 1999; Fukuda et al., 1999; Harada, 1995; Hightower and Moore, 2003) and in severe Hg exposures, deaths (attributed to toxicity in the Central Nervous System) occurred (Tamashiro et al., 1984; Tsubaki and Takahashi, 1986). Pregnant women, nursing mothers, infants, and young children are particularly sensitive populations. Developing fetuses are at increased risk of suffering adverse neurological effects from Hg exposure and can be born with cognitive impairments similar to those of cerebral palsy (ATSDR, 1999; Marsh et al., 1987).

Overall, except for the small number of identified hotspots of Hg contamination, mean Hg concentrations in Gulf fish seem to be relatively low (the mean and median Hg concentration for all species were both lower than the MAL) (Fig. 3). During our review, only 10% of the species means exceeded the MAL for Hg, the lowest percentage of exceedances for any of the four toxic metals. We also screened the compiled Hg data by using the more stringent ambient water quality standard of 0.3 mg/kg for CH_3Hg+ promulgated by the U.S. EPA (2001) for the protection of human health. Although we used this more stringent standard, still only 15% of the fish species means exceeded this protective value.

Spatial analysis of Hg exceedances of the MAL during our review of Gulf fish showed that several of the most elevated Hg concentrations observed were associated with industrial waste discharges from chlor-alkali facilities. For example, a chlor-alkali facility in the Banda Imam Petrochemical Complex in Iran released Hg effluents into the Musa Estuary through a sewage outfall, subsequently contaminating estuarine and marine sediment and organisms (Fard et al., 2015; Godarzi Nik et al., 2012; Haghighat et al., 2011). As reported by Haghighat et al. (2011), an Iranian government report identified this chlor-alkali facility, which operated intermittently for 15 years, as having released an estimated 31,000 kg of Hg into the estuary within a 3-month period. Because Hg concentrations in fish substantially exceed the MAL by a factor of 30–40 at this site, several researchers recommended either the government remediate this site (Haghighat et al., 2011) or reduce the Hg pollution load to the Musa estuary (Fard et al., 2015). Most recently, Hosseini et al. (2016) reported that the Musa Estuary is affected by 19 petrochemical plants, including the chlor-alkali and a superphosphate fertilizer plant. These authors concluded that the observed trend of decreasing concentrations of metals (including Hg) in fish tissue with increasing distance from this estuary is strong evidence that the source of the contamination is from the petrochemical complex (Hosseini et al., 2016). Another chlor-alkali facility in Shuwaikh, near Kuwait City, was the major point source for Hg releases into Kuwait Bay. The facility was estimated to have released 21 t of Hg during its operations from 1964 to 1985 (Al-Majed and Preston, 2000; BuTayban and Preston, 2004). As shown in Fig. 6, more than half of the species means that exceeded the MAL occurred in the northwestern portion of the Arabian Gulf (from the Musa Estuary south to the Kuwait Bay) and are likely associated with extensive contamination from these two chlor-alkali facilities.

Surveillance of Hg contamination in fish should focus on chlor-alkali facilities and other industrial plants and municipal wastewater treatment facilities that are known to discharge Hg or CH_3Hg+ compounds. Mercury-contaminated effluents are associated with combustion of petroleum products that contain concentrations ranging from 0.001 to > 5 mg Hg/kg (Wilhelm, 2001), wastes from Portland cement production, and from the manufacture or recycling of electronic devices, switches, batteries, light bulbs, and thermometers (ATSDR, 1999; Carrico, 1985; Johnson et al., 2008). Given the elevated Hg concentrations in fish collected close to some of these industrial facilities, it may be appropriate to close or restrict commercial, artisanal, and recreational fishing in some adjacent harvest areas to protect public health. Continued surveillance of Hg concentrations in fish at point of sale outlets near identified hotspots is also warranted as is further evaluation of contamination in local populations that consume Hg-contaminated fish through human health risk assessments.

4.5. Regulatory action limits versus human health risk assessments

Elevated metal concentrations detected in Gulf fish are problematic with respect to meeting the MAL standards set for fish sold in international commerce. However, these MALs are regulatory action limits and are not human health risk assessments per se that evaluate health risks of consuming metal contaminated fish based on the fish consumption rate, the preferred species of fish consumed, the concentration of the metal in the fish tissues, the reference dose (RfD in mg metal/kg of body weight/per day) and body weight (kg) of the consumer (age and gender specific); all of which influence acceptable fish consumption limits (Burger and Gochfeld, 2005; U.S. EPA, 2000b). Global fish consumption per capita has increased from an average of 9.9 kg (1960s) to 17.0 kg (2000s), 18.9 kg (2010), and 19.2 kg (2012) and preliminary results for 2016 (20.3 kg) and 2017 (20.5 kg) continue to show this increasing trend (FAO, 2018). This upward trend in seafood consumption also is likely to be occurring in the Gulf nations. Current data on fish consumption related parameters are necessary to adequately assess human health risks in the Gulf region.

Consumption of fish that contain elevated concentrations of any of these four toxic metals could potentially result in adverse human health effects, especially for sensitive populations such as pregnant and nursing women, infants, and young children. In addition, recreational or subsistence fishers are also at a higher risk because they tend to consume larger amounts of fish than the general population (Burger et al., 2014, 2015; U.S. EPA, 2000b). In a consumption study conducted in Saudi Arabia, Burger et al. (2014) reported that dietary intake of fish also differed greatly between the general Saudi Arabian population and the multinational and multicultural expatriate community living in Saudi Arabia. In this dietary study of fish consumption patterns, Burger et al. (2014) estimated that native Saudis were consuming 150 g of fish per week, while expatriates were consuming 397 g of fish per week — a consumption rate of 2.65 times higher for the expatriates. Future research on fish consumption patterns in other Gulf nations are needed to understand whether similar disparity exists between other native Gulf residents and their respective expatriate communities.

Coinciding with the increasing trend in fish consumption, environmental contamination of Gulf waters also has increased with the rapid increase in population and subsequent expansion of industrial development and municipal services to serve this increasing population. It is important that surveillance monitoring of these metals in fish continue Gulf-wide, but especially in those areas of extensive industrial development considered to be potential contamination hotspots. Continued surveillance monitoring will ensure that data are available to assess compliance of fish with international MALs and more importantly to conduct local human health risk assessments, as needed, for the general population, sensitive populations, and for above average fish consumers especially in areas identified as contaminant hotspots.

4.6. Selection of bioindicator fish species for use in the Gulf

We identified 13 fish species that met the criteria as bioindicator species (Table 10); however, it is important that the species selected are relatively abundant in the study area. As previously discussed, several of the potential bioindicators were listed as “Near Threatened”, “Vulnerable” and one species was rated as “Endangered”. These ratings by the IUCN should preclude these species from being used as bioindicators.

Several authors have reported that many Gulf fish species are currently overfished (Grandcourt, 2012; Sale et al., 2011; Vaughan et al., 2019), subsequently threatening sustainability of the commercial fishery. Al-Abdulrazzak et al. (2015) reported that catch data from Gulf marine fisheries were potentially underestimated from 1950 through 2010 by a factor of 2. Although Gulf nations have reported their industrial and artisanal fish catches, Al-Abdulrazzak et al. (2015) believe Gulf nations have misreported by-catch, recreational, subsistence, and illegal fish catches. These authors found that the by-catch alone could represent as much as 18% of the total fish landings from the Gulf. When selecting bioindicator species, it is important to consider the IUCN Red List rating, other fisheries population data, and if the populations of the selected bioindicator species are already being stressed by a combination of overfishing and habitat loss.

We recommended 13 bioindicator species (Table 10**)** for use in the Gulf; however, the selection of these species has several limitations. Our recommendations were based on the ability of the bioindicator species to accumulate two of the four metals of concern that we studied and we limited our selection to fish species that bioaccumulated these metals in edible muscle tissue because we were most interested in the potential human health implications. Other potential bioindicator species may be identified that efficiently bioaccumulate other metals and are also important commercial species as well as being consumed by various local Gulf populations. Given our study limitations, it is critically important to use the expertise of local fisheries scientists in the Gulf nations to ensure 1) that the fish species selected meet all of the criteria for bioindicators for the metal or metals of interest to the researcher and 2) that a healthy, stable, and harvestable fisheries population can be sustainably maintained into the future.

4.7. Improving comparability of metal contaminant studies in the Gulf

During this review, we identified variability in studies across national jurisdictions of the Gulf. This variability was associated with the number and chemical species of metals analyzed, fish tissue types selected for analysis, types of studies conducted, and the fish species selected to assess metal contamination. This variability, particularly in the methodologies used, makes data comparability and interpretation more difficult and diminishes the usefulness of data exchange among Gulf scientists. The approaches listed below for Gulf-wide consideration, would harmonize fish contaminant data and improve data comparability.

4.7.1. Adopt a standard protocol for fish sampling and analysis procedures

The multi-national implementation of standard protocols for fish sampling, analysis, and reporting procedures and adherence to rigorous quality assurance /quality control procedures such as those prescribed by the existing *Manual of Oceanographic Observations and Pollutant Analyses Methods* (ROPME, 1999), could improve comparability of fish contaminant data Gulf-wide. This manual, however, was only referenced in a few of the 85 reviewed papers and is now 20-years old. The ROPME nations may want to convene an expert scientific panel to review and revise this manual with updated procedures that would improve data quality and comparability.

4.7.2. Use a standard source for nomenclature of Gulf fish species

Using a standardized source for scientific and common fish names such as FishBase (Froese and Pauly, 2017) would help harmonize species identification of Gulf fish. Use of local names or identification of fish to the genus level only is not sufficient and should be avoided. Use of standardized nomenclature would allow researchers to compare metal concentration data for individual fish species seamlessly across all Gulf studies.

4.7.3. Select and use several bioindicator species Gulf-wide

We have recommended 13 bioindicator species for consideration in the Gulf. Selecting several widely distributed bioindicator species that represent different habitat designations would help provide data about metal concentrations in key species across the Gulf from a variety of diverse environmental and metal exposure conditions.

4.7.4. Use a standard method to convert DW to WW and present data in both units

Researchers should use and reference a standard method for converting DW to WW metal concentrations. Provision of the WW:DW ratio is recommended by ROPME (1999). Ideally, researchers should provide both the DW and WW metal concentrations for each metal in each species in their publications to help improve comparability of reported data, eliminate potential conversion errors by other data users, and facilitate data sharing Gulf-wide. All tables and figures should be clearly labelled to show whether data represent WW or DW metal concentrations.

4.7.5. Develop and use a standardized set of current international MALs

MAL values have been evolving since the mid-1980s, and many have been revised multiple times as additional information on the toxicology of the metals has become available. This review illuminated a major issue—there was great variability in the MALs used in many of the Gulf studies which affected how the researchers evaluated and interpreted their results. Using outdated MALs led to some authors reporting that the analyzed fish tissue did not exceed the MALs when in fact the concentrations were in exceedance of the most up-to-date MAL at the time their study was conducted. This was not unexpected as we found it both difficult and time consuming to track some of the most recent revisions issued by international agencies. To protect human health however, it is important that a single, agreed upon standardized set of current MALs be used for screening fish metal concentrations Gulf-wide.

4.7.6. Identify locations where fish are harvested and sold

Provision of accurate spatial information (latitude and longitude coordinates) can identify the locations of contaminant hotspots that are useful to the scientific community as well as fisheries managers, and which also can help public health officials restrict access to contaminated harvest areas when consumer health is at risk. In addition, from a public health perspective, it is important to identify precisely both the harvest areas and the local fish markets. Whether metal contaminated fish are identified by harvest area or at the point of sale, these locations should to be identified so that trends in the metal concentrations can be evaluated as appropriate over time. Today, global positioning system (GPS) hand-held units can be taken into the field or to a fish market to collect accurate locational information.

4.7.7. Report mean contaminant values for individual fish species at each site

Metal concentrations are best reported for individual species collected at each sampling location to provide species-specific information on metal contamination in the event the location is identified as a contaminant hotspot. Averaging metal concentrations across several species at a single site provides a general indication of the overall level of metal contamination in the fish from a specific harvest area, however, it does not identify which species are most efficient at bioaccumulating metals and thus pose a greater risk to human health. Similarly, averaging metal concentrations in an individual species across several sites potentially averages concentrations from both contaminated areas with those of relatively unpolluted areas. Neither of these data analysis procedures provides information that is useful for identifying contaminant hotspots or those species with elevated tissue concentrations whose consumption may need to be restricted or avoided by consumers.

4.7.8. Document analytical minimum detection limits (MDL) and develop method for using values below the MDL for calculating mean concentrations

Over the course of this review it was difficult to assess the effects of improvements in analytical detection limits used across the various studies particularly because analytical sensitivity improved greatly beginning in the late 1990s (Sturgeon, 2000). In addition to the analytical methods used, differences also may exist regarding how the various authors handled concentrations below the MDL when calculating and reporting the mean concentration for a given species. The mean can vary greatly depending upon how it is calculated. Using the

MDL or ½ MDL as the default concentration for values below the MDL results in a higher mean value than using zero as the default in the same calculation, especially when there are large numbers of values below the MDL (U.S. EPA, 2000a and b; Sturgeon, 2000). Unfortunately, information on the MDL was not always reported nor was information provided on what value was used for calculating the mean when the concentration was below the MDL.

Because fish are an important food source in the Gulf, there is a need to establish a consistent, up-to-date, multi-national approach to be able to better assess trends in bioaccumulation of toxic metals in Gulf fish now and into the future. Inconsistencies in the procedures and MALs used across jurisdictions hinders data interpretation. Improving the comparability of fish contaminant data could have the added bonus of reducing overall sampling and analyses costs for all Gulf nations and improve the value of data exchange. The consistent, multi-national approach that we recommend could be achieved within the existing ROPME framework.

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