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Arsenic and arsenic species in shellfish and finfish from the western Arabian Gulf and consumer health risk assessment

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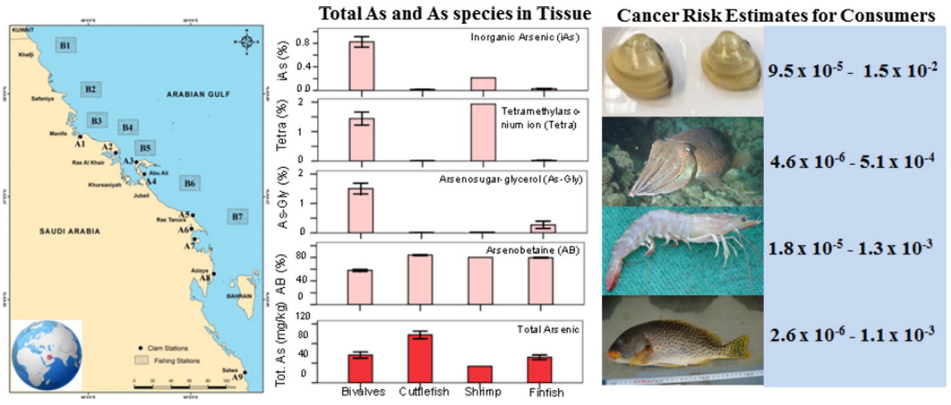
HIGHLIGHTS

- Arabian Gulf seafood contains relatively high concentrations of total arsenic.
- Non-toxic arsenobetaine forms the major fraction (~70%) of total arsenic.
- Toxic inorganic arsenic presents at only low levels (<1.84% of total As).
- No significant relationship between the tissue concentrations of total As and iAs.
- Health risk assessment shows non-negligible risks and the potential for hazards.

GRAPHICAL ABSTRACT



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ABSTRACT

This study reports the levels of total arsenic and arsenic species in marine biota such as clams (*Meretrix meretrix*; $N = 21$) and pearl oyster (*Pinctada radiata*; $N = 5$) collected from nine costal sites in Jan 2014, and cuttlefish (*Sepia pharaonis*; $N = 8$), shrimp (*Penaeus semisulcatus*; $N = 1$), and seven commercially important finfish species ($N = 23$) collected during Apr–May 2013 from seven offshore sites in the western Arabian Gulf. Total As and As species such as dimethylarsinic acid (DMA), arsenobetaine (AB), trimethylarsine oxide (TMAO), arsenocholine (AC), tetramethylarsonium ion (Tetra), arsenosugar-glycerol (As-Gly) and inorganic As (iAs) were determined by using ICPMS and HPLC/ICPMS. In bivalves, the total As concentrations ranged from 16 to 118 mg/kg dry mass; the toxic iAs fraction contributed on average less than 0.8% of the total As, while the non-toxic AB fraction formed around 58%. Total As concentrations for the remaining seafood (cuttlefish, shrimp and finfish) ranged from 11 to 134 mg/kg dry mass and the iAs and AB fractions contributed on average 0.03% and 81% respectively of the total As. There was no significant relationship between the tissue concentrations of total As and iAs in the samples. There was also no significant relationship between As levels in seafood and geographical location or salinity of the waters from which samples were collected. Based on our results, we recommend introducing a maximum permissible level of arsenic in seafood from the Gulf based on iAs content rather than based on total As. Our analyses of cancer risks and non-cancer hazards identified non-negligible risks and

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the potential for hazards; the greatest risks were identified for expatriate consumers of bivalves and high-end consumers of seafood. Despite this, many uncertainties remain that would be best addressed by further analyses.

1. Introduction

Although the high arsenic content of marine organisms, including that of popular seafood, was first observed almost 100 years ago (Jones et al., 1922; Chapman, 1926), the major chemical forms of this arsenic were first revealed following the identification of arsenobetaine in lobster in 1977 (Edmonds et al., 1977) and of arsenosugars in algae in 1981 (Edmonds and Francesconi, 1981). Subsequent research, based mainly on arsenic speciation analysis (Francesconi and Kuehnelt, 2004), has identified a wide range of naturally occurring arsenicals ranging from simple water-soluble forms such as dimethylarsinic acid (DMA) to complex arsenic-containing lipids (García-Salgado et al., 2012). Most of these arsenic species, however, occur at trace levels only, and the major arsenic species in marine organisms by far are arsenobetaine and arsenosugars.

Arsenic is a well-known toxic element but its toxicity depends on its chemical form. The inorganic forms of arsenic (iAs) are highly carcinogenic (Mandal and Suzuki, 2002; Wang et al., 2014a) and more recent epidemiological studies have also related iAs exposure to heart disease and diabetes (Wang et al., 2010; Wang et al., 2014b; James et al., 2015). On the other hand, the major forms of arsenic in seafood, namely arsenobetaine and arsenosugars, are considered non-toxic (Francesconi, 2010). Food as a source of iAs to humans is increasingly being investigated in arsenic risk assessment studies. In response to these concerns, the European Food Safety Authority has recently introduced a maximum permissible level of As in food that applies only to the iAs portion (ECR, 2015). This regulation acknowledges the special case of arsenic in seafood, where organoarsenic species predominate and iAs usually accounts for <1% of the total arsenic (EFSA, 2009).

Information on the chemical forms of arsenic, including iAs, and their concentrations in seafood is essential for consumer risk assessment (Lorenzana et al., 2009; Francesconi, 2010; Molin et al., 2012). The scientific opinions on arsenic in food, issued by the European Food Safety Authority (EFSA, 2009) and the FAO/WHO Joint Expert Committee on Food Additives (WHO, 2011), focus on dietary inorganic arsenic exposure - thus, arsenic speciation studies are essential to determine the toxic and nontoxic fractions of arsenic in seafood and for risk assessment.

Over one billion people all over the world rely on seafood as their primary source of animal protein. The global per capita fish consumption increased from an average of 9.9 kg in the 1960s to 17.0 kg in the 2000s, 18.9 kg in 2010 and 19.2 kg in 2012 (FAO, 2014). Total marine fisheries catches from the Arabian Gulf (hereafter "the Gulf") was estimated at around 400,000 t in 2010 (Al-Abdulrazzak et al., 2015). Recently, Burger et al. (2014) estimated the daily consumption rate for 12 commercially important finfish species from the Jeddah area of Saudi Arabia. The Gulf is characterized by elevated levels of salinity and sea surface temperature due to its shallow nature, restricted water exchange and high evaporation rates (Sheppard et al., 2010). Marine animals from high saline environments are likely to bioaccumulate high concentrations of arsenic (Larsen and Francesconi, 2003; Hong et al., 2014). Previous works (see references in Table 2) reported relatively high concentrations of total arsenic in biota collected from the western Gulf, compared to the published data from other parts of the world, and these levels were attributed to either the anthropogenic activities or natural biogeochemical processes in the region (de Mora et al., 2004). None of these studies, however, has investigated the arsenic species present in the biota from the Gulf. Globally, seafood consumers are exposed to persistent, bioaccumulative and toxic chemicals and there is an urgent need for improved replication and standardization of pollutant monitoring protocols for seafood (Bonito et al., 2015).

We report an investigation of the arsenic content in bivalves, squid, shrimp and finfish collected from sites within the western Gulf (Saudi waters), and examine the influence of sampling sites, salinity and animal type on the concentrations of the various arsenic species, with a focus on iAs. We also estimate the potential cancer risk and non-cancer hazards for native Saudi and expatriate consumers of selected seafood types from the study area resulting from iAs exposure.

2. Materials and methods

2.1. Study area and sampling

Sampling sites are shown in Fig. 1. Two bivalve species, the hard Asiatic clam *Meretrix meretrix* and the pearl oyster *Pinctada radiata* were handpicked from selected coastal sites (30–40 individuals from each site) along the Saudi Gulf coast in January 2014. These two species have been routinely used for environmental monitoring from the region (Naser, 2013). The Asiatic clam is a commercially important species inhabiting sand beds in the coastal bay systems. The clams (shell length = 30–40 mm) were collected from nine sites and pearl oysters (shell length = 40–50 mm) attached to the intertidal rocky substratum were collected from three sites during low tide and transported live to the laboratory where they were depurated by maintaining them in clean seawater for 24 h.

Samples of cuttlefish (*Sepia pharaonis*), shrimp (*Penaeus semisulcatus*) and seven finfish species were collected from seven offshore areas from Saudi waters while conducting experimental trawling during April–May 2013 (Fig. 1, Table 3). Soon after sampling, fish samples were washed in clean seawater, wrapped in polythene zip-bags and transported in ice chests to the lab where they were stored at −20 °C until analysis. In situ seawater parameters such as temperature, salinity, and pH were measured at the sampling sites using a multiprobe environmental monitoring system (YSI multi parameter water quality Sonde, model 6600 V2).

2.2. Sample preparation

Samples were prepared for analysis inside a clean laboratory with care taken to avoid cross contamination. Bivalve and fish samples were defrosted and total length and weight were measured. The whole soft tissue from bivalves was removed by using ceramic knives. The dorsal muscle samples from the finfish and edible portions from the cuttlefish and shrimp were collected by carefully removing the skin and any bony parts. Composite samples were prepared using the tissue samples from 3 to 6 animals from each site. The composite samples were spilt into three or (sometimes) two parts depending on the availability of tissue. All cleaned and separated tissue samples were minced, homogenized and freeze-dried; the dried samples were then crushed to make a powder and further homogenized before analyses.

2.3. Chemicals, standards and reference materials

Water (18.2 MΩ cm) provided from a Milli-Q Academic water purification system (Millipore GmbH, Vienna, Austria) was used throughout this work. Nitric acid (65% p.a., further purified by subboiling), pyridine (≥99%), trifluoroacetic acid (≥99.9%), ammonia solution (≥25% p.a.), hydrogen peroxide (30% p.a.) and formic acid (≥98% p.a.) were purchased from Carl Roth GmbH & Co (Karlsruhe.

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Fig. 1. Map showing the coastal (A1–A9) and offshore (B1–B7) sampling sites.

Germany). Malonic acid (>99%) was obtained from Sigma Aldrich (Vienna, Austria). Methanol HiPerSolv Chromanorm (LC-MS grade) was purchased from VWR International (Vienna, Austria). For the determination of the total As content by ICPMS, a 1000 mg As L^{-1} single element standard purchased from CPI International (Santa Rosa, CA, USA) was used. For speciation analysis, standards of arsenic species were prepared from stock solutions containing 1000 mg As L^{-1} of each of the following compounds: arsenobetaine, trimethylarsine oxide, arsenocholine, tetramethylarsonium iodide, and arsenosugar-glycerol (purity of compounds >99% by NMR and HPLC/mass spectrometry, previously synthesized in house), dimethylarsinate prepared from sodium dimethylarsinate (Fluka, Buchs, Switzerland), and arsenate prepared from Na_2HAsO_4 $7H_2O$ (Merck, Darmstadt, Germany). A characterized extract of *Fucus serratus* was used as a source of the anionic arsenosugars (Madsen et al., 2000).

2.4. Quality control

For quality control and method validation of total arsenic measurements, we used three certified reference materials: (i) IAEA-407 fish homogenate (International Atomic Energy Agency, Vienna, Austria) with a certified [As] = 12.6 ± 1.2 mg As kg^{-1}; we obtained [As] = 12.7 ± 0.2 mg As kg^{-1}, n = 4; (ii) BCR®-627 Tuna fish tissue (IRMM-Institute for Reference Materials and Measurement, Geel, Belgium) with certified [As] = 4.8 ± 0.3 mg As kg^{-1}; we obtained 5.01 mg As kg^{-1}; 5.05, 4.97; n = 2; and (iii) ERM®-BC211 Rice (IRMM) with certified [As] = 260 ± 13 μg As kg^{-1} (we obtained 260 μg As kg^{-1}; 254, 265; n = 2).

For method validation of arsenic speciation analysis, we used BCR®-627 Tuna fish tissue with certified content of dimethylarsinic acid of 0.150 ± 0.023 mg As kg^{-1} (obtained mean = 0.148 mg As kg^{-1}; 0.138, 0.157; n = 2), and arsenobetaine of 3.90 ± 0.23 mg As kg^{-1}

(obtained mean = 3.17 mg As kg^{-1}; 3.20, 3.13; n = 2); and ERM®-BC211 Rice with certified content of inorganic arsenic of 124 ± 11 μg As kg^{-1} (we obtained 113 μg As kg^{-1}; 112, 114; n = 2).

2.5. Instrumentation

ICPMS measurements were performed with an Agilent ICPMS 7500ce (Agilent Technologies, Waldbronn, Germany) equipped with a PC3 ESI cyclonic spray chamber (Elemental Scientific, Mainz, Germany) and a Burgener Ari Mist HP nebulizer (Burgener, Research International, Berkshire, UK). For HPLC measurements, the ICPMS was coupled to an Agilent 1100 Series HPLC system equipped with binary bump, solvent degasser, column oven and an auto sampler with a variable 100 μL injection loop.

Extraction solutions were centrifuged with a Hettich Rotina 420R (Hettich GmbH & Co. KG, Tuttlingen; Germany) centrifuge and acid digestions were performed with an Ultraclave IV microwave system (MLS GmbH, Leutkirch, Germany).

2.6. Determination of the total arsenic content by ICPMS

Portions (ca 100 mg) of samples were weighed to a precision of 0.1 mg directly into 12 mL quartz tubes. Nitric acid (2 mL) and water (2 mL) were then added and the samples were mineralized (digested) in the microwave system under an argon pressure of 4×10^6 Pa at 250 °C for 30 min. After mineralization and cooling to room temperature, the digest solutions were transferred to 15 mL polypropylene tubes (Greiner Bio-One International GmbH, Kremsmünster, Austria) and diluted with water to 10 mL; for analysis, the digested samples were further diluted 1 + 19 (*v*/*v*) with water. Nitric acid and an internal standard solution containing 100 μg L^{-1} Ge, Rh, In, Lu and Ir in 1% nitric acid were added to give final concentrations of 20% HNO_3 (*v*/*v*) and 10 μg L^{-1} Ge, Rh, In, Lu and Ir. Calibration standards were prepared in the same manner as digested samples containing 20% HNO_3 (*v*/*v*) and 10 μg L^{-1} internal standard for matrix matching. The arsenic concentration in the digests was determined by ICPMS. The ICPMS was operated with 12% optional gas (1% CO_2 in Ar) to enhance the arsenic signal and using helium (5 mL min^{-1}) as collision cell gas for removing polyatomic interferences from argon chloride ($^{40}Ar^{35}Cl$ on ^{75}As). The monitored masses were *m*/*z* 72, 74, 75, 77, 82 and 115. Calibration range was 0.1–500 μg As L^{-1}. Germanium (*m*/*z* 74) was used as internal standard to normalize for matrix effects on the arsenic signal. The additional internal standard elements (Rh, In, Lu and Ir) were used to matrix-normalize for other elements not reported in the current study.

2.7. Determination of arsenic species by HPLC/ICPMS

2.7.1. Extraction of inorganic arsenic and anionic organic arsenic species, and measurement by HPLC/ICPMS

The method applied is based on the method developed by Raber et al. (2012). Briefly, portions (200 mg) of the sample were weighed to 0.1 mg in 50 mL polypropylene tubes. Then 10 mL of a 0.1 M trifluoroacetic acid solution containing 1% (*v*/*v*) of a 30% (*w*/*w*) hydrogen peroxide solution were added. The samples were shaken in a GFL-1083 shaking water bath (Gesellschaft für Labortechnik, Burkwedel, Germany) for 1 h at 95 °C. After cooling to room temperature, the samples were centrifuged for 15 min at 4500 rpm (4750 g). The supernatant was removed and filtered through a 0.2 μm Nylon™ HPLC syringe filter, and the content of inorganic arsenic was determined by anion-exchange HPLC/ICPMS using a Hamilton PRP-X100 column (4.6 × 150 mm, 5 μm) at 40 °C with malonic acid buffers at 1 mL min^{-1}; Calibration standards were prepared in water; the calibration range was 0.2–10 μg As L^{-1} for As (V). The ICPMS was operated in no gas mode with 12% optional gas (1% CO_2 in Ar) to enhance the arsenic signal. Monitored masses were *m*/*z* 75, 77, 82 and 53. Integration times were 0.3 s, 0.3 s, 0.3 s and 0.05 s respectively. Note that the sample preparation oxidizes arsenic (III) species to arsenic (V), and hence provides a combined iAs value.

2.7.2. Extraction of water-soluble cationic organic arsenic species and measurement by HPLC/ICPMS

Portions (ca 100 mg) of freeze-dried samples were weighted to 0.1 mg in 15 mL polypropylene tubes. Then 10 mL of methanol/water (1/1, *v*/*v*) were added and the mixture was shaken overnight on a mechanical rotation device. Afterwards, samples were centrifuged for 15 min at 4500 rpm (4750 g); then the supernatant was removed and filtered through a 0.2 μm Nylon™ HPLC syringe filter (Markus Bruckner Analysentechnik, Linz, Austria). The extract was stored at 4 °C; it was diluted 1 + 9 (*v*/*v*) with water immediately prior to HPLC/ICPMS analysis. HPLC separation of the cationic organic arsenic species in the water/methanol extracts was performed with the silica-based cation-exchange column IonoSpher 5C (100 × 3 mm; 5 μm; Agilent Technologies, Waldbronn Germany) and a 10 mM pyridine buffer pH 2.6 (pH adjusted with formic acid) at 30 °C as mobile phase at a flow rate of 1 mL min^{-1}. Injection volume was 20 μL. Calibration standards were prepared in water; the concentration range was 1–100 μg As L^{-1} for dimethylarsinic acid (DMA), arsenobetaine (AB), trimethylarsine oxide (TMAO), arsenocholine (AC), and tetramethylarsonium ion (Tetra). As described above, the ICPMS was operated in no gas mode with 12% optional gas (1% CO_2 in Ar) and monitoring *m*/*z* 75, 77, 82 and 53, with integration times of 0.3 s, 0.3 s, 0.3 s and 0.05 s, respectively.

2.7.3. Extraction of lipid-soluble organic arsenic species and measurement by HPLC/ICPMS

We followed the method reported by Glabonjat et al. (2014) for analysis of clams for arsenolipids. A portion of 0.1 g of sample was extracted with 5 mL of DCM/MeOH (2 + 1, *v*/*v*); 4 mL of extract were evaporated to dryness and the residue was dissolved in 2 mL of DCM/Acetone (1 + 1, *v*/*v*); 1 mL of extract was purified on silica (3 fractions were collected: 5 mL of DCM/Acetone 1% formic acid, 3 mL of methanol 1% formic acid and 10 mL of methanol 1% aqueous NH_3); the 1% NH_3 ammonia fraction was evaporated to dryness and the residue was dissolved in 0.5 mL of ethanol and subjected to HPLC/ICPMS analysis (Reversed phase chromatography silica-based C8 and ethanol gradient; 20% split to ICPMS, carbon compensation and Ar/O_2 as optional gas).

2.7.4. Exposure assessment of iAs

We assessed carcinogenic risks and non-cancer hazards associated with iAs intake resulting from consumption of bivalves, cuttlefish, finfish and shrimp. Lifetime average daily doses (LADDs) of iAs resulting from consumption of the various types of seafood for native Saudis, expatriates, and the combined population were estimated using the formula:

$$\mathrm{LADD} = ([\mathrm{iAs}] \times \mathrm{IR})/\mathrm{BW}, \quad (1)$$

where [iAs] is our measured wet weight concentration of iAs in the respective type of seafood (in mg/kg), IR is the seafood intake rate (in kg/d), and BW is body weight (in kg BW). For finfish and bivalve samples, whose iAs concentrations were approximately log-normally distributed, median and 95th percentile values were used for central tendency and high-end estimation, respectively. Given the limited number of observations for cuttlefish, mean and maximum values were employed, and in the case of shrimp, the result of the single composite analysis was used. The rate of seafood consumption (IR) for finfish was estimated as the average rate of consumption for 12 commercially important finfish species from a published study of population seafood consumption behaviors in Saudi Arabia Jeddah area (Burger et al., 2014). People in their survey consisted of 53% male, 84% native Saudis and age ranged from <2 years old (4% of sample) to over 60 years (1% of sample), and 41% were between 20 and 40 years. They reported an average IRs of 68 g/d for native Saudis and 128 g/d for

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expatriates. Based on the results of Burger et al. (2014), the average and 90th percentile IR for the combined population were estimated as 91 g/d and 256 g /d respectively. Intake rates for bivalves, cuttlefish and shrimp were unavailable; thus, the IR for finfish was also used for each of these seafood types. Body weight values of 79.7 kg (Al-Baghli et al., 2008) and 60.0 kg (Ahmed et al., 2015; Ahn et al., 2009; Liu et al., 2007) were used for the native Saudi and expatriate subpopulations, and a weighted value of 72.5 kg was used for the combined population, assuming the population comprised 74% native Saudis (Salam, 2013).

2.7.5. Selection of quantitative toxicity metrics and characterization of risks and hazards

Controversy exists regarding the appropriate quantitative toxicity metrics for use in iAs risk assessment. The US Environmental Protection Agency's (EPA) Integrated Risk Information System (IRIS) program is currently in the midst of updating its oral cancer slope factor (CSF) and oral reference dose (RfD), a process that has involved multiple formal, lengthy reviews by the National Research Council (NRC) (NRC, 2014). In the meantime, the IRIS database currently houses a toxicological review for iAs that has a CSF (1.5 per mg/kg day^{-1}, corresponding to skin cancer) and RfD (0.0003 mg/kg day^{-1}) that have not been updated since the mid-1990s (US EPA, 2015a), thus not considering the past 20 years of epidemiologic evidence for various cancer and non-cancer endpoints. In 2010, EPA released a draft reassessment of iAs carcinogenicity only, and proposed a new CSF of 25.7 mg/kg day^{-1}, corresponding to lung and bladder cancers (US EPA, 2010). The draft reassessment was never finalized, and to date, neither the EPA nor the NRC has provided explicit guidance regarding the CSF that should be used in iAs risk assessment in the interim. For this reason, our assessment employed both CSFs, and presents a range of risk estimates generated using the older and newer slope factors. Cancer risk for seafood consumers was calculated as:

$$\text{Risk} = \text{LADD} * \text{CSF}. \quad (2)$$

Currently, no new proposed values are available for the RfD, despite a considerable body of literature examining potentially more sensitive endpoints (NRC, 2014). Given this, our assessment employs the existing EPA IRIS RfD mg/kg day^{-1}, in full recognition of its limitations. Non-cancer hazard was thus estimated as:

$$\text{HQ} = \text{LADD}/\text{RfD}, \quad (3)$$

where HQ is the hazard quotient, LADD is the lifetime average daily dose (calculated above), and the RfD is the EPA IRIS RfD.

3. Results and discussion

3.1. Total arsenic concentrations and organoarsenic species distribution in bivalves from coastal sites

The mean, minimum and maximum tissue concentrations (mg/kg dry wt) of total As and As species in clam (*M. meretrix*) and oyster (*P. radiata*) samples collected as part of our study are summarized in Table 1. The mean total As concentrations in clams (nine sites) and oysters (three sites) ranged from 17–100 mg/kg and 38–87 mg/kg, respectively. We found, however, no clear trend in these data in regard to geographical location or salinity of the waters from which they were collected.

Previous studies on arsenic in bivalve molluscs have shown a wide range of concentrations. Table 2 shows some representative results from these studies and compares them with the average values for our Gulf samples. The accumulation of high levels of arsenic by marine organisms is a well-known natural phenomenon, and there is no suggestion of anthropogenic contamination being a factor. The total As levels we observed in bivalves from the study area are higher than the values reported from relatively uncontaminated environments, and are comparable to the values reported from some of the contaminated sites in Taiwan, Canada and Europe (Table 2).

Our arsenic speciation data (Table 1), based on measurements by using HPLC/ICPMS, show that arsenobetaine is the major arsenic species in clams and oysters from the Gulf, a result consistent with the general pattern of arsenic species in bivalve molluscs worldwide (Francesconi and Kuehnelt, 2002). We also found Tetra to be a significant arsenic species in clams (1–3.2% of total As), but it was not quantifiable in oysters (Table 1). Clams are known to contain Tetra at higher levels than do other organisms, the reasons for which are not currently clear (Shiomi et al., 1987; Francesconi and Edmonds, 1998). The levels of AB in clams from our study, as expected, correlated strongly with total As levels ([AB] = 0.71[As] − 5.9 mg/kg, $R^2 = 0.98$), and the levels of Tetra also correlated, albeit less strongly, with both total As ([Tetra] = 0.019[As] − 0.04 mg/kg, $R^2 = 0.61$) and AB ([Tetra] = 0.026[AB] + 0.12 mg/kg, $R^2 = 0.60$). Other small cationic organoarsenic As species such as AC and TMAO were sometimes detected in the bivalves from the study area but were below quantifiable levels (Table 1).

All Gulf bivalves also contained the arsenosugar-glycerol (As-Gly); arsenosugars are usually a major arsenic species in bivalves, sometimes collectively present at levels matching the arsenobetaine levels. The only arsenosugar we report quantitatively is As-Gly because it was

Table 1
The tissue concentrations (mg/kg dry wt) of total As and As species in clam (*M. meretrix*) and oyster (*P. radiata*) samples collected from coastal sites in the Arabian Gulf. The percentage of As species to the total As values are given in parentheses next to the mean value. The minimum and maximum values are given below the mean value within parentheses. (N = number of composite samples, each comprising 30–40 individuals).

St Name	N	Salinity (psu)	Total As	DMA	AB	As-Gly	Tetra
Clam							
Manifa	2	50	43 (40–46)	0.19 (0.4%) (0.19–1.2)	26(60%) (24.1–27.1)	0.45 (1.1%) (0.36–0.55)	0.57 (1.3%) (0.54–0.61)
Ras Al-Khair	2	45	38 (36–41)	0.15 (0.4%) (0.13–0.16)	22(57%) (21 − 22)	0.39 (1%) (0.37–0.42)	0.63 (1.7%) (0.61–0.64)
Abu Ali North	2	45	100 (99–101)	0.41 (0.4%) (0.27–0.55)	67 (67%) (65–69)	2.5 (2.5%) (1.4–3.61)	0.97 (1.0%) (0.87–1)
Abu Ali South	2	52	94.4 (93–96)	0.23 (0.3%) (0.12–0.34)	60 (63%) (56–64)	1.6 (1.7%) (0.62–2.6)	2.0 (2.1%) (1. 9–2.1)
Ras Tanura	2	46	48 (47–48)	0.24 (0.5%) (0.24–0.25)	26 (55%) (25–27)	0.92 (1.9%) (0.88–0.96)	<dl
Tarut Bay	3	45	17 (16–18)	0.26 (1.5%) (0.24–0.27)	6.7 (40%) (6.3–7.3)	0.46 (2.8%) (0.25–0.67)	0.26 (1.6%) (<dl - 0.54)
Tarut Island	2	45	39 (36–41)	0.20 (0.5%) (0.19–0.21)	22(57%) (21–22)	0.33 (0.9%) (0.33–0.34)	1.3 (3.3%) (1.07–1.44)
Aziziyah	2	55	24 (23–25)	0.19 (0.8%) (0.18–0.2)	10.6 (44%) (9.6–12)	0.44 (1.9%) (0.42–0.47)	0.55 (2.3%) (0.47–0.63)
Salwa	4	60	97 (75–118)	0.23 (0.3%) (0.14–0.34)	62 (64%) (44–77)	1.4(1.5%) (1.1–1.9)	2.2 (2.3%) (1.9–2.5)
Oyster							
Manifa	2	50	38 (38–38)	0.11 (0.3%) (0.10–0.12)	25 (67%) (25–26)	0.14 (0.38%) (0.13–0.16)	<dl
Abu Ali North	2	45	49 (49–50)	0.21 (0.4%) (0.21–0.22)	29 (60%) (29–30)	0.39 (0.8%) (0.37–0.41)	<dl
Salwa	1	60	87	0.27 (0.3%)	64 (74%)	0.66 (0.8%)	<dl

dl = detection limit (0.01 mg As/kg dry wt).
Note: Traces of TMAO and AC were detected in some of the bivalve samples, but were not present at quantifiable levels.

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Table 2
Total arsenic concentrations (mg/kg, dry wt) and major organoarsenic species in bivalves, cuttlefish, shrimp, and finfish from various regions worldwide. The soft tissue of bivalve was analyzed while in the case of remaining species the muscle tissue was analyzed. Samples were collected from the wild unless otherwise indicated.

Study region	Species	Tot As	Major As species	Ref
Bivalves				
Arabian Gulf (Saudi Arabia)	*Meretrix meretrix*	58 (16–118)	AB	This study
	Pinctada radiata	52 (38–87)	AB	
Arabian Gulf (Saudi Arabia)	*M. meretrix*	77 (42–102)	NS	Fowler et al., 1993
	Pinctada margaritifera	48 (23–73)	NS	
	Sacostrea cucullata	17 (14–23)	NS	
Arabian Gulf (Bahrain and UAE)	*Pinctada radiata*	32.6 (4.5–73)	NS	de Mora et al., 2004
Arabian Gulf (Oman and UAE)	*Sacostrea cucullata*	16.7 (11–29.7)	NS	
India (Sundarban)	*Meretrix meretrix*	10.3	AB	Fattorini et al., 2013
Thailand, Map Ta Phut	*Perna viridis*[a,b]	10.9 (9.3–12.1)	AB	Rangkadilok et al., 2015
	Meretrix meretrix	10.7 (9–12.2)	AB	
Taiwan, Southwest Coast	*Crassostrea gigas*	9.9 (8.4–11.1)	AB	Liu et al., 2006
	Meretrix meretrix[b]	8.5 (7.6–10.7)	AB	Liu et al., 2007
Turkey, Izmir Bay	*Mytilus galloprovincialis*[a]	12.1 (11–15.5)	NS	Kucuksezgin et al., 2014
Hotspots in Europe	*Mytilus galloprovincialis*[a]	11 (8.1–16)	NS	Maulvault et al., 2015
Spain, North and Northwest coast	*Mytilus galloprovincialis*	10.1 (6.4–13.3)	NS	Besada et al., 2014
Nova Scotia, Canada	*Mytilus edulis*	69 (34–109)	AB	Whaley-Martin et al., 2012
Nova Scotia, Canada	*Mya arenaria* (contaminated)	1120 (1090–1140)	iAs	Koch et al., 2007
	Mya arenaria (Control)	37.8 (35–39.5)	AB	
North-eastern Adriatic (Gulf of Trieste)	*Mytilus galloprovincialis*	24.3 (14.7–29.9)	AB	Kristan et al., 2014
Fish, cuttlefish and shrimp				
Arabian Gulf (Saudi Arabia)	Marine fishes	52 (11–92)	AB	This study
	Cuttlefish (*Sepia pharaonis*)	98 (74–134)	AB	
	Shrimp (*Penaeus semisulcatus*)	34	AB	
	Marine fishes[a]	26.6 (0.8–161.5	NS	Attar et al., 1992
	Penaeus semisulcatus[a]	79 (30–176	NS	
Arabian Gulf (UAE, Qatar, Bahrain & Oman)	Grouper fish	3.4 (0.83–14.4)	NS	de Mora et al., 2004
	Spangled emperor fish	4.5 (2.5–10)	NS	
Red Sea and Arabian Gulf (Saudi Arabia)	Marine fishes[a]	9.8 (0.3–471)	NS	Burger et al., 2015
Kuwait (Arabian Gulf)	Coastal shark[a]	137 (80–205	NS	Moore et al., 2015
	Giant sea catfish[a]	77.5 (65–105)	NS	Al-Zaidan et al., 2015
Persian Gulf, Iran	Fishes	0.78 (0.84–2.4)	NS	Raissy and Ansari, 2014
Persian Gulf, Iran	Marine fishes[a,b]	1.7 (0.8–4.2)	NS	Saei-Dehkordi et al., 2010
Taiwan, Central	Cephalopods[a]	29.55 (1.95–115)	AB	Lin et al., 2008
	Fish[a]	9.8 (3.7–30.3)	AB	Lin et al., 2008
Turkey, Izmir Bay	Fish (*Mullus barbatus*)[a]	152 (15.5–110.5)	NS	Kucuksezgin et al., 2014
Hotspots in Europe	Fish (*Platichthys flesus*)[a]	32	NS	Maulvault et al., 2015
Spain	Cuttlefish (*Sepia officinalis*)	0.86[c] (0.45–2.15)	NS	Olmedo et al., 2013
	Fish (*Scomber scombrus*)[a]	0.95[c] (0.5–1.5)	NS	
	Shrimp (*Parapenaeus longirostris*)[a]	3.7[c] (1.9–4.42)	NS	
Northern Adriatic Sea	Ray species	113 (32.4–362)	AB	Šlejkovec et al., 2014

[a] Data converted to dry wt basis for comparison.
[b] Cultured or obtained from the local market.
[c] Median values; NS = Not Studied.

amenable to measurement by cation-exchange HPLC. The other three major arsenosugars, commonly reported in bivalves (Francesconi and Edmonds, 1997), are all anionic arsenic species. When we later employed anion-exchange HPLC for our samples, we applied extraction and HPLC conditions optimized for the quantification of iAs, and we did not quantify the anionic arsenosugars (see below). Table 1 shows that the sum of measured As cationic species accounts for about 50–75% of the extractable arsenic; the remainder is likely to be mainly anionic arsenosugars based on previous studies with bivalves (Francesconi and Edmonds, 1997).

We also looked for the presence of arsenolipids in the Gulf clam samples using the extraction and HPLC conditions reported by Glabonjat et al. (2014). The levels of total arsenolipid were low (<1%–4% of total As, median value 1%) and we did not identify the individual compounds. Analysis of the CRM Hijiki at the same time returned results comparable to those previously reported (Glabonjat et al., 2014), indicating that the procedure was working. It is possible, however, that a new group of arsenolipids, namely arsenic-containing phosphatidylcholines (Viczek et al., 2016), which were unknown to us at the time of these analyses, were present in the Gulf samples. These compounds recently were identified in fish roe (herring caviar) and are not amenable to the sample preparation steps applied in the current study.

3.2. Total Arsenic concentration and organoarsenic species distribution in cuttlefish, shrimp and finfish

The mean, minimum and maximum tissue concentrations (mg/kg dry wt) of total As and As species in cuttlefish (three sites), shrimp (one site) and finfish (seven sites) from our study are summarized in Table 3. The mean total As concentrations in cuttlefish and finfish ranged from 74–134 mg/kg and 11–92 mg/kg, respectively. The total As concentration in the shrimp sample was 34 mg/kg. Similar to the clam data, there was no clear trend in these data with reference to geographical location or salinity of the waters from which samples were collected.

Previous studies on arsenic in cuttlefish, shrimp and finfish have shown a wide range of concentrations. Table 2 gives some representative results from these studies and compares them with the average values for our Gulf samples. The accumulation of high levels of arsenic has been reported in marine fish from the Gulf and other regions such as the Northern Adriatic Sea and Izmir Bay (Table 2). The fairly high salinity of the western Gulf (42–60 psu) in combination with low phosphate and nitrate levels in seawater (Hashimoto et al., 1998) might at least in part explain the presence of high arsenobetaine and total As concentrations in bivalves, cuttlefish, shrimp and finfish from the study area, since no contamination of the area with arsenic has been reported.

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Table 3
The tissue concentrations (mg/kg dry wt) of total As and As species in cuttlefish, shrimp and finfish samples collected from different offshore sites. The percentage of As species to the total As values are given in parentheses next to the mean value. The minimum and maximum values are given below the mean value within parentheses. (N = number of composite sample).

Site name	Species	N	Fish length (mm)	Salinity (psu)	Tot As	AB	**As**-Gly	Tetra
Off Kafji	*Arius thalassinus*	3	322	43	65 (60–71)	47 (74%) (46–49)	<dl	<dl
Off Safaniya	*Arius thalassinus*	3	414	43	76 (69–80)	62 (82%) (58–66)	<dl	<dl
Off Manifa	*Sepia pharaonis* (cuttlefish)	3	200	45	117 (93–134)	101 (86%) (80–114)	<dl	<dl
	Parupeneus margaritatus	3	181	43	57 (48–66)	45 (80%) (39–53)	<dl	<dl
	Acanthopagrus bifasciatus	2	240	43	56 (20–92)	44 (79%) (14–74)	0.55 (0.98%) (0.49–0.61)	dl
	Rhabdosargus haffara	2	134	45	23 (22–24)	18 (76%) (16–20)	<dl	<dl
	Penaeus semisulcatus (shrimp)	1	143	45	34	27 (80%)	<dl	0.65 (1.9%)
Off Ras Al Khair	*Sepia pharaonis* (cuttlefish)	3	179	46	82 (74–98)	67 (82%) (61–77)	<dl	<dl
	Arius thalassinus	1	415	46	72	63 (88%)	<dl	<dl
Off Khursaniyah	*Sepia pharaonis* (cuttlefish)	2	230	44	93 (85–100)	78 (84%) (72–84)	<dl	<dl
Off Jubail	*Carangoides fulvogutatus*	3	699	44	56 (54–57)	47 (85%) (45–51)	<dl	<dl
	Nemipterus japonicus	4	260	43	36 (32–41)	29 (81%) (26–32)	0.13 (0.37%) (<dl-0.27)	<dl
Off Ras Tanura	*Acanthopagrus bifasciatus*	1	227	42	52	41 (80%)	0.7 (1.4%)	<dl
	Argyrops spinifer	1	275	42	11	8.2 (75%)	<dl	<dl

dl = detection limit (0.01 mg As/kg dry wt).
Note: Traces of TMAO and AC were detected in some of the samples, but were not present at quantifiable levels.
Species are finfish unless otherwise stated.

Our arsenic speciation data for cuttlefish, shrimp and finfish (Table 3), based on measurements made by using HPLC/ICPMS, show that arsenobetaine is the major arsenic species in the Gulf, a result consistent with the general pattern of arsenic species in seafood worldwide (Francesconi, 2010). The levels of AB, as expected, correlated strongly with total As levels in cuttlefish ([AB] = 0.89[As] − 5.1 mg/kg, R^2 = 0.98) and finfish ([AB] = 0.81[As] − 0.68 mg/kg, R^2 = 0.97) from the study area. Concentrations of other arsenic species in the samples were very low or below the detection limit (Table 3).

3.3. Inorganic arsenic in bivalves, cuttlefish, shrimp and finfish

The mean, minimum and maximum tissue concentrations (mg/kg dry wt) of total As and iAs in clams (nine sites), oysters (three sites), cuttlefish (three sites), shrimp (one site) and finfish (seven sites) from our study are summarized in Table 4. The observed iAs contents in the bivalves from the Gulf were very low and comparable to the values reported in bivalves from other parts of the world. For example, low iAs values were reported in clams (0.07–0.24 mg/kg wet wt) from American Samoa (Peshut et al., 2008), in oysters (0.08–0.29 mg/kg dry wt) from Taiwan (Liu et al., 2006), in clams (0.01–0.34 mg/kg dry wt) from the north east coast of India (Fattorini et al., 2013), in mussels (0.03–0.04 mg/kg wet wt) from Izmir Bay, Turkey (Kucuksezgin et al., 2014) and in clams (0.36 mg/kg dry wt) and mussels (0.31–0.51 mg/kg dry wt) from different contaminated sites in Europe (Maulvault et al., 2015). However, high levels of inorganic arsenic (up to 13.8 mg/kg ww) were reported in blue mussels from Norwegian Fiords (Sloth and Julshamn, 2008). Concentrations of iAs were considerably higher in clams (0.47 ± 0.2 mg/kg) compared to oysters (0.13 ± 0.05 mg/kg) from the study area. Similar to the total As data, there was no clear trend in iAs data with reference to geographical location or salinity of the waters from which they were collected.

The iAs formed around 0.6–1.7% (mean 0.82%) of total As in clams and around 0.2–0.28% (mean 0.25%) in oysters (Table 4). These values are low compared to the % iAs values reported in clams (86–98%)

Table 4
The tissue concentrations (mg/kg dry wt) of total As and inorganic arsenic (iAs) and the percentage of iAs species to the total As values in bivalves, cuttlefish, shrimp and finfish from the Arabian Gulf. The minimum and maximum values are given together with the mean value within parentheses. (N = number of composite sample).

Species	St name	N	Total As	iAs	% iAs
Meretrix meretrix	Manifa	2	43 (40–46)	0.58(0.51–0.65)	1.4 (1.3–1.4)
	Ras Al-Khair	2	38 (36–41)	0.41 (0.40–0.41)	1.1 (1–1.13)
	Abu Ali North	2	100 (99–101)	0.83 (0.76–0.89)	0.83 (0.75–0.9)
	Abu Ali South	2	94 (93–96)	0.58 (0.56–0.60)	0.62 (0.58–0.65)
	Ras Tanura	2	48 (47.2–48)	0.35 (0.32–0.37)	0.72 (0.68–0.77)
	Tarut Island	2	39 (36–41)	0.35 (0.32–0.37)	0.89 (0.88–0.9)
	Tarut Bay	2	17 (15–18)	0.28 (0.26–0.29)	1.7 (1.5–1.8)
	Aziziyah	3	24 (23–25)	0.19 (0.19–0.19)	0.80 (0.75–0.84)
	Salwa	4	97 (75–118)	0.63 (0.53–0.73)	0.67 (0.54–0.94)
Pinctada radiata	Manifa	2	38 (37–38)	0.11 (0.09–0.12)	0.28 (0.24–0.33)
	Abu Ali North	2	49 (49–50)	0.10 (0.10–0.11)	0.21 (0.2–0.22)
	Salwa	1	87	0.22	0.25
Arius thalassinus	Off Kafji	3	65 (60–71)	<dl	<dl
Arius thalassinus	Off Safaniya	3	76 (69–80)	0.01 (<dl-0.02)	0.01
Sepia pharaonis	Off Manifa	3	117 (93–134)	0.01 (dl-0.02)	0.01
Parupeneus margaritatus		3	57 (48–67)	0.01 (0.01–0.02)	0.02
Acanthopagrus bifasciatus		2	56 (20–92)	0.01 (0.01–0.01)	0.02
Rhabdosargus haffara		2	23 (22–24)	0.02 (0.02–0.03)	0.08
Penaeus semisulcatus		1	34	0.07	0.21
Sepia pharaonis	Off Ras Al Khair	3	82 (74–98)	0.03 (0.027–0.03)	0.03
Arius thalassinus		1	72	<dl	<dl
Sepia pharaonis	Off Khursaniyah	2	93 (85–100)	<dl	<dl
Carangoides fulvogutatus	Off Jubail	3	56 (54–57)	<dl	<dl
Nemipterus japonicus		4	36 (32–41)	0.01 (<dl-0.01)	0.01
Acanthopagrus bifasciatus	Off Ras Tanura	1	52	<dl	<dl
Argyrops spinifer		1	11	<dl	<dl

dl = detection limit (0.01 mg As/kg-dry wt).

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from a contaminated marine environment in Seal Harbor, Canada (Koch et al., 2007) in blue mussels (0.10–42%) from Norwegian Fiords (Sloth and Julshamn, 2008) and in bivalves (2–6%) from contaminated sites in Europe (Maulvault et al., 2015).

Inorganic As was detected in only 67% and 50% of the cuttlefish and finfish samples, respectively, analyzed from the study area. The iAs formed about 0.01–0.04% (mean 0.01%) of total As in cuttlefish and around 0.01–0.06% (mean 0.01%) in finfish (Table 4). After compiling published data from 20 studies, Schoof and Yager (2007) reported mean iAs concentrations ranging 0.01 to 0.02 mg/kg wet wt in finfishes and 0.04 to 0.05 mg/kg wet wt in shellfishes. Our results indicate that there is no workable relationship between total As and iAs content in the Gulf samples. Hence the method of deriving iAs values from total As data using a conversion factor approach would be misleading for the seafood samples from the study area.

3.4. Health risk assessment

3.4.1. Estimation of inorganic arsenic exposure

Given variation in measurable iAs across seafood type, considerable differences were observed in estimated LADDs, with highest exposures seen among bivalve and shrimp consumers for all populations (Table 5). Across populations, given differences in body weight and rate of seafood consumption, the LADDs were approximately twice as high for expatriates as for native Saudis. High-end consumers taken from the combined native Saudi and expatriate populations had LADDs more than four times higher than the native Saudi subpopulation. Exposures modeled using high percentile iAs concentrations, however, are intended to represent an upper bound on possible exposures; in light of the number of samples assessed and limited variability observed among sample iAs concentrations, these estimates carry considerable uncertainty.

3.4.2. Cancer risk estimates and hazard quotients

Patterns of estimated bladder and lung cancer risk mirrored those of LADDs. Risks were notably higher among bivalve consumers (9.5×10^{-5} to 1.5×10^{-2}) as compared to finfish (2.6×10^{-6} to 1.1×10^{-3}), cuttlefish (4.6×10^{-6} to 5.1×10^{-4}) and shrimp (1.8×10^{-5} to 1.3×10^{-3}) consumers (Table 5). All risk estimates relying upon EPA's proposed 2010 CSF were higher than 1×10^{-5}, with the majority exceeding 1×10^{-4}, a value used as the acceptable risk ceiling often used by the EPA Superfund program (USEPA, 2015a). Given the uncertainty surrounding the use of EPA's proposed 2010 CSF, however, we advise that these estimates be viewed with caution.

Nearly all HQs were below the value of 1, except for the high-end consumers of bivalves (95th percentile of iAs concentrations), indicating that hyperpigmentation, keratosis, and possible vascular complications are unlikely to be observed among seafood consumers in Saudi Arabia. Among the expatriates and the combined population with high rates of consuming bivalves at the 95th percentile of iAs concentrations, HQs were estimated at 1.20 and 1.99, respectively. It should be noted, however, that the RfD does not consider more recent epidemiologic literature regarding iAs exposure, and thus the possibility of non-cancer health outcomes resulting from these exposures cannot be dismissed.

Our assessments of risks and hazards must be interpreted with caution, given limitations in available knowledge. While our study is the first to present arsenic species data for selected seafood from the Gulf, for some types (especially shrimp and cuttlefish), we estimated parameters for use in the risk analyses from a limited number of samples. More confidence in risk estimation would be afforded by analysis of a larger number of samples. Also, since population seafood intake rates were not available for bivalves, cuttlefish and shrimp, we relied upon information about finfish consumption to inform our exposure and risk estimations.

The EPA IRIS quantitative toxicity metrics for inorganic arsenic are in the process of being revised, and in the interim, outdated values appear in their database. In our analysis, we estimated risk using both the older CSF value in the database (USEPA, 2015b) and a newer, proposed CSF (USEPA, 2010), which differ by a factor of 17. Given the ongoing discussion over the CSF and the wide range between the two values, we advise caution in interpretation of the estimated risk, especially for the higher-end estimates, which carry more uncertainty.

In addition, we relied exclusively on the older RfD (USEPA, 2015b) to examine non-cancer hazard, since a newer proposal for the RfD has yet to be released. Consequently, we have some confidence that our range of estimates of risk captures the true risks experienced by exposed

Table 5
The estimated lifetime average daily doses (LADD), cancer risks and hazard quotients (HQ) resulting from exposure to iAs among consumers of bivalves, cuttlefish, finfish, and shrimp.

		iAs	Native Saudi				Expatriate			
Seafood	Values	(mg/kg wet weight)[a]	LADD (mg/kg d^{-1})	Risk (CSF = 1.5 per mg/kg d^{-1})	Risk (CSF = 25.7 per mg/kg d^{-1})	HQ	LADD (mg/kg d^{-1})	Risk (CSF = 1.5 per mg/kg d^{-1})	Risk (CSF = 25.7 per mg/kg d^{-1})	HQ
Bivalves	median	7.40×10^{-2}	6.35×10^{-5}	9.53×10^{-5}	1.63×10^{-3}	0.21	1.58×10^{-4}	2.37×10^{-4}	4.06×10^{-3}	0.53
	95th %ile	1.69×10^{-1}	1.45×10^{-4}	2.17×10^{-4}	3.73×10^{-3}	0.48	3.61×10^{-4}	5.41×10^{-4}	9.27×10^{-3}	1.20
Cuttlefish	mean	3.55×10^{-3}	3.05×10^{-6}	4.57×10^{-6}	7.83×10^{-5}	0.01	7.58×10^{-6}	1.14×10^{-5}	1.95×10^{-4}	0.03
	maximum	5.60×10^{-3}	4.81×10^{-6}	7.21×10^{-6}	1.24×10^{-4}	0.02	1.20×10^{-5}	1.79×10^{-5}	3.07×10^{-4}	0.04
Finfish	median	2.00×10^{-3}	1.72×10^{-6}	2.57×10^{-6}	4.41×10^{-5}	0.01	4.27×10^{-6}	6.41×10^{-6}	1.10×10^{-4}	0.01
	95th %ile	1.18×10^{-2}	1.01×10^{-5}	1.51×10^{-5}	2.59×10^{-4}	0.03	2.51×10^{-5}	3.76×10^{-5}	6.45×10^{-4}	0.08
Shrimp	single	1.40×10^{-2}	1.20×10^{-5}	1.80×10^{-5}	3.09×10^{-4}	0.04	2.99×10^{-5}	4.48×10^{-5}	7.68×10^{-4}	0.10

		iAs	Combined (mean consumption)				Combined (90th %ile consumption)			
Seafood	Values	(mg/kg wet weight)[a]	LADD (mg/kg d^{-1})	Risk (CSF = 1.5 per mg/kg d^{-1})	Risk (CSF = 25.7 per mg/kg d^{-1})	HQ	LADD (mg/kg d^{-1})	Risk (CSF = 1.5 per mg/kg d^{-1})	Risk (CSF = 25.7 per mg/kg d^{-1})	HQ
Bivalves	median	7.40×10^{-2}	9.24×10^{-5}	1.39×10^{-4}	2.37×10^{-3}	0.31	2.61×10^{-4}	3.92×10^{-4}	6.72×10^{-3}	0.87
	95th %ile	1.69×10^{-1}	2.11×10^{-4}	3.16×10^{-4}	5.42×10^{-3}	0.70	5.97×10^{-4}	8.95×10^{-4}	1.53×10^{-2}	1.99
Cuttlefish	mean	3.55×10^{-3}	4.43×10^{-6}	6.65×10^{-6}	1.14×10^{-4}	0.01	1.25×10^{-5}	1.88×10^{-5}	3.22×10^{-4}	0.04
	maximum	5.60×10^{-3}	6.99×10^{-6}	1.05×10^{-5}	1.80×10^{-4}	0.02	1.98×10^{-5}	2.97×10^{-5}	5.09×10^{-4}	0.07
Finfish	median	2.00×10^{-3}	2.50×10^{-6}	3.75×10^{-6}	6.42×10^{-5}	0.01	7.07×10^{-6}	1.06×10^{-5}	1.82×10^{-4}	0.02
	95th %ile	1.18×10^{-2}	1.47×10^{-5}	2.20×10^{-5}	3.77×10^{-4}	0.05	4.15×10^{-5}	6.23×10^{-5}	1.07×10^{-3}	0.14
Shrimp	single	1.40×10^{-2}	1.75×10^{-5}	2.62×10^{-5}	4.49×10^{-4}	0.06	4.95×10^{-5}	7.42×10^{-5}	1.27×10^{-3}	0.16

iAs = inorganic arsenic; LADD = lifetime average daily dose; CSF = cancer slope; HQ = hazard quotient.
[a] Wet weight concentrations were estimated by multiplying dry weight concentrations by 0.2, since the moisture content in the samples was approximately 80%.

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persons; conversely, we have limited confidence in our evaluation of non-cancer hazard, but present HQ estimates for purposes of comparisons with other exposures.

4. Conclusion

Seafood species, including molluscs, crustaceans and finfish, collected from the Arabian Gulf contain high levels of arsenic, mostly present as non-toxic organoarsenic species such as arsenobetaine (around 71%). The toxic iAs arsenic form was present in the seafood species at only low levels (<dl – 1.84% of total As). No significant relationship was observed between the tissue concentrations of total As and iAs in the samples. Hence, the method of deriving iAs data from total As data using a conversion factor approach cannot be used for Gulf seafood samples, and is probably also inappropriate for marine samples from other areas as well. Our analyses of cancer risks and non-cancer hazards identified non-negligible risks and the potential for hazards; especially for consumers of bivalves sourced from the region; despite this, many uncertainties remain that would be best addressed by further analysis of seafood samples, better information about intake of various non-finfish seafood species among the Saudi population, and updated quantitative toxicology metrics that incorporate the knowledge gained from recent epidemiologic work regarding iAs exposure. With these in place, decisions about interventions aimed at exposure mitigation can be considered.

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