HUMAN HEALTH RISK OF HEAVY METALS THROUGH FISH AND SHELLFISH INGESTION: A CASE STUDY FOR THE ARABIAN GULF

CHOWDHURY S.

Department of Civil and Environmental Engineering, King Fahd University of Petroleum and Minerals (KFUPM), Dhahran 31261, Saudi Arabia.
E-mail: SChowdhury@kfupm.edu.sa

ABSTRACT

Human exposure to heavy metals through ingestion of fish and shellfish from the Arabian Gulf was predicted. Risks associated with such exposures were assessed. Concentrations of ten heavy metals: cadmium (Cd), arsenic (As), chromium (Cr), copper (Cu), mercury (Hg), manganese (Mn), nickel (Ni), lead (Pb), vanadium (V) and zinc (Zn) were characterized for 35 marine fish and shellfish species from the Arabian Gulf region for the period of 1988-2010. Ingestion patterns of seafood were developed. Cancer risks were estimated from arsenic (As) exposure through the oral route, while other metals were not reported to be human carcinogen through oral route. Hazard indices from these metals through fish ingestion were predicted. Chronic daily intakes (CDI) of these metals were estimated to be in the range of 1.55×10^{-06} – 5.72×10^{-04} mg/kg/day. This study estimated cancer risks in the range of 6.7×10^{-11} – 1.1×10^{-04} with an average of 2.1×10^{-06}. Cumulative hazard index was estimated to be 0.159 (range: 0.01–1.59). Approximately 0.18% of the exposed populations had higher hazard index than the U.S. Environmental Protection Agency regulatory limit of unity. The probability of one cancer incident in 1 million populations from lifetime exposure to arsenic through fish ingestion was predicted to be 0.49. Presence of other metals and inorganic/organic chemicals and complexes in fish and shellfish may impose additional risks to human. Through comprehensive understanding of cumulative exposures and risks from all contaminants, appropriate measures can be adopted to better protect human health in the Arabian Gulf region.

Keywords: Risk assessment, fish ingestion, heavy metals, uncertainty, hazard index

1. Introduction

Since 1967, oil spill events ranging up to 1.0×10^{6} tons of crude oil discharge in a single occurrence have been reported in the Arabian Gulf [1]. Approximately 12000 oil tankers move in the Gulf water every year, which increases the possibility of crude oil contamination through operation, and loading and unloading of crude oil and ballast water [2,3]. Concentrations of heavy metals in crude oil can be significant [3] while these metals have much longer half-lives and many can have sub-chronic and chronic effects to human through fish and shellfish ingestion [4,5]. Presence of heavy metals including barium (Ba), cadmium (Cd), calcium (Ca), cobalt (Co), copper (Cu), chromium (Cr), iron (Fe), nickel (Ni), lead (Pb), arsenic (As), mercury (Hg), vanadium (V), beryllium (Be) and zinc (Zn) have been reported in crude oil [3]. Crude oil from Venezuela, Arabian Gulf, Iran and Pakistan have reported variable concentrations of As, Cd, Cr, Cu, Co, Fe, Pb, Ni, V and Zn [6-8], which might have contaminated the Gulf water and subsequently, the marine fish and shellfish species. Ingestion of these seafood can pose elevated cancer and non-cancer risks to human [4,9]. Meinhold et al. [10] estimated potential cancer risk from two isotopes of radium (^{226}Ra and ^{228}Ra) in produced water for the open bays in Louisiana. A robust modeling system was developed using fuzzy rule-based approach to control human exposure to ^{226}Ra and ^{228}Ra in produced water in the East coast of Canada [4].

Despite the metals in crude oil and/or produced water can have potential toxic effects to human health due to chronic exposure through food chain [5,11], limited studies have reported human exposure and risk from metal contaminated fish ingestion in the Arabian Gulf region. Few metals, such as, As, Cd, Cr, Pb, Ni and Hg can have potential toxic and/or carcinogenic effects to human health [5]. In this study, concentrations of ten heavy metals (Cd, As, Cr, Cu, Hg, Mn, Ni, Pb, V and Zn) in 35 marine fish and shellfish species from the Arabian Gulf region were characterized for the period of 1988-2010. The metals were represented using appropriate statistical distributions. Human ingestion of seafood was assessed using the edible part concept developed in an earlier study of Chowdhury et al. [4]. Cancer and non-cancer risks to human were predicted using the slope factor (SF) and reference doses (RfD) of the metals from the U.S. Environmental Protection Agency [5]. Finally, importance of comprehensive understanding of risk from contaminated fish ingestion in the Arabian Gulf was outlined.

2. Methodology

Concentrations of Cd, As, Cr, Cu, Hg, Mn, Ni, Pb, V and Zn in the fish tissue from the Arabian Gulf were obtained from literature [12-16]. The data were characterized using statistical distributions to incorporate uncertainty in exposure assessment (Table 1). Human exposure to metal through fish ingestion was assessed following Chowdhury et al. [4] as:

$$CDI_m = x \times \frac{C_m \times FIR \times EF \times ED \times FR \times 10^{-3}}{BW \times AT} \qquad (1)$$

Where, CDI_m= chronic daily intake (mg/kg/day); C_m = concentration of metal (μg/g); FIR = fish ingestion rate (g/day); EF = exposure frequency (days/yr); ED = exposure duration (yrs); FR = fraction of contaminated fish ingested; 10^{-3} = conversion factor for μg to mg; BW = body weight (kg); AT = averaging time (days); x = fraction of edible parts in a fish. Chowdhury et al. [4] used a lognormal (3.455, 0.622) distribution for FIR in the Eastern coast of Canada. In the Arabian Gulf region, limited information is available on FIR. The Middle East and North Africa (MENA) region produced 3574,400 tons of fish in 2000-2002 [17] while the populations in the MENA region were 386 million. During this period, North America produced 6700,200 tons of fish while the populations were 320 million [17]. The per capita fish production in the Arabian Gulf region is 46% to that of North America. To approximate fish ingestion for the Arabian Gulf populations, the estimate of Chowdhury et al. [4] was multiplied by 46%. Upon availability of regional data, FIR can be updated in future. Details of the parameters are shown in Table 2.

In context to contaminated fish ingestion, Chowdhury et al. [4] used the value of FR as 50% based on the findings from several survey works in the U.S., which may not be applicable for the Arabian Gulf region, due to the fact that most of the fish in this region is produced from the seawater (e.g. Gulf water) [17]. In this study, contaminated fish ingestion was assumed to be 80% with the range of 70-90% of the total fish ingestion. Chowdhury et al. [4] reported the edible parts of a fish (x) to be 78% (range: 64-87%), which followed lognormal (4.36, 0.063) distribution. Further details on these parameters can be obtained from literature [4,18]. The human cancer risk can be estimated as: $CR_i = CDI_{mi} \times SF_i$, where, CR_i= cancer risk from the ith metal; CDI_{mi}= chronic daily intake of ith metal (mg/kg/day); SF_i = slope factor of ith metal (per mg/kg/day). The predicted cancer risks were adjusted using the early-life exposure adjustment factors [19] as: (i). for exposures from the first day of birth up until a child's second birthday, a 10-fold adjustment; (ii). for exposures from a child's second birthday up until their sixteenth birthday, a 3-fold adjustment; and (iii). for exposures after turning 16 years of age, no adjustment. The hazard indices were estimated following USEPA [18] as: $HI_i = CDI_{mi} / R_fD_i$, where, HI_i = hazard index from the ith metal; R_fD_i = reference dose for the ith metal (mg/kg/day).

Among the 10 metals (Table 1), inorganic As is a human carcinogen through ingestion route [5] while the other metals can pose non-cancer risks through ingestion route. However, few metals such as, Cd, Ni and Cr may pose cancer risk through inhalation route, which it is not applicable in this study [5]. In fish, As is generally found in forms of complexes, which may not pose the similar risk to that of inorganic As [1,5]. Schoof and Yager [20] documented that the carcinogenic form of As (e.g., inorganic As) in fish might be as little as 1% of total As in a fish. Concentrations of As in fish (Table 1) were multiplied by 1% for exposure and risk assessment.

Table 1: Metals in fish tissues (μg of metals/g of fish tissue)

Metal	Distribution	Average	Std. dev.	Range	Slope factor	Reference dose
Cd	LN(-4.247, 1.538)[a]	0.05	0.1	0.001 - 0.7		0.001
As*	W(0.904, 1.676)[b]	1.76	1.98	0.04 - 9.6	1.5	0.0003
Cr	LN(0.1103, 0.879)[a]	1.54	1.25	0.2 - 4.8		0.003
Cu	LN(1.909, 0.3843)[a]	6.60	3.61	0 - 16.4		0.005
Hg	W(0.986, 0.09889)[b]	0.10	0.10	0 - 0.48		0.0003
Mn	T(1.0, 4.54, 8.6)[c]	4.54	2.31	1.0 - 8.6		0.14
Ni	T(0.3,6.8,16.7)[c]	9.4	6.73	0.3 - 16.7		0.02
Pb	LN(-2.107, 1.569)[a]	0.42	0.89	0 - 4.31		
V	LN(0.753, 0.539)[a]	2.46	1.38	0.9 - 5.3		0.007
Zn	LN(2.716, 0.614)[a]	18.14	11.45	5.9 - 44.2		0.01

*Multiply by 1% to obtain carcinogenic form of As (Schoof and Yager, 2007); [a]the 1st and 2nd parameters represent location and scale of Lognormal distributions; [b]the 1st and 2nd parameters represent shape and scale of Weibull distribution; [c]the 1st, 2nd and 3rd numbers are the lowest, mean and highest values for the triangular distributions; Slope factor (per mg/kg/day; Reference dose (mg/kg/day)

Table 2: Parameters for exposure assessment (Chowdhury et al. [4])

Model parameters	Characterization
FIR (Fish ingestion rate g/day)	0.46 × LN (3.46, 0.62)
x (Edible part of fish %)	LN (4.36,0.063)
FR (Contaminated fish ingested %)	T(70,80,90)
EF (Exposure frequency days/yr)	T(330, 345, 360)
ED (Exposure duration, yr)	T(20, 30, 40)
BW (Body weight kg)	T(60,70,80)
AT (Averaging time, days)	T(21989, 25533, 29144)
C_m (Metal concentration in fish, (μg/g)	From Table 1

3. Results

Table 1 shows the metal concentrations in 35 marine fish and shellfish species. Concentrations of Cd, As, Cr, Cu, Hg, Mn, Ni, Pb, V and Zn were 0.001-0.7, 0.04-9.6, 0.2-4.8, 0-16.4, 0-0.48, 1-8.6, 0.3-16.7, 0-4.31, 0.9-5.3 and 5.9-44.2 μg/g of fish tissue (wet weight) respectively. These data were fit into the appropriate statistical distributions (Table 1). Using the distributions (Table 1), 5000 random data were generated for each metal, which were incorporated into exposure and risk assessments. The chronic daily intake of metals are shown in Table 3. Average CDI for Cd, As, Cr, Cu, Hg, Mn, Ni, Pb, V and Zn were 1.55×10^{-06}, 1.38×10^{-06}, 5.04×10^{-05}, 2.24×10^{-04}, 3.02×10^{-06}, 1.47×10^{-04}, 2.43×10^{-04}, 1.21×10^{-05}, 7.69×10^{-05} and 5.72×10^{-04} mg/kg/day respectively (Table 3). Human cancer risk from As was predicted to be in the range of 6.7×10^{-11} – 1.1×10^{-04} with an average of 2.1×10^{-06}. The exceedance probabilities of cancer risk are plotted in Figure 1.

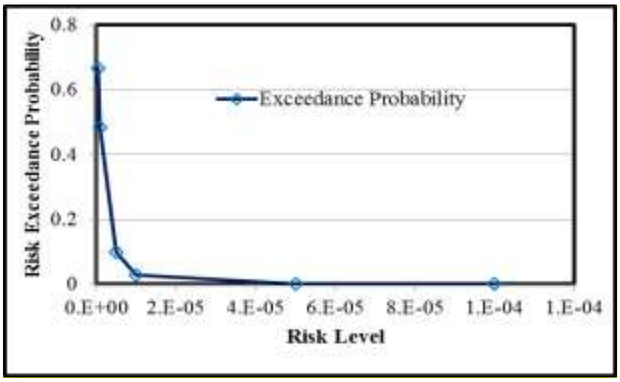

Figure 1: Cancer risk exceedance probability

Table 3. Chronic daily intake (CDI) of metals (mg/kg/day)

Metal	Average	Min	Max	Std. Dev
Cd	1.55E-06	1.40E-09	4.37E-04	7.82E-06
As	1.38E-06	4.49E-11	7.01E-05	2.37E-06
Cr	5.04E-05	9.29E-07	1.90E-03	7.59E-05
Cu	2.24E-04	1.10E-05	2.41E-03	1.96E-04
Hg	3.02E-06	1.87E-10	8.41E-05	4.19E-06
Mn	1.47E-04	6.51E-06	1.52E-03	1.23E-04
Ni	2.43E-04	4.57E-06	2.31E-03	2.18E-04
Pb	1.21E-05	6.88E-09	8.86E-04	3.54E-05
V	7.69E-05	2.06E-06	1.81E-03	8.40E-05
Zn	5.72E-04	1.04E-05	1.04E-02	6.33E-04

At risks of 1.0×10^{-06}, 5.0×10^{-06} and 1.0×10^{-05}, cancer risk exceedance probabilities are 0.49, 0.10 and 0.028, indicating the corresponding probabilities of 1, 5 and 10 cancer incidents, respectively, in 1 million populations (Figure 1). The cumulative hazard index (HI) from these metals was estimated to be 0.159 (range: 0.01 – 1.59). Among the 5000 simulated cases, nine (9) cases were observed to have HI greater than unity, meaning 0.18% cases with possible hazard [18]. For the individual metals, hazard indices of Cd, As, Cr, Cu, Hg, Mn, Ni, V and Zn have been estimated to be 0.0015 (1.39×10^{-06}–0.437), 0.0046 (1.5×10^{-07}–0.233), 0.017 (0.00031–0.633), 0.045 (0.002–0.48), 0.01 (6.23×10^{-07}–0.28), 0.001 (4.65×10^{-05}–0.011), 0.012 (0002–0.116), 0.011 (0.0003–0.259) and 0.057 (0.001–1.04) respectively. The HI for Pb was not estimated as the reference dose of Pb is not available in IRIS [5].

It is to be noted that average concentrations of As is 1.76 μg/g (range: 0.044–9.6 μg/g), which is relatively high while inorganic As poses cancer risk. Based on literature from different regions, inorganic As was assumed to be 1% of the total As in a fish. Few As complexes are highly absorbed through human gastrointestinal (GI) system [1], which can make more As to be biologically available and thus pose higher cancer risks. Understanding of As complexes in fish and their biologically available fractions are necessary to better explain cancer risks. Better understanding of other metals, radioactive materials and contaminants can also improve the exposure and risk assessment.

4. Conclusions

This study demonstrates that specific metal individually and in combination can pose elevated risks to human through contaminated fish and shellfish ingestion in the Arabian Gulf countries. Although the cancer risks and hazard indices are comparable to the risks in some other coastal regions (e.g., Gulf of Mexico, Eastern Coast of Canada, etc.), higher levels of arsenic, mercury, cadmium, lead and nickel can be a concern. In particular, higher levels of arsenic and its complexes in fish and shellfish need to be comprehensively investigated and the carcinogenic form of arsenic must be determined. Many other contaminants, including BTEX (Benzene, Toluene, Ethyl Benzene and Xylenes), PAHs (polycyclic aromatic hydrocarbons), NPD (Naphthalene, Phenanthrene and Dibenzothiophene including alkyl homologues) and NORM (Naturally Occurring Radioactive Materials) were reported in marine fish and shellfish species. Future study should be directed toward better understanding of the concentrations, forms and variability of these contaminants and the intake patterns of seafood. Despite few limitations, this study sheds light on the importance of comprehensive study for the oil-rich Arabian Gulf region.

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