Chapter

Heavy Metal Contamination in a Protected Natural Area from Southeastern Mexico: Analysis of Risks to Human Health

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Abstract

In this chapter, a little of the history of Carmen City, Mexico is addressed; this island is immersed in a Protected Natural Area and in the "Campeche Sound" an oil extraction site. Fishing natural resources were for many years the pillar of the development of the area; the most commercially important species are still shrimp, oysters and scales. Nowadays, although the volumes of capture have decreased considerably, different species of high commercial value are still extracted.
The considerable development of the oil industry has brought with its economic development and a better quality of life for its inhabitants; however, the ravages of pollution, rapid population growth, and deforestation have been the unwanted factor. This chapter addresses the effects of heavy metals on human health through a risk analysis, based on the criteria of the US Environmental Protection Agency (USEPA) that was carried out for different commercial species based on carcinogenic factors and not carcinogenic; the results show that the risk from consumption of these species is "potentially dangerous" for human health, especially in those species that, due to their eating habits (mollusks, bivalves, clams) tend to bioaccumulate heavy metals, such as cadmium, which it has been considered by the International Agency for Research on Cancer (IARC) as a risk factor; for this reason, the importance of periodically evaluating and monitoring oyster extraction banks, clams and, in general, all fishery products. Mexican legislation and various international legislations dictate the maximum permissible and tolerable levels of heavy metals in fishery products; the organisms considered in this study exceeded the permissible limits in copper and nickel, which represents a risk for human consumption.

Keywords: heavy metals, pollution, marine organisms

1. Introduction

Mexico is a mega-diverse country with 90,839,521 hectares of protected natural areas, of which Terminos Lagoon, classified as "Flora and Fauna Protection Area" has 705, 016 hectares that make it one of the largest areas in the country. Within its status as a Protected Natural Area, there are fishing activities and oil and gas extraction-conduction areas. Until a decade ago, the Campeche Sound contributed nearly 95% of the crude oil and 80% of the national natural gas; today, due to recent changes in the use of fossil energy, production has decreased, although it remains one of the most important companies in Mexico.

Campeche Sound in general, and Carmen Island in particular, have been zones of abrupt changes, beginning with the exploitation of shrimp, which in the years of 1969 to 1979, promoted the economic development of the area. Since 1976, a historical production of crude oil began for Mexico, bringing with it important changes in the population, social changes and therefore, environmental changes.

According to Cuellar et al. [1] in 1979 the company "Mexican Petroleum" (PEMEX) had a large number of facilities for the extraction and processing of crude oil and natural gas on the southwestern coasts of the Gulf of Mexico, as well as a total of 200 facilities for different purposes and 185 production platforms. These changes affected the fishing production and the lives of the inhabitants as they went from being a "fishing village" to industrial zones with an increase in the population and the services that were demanded. By 1970, there were more than 800 vessels with capacities ranging from three to fifty tons to process shrimp and more than twenty freezers and packers of the fishing product in the area, as well as four shipyards for the shrimp fleet; at present, all this activity has been in considerable decline, almost disappearing [2].

When the oil boom began, the first oil spills put fishing activity at risk and there have been very few studies in the area to determine the degree of impact of the oil industry on the deterioration of the environment; certain species such as white shrimp (*Litopenaeus setiferus*) are permanently banned to avoid completely depleting the resource; however, recent data and with the current crisis of the SARS-COV2 pandemic indicate that poaching activities have increased in the area, even with the capture of protected species; [https://www.novedadescampeche.com.mx/estado/campeche/urgente-estrategia-federal-del-control-de-la-pesca-ilegal].

The main fishery resources in this area are shrimp, oyster and scale. The oyster harvest currently has the certification of the Commission for the Protection against Sanitary Risks of Campeche (COPRISCAM, by its acronym in Spanish) in the Atasta lagoon; however, its production has been diminished due to the fishing and poaching of this resource. On the other hand, the clam was the main fishing resource in the Pom lagoon for more than four decades. Currently the catch levels show a notable decrease, which has been attributed to excessive overexploitation; some studies attribute it to pollution and deforestation in the mangrove area. According to Ramos and Villalobos [3], the mangrove ecosystems of the Terminos Lagoon Flora and Fauna Protection Area have registered in recent years, a rapid transformation towards ecosystems with low productivity and biodiversity. The causes of this rapid loss are deforestation, urbanization, industrialization, agricultural, fishing and aquaculture activities; and the alteration of the hydrological regime of the Grijalva-Usumacinta river basin.

The shrimp fishery does not show a better picture. This resource, which was exploited for many years, is now only one fifth of what was obtained in the 1980s. Historical data show that in 1972 the yield of pink shrimp (*Farfantepenaeus duorarum)* was 11,904 tons and in 2000 it was only 1,409 tons [4]. With regard to the seven-bearded shrimp (*Xiphopenaeus kroyeri)* from 1993 due to its overexploitation in the coastal marine strip and with the entry into force of Mexican standards NOM-004-PESC-1993 and NOM-002-PESC-1993 (Diario Oficial de la Federación 1994., Plan de Manejo Pesquero de camarón siete barbas *Xiphopenaeus kroyeri* en las costas de los estados de Campeche y Tabasco) its fishing has been regulated by fishing bans seasons.

Overall, the development of the oil industry, urbanization and overexploitation of marine species have had a strong environmental impact, as well as in the displacement of deep-sea fishing areas. However, very few studies have been conducted in the area that show the overall impact generated on the flora and fauna of this region. Studies have been reported on the impacts on benthic communities and their relation to the presence of hydrocarbons [5]; the studies show the presence and concentration of hydrocarbons in sediments and organisms [5–9]. There are numerous factors to be considered in the deterioration of an ecosystem, among them the great quantity of organic and inorganic substances that are generated not only by oil activity, but also by the entire related industry. In the years 2000–2001 alone, a total of 104,901 tons of sulfur oxides (SOx) and 1,747 tons of nitrogen oxides (NOx) were emitted into the atmosphere [1, 10]. There are currently no recent studies to compare these levels.

Among the inorganic contaminants that cause interest due to the adverse effects they can cause to living beings, heavy metals stand out, some of which have been cataloged as serious threats to human health because of their carcinogenic risk. Regarding the studies carried out to determine the degree of impact on the Campeche Sound, we can cite Vázquez et al. [11] who carried out oceanographic campaigns and comparative studies on the levels of Cd, Cr, Ni and V in marine sediments. In their study, they highlight that oil activity, fishing and marine traffic in the area substantially modify the levels of heavy metals; they also agree that the levels of organic matter have a direct influence on the distribution of metals in sediments; they conclude that metals can interact with organic matter in different ways forming phenomena of adsorption, ion exchange, coprecipitation and complexation.

Other studies have determined the levels of heavy metals in sediments and organisms along the Terminos, Atasta and Pom lagoons and in the Palizada, Candelaria and Chumpan Rivers. Aguilar et al. [12] attributed the levels of Cd, Cr, Cu, Hg and V detected in oysters (*Crassotrea virginica*) to anthropogenic activities; additionally, they calculated the condition index of the oyster (variable that indicates the condition of health) and attributed a decrease in it to the presence of heavy metals; likewise, the levels of Cd, Cr and Cu exceeded the permissible limits established for mollusks and fishery products in the Mexican norms NOM-031-SSA1–1993.

In another study, the concentrations of Cd, Fe, Cu, Pb and Zn were evaluated in oyster (*Crassostrea virginica*), crab (*Callinectes sapidus*) and shrimp (*Litopenaeus setiferus*). The results showed that both oyster and crab are foods that present high levels of Cd, Fe, Cu and Pb in comparison with shrimp; in this study all detected levels were within the permissible limits established by the Mexican Official Standards NOM-031-SSA1–1993 [13].

Regarding sediment studies, Montalvo et al., [14] analyzed the concentration of heavy metals in sediments of the Palizada River; the results showed a high relationship between the levels of metals found with the climatic season and the texture of the sediment. Later, Canedo et al. [15] evaluated the levels of heavy metals in sediments of the Terminos Lagoon; they concluded that the spatial distribution was influenced by river discharges and that the significant correlations found between B, Ba, Co, Mn, Ni and Zn are due to natural biogeochemical inputs; they also found heavy metal levels above background concentrations in sites near the Atasta Lagoon and considered this area vulnerable to heavy metal contamination.

1.1 Effects of heavy metals

Heavy metals exert a wide range of toxic effects in humans, aquatic and terrestrial life [12]. Different strategies have been developed to study the degree of contamination of an area, such as the use of organisms called sentinels (oysters, clams) that due to their feeding habits, their little or no mobility, their little capacity to regulate the concentrations of ions in the internal fluids and their high tolerance to the metal ions absorbed above the metabolic requirements [16], make them ideal for studies of contamination by heavy metals; likewise, studies on fish have been of considerable interest to understand the toxic effects and because they are an important source of nutrients for humans and have the potential to bioaccumulate heavy metals in their tissues [17, 18]. Food contamination can come from different sources: from contamination of the aquatic environment, during harvesting, transportation, handling or packaging.

1.1.1 Mercury (Hg)

Regarding the toxicity of heavy metals, Hg is distinguished because it does not have any biological function; its presence in the environment is due to anthropogenic causes; the natural causes of contamination by this element are not significant. It is an extremely toxic metal; organisms that have been exposed have few biological mechanisms for its elimination and it accumulates progressively through the food chain [19, 20]. The most common form of organic Hg is in the form of methyl mercury (MeHg). Usually levels above tolerance limits can alter the normal functioning of the central nervous system and affect the kidneys and the immune system [21]. Studies show that the toxicity attributed to it is associated with aging and cell death. Bryan and Langston's study [22] study on the oyster *Crassostrea virginica* showed evident embryonic abnormalities at concentrations of 5 to 10 μg/L, while the survival rates of clams, copepods, shrimp and crustaceans were affected by the increase in Hg levels.

1.1.2 Cadmium (Cd)

Cadmium is an element that has no natural source of generation so its presence in aquatic systems and organisms is entirely anthropogenic [23]. Cd does not have biochemical or nutritional functions; it is highly toxic to plants and animals. The International Agency for Research on Cancer points out the Cd and its compounds as carcinogenic. Cd intake pathways in organisms are gastrointestinal and respiratory; it has severe consequences in the blood by binding to high molecular weight proteins [24]; likewise, it has been reported that it can cause different alterations in the biology of living beings, since it accumulates mainly in the liver and can have a half-life of thirty years [25]. In phytoplankton species, growth inhibition was observed at concentrations as low as 1 μg/L [22]. Other species such as *Galaxias maculatus* exposed to acute concentrations showed deficiencies in metabolic rate and deteriorating oxygen consumption; also, stress parameters and decrease in liver catalase activity were observed [26]. In the *Henanese Sinopotamon* crab, a high deterioration of enzyme activity was found in the stomach, intestines, and hepatopancreas [27]. For *Crassotrea virginica* oyster, hepatological changes of the intestine, digestive gland and other organs were presented when exposed to Cd [28]. Due to its source of origin, the activities by which it can be generated are the industrial processes of fertilizer production, by-product of the smelting of other metals and in electronic devices [24].

1.1.3 Copper (Cu)

Cu is an essential element for the growth and metabolism of many living beings; when the levels are increased, it becomes a not very tolerable element [12]. This metal can cause harmful effects in fish, showing damage such as histopathological alteration and accumulation in different organs [29]. Other studies [22] presented experimental evidence that a considerable number of species are sensitive to concentrations of 1 to 10 μg/L of Cu, while at levels of 2 μg/L, the survival rate in young scallops was reduced; likewise, oyster and mussel embryos showed abnormalities in growth and development after exposure to 5 μg/L and the isopod crustacean *Idothea baltica* showed an increase in population mortality. Calabrese et al. [30] studied the acute toxicity of Cu in embryos of *Crassostrea virginica*; the results showed that at certain concentrations there was no development in more than 50% of the individuals under study.

1.1.4 Lead (Pb)

Pb can be in the environment in particulate form or formed into lead compounds; it can be generated as a result of human activities such as oil combustion, industrial processes and solid waste combustion; there are no natural sources of lead, its presence in the environment is anthropogenic [25]. It has been reported that in humans this metal can cause alterations of the nervous system, kidney problems and is related to the development of cancer. In exposed fish, it has been shown to decrease red and white blood cells and decrease hemoglobin levels [18]. The process of Pb accumulation in fish tissues causes oxidative stress; thus, this stress induces synaptic damage and neurotransmitter malfunction and influences immune responses [31].

1.1.5 Nickel (Ni)

Ni is a non-essential and toxic metal whose main source of exposure is food, highlighting fish and vegetables that are treated with wastewater. Its introduction to the aquatic environment is anthropogenic. The effects that it causes in different organisms were studied by Martin et al., [32] in embryos of Pacific oyster (*Crassostrea gigas*), embryos of laurel mussel (*Mytilus edulis Linnaeus*) and larvae of Dungeness crab (*Cancer magister Dana*) exposed to ten metals among them Ni; the effects caused in these species are the abnormal development in more than 50% of the studied individuals. In fish such as *Colisa fasciatius*, a freshwater teleoste, exposed to 45 ppm nickel sulfate, the adverse effects observed were leukopenia due to reduced numbers of lymphocytes and polycythemia, as well as a considerable delay in the rate of erythrocyte sedimentation of dying fish [33].

The conditions of the aquatic environment have a great influence on the transport and mobility of metals such as Ni, so Tamzin et al., [34] carried out their studies in saline waters, hoping that these conditions would decrease the impact on marine biota; however, despite the speciation of the metal in these saline environments it was determined that the physiology of the organisms is the main factor in the toxic impact, finding deterioration as inhibition of breathing and promotion of oxidative stress. In other studies, the mortality rate of African catfish, *Clarias gariepinus*, showed a linear trend with increasing concentration; the researchers concluded that the depression observed in hematocrit, hemoglobin and erythrocyte decreases in this hematological study can be used as an indicator of Ni-related stress in fish [35].

2. Methodology

2.1 Study area

Terminos Lagoon is the largest lagoon-estuarine ecosystem in Mexico by area and volume. The water body and immediately surrounding shorelands are fully incorporated into a National Flora and Fauna Reserve comprised of 705,016 ha of open water and associated wetlands and upland. Terminos Lagoon consists of about 200,108 ha of open water including associated lagoons and channels, with an average depth of 4 m, surrounded by about 259,000 ha of mangrove and cattail marsh. Of the surrounding 180,000 ha of land that is in some productive use, 90% is cattle ranching, 6% is agricultural, and 4% is urbanized, principally the City of Carmen. It is separated from the Gulf of Mexico by the Carmen Island, a 37 km long, 4 km wide barrier island with two mouths, of 3.2 and 3.8 km located to the east and west, respectively.

Terminos Lagoon was declared as a Federal Flora and Fauna Protection Zone in 1994 and is considered a "critical habitat" by the Mexican Environmental Agency [http://www.paot.org.mx/centro/ine-semarnat/anp/AN19.pdf] due to its importance as a refuge for marine species, mangrove forests, sea grass and, associated fluvial - lagoon delta system. Anthropogenic pressure mainly due to urban settlements, the disposal of wastewater in the lagoon and industrial activity based on the drilling and exploration of hydrocarbons, all these activities have been identified as a continuous threat to the quality of the ecosystems within the Terminos Lagoon.

This work summarizes the results of several investigations carried out in Terminos Lagoon Natural Protected Area where the content of heavy metals in a variety of aquatic organisms was analyzed. The sampling periods and collect sites are shown below, as well as the aquatic organisms used for the determination of heavy metals.

In 2009, during two sampling campaigns (rainy and dry seasons), the oyster (*Crassostrea virginica*) was collected at the mouth of three of the rivers that flow into the Terminos Lagoon: the Palizada River, the Chumpan River and the Candelaria River. At each site, three sampling points were established and 100 organisms were obtained from each one.

In 2013–2014 three different types of organisms were analyzed: the oyster (*Crassostrea virginica*) collected in two sites, Estero Pargo and Mouth of Atasta; shrimp (*Litopenaeus setiferus*) obtained by trawling in depths of less than 5 fathoms in the Terminos Lagoon; and the crab (*Callinectes sapidus*) collected at the mouth of the Palizada River. All the organisms were donated by the fishermen's cooperatives. 60 organisms of commercial size were obtained of each species.

In 2014, samples of three species of macrophytes (*Cyperus ligularis L., Lemna minor* and *Typha domingensis*) were collected and analyzed in the "Arroyo La Caleta", which is a natural water channel parallel to the coast that crosses Carmen City, with a variable extension between both banks. The main contribution of water enters through the west mouth of the Terminos Lagoon and does not present an outlet. Other contributions of water come from land and urban drainage. The system is 7.5 km long. In the case of *T. domingensis* and *C. ligularis L.*, the complete plants were cut, stored in plastic bags, and placed in refrigeration for later analysis in the laboratory. The samples of *L. minor* were collected in plastic bags in which water from the "Arroyo La Caleta" was left and, like the previous macrophytes, they were stored in refrigeration.

In 2017, the clam *Rangia cuneata* was collected at four sampling points in the Atasta Lagoon, which is a lagoon that empties into Terminos Lagoon. In total, eight composed samples were analyzed for this study. In the same lagoon (Atasta) but in 2018, during two sampling campaigns (rainy and dry seasons), catfish (*Ariopsis felis*) was obtained by fishing with cast nets eight. 30 composed samples were analyzed.

Table 1 summarizes information about of the analyzed organisms, their sampling location and year of collection, while **Figure 1** shows the Terminos Lagoon Natural Protected Area and the sampling locations of the organisms analyzed.

Species analyzed	Sampling location	Year of collection
Oyster (*Crassostrea virginica*)	Mouth of the Palizada[1], Chumpán[2] and Candelaria[3] rivers that flow into the Terminos Lagoon.	2009
Oyster (*Crassostrea virginica)*	Pargo Estuary[4] and Atasta Mouth[5]	2013–2014
Shrimp (*Litopenaeus setiferus*)	Terminos Lagoon	2013–2014
Crab (*Callinectes sapidus*)	Mouth of the Palizada River[1]	2013–2014
Macrophytes (*Cyperus ligularis L.*, *Lemna minor* and *Typha domingensis*)	Arroyo La Caleta[6]	2014
Clam *Rangia cuneata*	Atasta Lagoon[7]	2017
Catfish (*Ariopsis felis*)	Atasta Lagoon[7]	2018

*Superscripts indicates their location on the map (**Figure 1**).*

Table 1.
Organisms analyzed, their sampling location and year of collection.

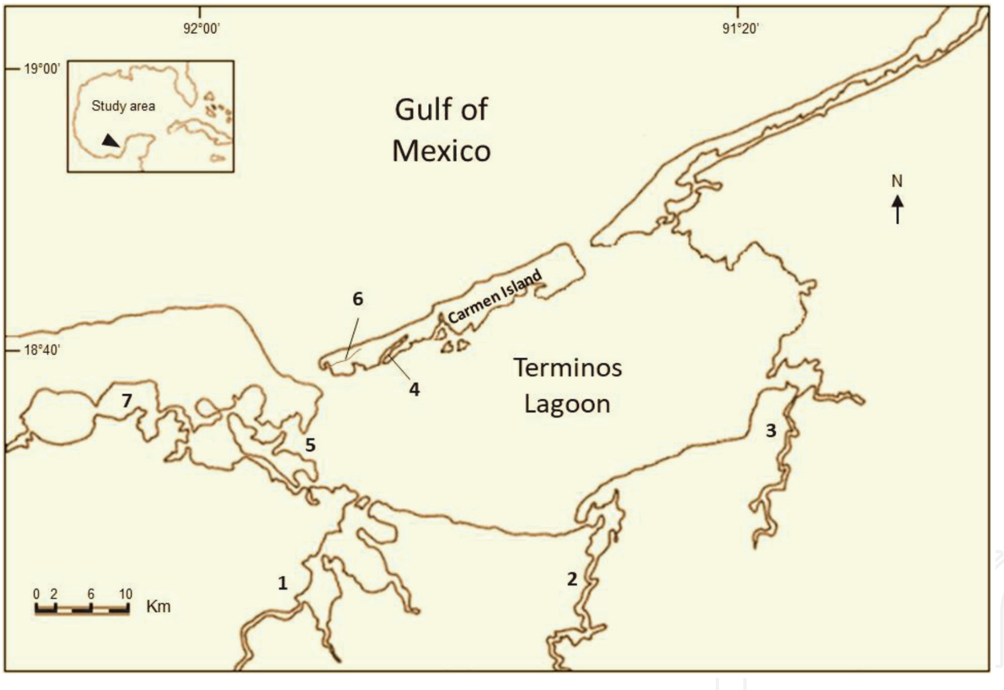

Figure 1.
Terminos Lagoon natural protected area and sampling locations of the organisms analyzed: 1) Palizada River; 2) Chumpan River; 3) Candelaria River; 4) Pargo estuary; 5) Atasta mouth; 6) arroyo La Caleta; 7) Atasta lagoon.

2.2 Sample processing and analysis

The methods used for the processing of tissues and extraction of heavy metals reported in the various studies considered for the evaluation in this work, have few variations or modifications according to "Official Mexican Standard" (NOM-117-SSA1–1994, *Test method for the determination of cadmium, arsenic, lead, copper, iron, zinc and mercury in food*) for food analysis that generally consisted of an acidic digestion of the tissues with a repetitive addition of concentrated HNO_3 and H_2O_2.

For bivalves: before extracting the tissues, they were purged during a period of 24 hours in a system with a controlled salinity of 20 psu. By so doing, the bivalves eliminated all the organic matter from their intestines that could have interfered with the results. Finally, they were shucked manually. Organisms were dried through the process of lyophilization for 24 hours and then were homogenized. Subsequently, an acid digestion was carried out, according to the official Mexican standards, as mentioned at the beginning of this section.

Bivalves samples were analyzed by Plasma Emission Spectroscopy (ICP), Perkin Elmer model 400 instrument was used, and standard solutions (J. Baker). For the evaluation of the analytical quality, the samples of oyster tissues were treated in duplicate and were analyzed in parallel with the standard certificates of "Standard reference materials oyster tissue" (SRM-1566b), with a recuperation percentage of between 84 and 94%.

For crustaceans and fish: composite samples were used for which the edible part was extracted from the organisms of each species, the tissues were homogenized with a food processor and a final sample of (20 $\pm$ 0.001 g) was taken. The digestion was carried out by adding 10 mL of HNO_3 to the tissues and placed on a heating grill at a controlled temperature, after the total destruction of organic matter, 2 mL of H_2O_2 was added to each sample in 30% solution, concentrating them up to a volume of 1 mL, finally, the concentrate was filtered through Whatman No. filter paper 32, measuring to a final volume of 20 mL for subsequent analysis. The tissue samples were analyzed in an atomic flame absorption equipment adapted with a Thermo-Scientific brand graphite furnace.

For macrophytes: samples were dried in a drying oven at a temperature of 65° C for 96 hours. The dried samples were dissected at the root, stem, and leaf. The digestion was carried out as mentioned for crustaceans and fish, following the methodology of the official Mexican standard NOM-117-SSA1–1994 (Test method for the determination of cadmium, arsenic, lead, tin, copper, iron, zinc and mercury in food). Macrophytes samples were analyzed by Plasma Emission Spectroscopy (ICP).

3. Metal concentration and risk analysis to human health

Table 2 shows the concentrations of metals found in different organisms, which are the basis for determining the risk factor analysis; as expected, organisms such as bivalve mollusks from the Candelaria, Chumpan and Palizada rivers, show the

Organism	Heavy metal levels in different organisms in μg g^{-1}					Location	Reference
	Cu	Hg	Cd	Ni	Pb		
Crassotrea virginica	56.630 (43.320–56.630)	ND	0.038 (0.020–0.038)	ND	0.137 (0.043–0.137)	Terminos Lagoon	Aguilar et al., 2014
Crassotrea virginica	60.5 (33.24–60.5)	1.1 (0.05–1.1)	2.4 (0.243–2.4)	ND	ND	Candelaria River	Aguilar et al., 2012
Crassotrea virginica	90.8 (30.50–90.8)	0.7 (0.01–0.7)	3.2 (0.23–3.2)	ND	ND	Chumpan River	Aguilar et al., 2012
Crassotrea virginica	176.5 (90.23–176.5)	0.5 (0.02–0.5)	3.0 (1.2–3.0)	ND	ND	Palizada River	Aguilar et al., 2012

Organism	Heavy metal levels in different organisms in μg g^{-1}					Location	Reference
	Cu	**Hg**	**Cd**	**Ni**	**Pb**		
Callinectes sapidus	57.800 (34.680–57.800)	ND	0.0687 0.0398–0.0687	ND	0.4253 (0.2644–0.4253	Terminos Lagoon	Aguilar Et al., 2014
Litopenaes setiferus	58.470 (41.620–58.470)	ND	0.015 (*ND-0.015)	ND	ND	Terminos Lagoon	Aguilar et al., 2014
Ariopsis felis	—	0.02934 (0.00159–0.02934)	1.2864 (0.00454–1.2864)	41.77 (0.33–41.77)	3.2097 (0.033–3.2097)	Atasta Lagoon	This Study
Rangea cuneata	308.2135 (6.1609–308.2135)	ND	0.74827 (0.2905–0.7482)	30.2055 (13.7574–30.2055)	ND	Atasta Lagoon	This Study

Table 2.
Heavy metals determined in different marine species.

National and international legislation governing heavy metal levels in fish in μg g^{-1}						
	Cu	**Hg**	**Cd**	**Ni**	**Pb**	**Year**
JECFA[1]	—	0.5	—	—	0.5	1989
WHO[2]	20	0.005	2	—	2	1996
USFDA[3]	—	—	—	70-80	—	1993
NOM 242[4]	—	1	0.5	—	1	2009
FAO[5]	30	0.5	0.5	—	0.5	1983
National and international legislation governing the levels of heavy metals in marine mollusks and crustaceans in μg g^{-1}						
	Cu	**Hg**	**Cd**	**Ni**	**Pb**	**Year**
NOM 242[6]	—	1	0.5	—	1	2009
USFDA[7]	—	—	4.0	—	1.7	1993
NAUEN[8]	32.5	—	—	—	—	1983
ISSC[9]	—	—	—	80	—	2007

[1]*JECFA, 1989 Evaluation of Certain Food Additives and Contaminants (Thirty-third Report of the Joint FAO/WHO Expert Committee on Food Additives) [meeting held in Geneva from March 21 to 30, 1988]. World Health Organization.*
[2]*WHO, 1996. Health criteria other supporting information. In: Guidelines for Drinking Water Quality p. 31–388.*
[3]*USFDA, 1993. Food and Drug Administration, Guidance for Nickel in Shellfish. DHHS/PHS/FDA/CFSAN/Office of Seafood, Washington DC, 1993.*
[4]*NOM-242-SSA-2009. Products and services. Fresh, refrigerated, frozen and processed fishery products. Health specifications and test methods.*
[5]*FAO,1983. Compilation of legal limits for hazardous substances in fish and fishery products Fish Circular 464:5–100.*
[6]*NOM-242-SSA1–2009 Products and services. Fresh, refrigerated, frozen and processed fishery products. Health specifications and test methods.*
[7]*USFDA, 1993. Guidance document for lead in shellfish. Center for Food Safety and Applied Nutrition. United States Food and Drug Administration, Washington, D. C.*
[8]*Nauen C.E, 1983. Compilation of legal limist for hazardous substances in fish and fishery products. FAO fisheries circular 764. United Nations Food and Agriculture Organization. Rome, Italy, 102 pp.*
[9]*ISSC,2007. National shellfish sanitation program. Guide for the control of molluscan shellfish. Interstate Shellfish Sanitation Conference. U.S. Food and Drug Administration, Department of Health and Human Services. Washington, D.C., 549 pp.*

Table 3.
Permissible levels of heavy metals in fishery products.

highest values in Cd, exceeding the limits established by the official Mexican Standards (NOM 242 and the USFDA standards **Table 3**). Likewise, Cu levels are high in clams *Rangea cuneata*, exceeding international specifications (**Table 3**); the Pb levels in *Ariopsis felis* exceed all the international specifications contemplated in this study. The reference values reported in national and international standards for heavy metals in marine fish and mollusks are shown in **Table 3**.

Concentrations of heavy metals in the macrophytes collected in "Arroyo La Caleta", where detected below the limit of quantification of the method (not detected ND) for B, Be, Fe, Mn and Si to 4671.29 μg g^{-1} for Fe. The average concentration of the metals analyzed presented the following order: Be < As < V < Mn < B < Si < Fe. Of the three species of macrophytes studied, the one that presented higher concentrations of metals was *Cyperus ligularis L.* accumulating higher percentages of Be, Fe, Mn, Si and V, followed by *Typha domingensis* which presented the highest concentration of As and average concentrations of the other metals, finally, *Lemna minor*. Regarding the structure analyzed in plants, the highest concentrations were found in the roots and to a lesser extent in the stem and leaves, which tells us about the ability to translocate (mobilize) the metals to the aerial parts as a strategy of adaptation to pollution due to heavy metals. In general, the highest concentrations found in macrophytes were related to the sites of highest anthropogenic activity [36]. It should be noted that, of all the collection sites in the Términos Lagoon Natural Protected Area analyzed in this study, the "Arroyo La Caleta" is considered one of the most contaminated because it receives wastewater from Carmen City, which is why the fish products obtained there have not been considered suitable for human consumption for several years. Due to the above, the study of the behavior of heavy metals in this location, was based on organisms with high levels of adaptation to heavy metal pollution that are not used for human consumption.

4. Estimation of the estimated daily intake, target hazard quotient, hazard index and target cancer risk

According to the USEPA [37–38] the estimate of potential risk to human health from the consumption of contaminated marine products is based on the estimated daily intake (EDI), the target hazard quotient (THQ), hazard index (HI) and target cancer risk (TR). The first parameter that was estimated is a function of the relation (EDI; μg kg^{-1} $week^{-1}$)

$$\mathrm{EDI} = \frac{(\mathrm{FIR})(\mathrm{MC})}{\mathrm{BWa}} \tag{1}$$

Where FIR is the food ingestion rate of fish or shellfish consumed by an adult; according to CONAPESCA, these data for Mexico, is up to 12 kg $year^{-1}$ or the equivalent of 230 g $week^{-1}$ for an adult with an average weight. For children aged 4 to 6 years, the recommended food ingestion rate is 100 grams per week (CONAPESCA: National Commission for Aquaculture and Fisheries, Fishery production statistics, www.conapesca.sagarpa.mx) or its equivalent of 5.214 kg $year^{-1}$.

According to studies by Araneda [39] the population group of adolescents is the one that shows a lower food ingestion rate of these foods; there is no data on intake in Mexico, but the recommendations indicate that an average adolescent between 14 and 17 years of age should consume between 240 and 300 grams of fish and seafood per week, the equivalent of an average of 15. 64 kg $year^{-1}$. Due to the scarcity of information in this population group, in this study it is considered that the average food ingestion rate for adolescents is 7.8 kg year^{-1}, which is considered a low consumption and that represents a value of 150 gr week^{-1}.

The parameter BWa is the reference body weight of an adult. In countries such as China, this data is 55 kg [40]; the average weight of a Mexican adult is 70 kg. The average weight for children between 4 and 6 years old is 16 kg [41] and for an average adolescent between 14 and 17 years old the weight considered is 54 kg. All the reference data are based on the characteristics and habits of the Mexican population without obesity problems. The MC parameter is the metal concentration (Cu, Pb, Ni, Hg) expressed in μg g^{-1}.

The estimated results for the EDI parameter are shown in **Table 4**. With the data from the population intake rates, we can estimate the THQ parameter which is a dimensionless amount and a relationship between the concentration of heavy metals in ingested food with other factors. According to USEPA [37–38] the THQ value should not exceed the numerical value of 1. Estimated values below 1 indicate that the contaminant levels do not cause adverse effects or potential non-carcinogenic risks in exposed persons during the estimated average life span of the Mexican population of 70 years.

The model for estimating the target hazard quotient (THQ) is determined by Eq. 2. The units were adequate for not using correction factors (**Table 5**).

$$THQ = \frac{(Efr)(EDtot)(FIR)(MC)}{(Rfd)(BWa)(Atn)} \tag{2}$$

Where Efr is the exposure frequency to the trace element, (365 days year^{-1}), EDtot is the exposure duration (average life span of 70 years), FIR is the food ingestion rate in grams per day for the respective food item (g day $^{-1}$), MC is the concentration of the trace element in the given food item (μg g^{-1}), Rfd: is the oral reference dose of the trace element (μg g-^{1} day^{-1}) (5×10^{-4} for Hg; 1×10^{-3} for Cd; 4×10^{-3} for Pb; 2×10^{-2} for Ni and 4×10^{-2} for Cu), BWa is the reference body weight (g), Atn is the averaged exposure time (Efr x EDtot).

The values obtained show a variable trend among the reference population groups, with children aged 4–6 years being those with the highest values of EDI for Cu and Ni; these values are directly related to weight.

4.1 Hazard index (HI)

The accumulated risk was evaluated by the individual sum of each of the THQ factors which represent the risk index (HI), which is shown in Eq. 3. These values, like THQ, must not exceed the numerical value of 1, otherwise it could indicate that there are considerable risk factors for the consumption of marine products reported in this study.

$$\mathrm{HI} = \sum \mathrm{THQ} \tag{3}$$

According to the estimated results (**Table 6**) no value calculated for THQ and HI exceed the parameters established to consider a risk to health by the intake of these contaminants from the consumption of fish and seafood.

The results indicate that the estimated HI values do not represent a risk for the reference population, since they do not exceed the comparison value of 1.

Estimated daily intake EDI (μg kg^{-1} $week^{-1}$)															
	Cu			Cd			Pb			Ni			Hg		
Organism	**Adults**	**Children (4–6 years)**	**Teenagers (14–17 years)**	**Adults**	**Children (4–6 years)**	**Teenagers (14–17 years)**	**Adults**	**Children (4–6 years)**	**Teenagers (14–17 years)**	**Adults**	**Children (4–6 years)**	**Teenagers (14–17 years)**	**Adults**	**Children (4–6 years)**	**Teenagers (14–17 years)**
Crassotrea virginica	186.070	353.937	156.256	0.1248	0.2375	0.10485	0.4501428	0.85625	0.37801	—	—	—	—	—	—
Crassotrea virginica	198.785	378.125	166.935	7.8857	15	6.622	—	—	—	—	—	—	3.6142	6.875	3.0351
Crassotrea virginica	298.342	567.500	250.540	10.514	20	8.8296	—	—	—	—	—	—	2.3	4.375	1.93148
Crassotrea virginica	579.928	1103.125	487.009	9.857	18.75	8.277	—	—	—	—	—	—	1.64285	3.125	1.3796
Callinectes sapidus	189.914	361.250	159.485	0.226	0.42937	0.189561	1.39741	2.6581	1.1735	—	—	—	—	—	—
Litopenaes setiferus	192.115	365.437	161.333	0.049	0.09375	0.041388	—	—	—	—	—	—	—	—	—
Ariopsis felis	143.171	313.180		4.226	1.878.04	3.54951	1.045	2.285	8.8563	137.244	261.0625	115.2542	0.096402	0.1833	0.08095
Rangea cuneata	1012.700	1926.33	850.440	2.4601	4.675	2.06392	—	—	—	99.246	188.78	83.344	—	—	—

Table 4.
Estimated daily intake (EDI), taking as a reference a population group between adults and children of different age ranges.

	Target hazard quotient (THQ)														
	Cu			Cd			Pb			Ni			Hg		
Organism	Adults	Children (4–6 years)	Teenagers (14–17 years)	Adults	Children (4–6 years)	Teenagers (14–17 years)	Adults	Children (4–6 years)	Teenagers (14–17 years)	Adults	Children (4–6 years)	Teenagers (14–17 years)	Adults	Children (4–6 years)	Teenagers (14–17 years)
Crassotrea virginica	0.1056	0.0884	0.0262	0.0005	0.0024	0.0007	0.0005	0.0022	0.0007	—	—	—	—	—	—
Crassotrea virginica	0.1127	0.0945	0.0280	0.0342	0.1500	0.0444	—	—	—	—	—	—	0.0291	0.1273	0.0377
Crassotrea virginica	0.1690	0.1418	0.0424	0.0457	0.2000	0.0593	—	—	—	—	—	—	0.0185	0.0820	0.0240
Crassotrea virginica	0.3287	0.2757	0.0817	0.0428	0.1875	0.0556	—	—	—	—	—	—	0.0132	0.0580	0.0172
Callinectes sapidus	0.1077	0.0903	0.0268	0.0009	0.0043	0.0013	0.0015	0.0067	0.0020	—	—	—	—	—	—
Litopenaes setiferus	0.1088	0.0913	0.0270	0.0002	0.0009	0.0003	—	—	—	—	—	—	—	—	—
Ariopsis felis	—	—	—	0.0184	0.0804	0.0239	0.0115	0.0502	0.0149	0.0299	0.1305	0.0387	0.0008	0.0034	0.0010
Rangea cuneata	0.5739	0.4815	0.1427	0.0107	0.0467	0.0139	—	—	—	0.02160	0.0944	0.0280	—	—	—

Table 5.
Estimated values of the target hazard quotient (THQ) in a reference population group.

Hazard index values (HI)			
Organism	**Adults**	**Children (4–6 years)**	**Teenagers (14–17 years)**
Crassotrea virginica	0.1065	0.0930	0.0276
Crassotrea virginica	0.1467	0.2445	0.0724
Crassotrea virginica	0.2148	0.3419	0.1013
Crassotrea virginica	0.3715	0.4632	0.1372
Callinectes sapidus	0.1101	0.1013	0.0300
Litopenaes setiferus	0.1091	0.0923	0.0273
Ariopsis felis	0.0298	0.1306	0.0784
Rangea cuneata	0.5846	0.5283	0.1567

Table 6.
Estimated values of the hazard index estimated for a reference population, expressed as the sum of all the individual HI factors.

4.2 Target cancer risk

The risk from carcinogens was indicated in this study by the TR values (**Table 7**). For the estimation of these parameters, the values of Region III were taken, where the population of Mexico is included according to USEPA criteria [37–38]. The equation that represents risk for carcinogenic factors is represented by the following expression:

$$TR = \frac{(Efr)(EDI)(FIR)(MC)(Cfo)}{(BWa)(Atn)} \qquad (4)$$

Where TR is the target cancer risk (dimensionless factor) and Cfo is the oral cancer slope factor; USEPA criteria [42] (μg g^{-1} bw day^{-1}). The other values are the same used for the estimation of EDI and THQ. In this study, the Cfo values used to estimate TR are 8.5×10^{-3} for Pb and 1.7 for Ni, a metal that is on the list of potent carcinogens [42]. The value of 2.59×10^{-4} was used for Cd which has been considered carcinogenic according to the International Agency for Research On Cancer (IARRC: http://www.iarc.fr/en/websites/index.php). Not all metals are considered within this classification so far; only Cd, Pb and Ni fall into this category. According to the New York State Department of Health [43] (NYSDOH) the categories of the target cancer risk (TR) are the following: TR $\leq 10^{-6}$ low risk, between 10^{-4} and 10^{-3} moderate risk, from 10^{-3} to 10^{-1} high risk and TR $\geq 10^{-1}$ very high risk that people may develop cancer at some point in their life after exposure to the metal.

The results of the target cancer risk for Cd and Pb show that the three study categories (adults, children, and adolescents) present a low risk of developing cancer from the ingestion of fish and shellfish. On the other hand, the target cancer risk calculated for Ni shows that the population group of children aged 4–6 years represents a very high risk, and a moderate risk is expected for adults and adolescents.

In certain cases, it is not advisable to limit the consumption of these marine products. It is more useful to be vigilant, as well as to limit the frequency of consumption when there is evidence of risk for the population. These actions make the difference in developed countries that have public policies and develop research

Target cancer risk (TR)									
	Cd			Pb			Ni		
Organism	Adults	Children (4–6 years)	Teenagers (14–17 years)	Adults	Children (4–6 years)	Teenagers (14–17 years)	Adults	Children (4–6 years)	Teenagers (14–17 years)
Crassotrea virginica	5.768E-11	2.087E-10	4.10308E-11	2.460E-08	8.920E-08	1.637E-07			
Crassotrea virginica	2.300E-07	8.325E-07	1.63668E-07	—	—	—	—	—	—
Crassotrea virginica	4.090E-07	0.004	2.90966E-07	—	—	—	—	—	—
Crassotrea virginica	3.595E-07	1.307E-06	2.55732E-07	—	—	—	—	—	—
Callinectes sapidus	1.886E-10	6.821E-10	2.37122E-07	2.371E-07	8.596E-07	1.686E-07	—	—	—
Litopenaes setiferus	8.987E-12	3.251E-11	1.34108E-10	—	—	—	—	—	—
Ariopsis felis	6.610E-08	2.391E-07	6.3933E-12	2.087E-10	5.577E-06	8.988E-05	0.458	1.658	0.325
Rangea cuneata	2.234E-08	8.086E-08	4.70213E-08	—	—	—	0.3308	1.867	0.158

Table 7.
Estimated values of the target cancer risk (TR) in a reference population.

through environmental agencies to develop models that can be applicable to different regions of the world and thus predict or estimate possible risks.

There are general recommendations in Mexico regarding the consumption of fish and seafood by children under four years of age. Certain countries such as Canada restrict consumption of species caught in rivers and lakes and recommend that consumption in the population group of children 1–4 years old be only 75 g month^{-1} and in children 5–11 years old be 125 g month^{-1}, as well as that pregnant women should not consume more than 150 g month^{-1} [44]. Some of these recommendations are in most cases based on economic interests.

Bellanger et al. [44], analyze in their studies the economic implications of exposing a population group to the toxic effects of heavy metals. In Mexico, consumption of fish and seafood is lower than that of other foods. In Mexico, fish products are governed by Mexican standards that limit the presence of heavy metals in their products; likewise, government institutions regulate the health of oyster and clam banks (due to the presence of pathogenic microorganisms), but there are no effective public policies focused on protecting the environment, stopping the deterioration of mangrove areas or monitoring and sanctioning poaching and depredation that are putting numerous species at risk.

5. Conclusions

Throughout this chapter, aspects of the region of the Campeche Sound were shown, and in particular the Terminos Lagoon, which only a decade ago produced more than 80% of the national production of crude oil, while at the same time numerous marine species of high commercial value were extracted from its waters.

Today, even though oil activity has decreased considerably, the effects of this industry, combined with population growth and the ineffectiveness of monitoring programs, still persist. The results of this study of the concentrations of heavy metals in *Crassostrea virginica*, *Rangea cuneata* and *Ariopsis felis* indicate that some of the values found are higher than those established in international and national legislation, so these fish products should not be consumed. The establishment of a monitoring program is suggested to identify the variations and conditions that favor the bioaccumulation process in exposed organisms.

Regarding the risk analysis carried out in this study, the values calculated for the target hazard quotient (THQ) and the hazard index (HI) indicate that the consumption of the studied species does not represent a risk for human health in any of the considered age groups; however, in relation to the TR that evaluates the potential risk for carcinogens, the results show worrying values, especially for the organisms that come from the Pom-Atasta lagoon system. In the two species evaluated (*Rangea cuneata* and *Ariopsis felis*), the TR values are considered "high risk" and "moderate risk", especially in the most vulnerable population group, children. For this reason, it is not recommended the consumption of these species by children under 4 years old and it is suggested to decrease their consumption in the adult and adolescent age groups. These actions are not intended to stigmatize the consumption of these products, but to have greater control and surveillance, especially in population groups of greater vulnerability.

Conflict of interest

The authors declare no conflict of interest.

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