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Biotransport of mercury and human methylmercury exposure through crabs in China – A life cycle-based analysis

Zhihao Zhang [a], Long Chen [b], Menghan Cheng [a], Maodian Liu [c,*], Xuejun Wang [a,*]

[a] Ministry of Education Laboratory of Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China
[b] Key Laboratory of Geographic Information Science, Ministry of Education, East China Normal University, Shanghai 200241, China
[c] School of the Environment, Yale University, New Haven, Connecticut 06511, USA

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ABSTRACT

Exposure to methylmercury (MeHg) has various toxic effects on humans. The evaluation of human MeHg exposure has previously focused on fish consumption. However, in this study, we found that MeHg levels in domestic crabs in China were also relatively high (range: 50–1400 ng/g, dry weight). The high MeHg levels in crabs and their high consumption do not match the risk assessment of MeHg, indicating an underestimated exposure risk, especially in MeHg-sensitive groups such as pregnant women. The annual crab MeHg content output in China was estimated to be 30 ± 27 kg. A total of 6.8% of the country's land area contributes 71% of the MeHg output. However, 66% of the output is redistributed to non-crab-producing regions via interregional food trade, posing risks to the population on a national scale. The daily intake of MeHg from crabs could easily exceed the reference dose (0.1 μg/kg of body weight per day) suggested by the United States Environmental Protection Agency with consideration of coexposure from fish, rice, and other food sources. We suggest that future MeHg exposure analysis includes crab MeHg as a coexposure pathway to estimate the dietary MeHg limit accurately and emphasize the influence of interregional food trade on MeHg exposure.

1. Introduction

Mercury (Hg) is a poisonous trace metal that travels in the environment via various pathways and biomagnifies along food webs (Clarkson and Magos, 2006). Although Hg occurs naturally in the environment, human activities such as mining and fossil fuel combustion have elevated the amount of Hg available from various reservoirs, significantly influencing land, water bodies, and the atmosphere (Krabbenhoft and Sunderland, 2013; Mason et al., 1994; Selin, 2009; Streets et al., 2017). More than 1.5×10^6 megagrams (Mg) of Hg has been released by human activities into the environment since 1850, increasing the amount of Hg in surface ocean water by a factor of two (Lamborg et al., 2014; Streets et al., 2017).

Methylmercury (MeHg) is one of the most toxic derivatives of Hg, and it reaches high concentrations in aquatic species at high trophic levels via biomagnification along food webs (Stein et al., 1996). Human exposure to MeHg leads to cardiovascular impairment in adults, and neurocognitive deficits and developmental delays in children after direct exposure and in fetuses and infants after indirect exposure from exposed mothers (Clarkson and Magos, 2006). Human MeHg exposure is primarily attributed to dietary intake, where fish consumption is the predominant exposure pathway due to high Hg levels and intake rates (Morel et al., 1998). Therefore, food selection is crucial for maintaining health in MeHg-sensitive individuals, such as children and women of childbearing age (Clarkson, 1997; Clarkson and Magos, 2006; Harada, 1995). Several common marine fish species have high MeHg levels, such as sharks, swordfish, and tuna (Food and Drug Administration (FDA, 2017). Previous studies on human MeHg exposure have focused on marine fish species at high trophic levels and underestimated the impact of Hg intake from other marine and freshwater products with potentially high Hg levels, such as crabs. Additionally, studies have shown that pregnant and postpartum women who are aware of the health risks of MeHg generally limit their consumption of marine fish based on the United States FDA guidelines (Lando et al., 2012). High MeHg levels in crabs have been widely overlooked and crabs are considered a quality nutrition source due to their high levels of glutamic acid and other dietary values (Chen et al., 2007). Children and female adults consume more crabs than male adults, indicating a higher health risk to Hg-sensitive groups (Yu et al., 2020). Recent studies have found that rice is also an important source of human MeHg exposure because of its high

* Corresponding authors.
E-mail addresses: maodian.liu@yale.edu (M. Liu), xjwang@urban.pku.edu.cn (X. Wang).

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intake, especially in certain Asian regions (Feng et al., 2008; Liu et al., 2019; Rothenberg et al., 2014; Zhang et al., 2010).

The Global Mercury Assessment 2018 suggested that the lack of good quality and nationally representative data in many geographical regions poses obstacles to reducing human Hg exposure (UNEP, 2019). Common edible crabs worldwide have high MeHg levels (Sunderland et al., 2018). For example, the red snow crab *Chionoecetes japonicus* in Japan has a mean MeHg level of 200 ng/g ww (wet weight basis) and a maximum MeHg level of 540 ng/g (Kakimoto et al., 2019). The king crab *Pseudocarcinus gigas* in Australia has a total Hg (THg, all forms of Hg) level of 220 ± 100 ng/g ww (Turoczy et al., 2001), compared to the Atlantic bluefin tuna (520 ± 30 ng/g ww, THg) and yellowfin tuna (200 ± 30 ng/g ww, THg) in the US (Burger and Gochfeld, 2011). Other studies have produced similar findings of total Hg levels, such as the red king crab (50 ± 24 ng/g ww) in Norway (Julshamn et al., 2015). In China, measurements of MeHg levels in crabs are relatively rare, besides the reported MeHg levels of 110 ± 76 ng/g ww for marine crabs and 53 ± 26 ng/g ww for freshwater crabs (Liu et al., 2020). Despite the high MeHg levels generally observed in crabs, Hg sources present in crabs have not been specified. Sediment is considered the dominant source of MeHg in crab biota because the geochemical conditions enable the sediment to be an active location for methylation via anaerobic bacteria (Benoit et al., 2003; Compeau and Bartha, 1985; Kwon et al., 2014). Water column MeHg levels have also been considered a predictor of biota MeHg, given their possible association with in situ MeHg formation (Buckman et al., 2021; Schartup et al., 2015a). Previous studies have utilized Hg isotope fractionation analysis and shown that the Hg sources in crabs are a combination of the water column, sediments, and unspecified external sources (Chandan et al., 2014; Balogh et al., 2015).

As reported in the *China Fishery Statistical Yearbook*, three crab species (the Chinese mitten crab, swimming crab, and mud crab) are domestically harvested in China, generating an annual production of 1.8 million Mg, 10 times that of sea bass (0.16 million Mg) or yellow croaker (0.18 million Mg), which are common marine fish species in China (Chinese Ministry of Agriculture (CMA, 2018). Crab production in China is concentrated in provinces with coastlines or major inland lakes, such as Jiangsu, with five provinces contributing 74% of the total production. Many previous studies regarding human MeHg exposure in China have not considered edible crabs as sources because of the relatively rare observations of Hg in crabs (Li et al., 2012; Ou et al., 2015; Zhang et al., 2010).

Here, we aimed to establish a national database of crab Hg concentrations, supplementing the lack of biomonitoring data for domestic crab species in China. Additionally, we depicted the temporal and spatial patterns of crab MeHg and THg levels, characterized the geographical origins of MeHg in crabs consumed by humans, and analyzed the regional changes from producing provinces to consuming provinces via interregional food trade. Chinese mitten crabs, swimming crabs, and mud crabs were quantitatively assessed for MeHg and THg concentrations. For these analyses, we assumed that all crabs were consumed domestically because the export rate was relatively low (0.59%) (CMA, 2018; UNSO, 2020). The geographical transport of crab MeHg and THg via trade was integrated with crab Hg output, cross-regional trade data, and the multi-regional input-output (MRIO) model. A life cycle-based analysis of crab Hg was conducted. MeHg accounts for more than 80% of the total Hg in living organisms; thus, the later discussion will generally focus on MeHg (Clarkson and Magos, 2006; Lavoie et al., 2013). The present study could be beneficial for assessing the contribution of edible crabs to human MeHg intake on a national scale and provide potential guidance for reducing the overall exposure levels.

2. Methods

2.1. Sample collection and preparation

Samples were collected from different regions in China from June to November 2018, the primary crab harvesting and consuming season in Asia. The sample sites (Fig. 1) were established along the southeast coast of China, which accounted for 82% of the total domestic crab production in 2017 (CMA, 2018). Only live crabs with credible origin information were collected. Samples were collected at multiple sites within a province to minimize the uncertainty caused by different water environments. In some low-production regions, it was difficult to collect samples from multiple sites, resulting in a small sample size. However, this did not influence the results substantially because such regions have a single harvesting environment and significantly lower crab production. The species considered in this analysis were the Chinese mitten crab (*Eriocheir sinensis*), swimming crab (*Portunus pelagicus*), and mud crab (*Scylla serrata*), comprising all documented crab species domestically harvested in China (CMA, 2018). The swimming crab and mud crab are marine crab species, and the Chinese mitten crab is a freshwater species. Crabs from wild and aquaculture environments were included in the present study. In 2017, crabs harvested from the wild and aquaculture accounted for 72% and 28% for marine species and 6% and 94% for freshwater species, respectively (CMA, 2018).

All samples were directly collected from the harvesting regions, delivered alive to the laboratory, and processed within 48 h. We documented live weight, body length (the longest horizontal width of the carapace), and sex. We opened the cephalothorax and removed the gills and intestine, retaining only the body muscle tissue and claw muscle tissue as the total edible parts of the crab (Andres et al., 2002; Turoczy et al., 2001). While we acknowledge that a certain portion of crab intestines is considered edible according to individual eating habits, such habits are not universal. Further, the Hg levels in crab intestines (3–5 ng/g ww) and its weight percentage in the entire body (13–16%) were low based on our experimental results (Supplementary Table S1). Therefore, we excluded intestines from the edible parts of crabs in the present study. The mass values of the total edible parts of each sample were documented for subsequent analysis of Hg intake. A total of 10 representative samples were separated into five body parts: body muscle, claw muscle, carapace (exoskeleton), gill, and intestine (including

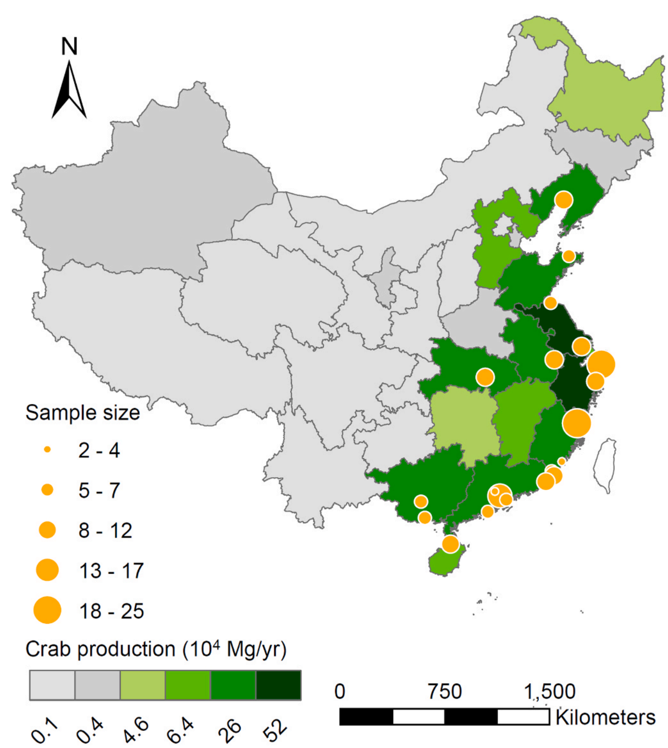

Fig. 1. Sample sites in different regions in the present study. The unit of sample sizes is the number of samples collected.

midgut gland, hepatopancreas, gonad, brain, and heart) to study the potential tissue variations in MeHg and THg distribution in crabs (Turoczy et al., 2001). All samples were placed in an oven at 60 °C to dry for an average of 4 h until a constant weight was achieved, and were then homogenized into an extra fine powder and stored in a laboratory freezer at − 20 °C until subsequent analysis (Liu et al., 2020).

2.2. Analytical methodology

MeHg and THg concentrations were measured in all samples. We measured the concentrations of THg using a Direct Mercury Analyzer 80 (DMA 80, Italy), as described previously (Barst et al., 2013; Maggi et al., 2009). MeHg was analyzed by methods described in the literature (Barst et al., 2013; Baumann et al., 2016; Mason et al., 2019). The homogenized samples were weighed and transferred to a water bath at 60 ± 2 °C for approximately 12–15 h with 4.5 N HNO_3 added for leaching. The extracted solutions were neutralized with 8 N KOH and buffered with 2 M CH_3COOH to a pH value of 4.7. To form MeEtHg, 30 μL of $NaBEt_4$ was added to the solution to protect the volatile product MeEtHg. The total volume of the final solution was 30 mL. After MeEtHg was purged and separated by gas chromatography, the MeHg concentration of the samples was determined using cold vapor atomic fluorescence spectrometry with a Tekran 2700 Analyzer (Tekran Instruments Corporation) (Li et al., 2016).

All chemicals were of reagent grade and used without further purification. The water used was deionized (resistivity ≥ 18.2 MΩ cm). Blanks and duplicates were processed for every sample set in the present study. The method blanks for MeHg and THg were 5.7 ± 0.28 pg/L and 0.038 ± 0.019 ng/g, respectively. The detection limits for MeHg and THg were 6.5 pg/L and 0.095 ng/g, respectively. The recoveries for ambient MeHg and THg were 107% ± 11% and 109% ± 7.0%, respectively. The detection limits of MeHg and THg were calculated using the mean concentrations of the method blanks plus triple standard deviations of the blanks, as described in the literature (Emmerton et al., 2013). THg was detectable in all samples. For MeHg analysis, only one intestinal sample of Chinese mitten crabs was below the detection limit. The concentrations were adjusted using individual standard spike recoveries. The final results of MeHg and THg in the crab samples are shown in Figs. 2 and 3. There were no outlier values in the present study,

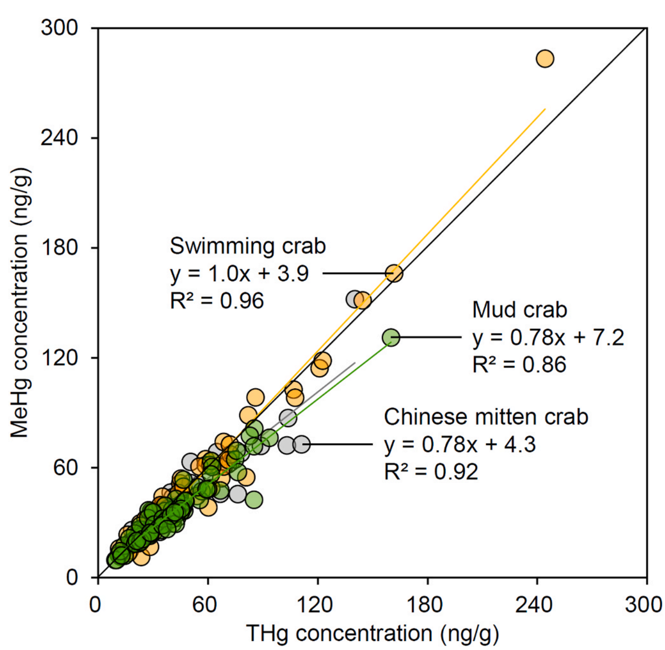

Fig. 2. Relationship between the concentration (wet weight basis) of Hg in three crab species.

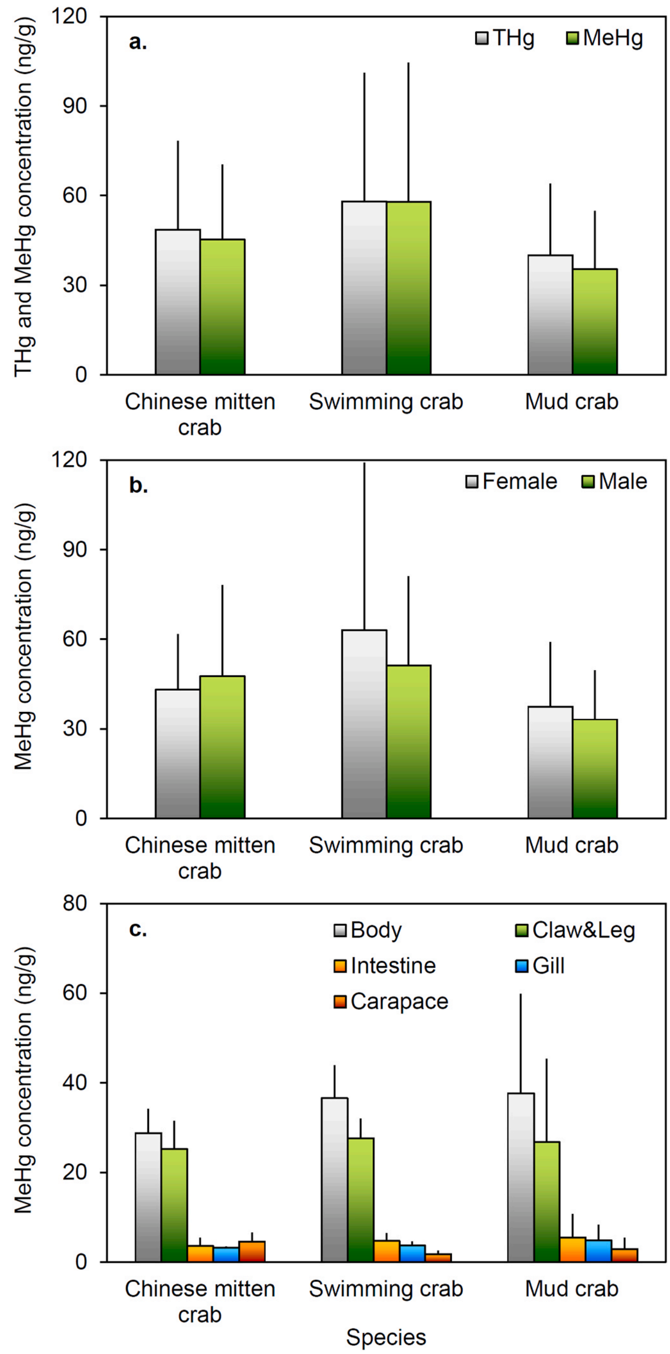

Fig. 3. Comparison of Hg levels in crabs. Panel a: Comparison of THg and MeHg in Chinese mitten crabs, swimming crabs, and mud crabs. Panel b: Comparison of MeHg levels in male and female crabs within species. The p-values (significance of correlation) for MeHg concentrations in female vs. male specimens for each crab species are 0.740 (Chinese mitten crab), 0.616 (swimming crab), and 0.262 (mud crab). Panel **c**: Comparison of MeHg levels in different body parts of crabs (body, claw and leg, intestine, gill, and carapace) within species. The p-values for MeHg concentrations in the body vs. intestine, gill, or carapace are all < 0.05 * .

indicating that no samples were found or included from localized polluted conditions (method: boxplot outlier in SPSS). The concentration data are presented in Supplementary Table S2. All concentration values mentioned in the present study were measured or converted to wet weight standards unless stated otherwise.

2.3. Biotransport of Hg via crab-related processes

We summarized the temporal and spatial patterns of annual MeHg output generated via domestic crab production in China. The crab production data were collected from the *China Fishery Statistical Yearbook* (CMA, 2018). The yearly output of MeHg was calculated as follows:

$$F_{jk} = P_{jk} \times C_{jk} \tag{1}$$

where, k stands for the crab species, Chinese mitten crab, swimming crab, or mud crab; j stands for the harvesting province; F_{jk} stands for the yearly output of MeHg (kg/yr) via the production of crab species k in province j; P_{jk} stands for the yearly production (kg/yr) of crab species k in province j; and C_{jk} stands for the MeHg concentration in crab species k of province j. The output is generated from domestic crab production because of a lack of data on imported crab products. We used provincial-level MeHg concentration values to generate MeHg output in the present study. In provinces with multiple water environments and crab production, we collected samples from different crab-producing regions within the province; however, we were unable to generate annual MeHg output with water environment-specific crab MeHg concentration values and used provincial averages instead, neglecting the MeHg differences possibly caused by water quality differences and other localized conditions. This was because the lowest level of crab production data in the *China Fishery Statistical Yearbook* is provincial, revealing no information on specific water environments. However, we assumed that the uncertainty was relatively low because there was no significant difference ($p > 0.05$) within one group of crabs categorized by species, feeding guild (wild or aquaculture), and province, as shown by our t-test results in SPSS.

The multi-regional input-output (MRIO) model has been used widely to understand the driving factors of environmental phenomena associated with interregional trade, such as carbon emissions (Lin et al., 2017), water footprint (Ewing et al., 2012), global biodiversity loss (Wilting et al., 2017), and MeHg exposure by food consumption (Liu et al., 2018). The MRIO model, as described in the literature, is a monetary multi-regional table describing product exchanges across regions, and for the present study, across Chinese provinces and regions (Lenzen et al., 2012; Liang et al., 2014; Lin et al., 2016). Here, we used the MRIO model to understand the redistribution of crab MeHg output via the interprovincial food trade and its influence on regional variations in human Hg intake. The mass flow analysis associated with MeHg and THg in Chinese domestic edible crab production was based on the total output monetary values of crab production in each province and the interprovincial trade of domestic crab production inferred from the MRIO model. We linked the crab MeHg and THg concentrations of each province with the MRIO table to calculate the crab Hg mass flow across provinces. The MRIO table does not have monetary flows specifically for crab species. Therefore, we estimated the flows for crabs in proportion to the monetary flows for the entire economic sector "Farming, Forestry, Animal Husbandry, and Fishery" in the Chinese MRIO table by the gross annual value of crab production to the gross annual value of the whole sector, assuming the flow for crabs presented the same pattern as the general sector. The calculation process was as follows:

$$NF_{ijk} = F_{ij} \times \frac{P_{ik}}{TP_i} \tag{2}$$

$$Per_{ijk} = \frac{NF_{ijk}}{\sum_j NF_{ijk}} \tag{3}$$

$$IF_{jk} = \sum_i (Per_{ijk} \times Pro_{ik} \times C_{ik}) \tag{4}$$

where, k stands for the crab category, marine or freshwater; i stands for the producing region (province or region of equivalent administrative unit); j stands for the consuming region; F_{ij} denotes the final demand (10^2 million \$/yr) of the economic sector "Farming, Forestry, Animal Husbandry, and Fishery" in region j from the producing region i, inferred from the Chinese MRIO table as described in the literature (Liu et al., 2014); NF_{ijk} denotes the final demand (10^2 million \$/yr) of the crab category k in region j from the supplying region i; P_{ik} denotes the total output monetary value (10^2 million \$/yr) of the crab category k in province i with data from *China Fishery Statistical Yearbook*; TP_i denotes the total output monetary values (10^2 million \$/yr) for the sector "Farming, Forestry, Animal Husbandry, and Fishery" in province i, inferred from *Chinese Agriculture Yearbook*; Per_{ijk} is the percentage (%) of the monetary output to province j in province i for the crab category k; IFjk denotes the MeHg or THg mass flow for crab category k via interregional food trade to province j; Pro_{ik} denotes the annual production (kg/yr) of crab category k in province i; and C_{ik} denotes the calculated MeHg concentration or THg concentration of the crab category k in province i.

A detailed depiction of the Chinese provinces and regions is shown in Supplementary Fig. S1. The complete interregional MeHg mass flow table is shown in Supplementary Figs. S2 and S3. Mass flow analysis allocates responsibilities and reveals Hg-related risks between producers (regions or individuals alike) and final consumers (Weinzettel et al., 2014). In the present study, we did not consider the export of crabs because the export rate was low (CMA, 2018; UNSO, 2020).

We summarized the mass flow of crab MeHg and THg based on the analysis of the life cycle of crabs to systematically depict the interlinked transport process of Hg. This helps to understand the specific environmental pathway of the Hg contained in crabs and the influence of crab production and trade on the regional distribution of exposure to MeHg. We depicted the process chain starting from crab harvesting (inland or coastal water), MeHg and THg biotransport via human commercial activities (local consumption or interprovincial food trade), MeHg and THg flow in different crab body parts (edible or inedible parts), human exposure to MeHg from consumption of edible parts, and environmental Hg intake as food waste from inedible parts. The analysis adhered to the mass balance principle, as follows:

$$\begin{aligned}
\sum_{jk} Inlandwater_{j,k} + \sum_{jk} Coastalocean_{j,k} &= \sum_{jk} Wild_{j,k} + \sum_{jk} Aquaculture_{j,k} \\
&= \sum_{jk} Localconsumption_{j,k} + \sum_{jk} Provincialtrade_{j,k} \\
&= \sum_{jk} Production_{i,j,k} \\
&= \sum_{jk} [M_{i,j,k} + \alpha_k \times I_{i,j,k}] + \sum_{j,k} [\beta_k \times I_{i,j,k} + G_{j,k} + C_{i,j,k}] \\
&= \sum_{jk} Edible_{j,k} + \sum_{jk} Foodwaste_{j,k}
\end{aligned} \tag{5}$$

where, i represents MeHg or THg; j represents the individual province or region of equivalent administrative unit; k represents the crab species; *M, I, G,* and *C* represent Hg production in separate crab body parts, meat (body and claw muscle tissue), intestine, gill, and carapace, as described in Section 2.1.; α_k represents the mass percentage of the edible part in the intestine in crab species k; and β_k represents the mass percentage of the inedible part in the intestine in crab species k.

2.4. MeHg exposure via crab intake

We calculated the probable daily intake (PDI) values and compared the results against the daily reference dose (RfD) specified by the USEPA to evaluate MeHg intake from domestic crabs under different dietary preferences of the Chinese population. The PDI and RfD values have been widely used in similar studies as effective tools for evaluating dietary MeHg intake (EPA, 1997; Li et al., 2012; Liu et al., 2020; Mason et al., 2019; Zhang et al., 2010). We divided the population into separate groups based on sex and age according to the *National Health Report*

(GASC, 2014) and calculated the corresponding PDI values with the MeHg concentrations and average body mass of the crab species as described in Section 2.1. and 2.2. The calculation process was as follows:

$$PDI_i = \frac{I_k \times C_k}{bw_i} \Big/ 1,000 \tag{6}$$

where, PDI_i is the per capita PDI value of MeHg (μg/kg of body weight per day) in group i, and I_k is the intake rate (g/day) of crab species k under the presumed intake preference.

To provide dietary suggestions and evaluate the potential risks of crab consumption, we designed different scenarios to generate dietary MeHg intake levels based on the crab choice and average mass value for the specific crab species, and compared the results with the RfD for MeHg specified by the EPA (1997). We modeled individuals in all groups ingesting one, two, or three crabs of one species daily, generating nine scenarios. The intake rates were calculated based on the modeled scenarios. C_k is the MeHg level (ng/g) of crab species k, and bw_i is the average body weight (kg) of individual in group i obtained from data from the *National Health Report* (GASC, 2014).

2.5. Statistical and uncertainty analysis

Statistical analysis was performed using IBM SPSS Statistics version 24 (IBM Corp., Chicago, IL, USA). Values are reported as mean ± standard deviation (SD). Two-sided t-tests were performed to compare the equality of means, and Levene's tests were performed to compare the equality of variances between groups. The significance level (p) was set to < 0.05 * and < 0.01 ** for all tests. The calculation of SD for each part of the mass flow analysis follows the functional correlation of the uncertainty propagation stated in the literature (Harvey, 2000):

$$S_{IF} = \sqrt{\sum_{i,k} (\alpha_{C_{ik}}^2 \times S_{C_{ik}}^2)} \tag{7}$$

where, k represents the crab species; i represents the province; S_{IF} represents the SD related to a certain intake value of THg or MeHg; C_{ik} represents the MeHg or THg concentration in crab species k in province i; α_{Cik} is the coefficient associated with C_{ik} in the calculation process of IF; and S_{Cik} is the SD related to it. During sample collection, samples could not be obtained from certain provinces with documented yet significantly low crab production. Estimation of the MeHg concentration in crabs in such provinces was made using the mean concentration of the same crab species in the same geographical region, as shown in Supplementary Fig. S1. In some provinces with low crab production, we collected qualified samples in low quantities, which may contribute to uncertainty. However, we estimate the uncertainty to be low since the sample need for such low-production and single water environment areas was naturally smaller. The coefficient of variation in the MRIO model described in Section 2.3 was quantified as 10% to cover the uncertainties of economic statistics and the input-output analysis (Lin et al., 2014).

3. Results

3.1. Hg concentration in crab products

MeHg was the primary form of Hg in all three species studied (Fig. 2), which was consistent with previous findings (Harris et al., 2003; Storelli et al., 2002). Swimming crabs had a MeHg/THg percentage of 99.6%, which was higher than that of mud crabs (88.4%) and Chinese mitten crabs (93.2%). Swimming crabs also had the highest MeHg concentration (57.9 ± 46.6 ng/g ww) compared to mud crabs (35.4 ± 19.4 ng/g) and Chinese mitten crabs (45.3 ± 25.1 ng/g, Fig. 3a). The variation in MeHg levels in swimming crabs was large, with a maximum value of 283 ng/g comparable to marine fish such as black sea bass in the US (125 ± 79 ng/g) (FDA, 2017). Specimens with MeHg concentrations > 200 ng/g originated mainly from Fujian, a province primarily producing wild marine crabs. The higher MeHg levels in the marine swimming crab might be attributed to the fact that its primary living habitat, the coastal ocean, is the predominant area of MeHg production (Schartup et al., 2015b). Additionally, swimming crabs that are sold in markets are primarily wild-caught (81.2% in 2017) (CMA, 2018) and are thus associated with a more complex food web, generally accumulating more MeHg than farmed species (Liu et al., 2018). In addition, regional water pollution can affect the crab MeHg levels. The northwestern Bohai Sea coast in northeastern China suffered Hg contamination from long-term metal smelting, resulting in elevated MeHg levels in the local water, sediment, and hydrophytes (Wang et al., 2009). Liaoning, within the polluted region, is a major crab-producing province. Unlike previous studies that commonly indicated high Hg levels in only marine species, MeHg levels in both freshwater and marine crab species in China are comparable to those of dominant commercially available marine fish, such as hairtail (76 ± 77 ng/g), yellow croaker (51 ± 38 ng/g), and grouper (26 ± 7.4 ng/g), and substantially higher than freshwater fish such as grass carp (4.3 ± 1.3 ng/g) (Table 1) (Liu et al., 2020; Zhang et al., 2010).

Sex appeared to be irrelevant for MeHg levels in all three species (Fig. 3b), whereas MeHg levels differed significantly in different crab body parts (Fig. 3c). Evaluation of the tissue-specific distribution of MeHg and other pollutants is important when estimating the potential risk of dietary intake. For example, arsenic-contaminated crabs may not pose as high a risk as Hg-contaminated crabs because the majority of arsenic accumulates in crab intestines, which are generally considered inedible as per human dietary habits (Julshamn et al., 2013, 2015). In the present study, MeHg had elevated concentration levels in the muscle tissues of the crab body, claw, and leg compared to other body parts (intestine, gill, carapace, $p < 0.01$ **). This is consistent with previous studies on the blue crab *Callinectes sapidus* in Florida, USA (Adams and Engel, 2014), *Carcinus maenas* in Portugal (Costa et al., 2011), and *Pseudocarcinus gigas* in Australia (Turoczy et al., 2001). This is partly because MeHg is incorporated into large peptides or proteins after entering the crab body, and readily accumulates in muscle tissues (Amlund et al., 2007). Other heavy metals, such as copper, zinc, and cadmium, generally bond with membranes or lipids inside living organisms and therefore accumulate mainly in the hepatopancreas, which contains high levels of metallothioneins (Karouna-Renier et al., 2007; Reichmuth et al., 2010). MeHg accumulates primarily in the edible parts of crabs. Hence, dietary intake serves as an important pathway of MeHg exposure, and a better-quantified evaluation should be provided.

3.2. Spatial and temporal variations of crab Hg output

Fig. 4 shows the annual variation in MeHg output from edible crab production, which exhibited an increasing trend from 2003 to 2015, with a minor decrease from 2015 to 2017 (Fig. 4a); and provincial production and variations in 2017 (Fig. 4b). Rapid economic development and diversification of the daily dietary structure for humans have led to an increase in crab production in China. In 2014, the total crab production of the three species reached 1.8×10^6 Mg, which was 50.6% higher than 2007 (CMA, 2018). The predominant freshwater crab-producing province, Hubei, had an annual production growth of 2.3×10^4 Mg/yr (mean, 2011–2018) and a maximum increase rate of 37% in 2014 (CMA, 2018). Therefore, the total crab MeHg output and related exposure risk increased correspondingly. However, the crab MeHg output reached a plateau from 2015 to 2016, followed by a minor decrease in 2017, which was primarily affected by national environmental protection policies in China (see the Discussion section).

In terms of spatial distribution, the crab MeHg output from southeastern coastal provinces was significantly higher than that in other inland regions. Jiangsu contributed 7.3 kg of MeHg in 2017, followed by Anhui, Fujian, and Zhejiang (4.2 kg, 4.0 kg, and 3.2 kg, respectively).

Table 1
MeHg and THg levels in different species in different countries.

Species Name	Category	Sample size (n)	THg (ng/g, ww[a])		MeHg (ng/g, ww)		Country	Refs.
			Mean	SD[b]	Mean	SD		
Yellowfin tuna	*Marine fish*	45	200	170	nd[c]	nd	USA	(Burger and Gochfeld, 2011)
Black sea bass	*Marine fish*	19	160	70	nd	nd	USA	(Burger and Gochfeld, 2011)
Atlantic croaker	*Marine fish*	63	120	70	nd	nd	USA	(Burger and Gochfeld, 2011)
The king crab	*Marine crustacean*	15	1100 (dw[d])	500	nd	nd	Australia	(Turoczy et al., 2001)
Red cod	*Marine fish*	6	nd	nd	149	79	New Zealand	(Sadhu et al., 2015)
Blue cod	*Marine fish*	7	nd	nd	74	30	New Zealand	(Sadhu et al., 2015)
Bluefin tuna	*Marine fish*	13	404	116	103	79	South Korea	(Park et al., 2011)
Cod	*Marine fish*	7	72	48	73	47	South Korea	(Park et al., 2011)
Red snow crab	*Marine crustacean*	39	210	nd	200	nd	Japan	(Kakimoto et al., 2019)
Hairtail	*Marine fish*	29	73	67	76	77	China	(Liu et al., 2020)
Yellow croaker	*Marine fish*	30	55	36	51	38	China	(Liu et al., 2020)
Grouper	*Marine fish*	12	31	11	26	7.4	China	(Liu et al., 2020)
Tilapia	*Freshwater fish*	11	6.5	0.77	4.0	1.1	China	(Liu et al., 2020)
Grass carp	*Freshwater fish*	21	6.2	1.4	4.3	1.3	China	(Liu et al., 2020)
Swimming crab	*Marine crustacean*	51	58	43	58	46	China	This study
Mud crab	*Marine crustacean*	82	40	24	35	19	China	This study
Chinese mitten crab	*Freshwater crustacean*	40	48	29	45	25	China	This study

Note: ww, wet weight basis; SD, standard deviation; nd, no data; dw, dry weight basis
Figure captions.

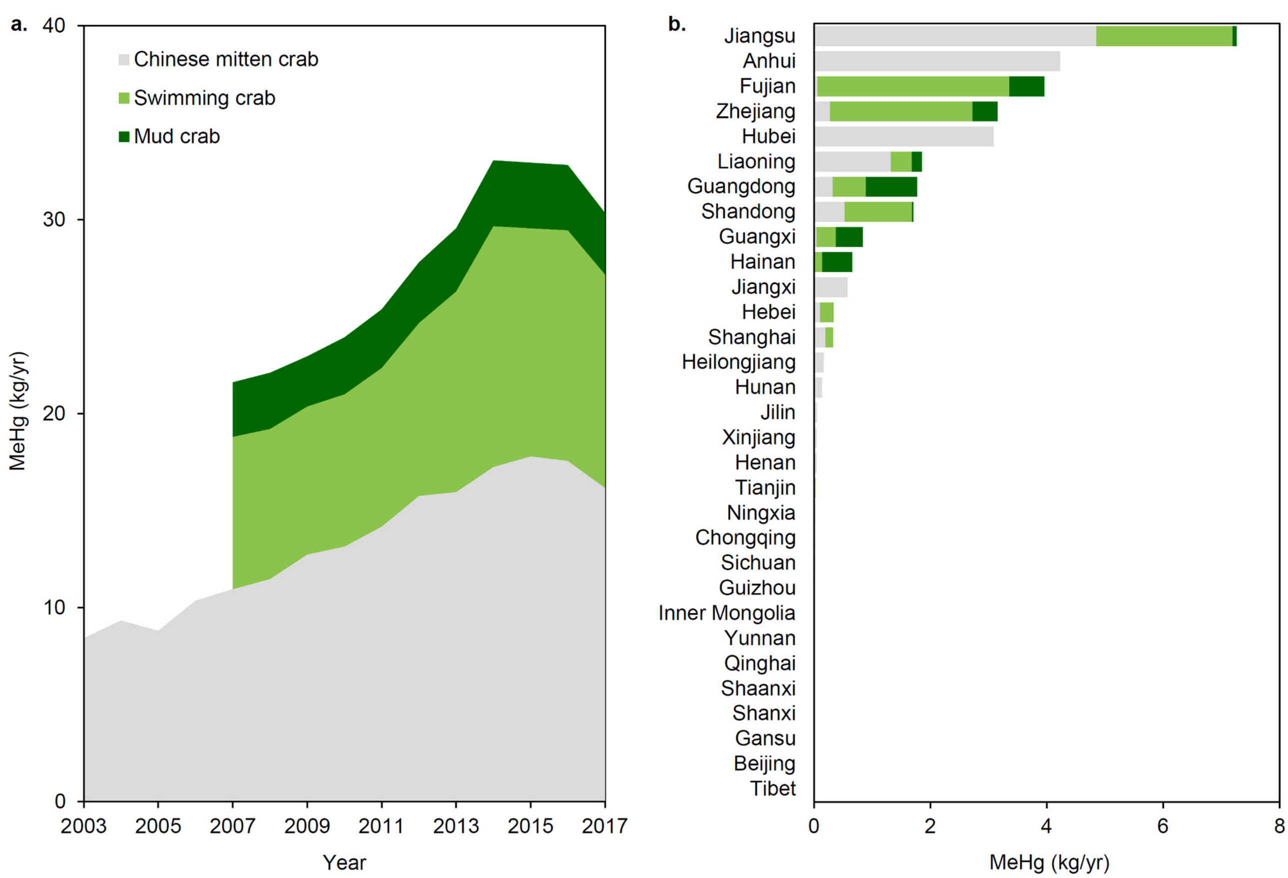

Fig. 4. Temporal and spatial patterns of MeHg generated via crab production in China. Panel a: The temporal pattern of MeHg output through the production of Chinese mitten crabs, swimming crabs, and mud crabs in China from 2003 to 2017. The inconsecutive data from 2003 to 2007 was due to lack of crab production data for swimming crabs and mud crabs in the China Fishery Statistical Yearbook for the indicated period. Panel **b**: Crab MeHg production in each provincial region of China in 2017.

The individual crab MeHg contributions from each province exhibited significant variations. The two major marine crab-producing provinces, Zhejiang and Fujian, contributed 48% of the total marine crab MeHg output in 2017, while Jiangsu and Anhui contributed 56% of the total freshwater crab MeHg output. In addition to coastal regions, Hubei, an inland province with multiple freshwater lakes, contributed 19% of the total freshwater crab MeHg output. The crab MeHg output of Anhui, Hubei, and coastal provinces in China reached 29 kg, 96% of the total crab MeHg output in 2017. The contribution of the other inland provinces was negligible. This is because the crab-producing areas are geographically concentrated along the southeast coastal provinces for marine species and inland provinces with large open lakes for freshwater species. Overall, the top five crab-producing provinces (Jiangsu, Anhui, Fujian, Zhejiang, and Hubei) accounted for 71% of the total crab MeHg production in China in 2017.

3.3. Biotransport of Hg associated with crab and potential human exposure

Our study found that cross-regional trade had a greater impact on the final consumption amount of crab MeHg in different provinces compared to the amount of crab produced. In 2017, 53% of crab-derived MeHg came from the inland freshwater environment, while the remainder was from the coastal marine environment (Fig. 5a). A total of 64% of the total crab MeHg output came from aquaculture (94%, 53%, and 19% from Chinese mitten crab, mud crab, and swimming crab, respectively), and only 36% was from wild capture (Fig. 5a). This finding indicates that farmed crabs, particularly freshwater species, are the dominant pathway for human MeHg intake via crab consumption in China. Therefore, controlling the feeding of aquacultured crabs and

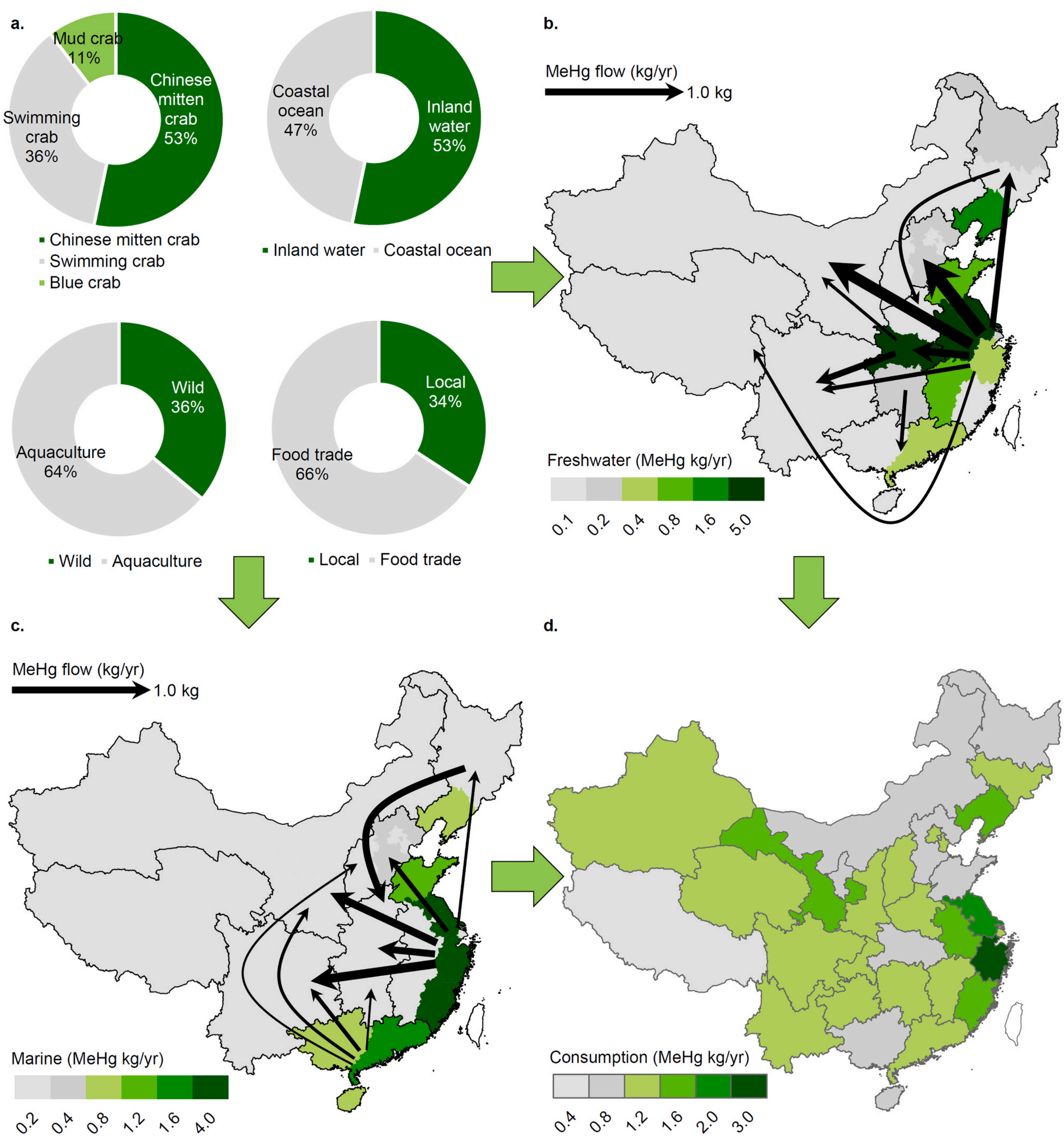

Fig. 5. Panels b and c: the modeled mass flow of freshwater and marine crab MeHg respectively, driven by the interregional food trade in China. Each arrow indicates one mass flow between two geographical areas in China (Northern China, Eastern China, Southern China, Central China, Northeast China, and Southeast China, Supplementary Fig. S1). The blackline bordered region where the starting point of the arrow falls indicates the production area, whereas the ending point indicates the final consumption area. The width of each arrow represents the relative amount of each mass flow. Complete data for provincial crab MeHg mass flows can be found in Supplementary Fig. S2 and S3. Panel d: The final crab MeHg consumption in each provincial region of China. Breakdown of crab MeHg exposure in China and its modeled MeHg mass flow. Panel a: Contribution of different source pathways of crab MeHg in China.

water quality management might be beneficial in reducing the MeHg levels inside crab tissues, and thus the overall MeHg exposure from crab consumption.

In 2017, 66% of the total crab MeHg output was redistributed outside the producing region, while only 34% was consumed locally (Fig. 5a–c). The final mean value of consumption of crab MeHg was 1.0 ± 0.4 kg/yr per province, whereas the average production was 1.0 ± 1.7 kg/yr, with a SD of 4.2 times the consumption. Therefore, cross-regional trade had an averaging effect on the provincial crab MeHg distribution during the consumption stage compared to the large variations in the primary production stage. The major crab-producing regions Zhejiang (2.5 kg MeHg), Jiangsu (1.7 kg MeHg), Liaoning (1.5 kg MeHg), and Fujian (1.4 kg MeHg) face the highest risk of MeHg exposure in terms of total consumption amount. Nonetheless, a considerable amount of crab MeHg was redistributed to other regions via the food trade. Crab MeHg was redistributed from the eastern China region to the northwestern and northern China regions for freshwater species (Fig. 5b) and coastal to inland regions for marine species (Fig. 5c). The final crab MeHg consumption in Beijing came solely from interregional trade input, reaching a value of 1.1 kg/yr, which was above the national average. The population of Beijing is 20 million (NBS, 2018), which is only 1.4% of the national population. Therefore, its per capita consumption is significantly higher than that of most other regions. Despite crab-producing being concentrated in a few regions, crabs are a popular food choice, can be consumed nationwide via the food trade, and pose the risk of exposure to a wider population (Fig. 5d). This is commonly found in non-crab-producing regions, such as Shanghai (1.2 kg/yr MeHg, 72% trade input). Overall, we found that regions with high gross domestic product levels face higher per capita crab MeHg exposure than less developed regions ($p < 0.05$ *, $p = 0.011$). Complete data for each

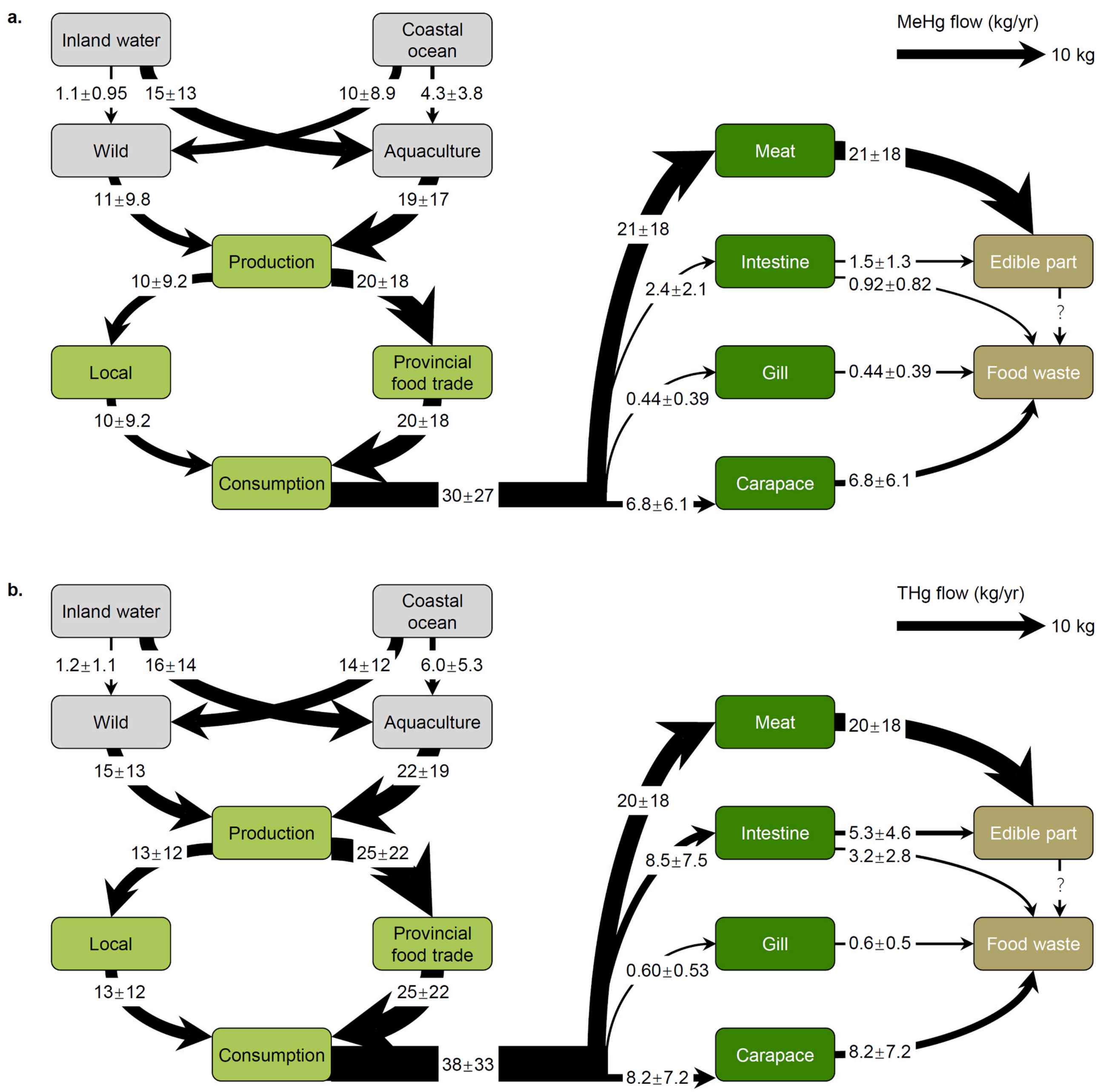

Fig. 6. The biotransport process of MeHg (Panel a) and THg (Panel b) through the production, trade, supply, and consumption of edible parts of crabs and through food waste management of crab residues. All bar labels indicate the MeHg or THg in crabs associated with the labeled criteria. The top-left bar labels "inland water" and "coastal oceans" indicate the MeHg content in crabs originating from inland water environment and coastal oceans, respectively.

interprovincial MeHg and THg flow can be found in Supplementary Figs. S2 and S3.

To understand the potential health risks posed by crab consumption, we calculated the PDI values for different population groups under assumed scenarios and compared the results with the RfD for MeHg (0.1 μg/kg bw/day) suggested by the EPA (EPA, 1997). According to our calculation, the PDI values of MeHg were 0.140 ± 0.015 and 0.094 ± 0.010 μg/kg bw/day when consuming one swimming crab and mud crab daily, respectively, for adults aged 20–60 years in China. These values exceeded the RfD. The PDI value of adults consuming one Chinese mitten crab daily was 0.059 ± 0.006 μg/kg bw/day, because the species had a smaller body mass than the other two species. Nonetheless, the PDI still reached 60% of the RfD, which was high enough to attract attention considering coexposure from other daily food sources. For sensitive groups such as pregnant women aged 20–34 years, the PDI values were 0.170 ± 0.003, 0.100 ± 0.002, and 0.070 ± 0.001 μg/kg bw/day when consuming one swimming crab, one mud crab, and one Chinese mitten crab daily, respectively. The above data considered only MeHg intake from crab consumption, neglecting exposure from other common food choices, such as rice and marine fish. Thus, crab MeHg should be considered in MeHg exposure assessments, especially for Hg-sensitive groups.

After production and food trade, most of the crab MeHg was consumed by humans via their dietary intake (Fig. 6). According to human dietary habits, at least 22.5 kg/yr of MeHg contained in crab body, claw, and leg tissues was consumed via dietary intake, accounting for 75% of total crab MeHg production. The remaining 8.2 kg/yr of crab MeHg output was treated as food waste and transported for waste disposal via landfills, incineration, and composting. Owing to its small amount, its environmental impact was neglected.

4. Discussion

Previous studies have considered marine fish as the predominant source of human MeHg intake globally (Bradley et al., 2017; Liu et al., 2018; Stein et al., 1996; Sunderland, 2007). Our findings showed that crabs might be a significant dietary source of human MeHg intake. MeHg levels in the three domestic crab species in China were relatively high compared to those in common marine fish in Asia, such as hairtail and yellow croaker, and higher than freshwater fish species, such as tilapia and grass carp (Table 1). In addition to high Hg levels in crabs, total crab consumption was also competitive with that of common fish species. In 2017, 1.8×10^6 Mg of domestic crabs is consumed, which is higher than hairtail (1.0×10^6 Mg), the marine fish species with the highest consumption in China. Crabs are not consumed as frequently and commonly as fish, indicating that the risk of exposure to MeHg from crabs is unevenly distributed across regions and periods. Particularly, in major crab-producing provinces with a large fishery production, such as Jiangsu, Anhui, and Zhejiang, the diet of the local population would rely heavily on fish and crabs, potentially causing high coexposure risks, especially in autumn, which is the crab market season. Moreover, dietary habits would play a significant role in crab MeHg exposure since as a non-staple food, the individual habits can diversify. In Zhejiang, the annual crab consumption is 13% of the total fish consumption (Yu et al., 2020). Given the MeHg levels in crabs are comparable to marine fish and significantly higher than freshwater fish species (Table 1), its contribution to human MeHg exposure is not negligible. However, little attention has been directed toward the MeHg contribution of crab species. In summary, the high MeHg levels in crabs, combined with their high consumption amount in the diet, should be considered in the risk assessment of Hg intake, as assessed for other seafood such as fish.

The crab Hg output in China has decreased marginally since 2015, after a decade of rapid increase. Hg emission in China has exhibited no significant changes in the last decade (Wu et al., 2016). Given the slow accumulation mechanism for Hg content in terrestrial Hg pools, Hg levels in crabs may not have increased significantly over the studied period (Smith-Downey et al., 2010). Therefore, the annual crab production serves as the driving factor of changes in crab Hg output, influenced by the market, weather, and local fishery policies. We suggest that in the future, the crab Hg output may start increasing again because the current crab production does not fulfill market demand. The recent decline in supply was due to the implementation of environmental policies (specifically the prohibition of farm net enclosures for inland water and closed fishing periods for estuarine and marine water), rather than a natural response to the decline in market demand. To restore the ecological balance in the local water environment heavily exploited for crab farming, Jiangsu Province released the *Notice on Removal of Net Enclosures in Lake Tai*, enforcing the full removal of crab farming nets in the area (SMG, 2018). Such policies significantly affect freshwater crab production, leading to a 7.9% reduction in 2017. However, it would not negatively affect the industry as it moved from aquaculture to wild capture (CMA, 2018). The earlier significant increase in crab Hg production was driven by market demand, which was not hampered by reducing crab production caused by national policies. Therefore, the actual market demand is higher than the current supply. There may be an increase in crab Hg consumption in the future; thus, crab Hg might be an important pathway for human MeHg exposure and requires further quantitative assessment.

The market season of fresh edible crabs is from July to November in Asia, and the total output of crab MeHg is primarily consumed by the population within this relatively concentrated time of the year. Such features indicate that people, especially Hg-sensitive groups, may experience intense short-term MeHg exposure in autumn via the daily dietary consumption of crabs. This may explain the high MeHg levels observed in human blood and hair in autumn and winter in China (Du et al., 2020). Thus, the exposure risk during the indicated market season might be underestimated for the general population. The mass value of edible parts in crabs was 110–220 g/crab for all three species considered in the present study. The PDI resulting from the consumption of one crab and rice and fish every day is 170% of the RfD specified by the EPA (CHNS, 2011; EPA, 1997; Gong et al., 2018; Liu et al., 2018). Such results are alarming for pregnant women during the crab market season, i.e., autumn, when the population may consume crabs daily. For women aged 20–34 years, the PDI value of one Chinese mitten crab (0.070 ± 0.001 μg/kg bw/day), one swimming crab (0.170 ± 0.003 μg/kg bw/day), or one mud crab (0.100 ± 0.002 μg/kg bw/day) is alarming given the RfD and potential coexposure from rice and fish (EPA, 1997; Zhang et al., 2010). Therefore, we suggest that children and pregnant women limit their crab consumption. Individual dietary differences should be considered in future studies. For example, populations in coastal provinces face a higher risk of coexposure due to the high consumption of seafood products and crabs during the market season. Additionally, some individuals consume crab intestines, which may cause coexposure to multiple heavy metals, such as arsenic, cadmium, and copper (Adams and Engel, 2014; Turoczy et al., 2001). Practical advice includes but is not limited to occasional crab consumption instead of daily consumption, small-sized crabs compared to large-sized crabs, sharing with the family instead of one crab each, and limiting fish consumption when already consuming crabs in a day's diet. Further studies should also explore the seasonal fluctuations in exposure to crab MeHg and other seasonal food sources to characterize and avoid the health risks of potential short-term MeHg intake.

In the present study, the interregional MeHg flow highlighted the displacement of crab MeHg from producing regions to non-producing regions; specifically, 66% of crab MeHg was redistributed outside the original crab-producing province. As per our trade analysis, special warnings should be provided to the public. First, for coastal provinces with high fishery and crab production, the coexposure risks the population faces from constant consumption of fish and other water products, together with possible daily crab consumption during the market season, are significantly high, and thus should be paid special attention, especially by pregnant women. Second, the inland populations with substantial crab MeHg imports, specifically Gansu and Beijing (Fig. 5), are generally not as informed as the coastal population about the high MeHg content in water products and its adverse health effects. Therefore, special warnings should be given to such populations. Human trade activities significantly influence the transport process of contaminants such as MeHg from sources to sinks, essentially reshaping their environmental pathways and eventual outcome in large quantities and over broad geographical regions. The government and related agencies should analyze the risk of emerging food selections via food imports and inform the local population of potential adverse health effects. Trade-influenced biotransport processes should also be considered in future studies.

As the first attempt to study annual MeHg output and its biotransport process in crabs, we provided a spatially specified depiction of the process chain from production, trade, and consumption to final exposure, and broadened the limited scope of previous studies that focused only on the consumption and exposure stage. The present study characterized the life cycle of crab Hg and is beneficial for targeting the specific transport stage to lower both regional and cross-regional health risks related to crab MeHg exposure. The present study has certain limitations. For the temporal analysis of annual crab MeHg and THg output, the measured Hg concentration values in crabs in 2018 were used to estimate the total Hg output from the previous year, which may contribute to uncertainty. Given the slow-changing nature of Hg emissions in China over the period and slow temporal accumulation trends of Hg content in water, soils, and biological tissues, we estimated the uncertainty to be low (Li et al., 2015; Wu et al., 2016; Zhou et al., 2018). We also estimated that the increasing trend presented in Fig. 4 would remain unchanged if the previous concentration data were available. The monetary flow data utilized in the MRIO model in the present study were derived from the "Farming, Forestry, Animal Husbandry, and Fishery" sector in the Chinese MRIO table, which might increase the uncertainty because this data were not species-specific, and thus caused uncertainties when using the model to depict crab MeHg mass flows across regions. However, the errors in estimating the crab commercial data converted from the sector in proportion to the total output rates were low (Lin et al., 2014). Additionally, when quantifying the total annual consumption of crab MeHg, we assumed that all crabs were consumed domestically and did not consider international exports, because there are no specific data on Chinese crab exports from national authorities. However, the export percentage was estimated to be only 0.59% of the production, and thus the error is very low (CMA, 2018; UNSO, 2020).

We suggest that future MeHg studies should examine other species apart from the frequently studied species of marine fish, shellfish, and marine mammals; shed light on food sources such as crabs, freshwater products, and rice to evaluate the coexposure risk due to human dietary preferences; and quantify the general MeHg intake accurately. Shrimp (marine), prawns (freshwater), and crabs are common edible crustacean species, accounting for 41%, 33%, and 27%, respectively, of the total crustacean production in China in 2017 (CMA, 2018). The MeHg levels in shrimps and prawns in China are 9–33 ng/g ww and < 2–15 ng/g ww, respectively, which is substantially lower than our results in crabs (Yu et al., 2020). However, given the high production of shrimps and prawns in China, the total MeHg output from edible shrimps and prawns and the effect of their coexposure should be explored in the future. For example, the production and consumption of crayfish, Procambarus clarki, has increased in recent years, and this species is widely associated with high concentrations of heavy metals (Peng et al., 2016). Food choices with high MeHg levels should also be considered. As stated in the Global Mercury Assessment 2018, high-quality and nationally representative data are lacking in many geographical regions, hampering our understanding of the risk of human Hg exposure (UNEP, 2019). Our study promotes this goal by providing Hg biomonitoring data on a national scale, and the related production-trade-consumption-exposure analysis helps gauge changes in human Hg exposure in temporal and spatial settings. We suggest that fish, crabs, and rice should be considered in future studies to predict the MeHg exposure levels more accurately and provide better guidance regarding food safety, especially for Hg-sensitive populations.

5. Conclusions

In summary, we found that the MeHg and THg levels in edible crabs were relatively high and comparable to those in marine fish at high trophic levels. Crab MeHg production is geographically concentrated, and five provinces (Jiangsu, Anhui, Fujian, Zhejiang, and Hubei) account for 71% of the total crab MeHg production in China. However, human MeHg exposure via crab consumption can be substantially displaced from producing regions to consuming regions via interregional food trade, posing health risks to the general population on a national scale. Given human dietary habits and the average size of edible crabs, MeHg exposure from crab consumption can be high, especially during the market season. The MeHg exposure from one crab consumption can easily exceed the daily RfD specified by the USEPA, considering the collective influence of MeHg from other food sources, such as fish and rice; thus, the exposure risk should be highlighted. We suggest that the Hg-sensitive population, particularly children and pregnant women, should be attentive to their consumption of crabs. Finally, we suggest that future studies should explore the collective impact of fish and other dietary choices of significant importance, such as crabs, rice, and other food options, and emphasize the influence of interregional food trade on Hg exposure in the human population.

CRediT authorship contribution statement

Zhihao Zhang: Conceptualization, Methodology, Investigation, Data curation, Writing - original draft. **Long Chen:** Methodology, Investigation, Data curation. **Menghan Cheng:** Investigation. **Maodian Liu:** Conceptualization, Methodology, Visualization, Writing - review & editing, Supervision. **Xuejun Wang:** Validation, Writing - review & editing, Supervision, Project administration.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2021.125684.

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