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Evaluation on the biomagnification or biodilution of trace metals in global marine food webs by meta-analysis[☆]

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ABSTRACT

The transmission and accumulation of trace metals in marine food webs have a profound influence on the structure and function of marine environment. In order to quantitatively assess the trophic transfer behaviors of eight common metals (As, Cd, Cr, Cu, Hg, Ni, Pb and Zn) in simplified five-trophic level marine food webs, a total of 9929 biological samples from 61 studies published between 2000 and 2019, involving 154 sampling sites of 33 countries/regions, were re-compiled using meta-analysis. Based on concentration-trophic level weighted linear regression and predator/prey comparison, the food web magnification factor (FWMF) and the biomagnification factor (BMF) were calculated, respectively. The results showed dissimilar trophic transfer behaviors of these metals in global marine food webs, in which As and Ni tended to be efficiently biodiluted with increasing trophic levels (FWMFs < 1, $p < 0.01$), while Hg, Pb and Zn trophically biomagnified (FWMFs > 1, $p < 0.05$). However, Cd, Cr and Cu presented no biomagnification or biodilution trend ($p > 0.05$). The values of FWMFs were ranked as: Hg (2.01) > Pb (1.81) > Zn (1.15) > Cu (1.13) > Cr (0.951) > Cd (0.850) > Ni (0.731) > As (0.494). In terms of specific predator-prey relationship, Pb showed significant biodilution from tertiary consumers (TC) to top predators (BMF < 1, $p < 0.05$), whereas Cd and Cu displayed obvious biomagnification from primary consumers (PC) to secondary consumers (SC) (BMFs >1, $p < 0.05$). Additionally, when Cu and Zn were transferred from SC to TC, and primary producers to PC, clear biodilution and biomagnification effects were observed, respectively ($p < 0.05$). Further analysis indicated that the average concentration of Hg in five-trophic level marine food webs of developed countries (0.904 mg kg^{-1} dw) was more noticeable ($p < 0.05$) than that of developing countries (0.549 mg kg^{-1} dw).

1. Introduction

Marine heavy metal pollution is an increasingly prominent environmental problem (Valavanidis and Vlachogianni, 2010). Marine environment is more likely to be depository site of heavy metals, as heavy metals can be poured into the sea in various ways, such as industrial effluents, atmospheric deposition and mining operations (Radhalakshmi et al., 2014). In marine environment, arsenic (As, metalloid element), cadmium (Cd), chromium (Cr), copper (Cu), mercury (Hg), nickel (Ni), lead (Pb) and zinc (Zn) are common trace metals with different bio-toxic levels. Among them, Cr, Cu, Ni and Zn are essential elements to construct bio-macromolecules (e.g. Cu/Zn superoxide dismutase), while As, Cd, Hg and Pb are non-essential elements with strong toxicity at low concentrations (Ishaque et al., 2006). Except for As (toxicity reduced by conversing inorganic As into arsenobetaine (AsB)), organisms are generally lack of effective metabolic mechanisms to

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handle these metals which sometimes can be converted into more toxic compounds (e.g. methylmercury (MeHg)) and transferred through food webs (Ubillús et al., 2000; Caumette et al., 2012). More seriously, toxicity-enhancement (synergism) for the mixture of trace metals was evidenced, posing a great threat to marine environment (Ishaque et al., 2006). In marine environment, dietary uptake is the main pathway of trace metal accumulations (Wang and Rainbow, 2000), with food webs serving as the vehicle (Moloney et al., 2011). Therefore, analyses on the trophic transfer rules of trace metals in marine food webs are of great significance to habitat protection and pollution recovery.

Recent researches related to the trophic transfer behaviors of trace metals have been generally conducted in a given sea area because of time and geographical constraints. However, the marine trophic relationships are extremely complicated and various, and the accumulation of trace metals is also influenced by many factors, such as community structure, environment condition and temporal-spatial differences (Friant and Koerner, 1981; Weng and Wang, 2014; Sakata et al., 2015), which bring great challenges to obtain a highly representative result about whether trace metal is trophically biomagnified or biodiluted in global marine food webs. Fortunately, the pioneering application of meta-analysis in the field of large-scale ecology research provides the possibility to explore this issue (Gurevitch and Hedges, 1993).

Meta-analysis is a very powerful tool to obtain scientific evidences by giving different weights to independent experiments with the same topic and combining them quantitatively, which not only synthesizes relevant studies, but also reduces the impact of individual study on final conclusions (Gurevitch and Hedges, 1993). The meta-analytic method contributed a lot to explore the trophic transfer behaviors of metals in specific food webs. Rowan and Rasmussen (1994) meta-analyzed 38 marine studies documenting the changes of radioactive cesium (^{137}Cs) concentrations with the increase of trophic levels, indicating their positive correlation. A total of 8 studies on the concentrations of Cd in herbivorous and carnivorous animals were meta-analyzed, showing that there was a significant biomagnification of Cd in the transmission from herbivorous to carnivorous animals (Veltman et al., 2007). Lavoie et al. (2013) meta-analyzed 69 studies (17 marine studies involving 33 sample sites) related to the concentrations of Hg in different types of food webs and trophic organisms, identifying the near-universal biomagnification effect of Hg. Li et al. (2015) demonstrated that most aquatic plants could significantly bioaccumulated Cd, Cu and Zn from water, using meta-analysis based on 15 studies on the absorption capacity of aquatic plants for heavy metals. However, there is still lack of global-scale meta-analysis-based study on the trophic transfer behaviors of multiple metals in marine food webs. Based on the reliability and robustness, therefore, this meta-analysis was performed aiming to (1) determine whether biomagnification or biodilution of As, Cd, Cr, Cu, Hg, Ni, Pb and Zn occurred in global marine food webs; (2) evaluate the trophic transfer behaviors of these metals between two adjacent trophic levels; and (3) examine the concentration differences of these metals in marine food webs of developed and developing countries.

2. Materials and methods

2.1. Protocol registration and literature collection

This meta-analysis was conducted following the Preferred Reporting Items in Systematic Reviews and Meta-Analyses (PRISMA) guidelines (Liberati et al., 2009). The review protocol was prospectively registered in International Prospective Register of Systematic Reviews (PROSPERO), with the registration number of CRD42019137091.

Referring to previous meta-analytic studies (Kozlowsky-Suzuki et al., 2012; Lavoie et al., 2013; Govers et al., 2014), various databases including *the Cochrane Library*, *Web of Science* and *Medline* were searched using the key terms of *heavy metal*, *trace metal*, *marine organism*, *marine foodweb*, *marine food web*, *marine food chain*, *biomagnification*, *biodilution*, *bioaccumulation*, identifying potential literature documenting metal concentrations in marine organisms. Gray literature (e.g. conference papers, dissertations, references listed, etc.) was also searched via *Index to Scientific and Technical Proceedings* and *Baidu Scholar*. In order to avoid the influence of man-made and temporal variations, and truly reflect the trophic transfer behaviors of metals nearly the past 20 years, this meta-analysis only included field marine sampling studies published from 2000. Two researchers independently reviewed and screened the retrieved papers. Disagreements were resolved by discussion or consulting a third researcher. After final screening, a total of 61 high quality studies (see Supporting *Information*, Tables S1 and S2), involving 154 sampling sites of 33 countries/regions (Fig. 1), were included in this meta-analysis.

2.2. Trophic structure of marine food webs

To facilitate the construction of marine food webs, a simplified model of five-trophic level marine food webs was established by categorizing biological samples from included studies into five groups (i.e. primary producers (PP), primary consumers (PC), secondary consumers (SC), tertiary consumers (TC) and top predators (TP)). The method of grouping samples was mainly referenced by the descriptions in Kozlowsky-Suzuki et al. (2012) and Liu et al. (2019). Phytoplankton and macroalgae were the predominant communities of PP. Zooplankton, herbivorous molluscs and herbivorous fish were considered as PC. Additionally, polychaeta were also chosen as PC because of their great contribution as the prey of demersal consumers. Crustaceans, cephalopods and zooplanktivorous fish were classified as SC based on their dietary compositions. Afterward, carnivorous fish and omnivorous fish were identified as TC. The food habits of fish were determined according to available information of included studies or relevant references. Finally, seals, sea birds, penguins and sharks usually occupied a higher trophic level, and thus were categorized as TP. All species were independently classified by two researchers, with disagreements resolved by discussion or consulting a third researcher. It was worthy to note that in real marine food webs, the same species might occupy dissimilar trophic levels in different food chains leading to diversity in trophic relationships (Vanderklift et al., 2006), and various factors (e.g. size, age, gender, hydrological condition etc.) were likely to cause certain biases of determining the trophic transfer behavior of metals in marine food webs (Gupta and Singh, 2011). In this study, the risks of biases were minimized by combining the advantages of big-data (increasing sample size to reduce sampling error) and meta-analysis (giving weight to reduce the impact of extreme data).

2.3. Data pre-processing

To quantitatively assess the trophic transfer behaviors of trace metals in marine food webs, data pre-processing was a crucial step prior to data analysis. The details were as follows: (i) the units of trace metal concentrations were unified as mg kg^{-1} in dry weight (mg kg^{-1} dw); (ii) in some cases, if the moisture percentage (m) was not measured, factors 5 and 10 (f) were applied for calculating biomass conversions from wet weight to dry weight for non-plankton and plankton respectively, while if the m was measured, the f was deduced by $f = 100\%/(100\% - m)$ (Ibelings and Havens, 2008; Kozlowsky-Suzuki et al., 2012); (iii) the mean

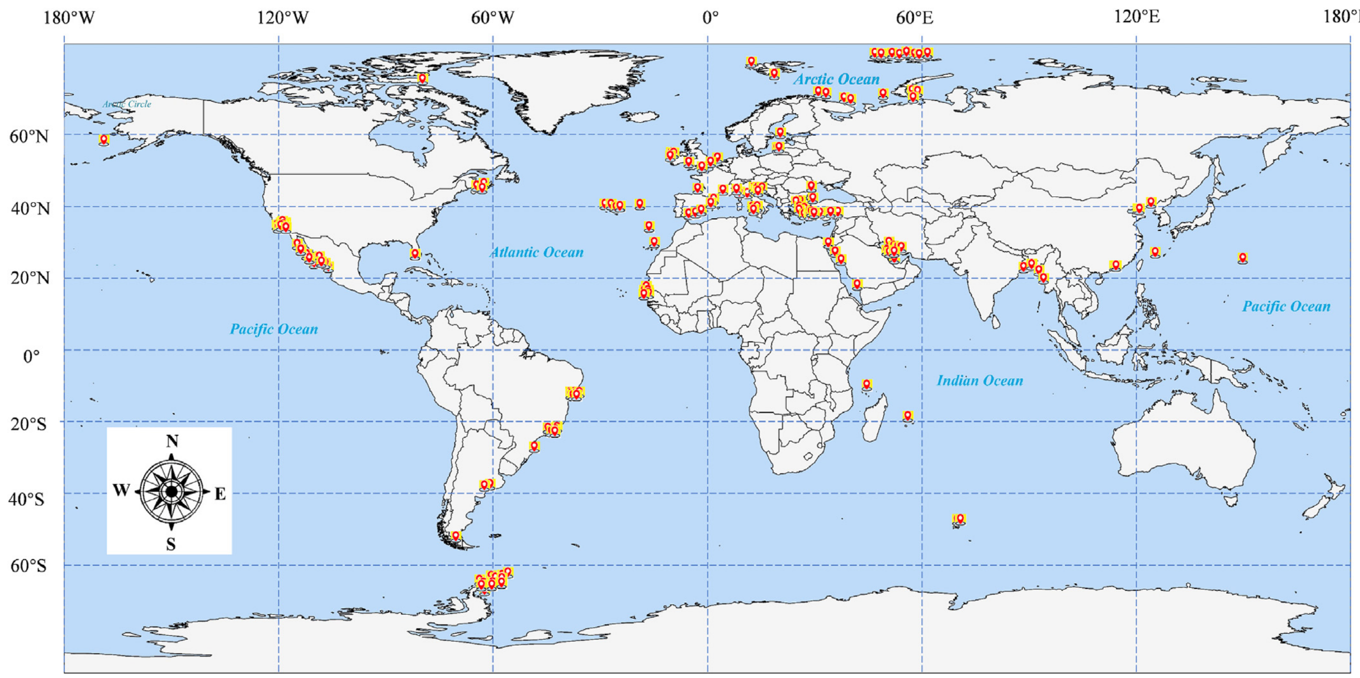

Fig. 1. World map with sampling sites of included studies. A total of 61 studies were included with 154 sampling sites of 33 countries/regions.

concentration of all available tissues was used to evaluate the accumulation of metals in whole body (Kozlowsky-Suzuki et al., 2012); (iv) for a few samples whose trace metal concentrations were not quantifiable because of the limits of quantification (LOQs), one-half of LOQs were used to estimate their concentrations according to the Cochrane Handbook v.5.2 (Higgins and Green, 2011); (v) mean ($\bar{x}$) and standard error (*SE*) were needed to combine data, however, some literature only reported the median ($x_{0.5}$), minimum (a), maximum (b), and sample size (n). In these cases, $\bar{x}$ and *SE* could be estimated using the formulas (S1–S6, see Supporting Information) (Hozo et al., 2005). (vi) if only standard deviation (*SD*) was reported, *SE* could be estimated by formula (S7, see Supporting Information) (Higgins and Green, 2011).

2.4. Data analysis

The STATA v.12.0 software (Stata Corporation, College Station, USA) was used to combine the independent studies. The random-effects model based on the method proposed by DerSimonian and Laird (1986) was used to pool relevant data and account for anticipated high heterogeneity among animal studies. To determine whether biomagnification or biodilution of each metal occurred in the global marine food webs and at two adjacent trophic levels, two types of trophic transfer terms, the food web magnification factor (FWMF) and the biomagnification factor (BMF), were calculated, respectively (Fisk et al., 2001; Hop et al., 2002; Liu et al., 2019).

The method determined the FWMF based on the relationship between the natural logarithm of metal concentrations and trophic levels, as:

$$\mathrm{LnC} = k\,(\mathrm{TL}) + d \qquad (1)$$

$$\mathrm{FWMF} = e^{k} \qquad (2)$$

where C was the metal concentration; k was the slope of weighted linear regression; *TL* was the abbreviation for trophic level; d represented the regression intercept. The weighted linear regression was performed in Minitab v.19 (Minitab Inc., State College, PA, US). The weight of each individual study was given by STATA v.12.0 software automatically via random-effects model.

The biomagnification factor (BMF) was calculated using:

$$\mathrm{BMF} = C_{\mathrm{predator}} / C_{\mathrm{prey}} \qquad (3)$$

where $C_{predator}$ and C_{prey} were the metal concentrations of the predator and the prey, respectively.

Metals with the values of FWMF and BMF statistically greater than 1.0 (via *t*-test) showed biomagnification in entire food webs and from prey to predator, respectively, while values statistically less than 1.0 indicated biodulution. However, if values were not statistically different from 1.0, no obvious biomagnification or biodilution trend was identified in this metal (Hoekstra et al., 2003).

According to the Cochrane handbook version v.5.2 (see Chapter 9.6.3.1), the differences of concentrations of each metal among classed groups and countries (i.e. developed and developing countries) were examined using meta-regression techniques based on random-effects model (Higgins and Green, 2011). The classification criteria of developed and developing countries were based on World Economic Situation and Prospects (2019). The statistical uncertainty was quantified in 95% confidence intervals (CIs), and the significant level was set at $p < 0.05$.

3. Results

3.1. Average concentrations of trace metals

Diverse bioaccumulation characteristics of classed groups (i.e. PP, PC, SC, TC and TP) were detected for each trace metal (Fig. 2). Among them, the average concentration of As in marine food webs was 6.93 mg kg^{-1} dw, and the lowest concentration was observed in TP, reducing to 1.45 mg kg^{-1} dw, which was significantly lower than those of other groups except for TC ($p < 0.05$). The average

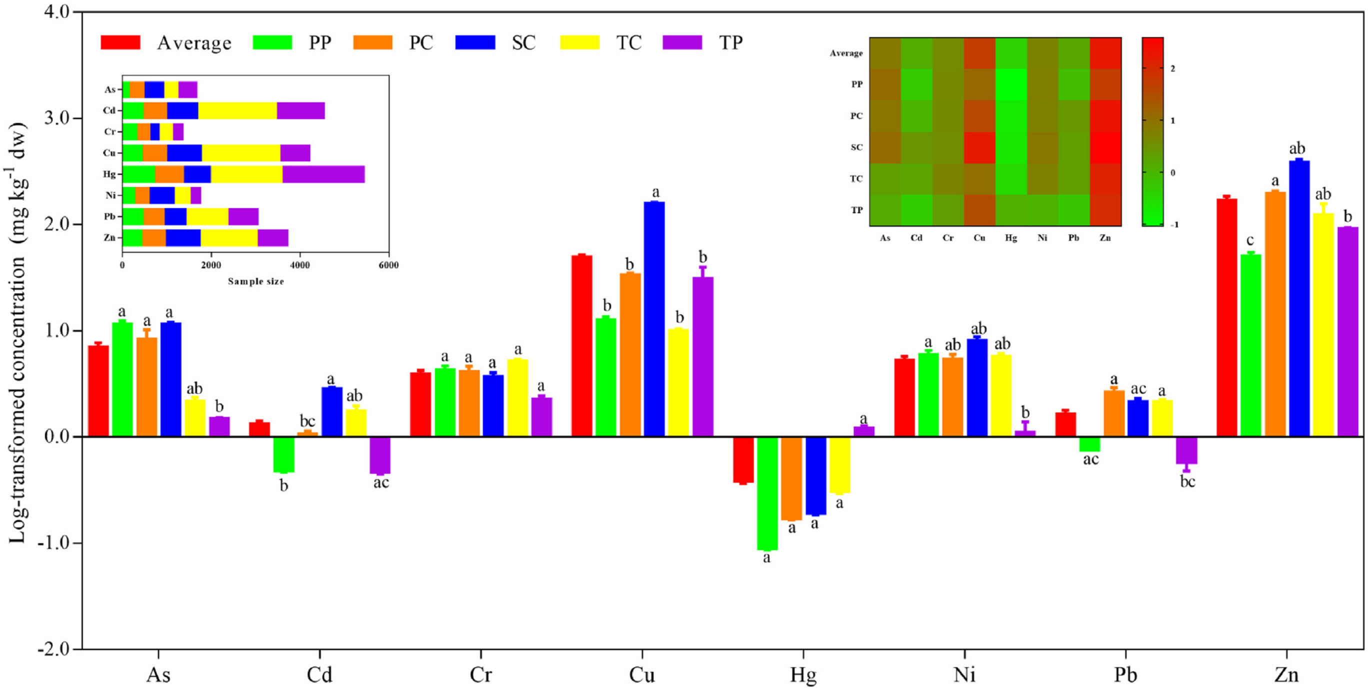

Fig. 2. The Log-transformed concentrations of metals in classed groups. The Log-transformed concentrations were summarized in the heat map in which the variation of color from green to red represented gradual increase of metal concentrations. The sample size of each metal/group was displayed in the left embedded graph. Different letters indicated significant differences at the 0.05 level among groups detected by meta-regression techniques. Data were presented as mean ± standard error (SE). **Abbreviations**: PP, primary producers; PC, primary consumers; SC, secondary consumers; TC, tertiary consumers; TP, top predators. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

concentration of Cd was 1.32 mg kg^{-1} dw. In SC, Cd concentration was as high as 2.83 mg kg^{-1} dw, which was obviously greater than those of PP and PC ($p < 0.05$). For Cr concentration, the mean value was 3.85 mg kg^{-1} dw, and no remarkable difference was observed among groups ($p > 0.05$). In terms of Cu, the average concentration was 48.5 mg kg^{-1} dw. Cu was peaked at 157 mg kg^{-1} dw in SC, which was markedly higher than those of other groups ($p < 0.05$). For Hg, the average concentration was 0.392 mg kg^{-1} dw. TP was the main Hg enrichment group with Hg accumulation reaching 1.19 mg kg^{-1} dw, which was higher than those of other groups with a p value (0.054) approaching the statistical significance (0.05). The average concentration of Ni was 5.18 mg kg^{-1} dw. In TP, Ni concentration was lower than other groups, but without significant difference except for PP ($p > 0.05$). The average concentration of Pb was 1.63 mg kg^{-1} dw. The Pb concentration in TP was lower than PP and SC ($p > 0.05$), and significantly lower than PC and TC ($p < 0.05$). Of the eight trace metals, Zn was the most abundant, with the average concentration up to 167 mg kg^{-1} dw. The lowest concentration of Zn was 50.1 mg kg^{-1} dw in PP, which was significantly lower than those of other groups ($p < 0.05$). The sum of the average concentrations of essential elements (Cr, Cu, Ni and Zn) in the food webs was 225 mg kg^{-1} dw, which was extremely higher than that of non-essential elements (As, Cd, Hg and Pb), 10.3 mg kg^{-1} dw ($p < 0.01$) (Fig. 3).

3.2. FWMFs and BMFs of trace metals

The weighted linear regression between Ln-transformed metal concentrations and trophic levels identified different trophic transfer behaviors of these metals in global marine food webs (Fig. 4). Among them, As ($R^2 = 0.393$, $p < 0.01$; FWMF = 0.494) and Ni ($R^2 = 0.110$, $p < 0.01$; FWMF = 0.731) displayed trophic level-dependent biodilution processes in the entire food webs, while Hg ($R^2 = 0.430$, $p < 0.01$; FWMF = 2.01), Pb ($R^2 = 0.127$, $p < 0.01$; FWMF = 1.81) and Zn ($R^2 = 0.0310$, $p < 0.05$; FWMF = 1.15) showed significant biomagnification trends. However, Cd ($R^2 = 0.0230$, $p > 0.05$; FWMF = 0.850), Cr ($R^2 < 0.01$, $p > 0.05$; FWMF = 0.951) and Cu ($R^2 = 0.0170$, $p > 0.05$; FWMF = 1.13) did not exhibit obvious trends with increasing trophic levels. The values of FWMFs of eight metals were ranked as: Hg (2.01) > Pb (1.81) > Zn (1.15) > Cu (1.13) > Cr (0.951) > Cd (0.850) > Ni (0.731) > As (0.494).

Dissimilar transfer behaviors of these metals at two adjacent trophic levels were exhibited (Fig. 5). Arsenic, Cr, Hg and Ni

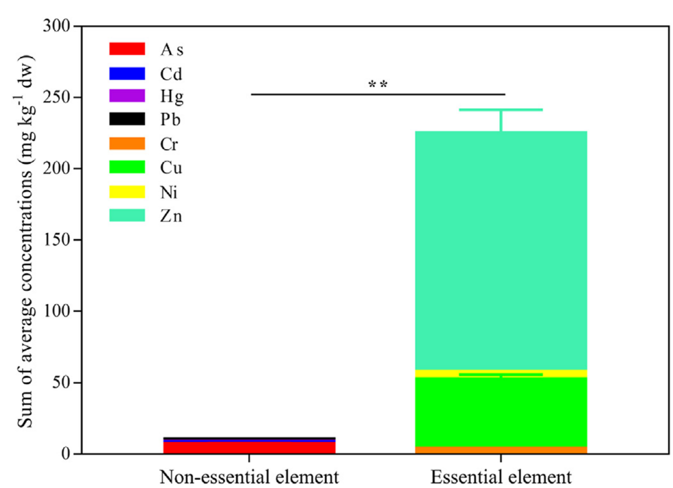

Fig. 3. The comparison of the sum of average concentrations between essential elements and non-essential elements. Essential elements included Cr, Cu, Ni and Zn, while non-essential elements were As, Cd, Hg and Pb. The difference was measured using meta-regression. Data were presented as mean ± SE. ** represented $p < 0.01$.

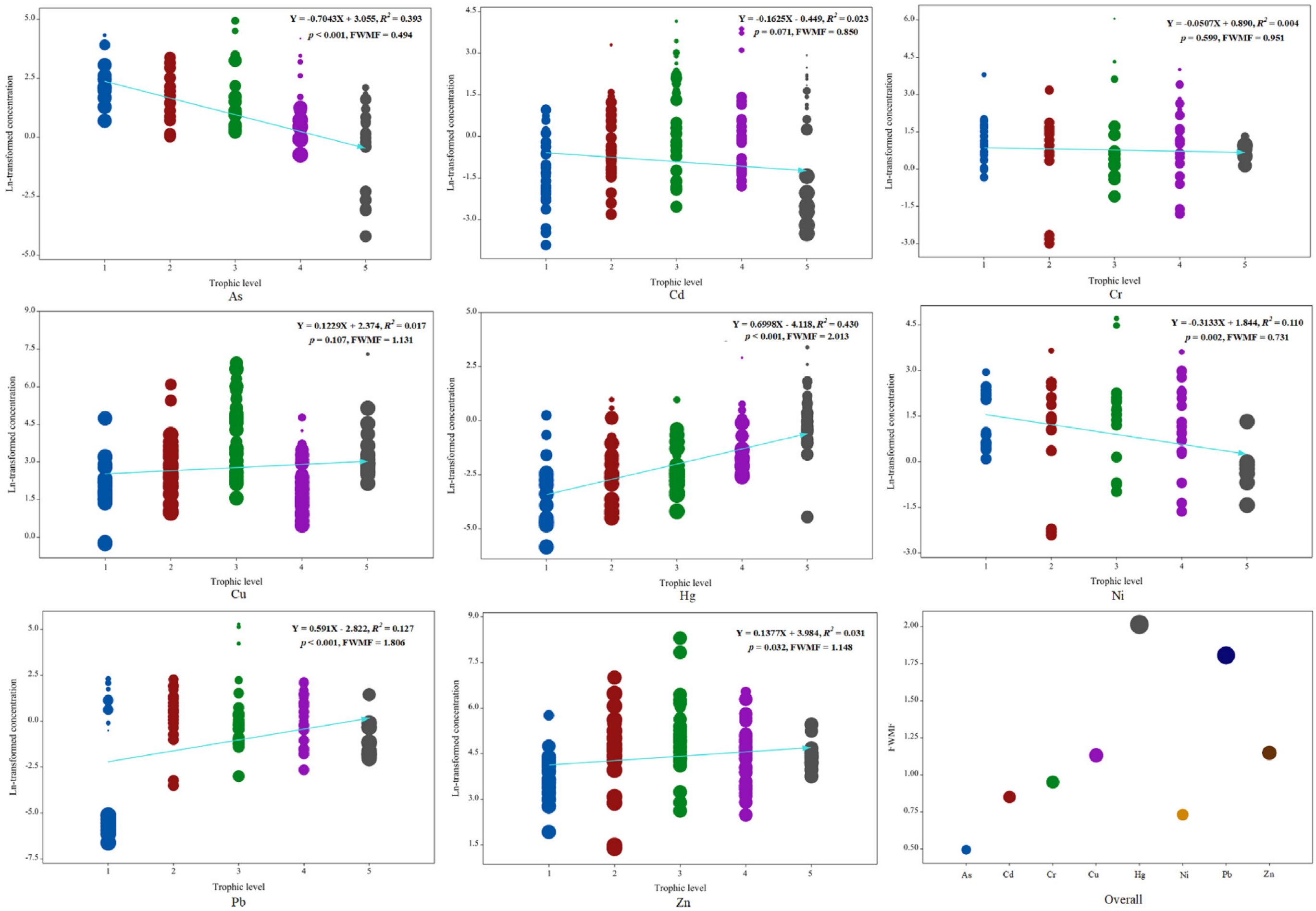

Fig. 4. The weighted linear regression between Ln-transformed concentration and trophic level. The bubble size was set at the weight of each study. The weights were obtained in STATA v12.0 using the random-effects model based on the method proposed by DerSimonian and Laird (1986). The random-effects model gave weight to each study based on the inverse of the variance adjusted by the heterogeneity (τ^2) among studies. In the fitting formula, Y represented Ln-transformed concentration, and X denoted the trophic level. The food webs magnification factor (FWMF) was calculated by the linear slope-power of natural logarithm. The values of R^2 and p were the coefficient of determination and the significant test of regression coefficient (via t-test), respectively.

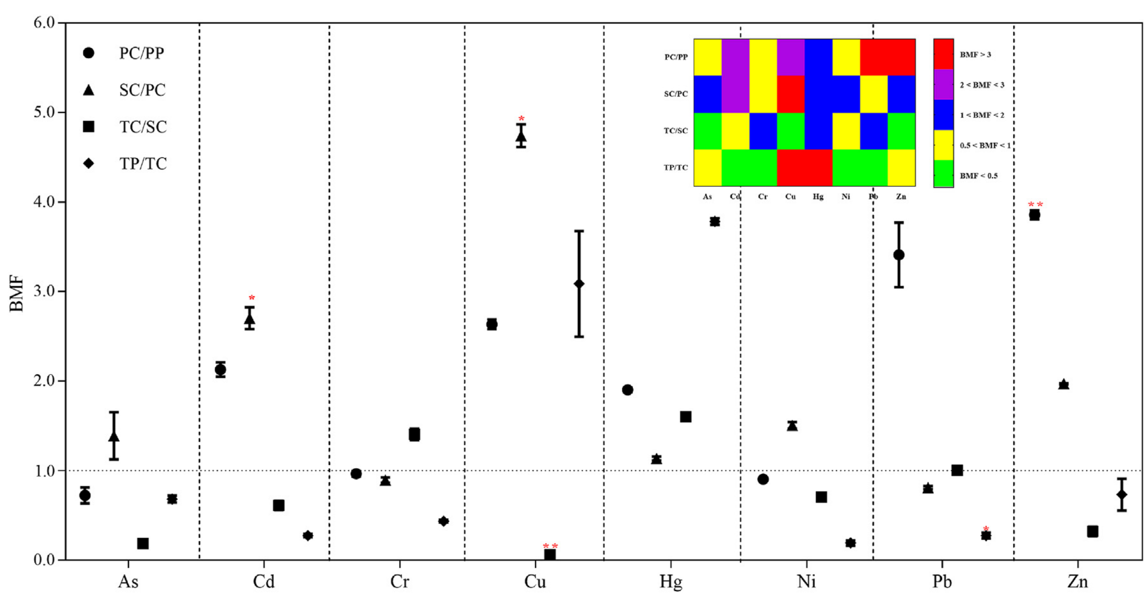

Fig. 5. The BMF values in each trophic transfer link. The oblique lines between two adjacent classed groups were the ratio of metal concentrations of the predator and the prey. The heat map displayed the values of the biomagnification factors (BMFs) in five intervals. Data were displayed as mean ± SE. *represented p < 0.05, and ** indicated p < 0.01.

exhibited no clear biomagnification or biodilution trend in the transmission between two adjacent trophic levels ($p > 0.05$). Interestingly, in every transfer link, Hg tended to be accumulated (BMFs > 1). For Cd, there was an apparent biomagnification along the trophic transfer from PC to SC ($BMF_{SC/PC} = 2.70$, $p < 0.05$). In the whole trophic transfer routes of these metals, Cu exhibited the most obvious biomagnification and biodilution in the transfer from PC to SC ($BMF_{SC/PC} = 4.74$, $p < 0.05$) and SC to TC ($BMF_{TC/SC} = 0.0629$, $p < 0.01$), respectively. Biodilution and biomagnification trends of Pb and Zn occurred respectively in the transmission from TC to TP ($BMF_{TP/TC} = 0.276$, $p < 0.05$) and PP to PC ($BMF_{PC/PP} = 3.86$, $p < 0.01$).

3.3. *Metal concentrations in marine food webs of developing and developed countries*

The comparison of metal concentrations in marine food webs between different development level countries revealed that the average concentration of Hg in five-trophic level marine food webs of developed countries (0.904 mg kg^{-1} dw) was significantly higher ($p < 0.05$) than that of developing countries (0.549 mg kg^{-1} dw) (Fig. 6). However, the average concentrations of other trace metals had no visible distinction ($p > 0.05$) (Fig. 6). Further analyses indicated that Hg concentration in PP of developed countries was obviously higher than that of developing countries, which was not only observed in global marine food webs ($p < 0.05$) (Fig. 7A), but also in a specific-area (Mediterranean Sea) ($p < 0.01$) (Fig. 7B). However, similar phenomenon was not found for other metals in global marine food webs ($p > 0.05$) (Fig. 7C).

4. Discussion

4.1. *As and Ni trophically biodiluted in global marine food webs*

Inconsistent trophic transfer behaviors of As were documented in previous studies (Asante et al., 2010; Hargreaves et al., 2011; Rahman et al., 2012; Liu et al., 2017; Trevizani et al., 2018; Shilla et al., 2019). Arsenic biodilution was the dominant process in aquatic food webs as reported by Hargreaves et al. (2011), Liu et al. (2017) and Trevizani et al. (2018), while As biomagnification was recorded in Asante et al. (2010), Rahman et al. (2012) and Shilla et al. (2019). This inconsistency demonstrated that the assimilation and enrichment ability of As was species-specific, which was evidenced by the lower concentrations of As at higher trophic levels in this study. The strong ability of higher trophic level species to biotransform inorganic As into less toxic AsB was identified in many studies (Bears et al., 2006; Zhang et al., 2012; Zhang et al., 2016), indicating that inorganic As was usually diminished with the increase of trophic levels, while AsB was increased. Synthetically, As presented a biodilution trend in global marine food webs.

The biodilution of Ni was observed in the entire food webs, whereas, the opposite trend occurred in the transmission from PC to SC. The same phenomenon was reported by Cardwell et al. (2013), showing that Ni might be biomagnified in specific and shorter food chains, especially, at the highest trophic level consisting of gastropods. Additionally, decreases of Ni concentrations in higher trophic organisms were observed in this study ($BMF_{TC/SC} < 1$ and $BMF_{TP/TC} < 1$). From an evolutionary perspective, species occupying higher trophic levels generally have effective metabolic mechanisms to regulate the metal concentrations in the body (Liu et al., 2019). However, there was no significant difference of Ni concentrations between PC, SC, TC and TP, which meant that Ni, an essential trace metal, was just absorbed and utilized by aquatic organisms in a very limited way. In a comparative genomic analysis of Ni utilization by Zhang and Gladyshev (2010), eukaryotes did not contain enough Ni-dependent metalloproteomes (e.g. urease and methionine synthase), revealing a restricted Ni utilization in eukaryotes.

4.2. *Hg, Pb and Zn trophically biomagnified in marine food webs*

In organisms, Hg and its compounds are hardly eliminated, and show strong genotoxicity (De Flora et al., 1994; Clarkson and Magos, 2006). It was recognized that biomagnification of Hg existed in almost all types of food webs (e.g. aquaculture pond, lake, ocean, stream, etc.), and bioaccumulated gradually with the increase of trophic levels (Lavoie et al., 2013; Squadrone et al., 2016). The trophic-level-dependent bioaccumulation of Hg was further identified in this study (all BMFs > 1). Hg pollution in marine and freshwater environments deserves special attention, as inorganic Hg is more easily enriched in muscle proteins to inactivate sulfhydryl-related enzymes (e.g. succinate dehydrogenase and

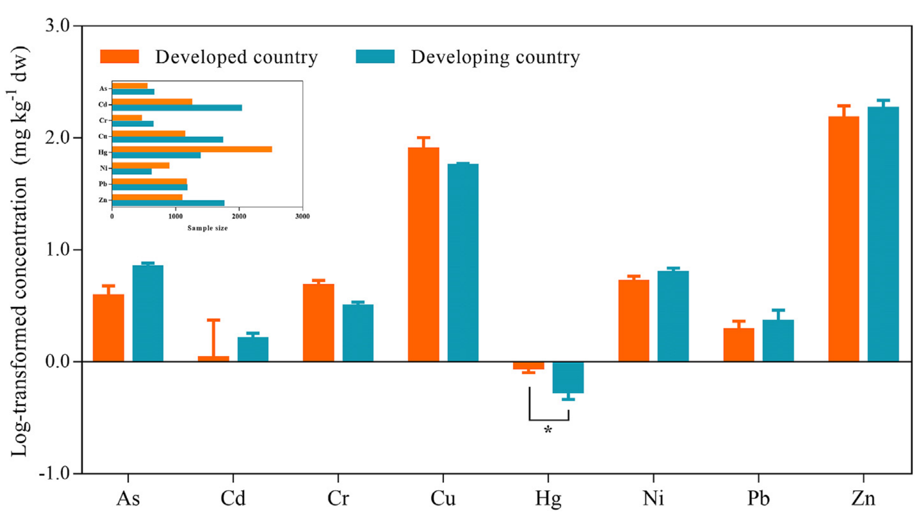

Fig. 6. The comparison of metal concentrations in marine food webs between developed and developing countries. The developed countries included Belgium, Canada, France, Ireland, Italy, Japan, Poland, Portugal, Romania, Spain, Sweden, United Kingdom and United States. The developing countries involved Argentina, Bangladesh, Brazil, Chile, China, Egypt, Iran, Mexico, Qatar, Saudi Arabia, Senegal, Turkey and United Arab Emirates. The sample sizes were displayed in the embedded graph. Data were presented as mean ± SE. *indicated $p < 0.05$. The difference of each metal was measured by meta-regression.

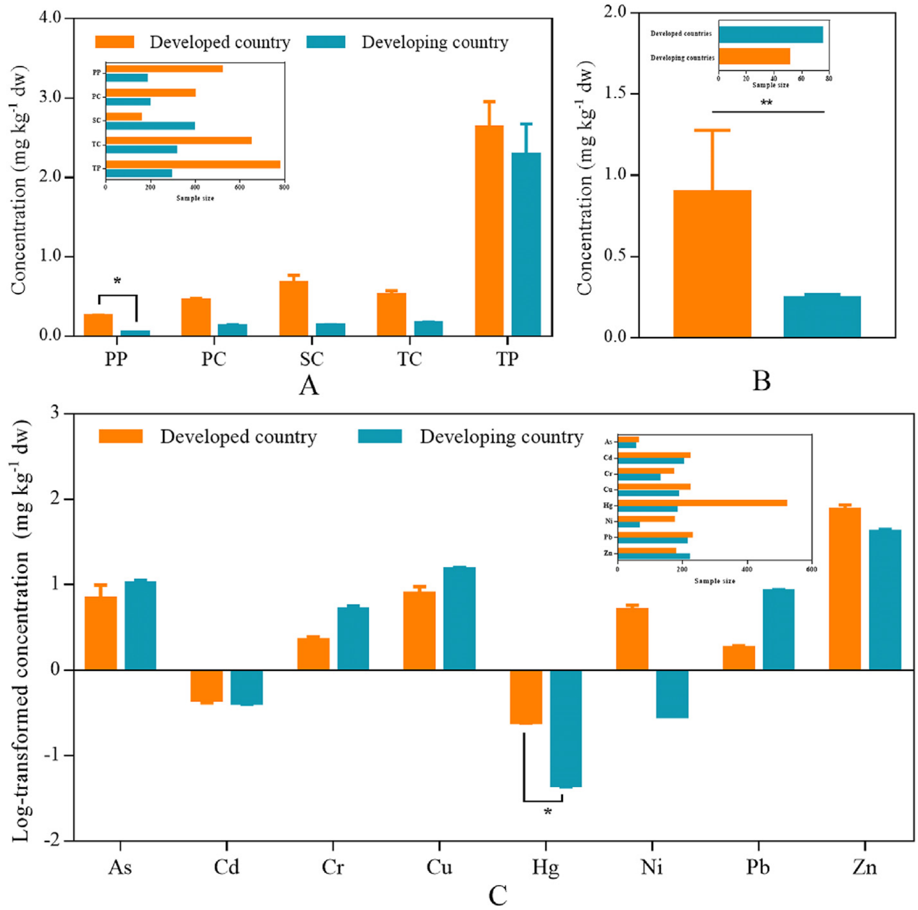

Fig. 7. The comparison of classed groups in marine food webs between developed and developing countries. (A) represented the comparison of Hg concentrations between classed groups in global marine food webs. (B) referred to the compassion of Hg concentrations between PP in the marine food webs of developed and developing countries along the Mediterranean Sea, with Italy and Turkey as examples. (C) showed the comparison of eight metals between PP in global marine food webs. The sample sizes were displayed in the embedded graph. Data were presented as mean ± SE. *indicated $p < 0.05$, and **indicated $p < 0.01$. The difference of each metal was measured by meta-regression.

cytochrome oxidase) after high-affinity binding to sulfhydryl of enzymes catalytic sites, thereby resulting in cell metabolic abnormalities and death (Ynalvez et al., 2016). More seriously, inorganic Hg is prone to be converted into higher toxic MeHg by bacterial action or physic-chemical processes in marine sediments/anoxic waters, and the latter has stronger sulfhydryl-binding ability than the former (Wang, 2012; Zhong and Wang, 2009; Liu et al., 2013; Liu et al., 2016).

Pb concentration had a significant decrease at the highest trophic level, reflecting the powerful eliminating ability at TP group. Several studies also indicated that the concentrations of Pb in TP individuals were usually below the background exposure levels (Andreani et al., 2008; Buekers et al., 2009). It is still unclear why Pb concentration presents a significant decline at TP. However, Scheuhammer (1987) pointed out that the existence of antagonism between Pb and calcium (Ca), and the accumulation and toxicity of Pb in mammals and birds could be dramatically modified by dietary Ca levels. In marine environment, organisms at the highest trophic level generally have relatively more opportunities for Ca uptake (Niebuhr, 1983; Dee Boersma et al., 2004).

Zn was the most prominent element in food webs (Nfon et al., 2009; Zhao et al., 2013). However, the trophic transfer behavior of Zn in different studies was inconsistent. The evidences of Zn biomagnification were identified by Zhao et al. (2013), Ruelas-Inzunza and Páez-Osuna (2008) and Shilla et al. (2019), whereas the opposite trend was reported by Nfon et al. (2009), Cardwell et al. (2013) and Sakata et al. (2015). This might be explained by the fact that Zn concentration was easily regulated by physiological metabolism activities (Mills, 1987), and the bioavailability of Zn varied significantly in different organisms due to seasonal, regional and interspecific variations (Ruelas-Inzunza and Páez-Osuna, 2000). In most cases, Zn tended to enrich in lower trophic levels, especially in invertebrates, because they often lacked regulation, excretion and detoxification mechanisms of Zn (Barwick and Maher, 2003), which was also confirmed in this study ($BMF_{SC/PC} = 1.97$, $BMF_{TC/SC} = 0.323$).

4.3. Cd, Cr and Cu exhibited no significant change along food webs

Cd is usually considered as a non-essential element with high toxicity and non-biological function (Mesonero et al., 1996; Das et al., 1997). However, Lane and Morel (2000) provided evidences of biological roles for Cd in enzymes synthesis in phytoplankton, which might correspond to the low Cd concentration in PP. In this study, there was an efficient accumulation of Cd in transferring process from PC to SC, however, further biomagnification in higher trophic levels appeared unlikely. Consistently, numerous studies indicated potential biomagnification of Cd in crustaceans, but few evidences were observed in fish, as the former could effectively sequester dietary Cd and store it in detoxified form, whereas fish had very low assimilation efficiencies in this process (Wang, 2002; Marsden and Rainbow, 2004). There were many controversies regarding the transfer behavior of Cd in food webs, due to different sampling sites and species composition (Amiard et al., 1980; Ward et al., 1986; Bargagli, 1993; Ruelas-Inzunza and Páez-Osuna, 2008; Zhao et al., 2013; Liu et al., 2019). In this study, Cd presented no obvious biomagnification or biodilution trends in global marine food webs.

The low and stable concentrations of Cr at different trophic levels might be associated with its low absorption (0.4–2.0%) (Pechova and Pavlata, 2007) and low bioavailability (<3%) (Lyons, 1994). Liu et al. (2019) suggested that Cr(III) and Cr(VI) were the main forms of Cr in marine environments, and the trophic transfer behavior of Cr was determined by its metal form, that was, the biomagnification of Cr would appear when Cr(VI) was the major form in marine environments. Levina and Lay (2008) also indicated that Cr(VI) compounds were more easily absorbed and accumulated into cells. These findings were in accordance with previous reports. It was confirmed that Cr(VI) could cross cell membranes easily with strong oxidative capacity, reacting with nucleic acids and protein inside cells (Pechova and Pavlata, 2007). In contrast, Cr(III) crossed cell membranes hardly, with low reactivity (Mertz, 1992), and could be easily adsorbed by particles in water, depositing in sediments (Rowbotham et al., 2000), which might limit the accumulation of Cr(III) in organisms from prey and environment, respectively.

SC was the main accumulated trophic level of Cu, with the BMF value astonishingly reaching 4.74 from PC to SC. The strong Cu-accumulation ability of SC was documented by many studies (Keil et al., 2008; Marsden and Rainbow, 2004; Olmedo et al., 2013; Liu et al., 2019). Olmedo et al. (2013) suggested that the presence of hemocyanin (a Cu-containing respiratory protein discovered in crustaceans and cephalopods) could account for the remarkable Cu concentration in SC. Meanwhile, a very interesting phenomenon was reported by Keil et al. (2008). In crustaceans, the high accumulation of Cu was usually accompanied by high accumulation of Cd (Keil et al., 2008). This could be explained by the efficient uptake mechanisms for Cu that cannot discriminate between Cu and Cd in crustaceans. The similar phenomenon was also identified in the present work (SC had the highest concentration of Cd up to 2.83 mg kg^{-1} dw). Moreover, regarding the reason for minimum $BMF_{TC/SC}$, Dang et al. (2009) pointed out that the bioaccumulation of Cu in marine fish could be regulated by the Cu uptake kinetics, exhibiting the characteristics of low assimilation efficiency and high efflux rate constant.

4.4. Limitations of FWMF and BMF

FWMF and BMF are commonly used for evaluating the trophic transfer behaviors of pollutants (Hoekstra et al., 2003). However, either FWMF or BMF has its own unavoidable limitations. FWMF was calculated from the slope of linear regression representing the entire food webs, which might over- or underestimate some trophic transfer data. BMF based on predator/prey comparison is underpowered for examining complicated predator-prey relationships (Fisk et al., 2001; Hop et al., 2002). To minimize the risk of misjudgement about determining the occurrence of biomagnification or biodilution, some corrective means were proposed in actual experiments. For example, if the preys were morphologically identifiable in the stomach of the predator, BMF was corrected by the dietary proportion. And, if the trophic level of the measured species was not an integer or the species occupied different trophic levels in the food webs, BMF was corrected by the trophic position (Phillips and Koch, 2002; Phillips et al., 2005; Fisk et al., 2001; Haukås et al., 2007). In this study, the BMF correction was not conducted, due to hardly available dietary proportion data and the construction of nearly single trophic relationship. Besides that, in this study, the FWMF was corrected by the weight of individual study using weighted linear regression that could effectively deal with the heterogeneous variances among studies (McCullagh and Nelder, 1989; Jaeger, 2008).

According to Kozlowsky-Suzuki et al. (2012), the average value of BMFs (BMF_{avg}) was regarded as an indicator to represent the transfer behavior of microcystins in the entire food webs. In this study, the BMF_{avg} was also calculated, which obtained highly consistent trends of transfer behaviors of most metals evaluated by FWMF (Fig. 8). Franklin (2016) suggested that BMF_{avg} could represent the FWMF over the whole food webs. However, this approximate equation (i.e. FWMF $\approx$ BMF_{avg}) was not applicable for all cases in this study (e.g. for Cu), as BMF_{avg} was very sensitive and easily influenced by the extreme data (Fig. 8). Actually, the potential differences were due to different evaluation perspectives of the two measures. FWMF focused more on the overall performance, while BMF was more powerful in specific predator-prey relationship (Hop et al., 2002).

4.5. Environment-friendly rather than environment-sacrificing development

Over the past few decades, developed countries attached much importance to the treatment of heavy metal pollution and formulated strict environmental protection policies after experiencing the tragic history of Minamata disease and obtained certain achievements (Matsuo, 2000). This study indicated that in the recent twenty years, under the influence of multiple factors (e.g. industrial upgrading, industrial transfer and ocean currents), the accumulation of some industrial metals (e.g. As, Cd, Ni and Pb) in marine food webs of developed countries showed lower trends than that of developing countries. However, Hg, a persistent and difficult-to-excrete pollutant, still accumulated significantly in global marine food webs of developed countries. The level of Hg in marine environment was primarily influenced by geochemical conditions and anthropogenic activities (Kotnik et al., 2014). In this study, the significant bioaccumulation of Hg in PP in developed countries along the Mediterranean Sea might imply that anthropogenic activities were the predominant factor affecting the level of Hg in marine food webs. Similarly, as reported by Zou et al. (2007), anthropogenic sources contributed 87.5% to the accumulation of Hg in the marine sediment of Hong Kong. Moreover, the significant difference of Hg concentration in marine food webs between developed and developing countries might also be related to the highest FWMF of Hg, meaning that the concentration of Hg in marine food webs was easily influenced by the level of Hg in marine environment. Therefore, the obvious bioaccumulation of Hg in marine food webs of developed countries might be accounted for more emission of Hg by anthropogenic activities and the highest FWMF of Hg in marine food webs. To some extent, the noticeable bioaccumulation of Hg in marine food webs of developed countries reflected the consequence of the traditional development model of “pollution first, treatment later”, suggesting that economic growth should not be achieved at the expense of environment, and more attention should be paid to curbing pollution from the source rather than later treatment.

5. Conclusions

The different trophic transfer behaviors of eight common metals in global marine food webs were identified in this study based on 9929 samples from 154 sampling sites of 33 countries/regions. As

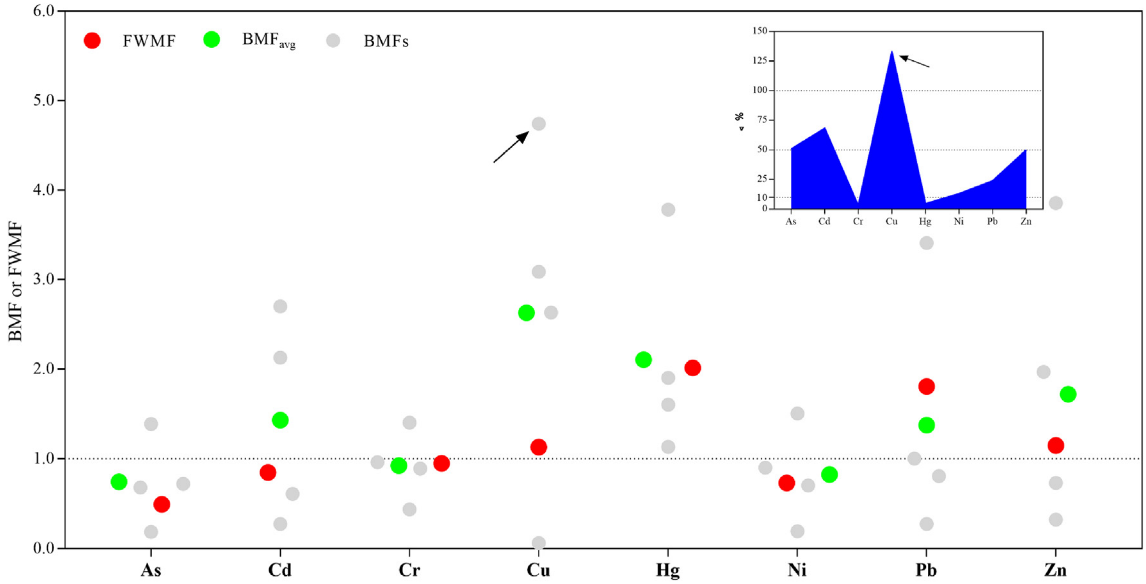

Fig. 8. The comparison of FWMF and BMF$_{avg}$. The BMF$_{avg}$ was the average of the BMF of every trophic link. In the embedded graph, Δ represented the relative differences between BMF$_{avg}$ and FWMF, which was calculated by $|(BMF_{avg}\text{-}FWMF)/FWMF| \times 100\%$.

and Ni tended to be biodiluted with increasing trophic levels, while Hg, Pb and Zn appeared to be biomagnified. However, no biomagnification or biodilution trend was observed for Cd, Cr and Cu. In terms of specific transfer link, there were significant biodilutions of Pb and Cu transferring from TC to TP, and SC to TC, whereas biomagnifications of Cd and Cu, and Zn occurred in the trophic transfer from PC to SC, and PP to PC, respectively. Moreover, the consequence of traditional development model of "pollution first, treatment later" was vividly manifested by the noticeable accumulation of Hg in marine food webs of developed countries, indicating that environment-friendly development is a promising way to prosper instead of environment-sacrificing development.

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.

CRediT authorship contribution statement

Tao Sun: Methodology, Formal analysis, Writing - original draft. **Huifeng Wu:** Supervision, Writing - review & editing, Project administration. **Xiaoqing Wang:** Formal analysis. **Chenglong Ji:** Writing - review & editing. **Xiujuan Shan:** Funding acquisition. **Fei Li:** Writing - review & editing.

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Appendix A. Supplementary data

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