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Metals and arsenic in sediment and fish from Cau Hai lagoon in Vietnam: Ecological and human health risks

Thi Ai My Tran [a, b, *], Martine Leermakers [a], Thai Long Hoang [b], Van Hop Nguyen [b], Marc Elskens [a]

[a] Department of Analytical, Environmental and Geo-Chemistry, Vrije Universiteit Brussel, 1050, Brussels, Belgium
[b] Department of Chemistry, University of Sciences, Hue University, 77 Nguyen Hue, Hue, Viet Nam

HIGHLIGHTS

- EF and I-geo values indicate that surface sediments are contaminated by As and Bi.
- Ecological risk (ER) are ranked as $ER_{As,Cd} > ER_{Pb} > ER_{Ni} > ER_{Cr,\ Cu,\ Zn}$.
- Farm fishes are slightly more contaminated by As than wild fishes.
- THQ and TR values reflect a potential health risk for regular fish consumers.

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ABSTRACT

Concentrations of Al, As, Bi, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, U, V and Zn were quantified in surface sediments collected from 13 different sampling sites from Cau Hai lagoon in Central Vietnam, and in 8 wild and farmed-fishes involving both pelagic and benthic species. Multivariate analysis shows that these trace elements are mainly associated with lithogenic matter, and are most likely the result of alteration and erosion processes in the lagoon. Enrichment factors and geo-accumulation indices reveal substantial sediment enrichments for both As and Bi with respect to the mean composition in the upper continental crust. As is enriched in the edible portion of fish tissue with values up to 10 times higher than the allowed limits set up by Health Canada. Target hazard quotient and target carcinogenic risk for As were assessed through fish diet and were greater than 1 and 10^{-4}, respectively, indicating potential health risks for fish consumers in Cau Hai lagoon.

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1. Introduction

Coastal lagoons are generally oriented parallel to the littoral, characterized by shallow water bodies, separated from the ocean by a land barrier but connected by one or more restricted water entrances (Kjerfve, 1994; Isla, 1995). Thus lagoons exhibit high salinity and are affected by tidal mixing inducing particle sedimentation. Human activities, surrounding agricultural areas and aquaculture within the lagoon significantly impact the fate and behavior of the lagoon ecosystem (González-Macías et al., 2006; Oyeyiola et al., 2013). According to European Water Framework Directive (2000/60/EC) (European Community, 2000), and other publications (Anthony et al., 2009; Oyeyiola et al., 2013) lagoons are part of ecosystems requiring continuous monitoring and conservation.

Metals and metalloids from natural and anthropogenic sources and applications have led to their wide distribution in the lagoon and estuarine environment, raising concerns over potential effects on human health (Costa-böddeker et al., 2017; Islam et al., 2017; Mugoša et al., 2016). Suspended matter play a key role for metals dynamics in these ecosystems because most are preferentially adsorbed on fine particles, and therefore may accumulate in bottom sediments via sedimentation (Strady et al., 2017a). High levels of metals and metalloids in sediments can be mobilized into interstitial water, and diffuse back to the surface water (although slowly) through diagenetic processes and eventually reach animals and humans through trophic chains (Muñoz-Barbosa and Huerta-Diaz, 2013). The consumption of fish is an essential element of the balanced human diet in the lagoonal ecosystem. Occupying the top

* Corresponding author. Department of Analytical, Environmental and Geo-Chemistry, Vrije Universiteit Brussel, 1050, Brussels. Belgium.
E-mail address: My.Tran.Thi.Ai@vub.be (T.A.M. Tran).

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of aquatic food chains, birds can be become contaminated (Alamdar et al., 2017). The analysis of metal concentrations in fish therefore makes it possible to estimate the exposure and risk of consumers and fishermen populations in lagoonal systems. Trace metals and arsenic may pose a health and environmental problem in water and sediments in Southern (Hoang et al., 2010; Strady et al., 2017a, 2017b) and Northern Vietnam (Ho et al., 2012).

Cau Hai lagoon is part of Tam Giang – Cau Hai, which is the largest coastal lagoonal systems in Southeast Asia with surface area of 216 km^2 and length of 70 km (Giuliani et al., 2011). However, the rapid increase of aquaculture in the region, especially inside the lagoon and the uncontrolled exploitation of natural aquatic resources has strongly impacted the Cau Hai lagoon, and depleted its natural resources (Nguyen et al., 2007). The lack of information about metals' sources and dynamics in Central Vietnam lagoons is a major challenge for policy makers on environmental protection and coastal ecosystems. Recent studies (Giuliani et al., 2011; Romano et al., 2012) on metals in soils and sediments in Central Vietnam did not assess the potential of ecological and human health risks induced by metals and metalloids. This study aims at (i) understanding the sources and distributions of metals and metalloids in sediments, (ii) assessing their impacts on the ecological environment, (iii) providing additional data on and (iv) assessing the potential human health risks for regular fish consumers in Cau Hai lagoon based on metals and metalloids distribution in the edible fish portion.

2. Materials and method

2.1. Study area and samples collection

Located in southern Vietnam and covering an area of 112 km^2, Cau Hai lagoon is the largest area of this lagoonal system (Thua Thien Hue, 2012), and presents all the physical features of a coastal lagoon. The lagoon connects to the sea by Tu Hien mouth and receives fresh water from Dai Giang and Truoi rivers, from the connected lagoon systems and runoff from the surrounding regions (Fig. 1A). The brackish water is beneficial to the development of the biodiversity of aquatic organisms which are exploited in turn by the local people. Sediment samples were taken from 13 sites in two campaigns (rainy and dry seasons) in 2015 and one campaign in the 2016 rainy season (Fig. 1B). The sampling design was the following:

- 7 random points in Cau Hai lagoon (S2, S3, S4, S5, S6, S7, S10)
- 3 intersection points (S1: between Tu Hien mouth and Cau Hai lagoon; S8: between the Dai Giang and Truoi rivers and Cau Hai lagoon; S9: between Thuy Tu lagoon and Cau Hai lagoon)
- 1 point in the Thuy Tu lagoon, before discharging into the Cau Hai lagoon (S11)
- 2 river mouth points (S12 and S13 at the Dai Giang and Truoi rivers, respectively)

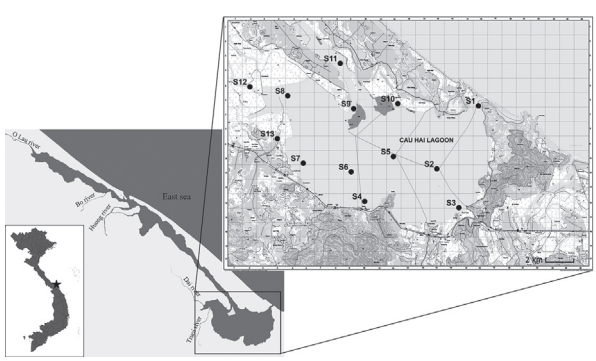

Fig. 1. Map of Tam Giang-Cau Hai lagoon (A); sediment samples collection in Cau Hai lagoon (B).

The surface sediments (upper 0–10 cm) were collected with a stainless-steel grab sampler, and then placed into chemically clean amber glass bottles. All the samples were immediately sealed and transportation to the laboratory. The samples were then air-dried, slightly crushed, homogenized and passed through a 250 μm stainless steel sieve and storage at the temperature room until analysis.

A total of twenty-three fish samples from the lagoon and the aquaculture farms were taken between March 2015 and August 2016. In the two first campaigns (March and July, 2015), six wild fish samples, including specific benthic specie (*Oxyurichthys papuensis*) and middle layer fish species (*Moolgarda pedaraki, Terapon jarbua*) were taken from fishing boats at the Cau Hai lagoon. Additionally, seventeen fish samples including *Lutjianus argentimaculus, Caranx bucculentus, Epinephelus coioides, Lates calcarifer, Anthopagrus latus, Lutjianus argentimaculus* sp. were taken from two farms at the Cau hai lagoon in Tu Hien and Vinh Hung communes in February and August 2016 (see Table 1 for more details). All fish samples were kept at 4 °C in cool containers during transportation to the laboratory and then lyophilized after removal of skin, bones and viscera. All samples were analyzed at the Analytical, Environmental and Geochemistry Laboratory from VUB, Belgium. Trace elements were measured by ICP-MS after microwave digestion.

2.2. Sample analysis

Approximately 200 mg of homogenized sediment were weighted in digestion Teflon vessels, in which 4 mL concentrated HF (40%), 6 mL distilled concentrated HNO_3 (65%) and 2 mL concentrated HCl (37%) super-pure grade were added. Digestions were performed according to USEPA method 3052 (1996) using a microwave oven (Multiwave Go, Anton Paar). The Teflon bombs were heated to 180 °C in a controlled heating step during 10 min

Table 1
Fish samples (species, weight, length, locations).

Species		Number of samples	Weight (kg/individual)	Length (cm/individual)
Wild fish	*Oxyurichthys papuensis*	1	0.010 ÷ 0.012	10 ÷ 11
	Moolgarda pedaraki	3	0.10 ÷ 0.11	20 ÷ 22
	Terapon jarbua	3	0.05 ÷ 0.07	10 ÷ 11
Farmed fish	*Lutjianus argentimaculus*	4	0.80 ÷ 0.90	30 ÷ 35
	Caranx bucculentus	2	1.00	35
	Epinephelus coioides	2	1.30 ÷ 1.70	35 ÷ 47
	Lates calcarifer	2	0.50 ÷ 1.50	32 ÷ 40
	Anthopagrus latus	3	0.20 ÷ 0.60	18 ÷ 25
	Terapon jarbua	3	0.10 ÷ 0.12	14 ÷ 18

and held at 180 °C for 9.5 min. After cooling 30 mL H_3BO_3 4% superpure grade were added to the digested samples to complex excess HF (Wilson et al., 2006).

Regarding fish samples, 6 mL distilled concentrated HNO_3 (65%) was added to 200 mg of homogenized lyophilized fish tissue weighed in digestion Teflon vessels. Fish samples were digested in a microwave oven Multiwave Go, Anton Paar as described in Rumisha et al. (2016). After cooling 40 mL MiliQ water were added to the digested fish samples.

The final digested solutions were decanted into a Teflon bottle for storage until measurement by Inductively Coupled Plasma-Sector Field Mass Spectrometry (ICP-SFMS) with an ELEMENT II (Thermo Instruments) using a concentric nebulizer (0.4 mL min^{-1}), a cyclonic Petier cooled spray chamber, quartz injector and Ni cones. A multi-element standard (ICP-MS Calibration Standard 2 (VWR, 10 mg L^{-1}) was used for calibration standards of Al, As, Bi, Cd, Co, Cu, Cr, Fe, Mn, Ni, Pb, U, V, Zn. A single element standard (Alfa Aesar Spectrapure) 1000 mg L^{-1} was used for calibration standards of Mo. Quality control samples were run in each analysis. More details about the method can be found in Gao et al. (2013).

2.3. Quality control analyses

The accuracy of the analytical procedure was tested through the analysis of certified reference material LGC6139 (River Clay Sediment) and DORM-4 (Fish protein). The recoveries of Al, As, Cd, Cr, Cu, Fe, Mn, Ni, Pb and Zn in LGC6139 ranged from 81 to 123%. Meanwhile, the recoveries of Al, As, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb and Zn in DORM-4 ranged from 82 to 111%. Confidence intervals (95%) were calculated for 4 replicates of LGC6139 and DORM-4, and the certified reference values of the elements analyzed were within these confidence limits. Precision on TE determinations were less than 7.5% for the high resolution inductively coupled plasma mass spectrometry.

2.4. Enrichment factor (EF) and geo-accumulation index (Igeo)

The enrichment factor (*EF*) and the geo-accumulation index (I_{geo}) are widely used indicators for assessing trace element (TE) enrichment in surface sediment and soil (Barbieri, 2016). The TE concentration of a given element was normalized to the concentration of a reference element, generally chosen because the latter is of natural origin (Ackermann, 1980). Typical reference elements are Al, Fe, Mn, Rb, organic carbon and grain size (Ackermann, 1980; Barbieri, 2016). Al and Fe are mainly associated with natural lithogenic sources (Huang et al., 2014; Muñoz-Barbosa and Huerta-Diaz, 2013), both of which have been applied in numerous studies (Cheng and Yap, 2015; González-Macías et al., 2006; Wang et al., 2015; Zhuang and Gao, 2015). In this study, Al, Fe and Mn were correlated and the EF-values were calculated with respect to Al according to Yang et al. (2017):

$$EF = \frac{C_M/C_{Al}[Sample]}{C_M/C_{Al}[Backgound]} \quad (1)$$

Because background values of major and minor elements for sediment in Cau Hai lagoon are not available, the worldwide composition of TEs in the upper continental crust was used (Wedepohl, 1995). EF-values are ranked into five categories ranging from minimal (EF < 2) to extremely high enrichment (EF > 40) (Barbieri, 2016).

The geo-accumulation index (I_{geo}), introduced by Muller (1969), has been applied (Lin et al., 2008; Salas et al., 2017; Strady et al., 2017b) to characterize TE contamination in the 2 μm grain size fraction according to:

$$I_{geo} = log_2 \frac{C_M[Sample]}{1.5{*}C_M\ [Background]} \quad (2)$$

where 1.5 is a correction factor considering differences in background values due to lithologic variations (Fontolan and Covelli, 1997; Rodríguez-Barroso et al., 2010). I_{geo} classification entails six classes ranging from uncontaminated ($I_{geo} < 0$) to extremely contaminated sediment ($I_{geo} \geq 5$) (Barbieri, 2016).

2.5. Ecological risk index (RI)

The ecological risk index (RI), introduced by Hakanson (1980) for the analysis of contamination in lakes and coastal systems was applied to the Cau Hai lagoon by considering the potentially harmful elements As, Cd, Cr, Cu, Ni, Pb and Zn:

$$RI = \sum_{i=1}^{7} TEF_i \times CF_i \quad (3)$$

where $CF_i = \frac{C_M\ [sample]}{C_M[background]}$ and TEF_i is the dimensionless toxic equivalent factor for a given element: Cd - 30, As - 10, Ni and Pb - 5, Cu and Cr - 2 and Zn - 1 (Chen et al., 2016; Giri and Singh, 2015; Kabir et al., 2011; Zhang et al., 2012). PCBs and Hg are not considered in this paper, since their concentrations were below or close to detection limits but have included Ni. RI classification entails four classes that can be interpreted as follows: RI < 150, low ecological risk; 150 ≤ RI < 300, moderate ecological risk; 300 ≤ RI < 600, considerable ecological risk and RI ≥ 600, very high ecological risk (Hakanson, 1980).

2.6. Estimated daily intake (EDI), non-carcinogenic and carcinogenic risk

Estimated daily intake (EDI) (mg kg^{-1} d^{-1}) was calculated from estimated fish consumption according to the researches (Ahmed et al., 2015; Alamdar et al., 2017; Cheng and Yap, 2015; Copat et al., 2012; Giri and Singh, 2015):

$$EDI = \frac{C_M \times CR}{B_W} \quad (4)$$

where C_M is element concentration in fish sample (mg kg^{-1} ww), CR is daily fish consumption rate (kg d^{-1}) and B_W is body weight for an adult (average 57.7 kg for Asian ethnicity (Walpole et al., 2012). The average daily fish consumption rate for a person was chosen as 0.142 kg d^{-1} based on a survey of 137 fisher households in Cau Hai lagoon.

For non-carcinogens, target hazard quotients (THQs) were calculated following the USEPA integrated risk analysis (1989):

$$THQ = \frac{EDI}{R_fD} \quad (5)$$

where R_fD is the oral reference dose or Tolerable daily intake (mg kg^{-1} d^{-1}) and was derived from Integrated Risk Information System - IRIS (USEPA, 2007). The R_fD represents an estimate of the daily exposure to which a human population may be continually exposed without a significant risk for deleterious effects. If THQ is less than 1, the exposed population is unlikely to experience obvious adverse effects. If THQ is equal to or higher than 1, there is a potential health risk, and mitigation strategies should be implemented.

Carcinogenic risks are assessed as the probability that an individual develops cancer during lifetime exposure to this potential

carcinogen. Target cancer risk (TR) was calculated following USEPA (1989):

$$TR = EDI \times SF_0 \quad (6)$$

where SF_0 is the oral carcinogenic slope factor derived from IRIS (USEPA, 2007) database. In general, default approaches are used for genotoxic and carcinogenic substances. For risks calculated to be linear at low doses, acceptable risk levels ranged from 10^{-6} to 10^{-5} (SCHER et al., 2009).

2.7. Statistical analyses

All statistical analyses were performed using the XLSTAT software, Version 2017.6 from Addinsoft. Agglomerative hierarchical clustering (AHC) and principal component analysis (PCA) were used to explain the spatial distribution of trace elements in sediment and fish samples. AHC was achieved using Ward linkage and Euclidian distance. The extraction procedure was based on a normalized PCA using the correlation matrix with Varimax rotation and a Kaiser normalization. All the statistical tests were performed at a significance level of 5% ($\alpha = 0.05$)

3. Results and discussion

3.1. Distribution of trace elements in surface sediments

Results for trace element (TE) concentrations in sediments are summarized in Table 2. A two factor ANOVA indicates that there is a statistically significant difference between sampling sites ($p < 0.05$) but not between sampling periods ($p > 0.05$). Accordingly, the data were seasonally aggregated by considering the median TE concentration at each sampling point. Agglomerative hierarchical clustering (AHC) was used to differentiate the sampling sites regarding the median distribution of TEs. As shown in Fig. 2A, AHC yields three groups with the sites classified as zone 1 (S1 & S2) and zone 2 (S3 to S9) being more contaminated by Al, As, Co, Cr, Cu, Fe, Mn, Ni, Pb and Zn than zone 3 (S10 to S13). On an average basis,

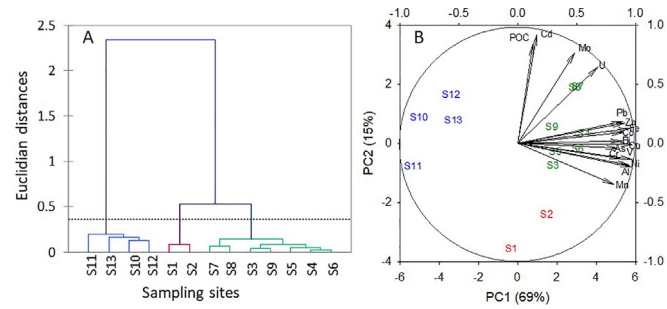

Fig. 2. AHC Dendrogram (A) for the spatial distribution of TEs in the surficial sediments. AHC was computed using Ward linkage and Euclidian distance. PCA bi-plot (B) obtained after Box-Cox transformation. The factor loadings (vectors) represent the correlations between the original variables and the principal components (top and right axes). The correlation circle is in black. In the factor scores (bottom and left axes), numbers represent sampling sites with group 1 in red group 2 in green and group 3 in blue according to AHC results.

zone 1 has lower Cd, Mo and U median concentrations than that of zone 2 and 3. Sites included in zones 1 and 2 are rather located in the south-eastern part of the Cau Hai lagoon, while sites from zone 3 are on the outer edge of the lagoon. The bi-plot (Fig. 2B) shows the factorial plans corresponding to the first two axes. These components are responsible for the bulk of the variance with PC1 and PC2 explaining 69% and 15% of the variations, respectively. The coordinates of the original variables in the plane express their correlations with the new principal components. Variables carrying similar information or varying in a comparable way are grouped together. Bi, Cr, Cu, Ni & Pb are mainly correlated with Al ($R \geq 0.834$; $p < 0.001$) and As, Co, U, V & Zn with Fe ($R \geq 0.850$; $p < 0.001$). On the other hand, a weak ($R = 0.50$) but significant ($p < 0.05$) correlation is observed between the organic matter (POC) and Cd & U. Moreover, there is a clear segregation of variables along the PC axes with Al, As, Bi, Co, Cr, Cu, Fe, Mn, Ni, Pb, V & Zn mainly associated with PC1, and Cd & Mo with PC2. For all variables, the quality of the representation in the loading plot is > 0 and < 1 (Fig. 2B). The

Table 2
Median concentrations (±median absolute deviation) of trace elements in surficial sediment (mg kg^{-1} dry weight (dw)).

Sites	Al	As	Bi	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	U	Zn
S1	70799 ±9553	13.9 ±1.3	0.7 ±0.3	0.16 ±0.04	13 ±0.3	68 ±6.0	15 ±1.3	33334 ±2310	622 ±7.6	26 ±2.1	36 ±11	3.2 ±0.7	79 ±11
S2	80092 ±10320	16.2 ±1.8	0.9 ±0.04	0.17 ±0.01	20 ±2.9	80 ±4.9	15 ±1.2	47275 ±4978	826 ±66	29 ±3.1	40 ±1.4	4.8 ±0.6	99 ±5.5
S3	82986 ±5277	15.3 ±0.8	1.0 ±0.01	0.23 ±0.03	14 ±0.7	77 ±4.3	17 ±1.2	42910 ±3766	560 ±6.6	28 ±2.0	45 ±0.5	7.0 ±0.6	85 ±4.0
S4	92843 ±9065	19.0 ±0.4	1.4 ±0.2	0.26 ±0.05	18 ±2.1	90 ±6.5	19 ±1.2	46914 ±2512	583 ±31	30 ±2.3	52 ±6.8	7.5 ±0.7	100 ±4.2
S5	74353 ±774	18.5 ±1.2	1.1 ±0.1	0.23 ±0.02	19 ±2.8	84 ±2.3	14 ±0.2	49882 ±4142	781 ±80	29 ±0.7	45 ±0.7	6.5 ±1.0	93 ±3.2
S6	97146 ±2115	16.6 ±0.1	1.0 ±0.2	0.24 ±0.06	21 ±0.7	98 ±9.7	17 ±0.3	50530 ±1114	638 ±4.3	32 ±1.9	45 ±9.4	6.7 ±1.2	105 ±6.8
S7	80872 ±1593	15.2 ±1.5	1.3 ±0.07	0.37 ±0.01	18 ±2.4	86 ±7.9	20 ±2.2	49798 ±847	570 ±29	27 ±0.6	52 ±4.8	9.4 ±0.5	108 ±0.3
S8	76749 ±10089	26.1 ±0.2	0.9 ±0.1	0.31 ±0.01	22 ±0.4	83 ±5.6	17 ±3.5	63511 ±4829	543 ±119	26 ±1.3	46 ±8.4	8.4 ±0.4	123 ±7.2
S9	70505 ±8579	14.7 ±6.5	1.1 ±0.4	0.32 ±0.08	16 ±5.5	67 ±14	15 ±2.9	39918 ±12186	661 ±181	29 ±11.3	50 ±19	6.4 ±2.3	95 ±29
S10	40582 ±859	8.2 ±0.4	0.5 ±0.03	0.27 ±0.01	10 ±0.4	44 ±1.3	9 ±0.4	28359 ±430	330 ±6.7	15 ±0.4	23 ±1.4	3.7 ±0.1	66 ±2
S11	43612 ±12821	8.6 ±1.9	0.7 ±0.2	0.17 ±0.01	8 ±2.6	62 ±11	8 ±1.0	22362 ±6267	278 ±64	14 ±3.0	26 ±4.6	4.3 ±1.1	47 ±5.8
S12	43427 ±1343	10.3 ±2.4	0.5 ±0.05	0.35 ±0.01	14 ±3.4	50 ±7.2	10 ±1.1	37065 ±9440	303 ±48	17 ±0.2	34 ±2.2	5.6 ±0.8	78 ±6.2
S13	58896 ±297	5.6 ±1.2	0.7 ±0.05	0.28 ±0.09	9 ±0.7	63 ±16	11 ±3.2	25529 ±5055	513 ±2.9	17 ±0.6	32 ±0.4	5.8 ±2.2	58 ±13

further away a vector lies from the origin, the closer it is near the correlation circle, the stronger its influence on the PCA model. Accordingly, the percent of variance explained by the new components for TEs ranges from 72% to 98%. This first analysis indicates that TEs examined in this study are associated with the lithogenic fraction (aluminosilicates and iron oxyhydroxide) rather than with biogenic material. The resulting EF values (Eq. (1)) indicate that As, Bi, Cd, Cr, Pb, Zn and U are enriched in the surface sediments of the Cau Hai lagoon (Fig. 3). The EF values for As & Bi are the highest among TEs and can be considered as moderate to severe enrichments (5–10), while the other values show minor enrichments (<3). The geoaccumulation index (Eq. (2)) also supports these observations, and suggests that surface sediments are moderately to heavily contaminated with respect to As & Bi ($2 < I_{geo} < 3$). As & Bi are two related elements of the pnictogens, and usually found in trace quantities in nature. While arsenic is well known as a toxic element, and associated to human health threat in Vietnam (e.g. (Berg et al., 2007, 2001; Pham et al., 2017; Postma et al., 2017, 2007; Tobias and Berg, 2011)), metallic Bi is not considered toxic, and does not per se constitute a real threat to the environment. The As enrichment reported in our study is consistent with previous researches on the Tam Giang-Cau Hai Lagoon in surface and subsurface sediments (Giuliani et al., 2011) and in most coastal lagoons from central Vietnam (Romano et al., 2012). Interestingly, higher concentrations of As were also reported in southern Vietnam in the Mekong Delta (Hoang et al., 2010) and in sediments from the Tien River (Strady et al., 2017a).

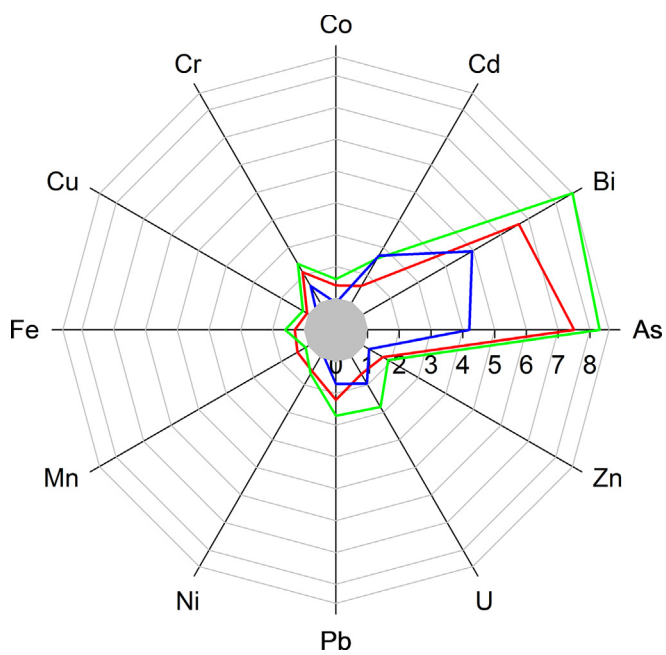

Fig. 3. Enrichment factors (EF) of TEs in surficial sediment. The circle in grey represents the background value with group 1 in red, group 2 in green and group 3 in blue according to AHC results (see Fig. 2).

3.2. Potential ecological risk of As and metals in sediments

Two common sediment quality guidelines (SQGs) were used to assess the eco-toxicological significance of As, Cd, Cr, Cu, Ni, Pb and Zn in Cau Hai lagoon sediments. Table 3 provides the Interim Sediment Quality Guidelines (ISQGs) represented by the threshold Effect Level (TEL) and Probable Effect Level (PEL) concentrations, which were developed by the Canadian Council of Ministers of the Environment's for the protection of aquatic systems (CCME, 2002), and the effects range-low value (ERL) and effects range-median value (ERM) guidelines adapted from Long et al. (1995). The lowest values (i.e., ERLs or TELs) are the concentrations that should protect wildlife living in the sediment, while in contrast, ERMs and PELs represent concentrations beyond which adverse effects are likely to occur (Long et al., 1998). Several TEs (i.e. As, Cr, Ni, Pb) equal or exceed the ERL limits, but none exceed the ERM ones (Table 3). While sediments from zone 2 are to be considered as an occasional threat to wildlife due to the presence of Cr and Pb, most of the sediments within the lagoon are likely to be associated with a toxic effect due to concentrations in As (zones 1 to 3) and Ni (zones 1 to 2). The SQG approach focuses on single element regardless of the presence of other TEs. Yet sediment toxicity often results from wildlife exposure to a mixture of elements. The potential ecological risk index (Eq. (3)), as introduced by Hakanson (1980), was used to assess the ecotoxicological significance of a mixture of As, Cd, Cr, Cu, Ni, Pb and Zn. Calculated RI-values are greater than 150 in zone 1 and 2 suggesting a moderate ecological risk ($150 < RI < 300$). A pairwise multiple comparison indicates that zone 2 has a significantly higher RI than that of Zone 1 and Zone 3, respectively (Holm-Sidak method, $p < 0.05$). Regarding the ecological risk factor per element (*i.e.*, $ER_i = TEF_i \cdot CF_i$), the series is ordered as follows $ER_{As,Cd} > ER_{Pb} > ER_{Ni} > ER_{Cr,\ Cu,\ Zn}$. As and Cd are the main factors causing a potential ecological risk, and their median values per zone can rise up to 80 (Fig. 4). All of the sampling sites have medium to strong ER_i-values ($30 < ER_i < 120$) for As and Cd, whereas other TEs only exhibited slight values ($ER_i < 30$).

3.3. Distribution of trace elements in fishes

Species including *Oxyurichthys papuensis, Moolgarda pedaraki, Lutjianus argentimaculus, Caranx bucculentus, Epinephelus coioides, Lates calcarifer, Anthopagrus latus* and *Terapon jarbua* are very commonly used by human communities in Thua Thien Hue province and in surrounding area, Da Nang city. Tissue concentrations of TEs in the fish tissues collected from the Cau Hai lagoon are summarized in Table 4. A Kruskal-Wallis rank ANOVA indicated that

Table 3
Median concentration (min – max) of selected trace elements in surficial sediments and sediment quality guidelines (mg kg^{-1} dw).

	As	Cd	Cr	Cu	Ni	Pb	Zn
Median Min-Max	15 (6–20)	0.3 (0.2–0.4)	77 (44–98)	15 (8–20)	27 (14–32)	45 (23–53)	93 (47–123)
TEL[a]	7.2	0.7	52	19	–	30	124
PEL[b]	42	4.2	160	108	–	112	271
ERL[c]	8.2	1.2	81	34	21	47	150
ERM[d]	70	4.6	370	270	52	218	410

[a] Threshold effect level and.
[b] Probable effect level concentrations (CCME, 2002).
[c] Effects range-low and.
[d] Effects range-median concentrations adapted from (Long et al., 1995).

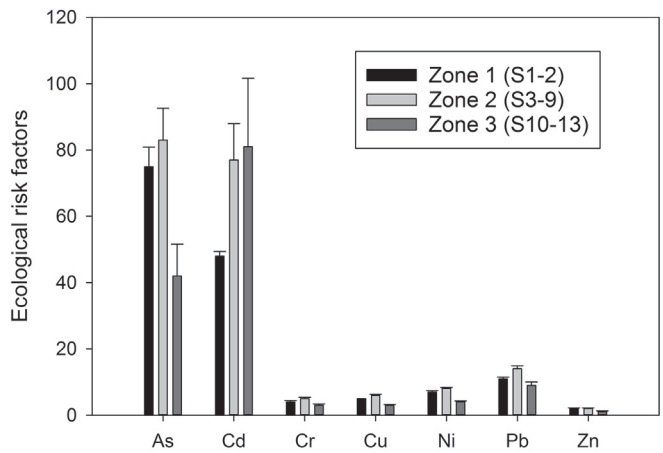

Fig. 4. Ecological risk factors ($ER_i = TEF_i \cdot CF_i$) for As, Cd, Cr, Cu, Ni, Pb and Zn. Zones 1 to 3 were determined according to AHC results (see Fig. 2). Error bars represent the median absolute deviation per zone.

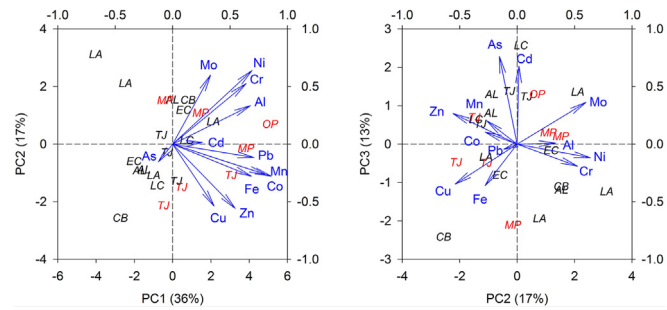

Fig. 5. PCA bi-plot obtained after Box-Cox transformation. The factor loadings (vectors) represent the correlations between the original variables and the principal components (top and right axes). The factor scores (bottom and left axes) represent the fish species with *Anthopagrus latus (AL); Caranx bucculentus (CB); Epinephelus coioides (EC); Lates calcarifer (LC); Terapon jarbua* (TJ); *Moolgarda pedaraki (MP); Oxyurichthys papuensis (OP).*

there are not statistically significant differences between the two aquaculture farms, for all investigated TEs and fish species ($p > 0.1$). Yet a PCA provides a better visualization of how TEs are distributed amongst fish species (Fig. 5). The bi-plot shows the factorial plans corresponding to the first three axes with PC1, PC2 and PC3 explaining 36%, 17% and 13% of the variations, respectively. There is a clear segregation of TEs along PC1 & 2 axes with a bundle of vectors mainly associated with Al (i.e., Cr, Mo, Ni with $R > 0.5$, $p < 0.01$) and Fe and/or Mn (i.e., Co, Cu, Pb, Zn with $R > 0.5$, $p < 0.01$) and other TEs such as Cd and As, which are rather associated with PC3. The farm fishes are scattered homogeneously around the biplot centroid and appears slightly more contaminated by As than wild fishes. The dispersion of the latter is greater and essentially directed along the axis PC1 with *Oxyurichthys papuensis (OP)*, a benthic species being more contaminated by TEs adsorbed on aluminosilicates, iron and manganese oxyhydroxides.

3.4. Assessment of human health risks

Based on the ecological risk calculations, three trace elements associated with a potential ecological risk were identified: As, Cd and to a lesser extent Pb. We therefore calculated the As, Cd and Pb concentration in fish protein based on a report by USDA (2018), which supplied the average protein in fish (edible portion) as 22%. The maximum levels of As and Pb in edible fish established by Health Canada (HC, 2017) are 3.5 and 1.5 (mg kg^{-1} fish protein), respectively. Concentrations of arsenic (mg kg^{-1} of fish protein) far exceed this limit for all investigated fish species (Fig. 6) with a maximum up to 39.1 mg kg^{-1} protein in *Lates calarifer*. Lead concentrations are below the maximum limit but with a maximum of up to 1.2 mg kg^{-1} protein for the benthic species *Oxyurichthys papuensis*. Regarding Cd, maximum up to 0.04 mg kg^{-1} protein was found in *Lates calarifer* (results not shown). There is, to our knowledge, no reported maximum level for Cd in the edible fish portion, Health Canada (HC, 2010) supplies a tolerable daily intake (TDI) of 0.001 (mg kg^{-1} d^{-1}). EDI values obtained using Eq. (4) range from 6×10^{-6} to 2×10^{-5} (mg kg^{-1} d^{-1}), and remain well below this limit. Therefore, Cd is unlikely to pose a threat to human consumption of fish in Cau Hai lagoon. Similar conclusions can be drawn for Pb with EDI-values ranging from 8×10^{-5} to 7×10^{-4} while the reported TDI is around 3.6×10^{-3} (HC, 2007). Non-carcinogenic (THQ, Eq. (5)) and carcinogenic risks (TR, Eq. (6))

Table 4
Median concentrations (±median absolute deviation) of trace elements in fish tissues (mg kg^{-1} dw).

Farmed fish	Al	As	Cd	Co	Cr	Cu	Fe	Mn	Mo	Ni	Pb	Zn
AL[a]	6.7 ±3.4	7.4 ±0.7	4.6E-3 ±2.1E-5	1.4E-2 ±6.3E-5	3.2E-1 ±7.7E-2	1.0 ±0.1	11 ±1	0.7 ±0.1	1.7E-2 1.4E-2	1.2E-1 ±6.0E-2	4.7E-2 ±2.4E-2	18 ±1
CB[a]	8.0 ±1.5	1.4 ±0.1	3.4E-3 ±1.4E-3	6.0E-3 ±1.1E-3	6.5E-1 ±6.0E-2	1.8 ±0.2	29 ±3	0.4 ±0.1	2.4E-2 1.0E-2	2.3E-1 ±1.2E-2	3.2E-2 ±2.4E-3	18 ±1
EC[a]	9.4 ±2.2	2.3 ±0.4	3.6E-3 ±1.3E-3	8.3E-3 ±1.3E-3	4.6E-1 ±2.1E-1	1.1 ±0.2	11 ±1	0.4 ±0.2	2.0E-2 ±1.2E-2	1.4E-1 ±8.2E_2	8.6E-2 ±2.4E-2	20 ±3
LC[a]	9.8 ±0.1	8.6 ±2.4	8.6E-3 ±1.5E-3	9.9E-3 ±2.7E-3	1.4E-1 ±4.1E-2	1.5 ±0.2	15 ±3	0.8 ±0.6	2.4E-2 ±1.5E-2	1.8E-1 ±9.7E-2	1.1E-1 ±3.2E-2	20 ±2
LA[a]	8.8 ±1.4	2.2 ±1.2	4.9E-3 ±1.5E-4	6.1E-3 ±2.4E-3	2.3E-1 ±5.7E-2	0.8 ±0.1	20 ±12	0.3 ±0.1	1.6E-2 ±8.5E-3	1.8E-1 ±9.9E-2	5.2E-2 ±2.7E-2	16 ±3
TJ[a]	10.2 ±0.2	2.0 ±0.2	7.3E-3 ±8.3E-5	9.7E-3 ±2.4E-3	1.7E-1 ±2.9E-2	1.2 ±0.1	15 ±1	0.4 ±0.1	2.9E-2 ±1.5E-2	1.0E-1 ±3.6E-3	3.2E-2 ±8.3E-3	30 ±1
Wild fish	**Al**	**As**	**Cd**	**Co**	**Cr**	**Cu**	**Fe**	**Mn**	**Mo**	**Ni**	**Pb**	**Zn**
MP[a]	37 ±19	2.6 ±1.0	4.7E-3 ±2.3E-3	1.4E-2 ±7.1E-3	4.4E-1 ±1.3E-1	1.0 ±0.2	25 ±9	0.9 ±0.4	2.8E-2 ±5.2E-3	3.2E-1 ±1.1E-1	8.4E-2 ±3.3E-3	16 ±3
OP[a]	84 NA	2.1 NA	7.4E-3 NA	7.4E-2 NA	7.7E-1 NA	0.8 NA	39 NA	5.2 NA	5.3E-2 NA	6.0E-1 NA	2.7E-1 NA	58 NA
TJ[a]	7.3 ±0.9	1.5 ±0.3	5.0E-3 ±1.1E-5	1.2E-2 ±2.7E-5	1.2E-1 ±2.5E-4	1.3 ±0.1	16 ±4	0.9 ±0.3	6.0E-3 ±2.9E-3	8.9E-2 ±3.5E-2	2.0E-1 ±3.0E-2	36 ±1

[a] Anthopagrus latus (AL); Caranx bucculentus (CB); Epinephelus coioides (EC); Lates calcarifer (LC); Lutjianus argentimaculus (LA); Terapon jarbua (TJ); Moolgarda pedaraki (MP); Oxyurichthys papuensis (OP).

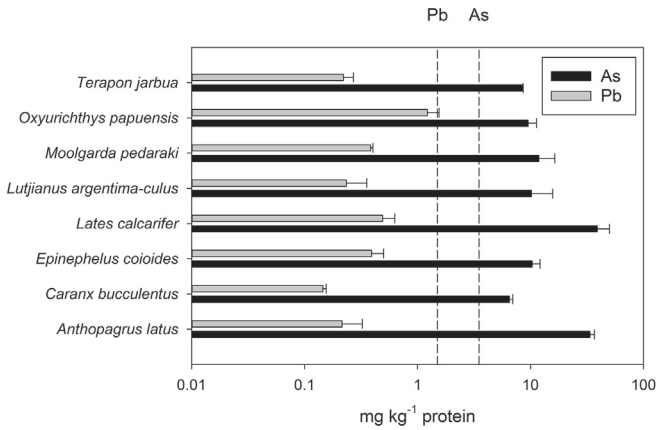

Fig. 6. As and Pb levels in edible fish tissues (mg kg^{-1} protein). Dot lines represent the maximum edible fish levels establsihed by Health Canada (2017). Error bars represent the median absolute deviation per fish specie.

were, therefore, assessed for As only. The implementation of Eqs. (5–6) requires the knowledge of the reference dose (R_fD) and the oral carcinogenic slope factor (SF_0), both of which being obtained from the IRIS database (USEPA, 2007). In addition, according to the guidance document on As in Shellfish (USFDA, 1993), the tolerable daily intake of As has to be based on exposure to inorganic As forms such as arsenide and arsenate. Since most of the arsenic present in shellfish is its organic form (less toxic), the recommended assessment method is to assume that inorganic As accounts for about 10% of the total As in shellfish. This guideline was applied to our study for both THQ and TR calculations. As shown in Table 5, all individual THQs are higher than 1 reflecting a potential health risk for regular fish consumers (farmed-fish communities) with the highest values in *Lates calarifer* and *Anthopagrus latus*. Similarly, TR values ranging from 7×10^{-4} to 3×10^{-3} suggests a carcinogenic risk to fish consumers from the Cau Hai lagoon. These results obviously raise also the question of the innocuity of the fish sauce made from fish species caught in the lagoon, especially for consumers or populations being treated for iron deficiency anemia, and using the fish sauce as iron fortification (Rodriguez et al., 2009).

4. Conclusion

Analysis of surface sediment samples collected in the Cau Hai lagoon showed that trace elements such As, Co, Cr, Cu, Ni, Pb and Zn are mainly associated with the lithogenic fraction (aluminosilicates, iron and manganese oxyhydroxides) suggesting a geological rather than an anthropogenic origin. These sediments are significantly enriched in As and Bi with respect to the average composition in the upper continental crust, and that As enrichment, especially could constitute a significant ecological risk for wildlife. Several other trace elements such as Cr, Ni and Pb equal or exceed the Threshold Effect Level (TEL) sediment guideline concentrations, but apparently, none exceed the Probable Effect Level (PEL) concentrations. A potential ecological risk has also been identified for Cd, but this is primarily related to the relatively high toxic equivalent factor used to characterize this element, and does not reflect a worrying concentration in the sediment.

Analysis of fish tissues showed that As by far exceeded the authorized limit of 3.5 mg kg^{-1} protein in the edible portion for all investigated fish species with a maximum up to 39 mg kg^{-1} protein, reflecting potential health risks for regular fish consumers in the Cau Hai lagoon.

Table 5
Target hazard quotient (THQ) and target cancer risk (TR) caused by As in different fish species from Cau Hai lagoon.

Fish species	THQ (As[a])	TR (As[a])
Oxyurichthys papuensis	1.7	0.000775
Moolgarda pedaraki	2.1	0.000964
Lutjianus argentimaculus	1.8	0.000828
Caranx bucculentus	1.1	0.000529
Epinephelus coioides	1.8	0.000842
Lates calcarifer	6.8	0.003174
Anthopagrus latus	5.8	0.002729
Terapon jarbua	1.5	0.000699

[a] 10% of total As concentration (mg kg^{-1}dw) in fish tissues (see in Table 4) was applied for THQ and TR assessment.

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