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Metals concentrations and ecological risk assessments of fish farms sediments in Kafr El-Shaikh, Egypt.

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ABSTRACT

As a result of the freshwater sacristy and growing demands for fish production, the usage of fresh Nile water (Ri-W) for fish cultivation has been banned and replaced by the usage of agricultural drainage water (ADW). The agricultural drains collect wastewater from various sources that pose a potential for metal pollution. Subsequently, affect the quality of water and sediments of the receiving fish farm and poses health hazards to cultured fish and man. Farms' sediments act as a reservoir for metals and an internal source of metal. Thus, the evaluation of its metal contents and their indications are preferred for tilapia culture management. The present study investigates the levels of 7 metals (essential, and non-essential) in the surficial sediments of three fish farms irrigated with two different water sources (RiW, and ADW). Followed by an evaluation of sediment pollution and potential ecological risks using pollution indices (PI) and sediment quality guidelines (SQGs). The results indicated that the mean levels of the metals at the three fish farms follow the decreasing order of Fe (> 50,000 ppm) > Mn (700 ppm) > Zn (110 ppm) > Cr (75 ppm) > Cu (55 ppm) > Cd = Pb (not detected). According to PI, all metals were depleted to mineral levels (enrichment factor (EF) < 2) and showed unpolluted status and almost at baseline levels (< 1) relative to geo-accumulation index (I geo), contamination factor (CF), and pollution loading index (PLI). According to ecological risks, the adverse effects due to Zn and Mn were unlikely to be noticed, in contrast, a slight risk may occur due to Cu, and Cr. There were no restrictions on using ADW for fish cultivation, where sediments were not considered a source of secondary metal pollution. Continuous monitoring and evaluation of the pollution status of farm sediments are recommended.

INTRODUCTION

One of the consequences of the unabated, indiscriminate, and uncontrolled development of industrialization and urbanization is the increase of levels of hazardous inorganic pollutants (including metals) emissions into the aquatic environment (Abdel-Halim *et al*. 2016; Anbuselvan *et al*. 2018). Although metals are naturally occurring elements that are found throughout the earth's crust, anthropogenic activities are also responsible for their abundance and distribution in the environment. Metals are entered the aquatic environment e.g. via point and non-point sources mainly from industrial, agricultural, and municipal waste effluents (Ullah *et al.* 2016; Benson *et al.* 2018).

From a biological point of view, metals can be sorted out according to their environmental impact and toxicity (Maret 2016; Shaaban *et al*. 2021b, c, a) Some of those elements are labeled as essential for life, such as Fe, Cu, Ni, Co, Mn, Cr, and Zn, and serve as micronutrients (Terech-Majewska *et al.*, 2016). Other metals like As, Cd, Pb, and Hg are termed nonessential, because they have no known biological and/or physiological functions, and they are potentially toxic to organisms (Bhat and Khan, 2011; Stankovic *et al*., 2014).

Sediments are important sinks, reservoirs, and carriers of metals and they can reflect the current quality of the aquatic system as well as the historical development of certain hydrological and chemical parameters (Soliman *et al*., 2019; Wu *et al*., 2014). Sediments can adsorb metals in water, but they cannot permanently fix them. Where, some sediment-bound metals may be remobilized and released back to surrounding waters because of changing environmental conditions of sediment or surrounding water such as redox potential, acidification, electrical conductivity (EC), pH, temperature, sediment particle size, chelating agents, etc., (Dong *et al*., 2014; Goretti *et al*., 2016; Zhang *et al*., 2016) causing secondary pollution to the water environment, producing unfavorable effects on living organisms (Xu *et al.* 2017). Thus, sediments play a convenient role in the assessment of metal pollution related to anthropogenic activities (Deng *et al*., 2022), specially sediments display less variations over time than dissolved metals in overlying water columns, they are preferred as a pollution monitoring approach (Abdel-Satar *et al.*, 2022).

The pollution of persistent and non-biodegradable metals in the aquatic environment is a growing problem worldwide and is currently reaching an alarming rate (Ogoyi *et al.*, 2011; Sharifuzzaman *et al*., 2016), due to their drastic environmental impact on all aquatic organisms. Upon their accumulation in sediments as well as bioaccumulation and biomagnification into food chains, high concentrations of metals implicate aquatic flora and fauna affecting ecological balance, in addition to the quantity and quality of aquatic organisms (Kaoud *et al*., 2014; Shreadah *et al*., 2015). Subsequently, they become a threat to man and pose health hazards, especially for those who depend directly or indirectly on aquatic habitats for fish and water supplementation (Wang *et al*. 2018).

In recent decades, fish farming has expanded significantly in response to the growing demand for aquatic products in developing countries (Nasr-Eldahan *et al*., 2021). Fish production from the aquaculture sector exceeds that of natural fisheries resources (FAO, 2018), where aquaculture produces about 57 % of the worldwide fish requirements (Ottinger *et al*., 2016). Moreover, Egypt is considered one of the major fish producers and succeeded to get a yield of farmed fish (about 80% of the total Egyptian fish production) more than captured fisheries (FAO, 2016; GAFRD, 2019). Nile tilapia (*Oreochromis niloticus, O. niloticus*) is the most significant and familiar farmed species in Egypt (Abdel-hakim *et al*., 2016).

As a mitigation step of freshwater sacristy, water resources shortage, and growing demands for fish production, the Egyptian Law 124 / 1983 banned the use of freshwater for fish cultivation (Ghanem and Haggag, 2015). Only governmental hatcheries and some governmental farms are allowed to use fresh Nile water (Ri-N) directly from the irrigated drains. About 90% of fish farms depend mainly on agricultural drainage waters (ADW), as a feeding water source to preserve current freshwater. (Soltan *et al.,* 2016). These complimentary water sources have various physicochemical properties which subsequently affect the quality of farm water and sediment in addition to cultured fish. Thus the main objectives of the present study are 1) to provide a clear picture of the distribution characteristics of the seven metals (essential and nonessential) in the sediments of Tilapia fish farms irrigated by different water sources [the agricultural drainage water (ADW), and the fresh Nile water (Ri-N)]; and 2) to evaluate the potential ecological risk levels on cultured fish using available Sediment Quality Guidelines (SQGs) by using the statistical indices like Enrichment Factor (EF), Geo-accumulation Index (I_{geo}), Contamination Factor (CF), and Pollution Load Index (PLI).

MATERIALS AND METHODS

1. Study area

Kafr El-Shaikh governorate is categorized as the highest -nearly 50 % of the total aquaculture production- fish producer among all Egyptian governorates (GAFRD, 2019). The tilapia farms are allocated around the drainage system of Burullus Lagoon. Drain-7 is one of the main Burullus Lagoon drains, it ranks as the second discharging drain surrounding the Lagoon (Assar *et al.*, 2016), and mainly collects the agriculture, industrial, domestic wastes, and effluents of fish farms from the surrounding areas (El-Amier *et al.*, 2017; El-Zeiny and El-Kafrawy, 2017). The water of Drain-7 was classified as the most polluted drain due to its high metal levels relative to other drains surrounding Burullus Lagoon (Yones *et al*., 2012 and El-Batrawy *et al*., 2018).

Three fish farms of *Oreochromis noliticus* were selected. The first two farms are representative of farms irrigated with ADW. They are situated at the upstream and downstream parts of the agricultural drain; Drain-7, they were termed Upstream Farm (FU) and Downstream Farm (FD), respectively. The third fish farm was away from drainage water (Drain-7) and was located at the agricultural irrigation canal of Nile water (Fig. 1), and it is represented as a control farm. Two ponds from each fish farm were chosen and from each pond, three sediment samples were collected to cover the sediment at the feeding water inlet and outlet in addition to one sample at the centre of each pond.

2. Sampling, preparation, and analysis

Sixteen surficial sediment samples were collected from the three fish farms (FU, FD, and FC) during the fish harvesting season (October 2018), using a stainless-steel grab sampler. The collected samples were immediately stored in a labeled self-sealed polyethylene bag and then kept in an icebox for transportation to the land lab for analysis.

The description of color, texture, shells, or other coarse fragments of the collected sediment samples were recorded. Firstly, each wet sample was divided into two portions, one of them was spread on a clean polythene sheet and left to dry in the air in a cleanroom. After dryness, the sediments were subsampled and then stored in an air-tight plastic vial for later analysis (dry weight). The other portion was used for the determination of the water content.

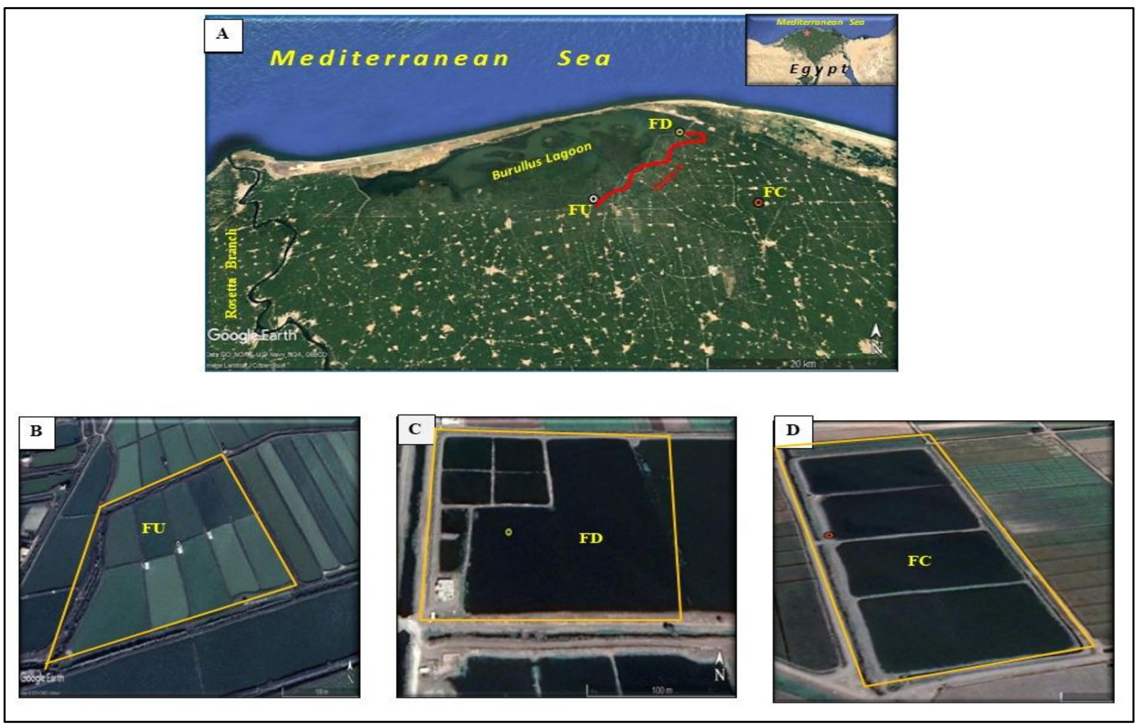


Fig. (1). Satellite images showing the three selected fish farms; (A) three farms locations; (B) the upstream farm (FU), (C) downstream farm (FD), and (D) the control farm (FC) at Kafr El-Shaikh governorate, Egypt.

- **Granulometric and chemical characteristics:**

The granulometric analysis was commenced and based on the determination of two main fractions sand and mud by separating the coarser (both sand and gravel) fractions above 4ϕ (0.063 mm), from finer (mud) fractions below (0.063 mm) using standard sieves (4ϕ) mounted on an electric shaker machine (Labor-technique) and 10 minutes was applied as a standard time of sieving (Folk, 1974). The percentage of water content was determined by the difference in weights between wet (W_w) and dry (W_d) sediment samples using the formula of: [(W_w-W_d/W_w) ×100]. The organic carbon content was determined by the oxidation of dry sediment with chromic acid (El-Wakeel and Riley, 1975). The total carbonate percent was determined according to (Vogel, 1978), using the diluted hydrochloric acid HCl 10%.

- ***Metals content:***

Bioavailable (mobile) and total metal concentrations (including Cu, Zn, Mn, Fe, Cr, Cd, and Pb) were analyzed in each sediment sample. Bioavailable metals (Bioav-M) were determined according to Standard Practices for Extraction of Trace Elements from Sediments (ASTM, 2003). A certain weight of the dry sample was extracted by shaking overnight using the diluted HCl (5%). Then the filtered extracted solution was subjected to the Bioav-M measurements using Inductively Coupled Plasma–Optical Emission Spectroscopy (ICP–OES, model ICP–OES 5100 vdv).

On the other hand, the total metals (T-M) content was analyzed according to Oregioni and Aston, (1984) technique; by digestion of dry sediment with a mixture of (nitric: perchloric: hydrofluoric acid, 3: 2: 1). All reagents used were of analytical grade and all solutions were prepared with double-distilled water (DDW). Blanks and duplicates were regularly employed during testing.

3. Pollution indices

Different types of indices were applied to assess the sediment quality including the enrichment factor (EF), geo-accumulation index (I_{geo}), metal pollution index (MPI), contamination factor (CF), and pollution Loading Index (PLI).

- Enrichment factor (EF)

Such technique was greatly applied by normalizing a metal concentration to the texture or compositional characteristics of sediments. Fe was used as a geochemical normalization element to alleviate the variations produced by heterogeneous sediments (Gu *et al*., 2013; Zhuang and Gao, 2014). EF was calculated using the equation of

$$\boldsymbol{EF = \frac{(M/Fe)\ Sample}{(M/Fe)\ crust}}$$

Where, (M/Fe) $_{sample}$ is the ratio of the metal and Fe concentration of the sample, and (M/Fe) $_{crust}$ is the ratio of the metal and Fe in the crust.

- Geo-accumulation index (I_{geo})

Praveena *et al.* (2010); and Sabo *et al.* (2013) stated that I_{geo} was, first, used by Muller (1969) by applying the formula of $\mathbf{I_{geo} = Log\ 2\ (C_n/1.5\ B_n)}$. Where C_n is the measured concentration of metal in sediment, B_n is the geochemical background value in average shale (Turekian and Wedepohl, 1961) of the element, n, 1.5 is the background matrix correction in factor due to lithogenic effects. The I_{geo} consists of six grades ranging from unpolluted to very highly polluted as mentioned in Table 1.

- Contamination factor (CF)

The degree to which sediment is contaminated is often expressed as CF (Sabo *et al*., 2013) from the equation of $\mathbf{CF} = \frac{\boldsymbol{C\ metal}}{\boldsymbol{C\ background\ value}}$. Where C_{metal} is the total metal concentration and C $_{background}$ is the average background value of the element from geologically similar and uncontaminated area. The used geochemical background values in the shale of the metals were earlier reported by (Turekian and Wedepohl, 1961). The CF value could fall into either of the levels of contamination. Where CF ranged from < 1 to > 6 as illustrated in Table 1.

- Pollution Load Index (PLI)

The PLI was calculated by obtaining the n-root from the n-CFs that were obtained for all the metals (Tomlinson *et al*. 1980), using the formula of $\boldsymbol{PLI} = \sqrt[n]{(\boldsymbol{CF1\ x\ CF2\ x\ CF3\ x \dots\dots\dots x\ CFn})}$. Where n is the number of metals, CF is the contamination factor. The PLI values were varied from 0 (unpolluted) to 10 (highly polluted) (Table 1).

Table 1. Terminologies for pollution classes on single and integrated indices.

EF classes		CF classes		I_{geo} classes			PLI	
EF value	**Pollution**	**CF value**	**Pollution**	I_{geo}	I_{geo} class	**Pollution**	**PLI**	**Pollution**
EF < 2	Depletion to mineral	CF < 1	Low	< 0–0	0	Unpolluted	0	Perfection
2 ≤ EF <5	Moderate	1 ≤ CF ≥ 3	Moderated	0–1	1	Unpolluted to moderated	< 1	Baseline levels
5 ≤ EF < 20	Significant	3 ≤ CF ≥ 6	Considerable	1–2	2	Moderated polluted	> 1	Polluted
20 ≤ EF < 40	Very high	CF > 6	Very high	2–3	3	Moderated to high polluted		
EF > 40	Extremely high			3–4	4	Highly polluted		
				4-5	5	Highly to extremely polluted		

4. Statistical analysis

The granulometric, and metals levels of the three fish farms were tested for spatial significant differences by operating one-way ANOVA analysis and Tukey Duncan's multiple range tests, using IBM SPSS Statistics (version 25). The statistical significance was reported at the $P < 0.05$ level.

RESULTS AND DISCUSSION

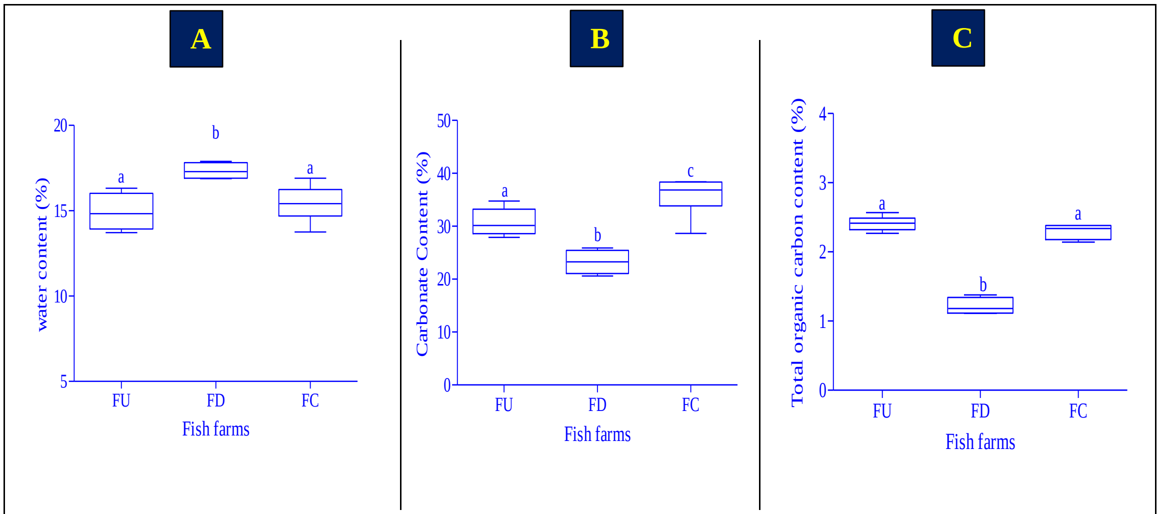
1. Granulometric and chemical characteristics of sediments

The granulometric analysis of sediments is a critical factor affecting the mobility and dispersion of metals in the fish farm environment. The grain size distributions for the sediments of the selected fish farms (FU, FD, and FC) were almost muddy (composed of 100% mud), as sediment of most surrounding drains discharge in Burullus Lake (Younis *et al*., 2014) (Masoud *et al*., 2011). The water content percentages were 14, 18, and 15 % for FU, FD, and FC, respectively (Fig. 2a).

Generally, the carbonate content showed no clear variation between the three fish farms (Fig. 2b), and sediments of FC showed a relatively high carbonate content (about

36 %). However, the present results were lower than those values reported in the surrounding area of Drain-7 and Burullus lagoon (El-Amier *et al.*, 2017; Masoud *et al*., 2011). That decline could be attributed to the muddy sediments, that matched the previously recorded relation between the grain size and carbonate levels in sediment, the finer-grained sediments were mainly poor with carbonate content (Abdel-Moati and El-Sammak, 1997).

Van Geest *et al*. (2011) stated that the total organic carbon (TOC) may influence metal bioavailability, and the accumulation of OC in surface sediments was a significant factor controlling the mineralization processes and the exchange of metals between the sediment and overlying water (Seiter *et al.*, 2004). The current TOC concentrations in the study area showed a narrow range of fluctuation between 1.2 % (at FD) and 2.4% (at FC) as shown in Fig. (2c). it is worthy to mention that the present TOC values were close to those values previously recorded at Burullus, Manzallah, and Bardawel lagoons with mean values of 1.9, 2.5, and 1.0, respectively (Shreadah *et al*., 2012).



Means with the same letter within the same histograme for each metal are not significantly different ($P < 0.05$).

Fig. (2). The chemical characteristics of sediment samples (a) water content (%), (b) carbonate content (%), and (c) total organic carbon (%) of the three selected fish farms (FU, FD, and FC) at Kafr El-Shaikh governorate, Egypt.

2. The abundance of metals in fish farm sediments:

The results of total metals and their bioavailable (mobile) fraction of essential metals (include Cu, Zn, Mn, and Fe) and non-essential metals (include Cr, Cd, and Pb) are presented in Fig. 3. It is worthy to highlight that the concentrations of Cd and Pb were below the detection limits of 0.012 and 0.003 ppm.

The abundance of both total (T) and bioavailable (Bioav) metals was mimic to each other and following a decreasing order Fe > Mn > Zn > Cr > Cu > Cd = Pb (not detected). Statistically, T-metals in the surficial sediments of the study area can be categorized into three main groups according to their levels of concentration. The first group contained extremely elevated levels of Fe with concentrations > 51,000 ppm. The second group contained Mn, Zn, Cu, and Cr with an intermediate concentration from 41 – 790 ppm. The last group contained Cd, and Pb (not detected).

Spatially, sediments of FD exhibited relatively high concentrations of most metals (T- and Bioav-M) when compared with the other farms, while FC showed the lowest levels for all Bioav-M fractions.

A comparison between the present metals' contents in the sediments of the study area with their corresponding ones in standard shale sediments and continental crust is shown in Table 2. The results showed that the concentration means of T-Fe were the highest at FC (57067±135 ppm), while the highest Bioav-Fe fractions were recorded at FD (12899 ±1683 ppm). The majority of Fe levels were more than the freshwater guidelines (EPA, 2006). That elevated levels of Fe were because Fe is the most abundant element in the earth's crust, it ranks the 10^{th} most abundant constituent and represented about 34.6 % of the total mass of the Earth's crust (Edwards 2010). The current values were close to those corresponding values of Fe in the shale and continental crust.

The essential micronutrients such as T-Mn, and -Cu were recorded in the sediments of the three fish farms at levels higher than those freshwater guidelines (EPA, 2006). On other hand, T-Mn content in the three fish farms sediments was lower than those obtained in standard rocks (Table 2). While T-Cu levels in sediments of FU and FC (< 55 ppm) were close to standard rocks, in contrast, the FD attained relatively elevated levels of Cu (average 64 ppm). The last studied essential metal concentrations (Zn) fluctuated between 68 and 156 ppm. However, FC sediments showed the least levels of T-Zn (with an average of 90 ±19 ppm), it was higher than those values of standard rocks and close to the freshwater guideline (EPA, 2006).

Regarding non-essential metal T-Cr, its concentration levels (mean concentration < 81 ppm) were lower than those values of standard rocks (Taylor, 1964; Turekian and Wedepohl, 1961) and more than freshwater guidelines (EPA, 2006). Concerning Pb and Cd, the hazardous metals with potential consequences on the environment, endogenous biota, and human health (Abdel-Satar *et al*., 2022), their concentrations were below the detection limits and lower than guidelines of freshwater and standard rocks (Table 2).

3. Assessment of sediment pollution and ecological risks

Several types of pollution indicators were employed to evaluate the sediment quality of fish farms and the suitability of tilapia culture. The best indices include three single indices, namely, Enrichment Factor (EF), Geo-accumulation Index (I geo), and Contamination Factor (CF). While Pollution Load Index (PLI) is an integrated index that was also applied (Abubakar *et al.*, 2018; Duncan *et al*., 2018).

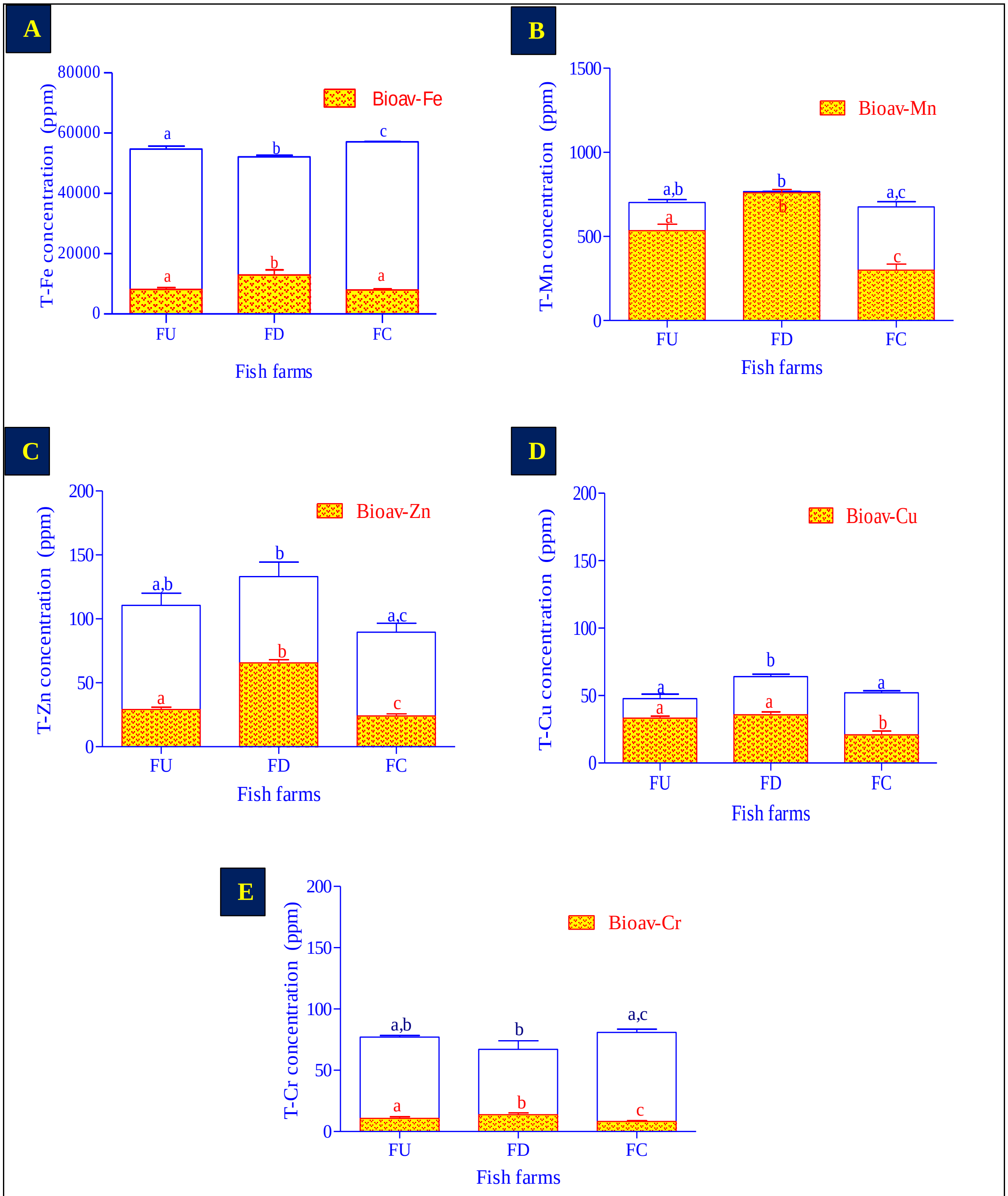
The objective of the indices is to transform complex sediment quality data into understandable and usable information for the public. Several authors (Naifar *et al.*, 2018; Praveena *et al*., 2010; Sabo *et al*., 2013) have proposed pollution impact ranges to convert the calculated numerical results into comprehensive descriptive bands of pollution ranging from low to high intensity.

The results of EF (Fig. 4a) revealed all investigated metals of the surficial sediments of the three fish farms were depleted to mineral levels (EF values < 2). According to the calculated I_{geo} values, most metals showed a class 0 of the geo-accumulation index (values < 0), with unpolluted indication. While I_{geo} values of Zn and Cu at sediments of farms irrigated with agricultural drainage water were of class 1 (values were from 0 to 1), reflecting the condition from unpolluted to moderated polluted (Fig 4b). The order of the computed CF values was Pb (0.0) < Cd < Mn < Cr < Fe < Cu < Zn (1.7), meaning that fish farms sediments were low contaminated by Pb, Cd, and Mn, and from low to moderately contaminated by the rest of metals (Fig 4c). The overall and integrated index of PLI (Fig. 4d) revealed that the three farms were almost at baseline levels (~1).

On other hand, the adverse effects of the pollutant on benthic aquatic organisms were usually determined by classifying the sediments according to sediment quality guidelines (SQGs). SQGs are commonly accomplished to make a preliminary evaluation of sediment toxicity in the absence of direct biological effects data. Where the establishment of biological response of sediment-bound contaminants requires an assessment of their toxicity and bioaccumulation.

An example of SQGs was suggested and referred to as the threshold effect level (TEL) and the probable effect level (PEL) which provides a reliable basis for assessing sediment quality conditions in aquatic ecosystems (MacDonald *et al.*, 2000). Applying the SQGs to sediments of the present study (Table 2), it was shown that the three fish farms had values of Zn, and Mn below the TEL level which means the harmful effects due to Zn and Mn are unlikely to be observed. Unlike, Cu, and Cr, their concentration values were above TEL and below PEL which reflect adverse effects that may occasionally occur for sensitive organisms, but only a slight risk may have taken place.

Moreover, Persaud *et al*., (1993), Batts and Cubbage (1995), Suter and Tsao (1996), and Jones and Suter II (1997) summarized ecological sediments guidelines and define three levels of chronic, long-term effects on benthic organisms as 1) No-Effect Level: no toxic effects have been noticed on fish or sediment welling organisms; as well as no predicted biomagnification through food chain; 2) Lowest-Effect Level: reveals a contaminate level that can be tolerated by most benthic organisms; 3) Severe-Effect Level: shows the expected distinct disturbance of sediment-dwelling organisms. Consequently, comparing the results of the present study with those guidelines (Table 2), it was noticed that majority of studied metals values were between the lowest- and severe- effect levels. While Zn values were below the lowest effect levels, addition to Cd, and Pb values showed no-effect levels.



Letters in blue and red colours refers to one-way ANOVA reults for total- and bioavailabale- metal farction, respectively. Means with the same letter within the same histograme for each metal are not significantly different ($P < 0.05$).

Fig. (3). The mean concentration values of total metals and its bioavailable fraction of (A) iron, Fe, (B) manganese, Mn, (C) zinc, Zn, (D) cupper, Cu, and (E) chromium, Cr of the three fish farms sediments.

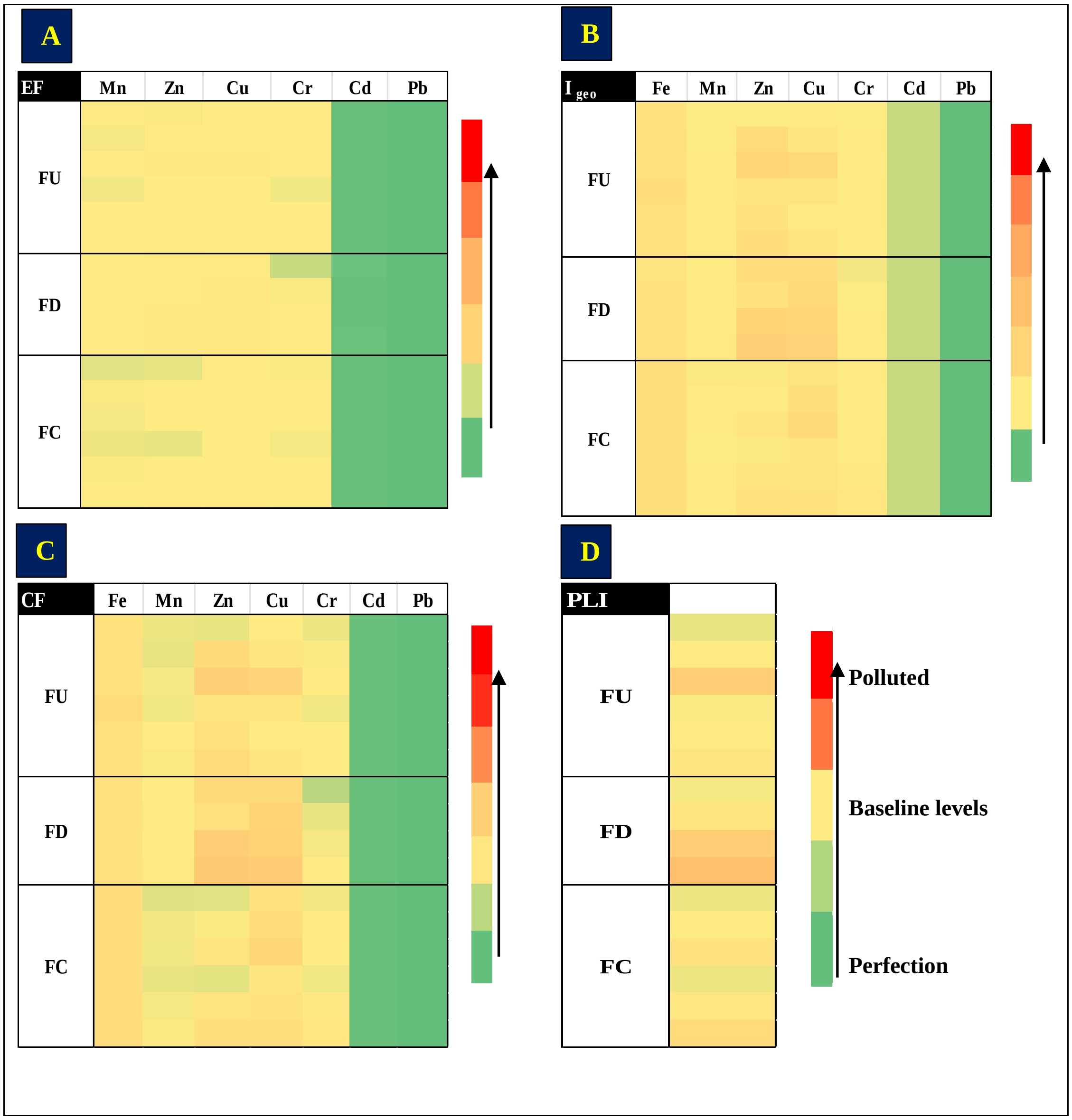


Fig. (4). Heat map shows different pollution indices (a) enrichment factor, EF, (b) geo-accumulation factor, I_{geo}, (c) contamination factor, CF, and (D) pollution loading index, PLI, of the study area.

Table 2. Comparison between mean levels of total-metals concentration in the study area and sediment quality benchmarks for freshwater.

Sediment quality benchmarks	Category	Cu	Pb	Cd	Cr	Zn	Mn	*Fe*	Reference
Fish farms	**FU**	48	BDL	BDL	77	111	702	54670	Present study
	FD	64	BDL	BDL	67	133	766	52137	
	FC	52	BDL	BDL	81	90	676	57076	
Standard rocks	**Shale**	45	20	0.3	90	95	850	47200	(Turekian and Wedepohl, 1961)
	Continental crust	55	12.5	0.2	100	70	950	56300	(Taylor, 1964)
Freshwater sediments	**TEL**	28	34	0.6	56	159	1673	-	ARCS (EPA, 1996)
	PEL	78	396	11.7	159	1532	1081	-	
	Low	16	31	0.6	26	120	460	-	Ontario MOE (Persaud *et al*., 1993)
	Severe	110	250	10	110	820	1110	-	
	-	31.6	35.8	0.99	43.4	121	460	20000	(EPA, 2006)

BDL= Below detection limit (<0.012, and < 0.003 ppm for Cd, and Pb, respectively)

ARCS = Assessment and Remediation of Contaminated Sediments Program; Ontario MOE = Ontario Ministry of the Environment.

TEL = threshold effects level PEL= probable effects level NEL = No Effect level

Low = lowest effect level and is the 5th percentile of the screening level concentration; Severe = severe effect level and is the 95th percentile of the screening level concentration

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