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Research Article
Published: 13 Apr, 2018

SF Journal of Environmental and Earth Science

# Occurrence and Risk Assessment of 22 Inorganic Contaminants in Urban Sewage Sludge and Lake's Sediment in the Republic of Benin

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## Abstract

The occurrence and risk assessment of 22 inorganic elements have been evaluated in urban sewage sludge and Lake's surface sediments in the Republic of Benin. Broad range concentration of elements was detected [1.43E-02 mg $kg^{-1}$ (Tl) to 2.68E+04 mg $kg^{-1}$ (Fe)] in sediment and [3.00E-02 mg $kg^{-1}$ (Tl) to 4.77E+04 mg $kg^{-1}$ (P)] in the sludge samples. The calculation of enrichment factor (EF) revealed that majority of studied elements were enriched in both sediment and sludge; and could reach extreme enrichment ($1 \leq EF < 2300$ and $1 \leq EF < 3000$) in the sediment and sludge samples, respectively. Principal components analysis showed that anthropogenic influence would explains such situation. Further results with respect the degree of contamination (DC) of both and sediments showed that sediments were within moderate DC ($DC < 6$) (H1, LN1, LN2) and considerable DC ($12 < DC < 24$) (LN3 and LN4). Sludge samples ($S_i$) and one sediment sample by a hospital effluent discharge point (H2) were within very high DC ($DC > 24$). Summing up, the overall pollution state of sludge and lake's sediments is of concern. It also raises concern on the quality of the treatment process in the WWTP and it constitutes a potential threat to the aquatic ecosystem and limits the valorization of the sewage sludge.

**Keywords: Sediment; Lake; Sewage sludge; Inorganic pollutants; Risk assessment; Republic of Benin**

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Received Date: 01 Mar 2018
Accepted Date: 09 Apr 2018
Published Date: 13 Apr 2018

Citation: Ifon BE, Yessoufou A, Suanon F, Togbe FCA, Frank Yovo, Dimon B, et al. Occurrence and Risk Assessment of 22 Inorganic Contaminants in Urban Sewage Sludge and Lake's Sediment in the Republic of Benin. SF J Environ Earth Sci. 2018; 1(1): 1010.

ISSN 2643-8070



## Introduction

Controlling pollution in the environment is among the top list of environmental challenges. The pollution of water reserves including rivers, lakes and sea has reinforced the need of protecting natural resources and ecosystem. The need of strict control of discharging wastes into the environment is of utmost importance. Inorganic pollutants, such as heavy metals, are of special concern because of their persistence [1]. Unlike organic contaminants, heavy metals are not biodegradable [2] and tend to accumulate in living organisms such as plants once reach the soil or fishes in the aquatic environment. Furthermore, at their higher concentrations metals threaten human health since many heavy metal ions are known to be toxic or carcinogenic [3,4]. Heavy metals can be responsible for various health hazards in humans. For example according to [5], Cr, Cu, and Zn can cause non-carcinogenic health hazards such as neurologic impairment, headache and liver disease, when they exceed their safe threshold values. It has also been reported that chronic exposure even at low doses of cancer-causing heavy metals may cause different types of cancer [6]. Occupational exposure to dusts and mists containing hexavalent chromium increased the risk of lung cancer death [7]. The occurrence of inorganic pollutants in soil, sediments and water could be attributed to numerous anthropogenic activities such as urbanization, industrialization, transportation, abusive use of synthesized fertilizer, insecticide, and pesticide, improper disposal of sewage and solid wastes as well as natural processes such as precipitation inputs, erosion and weathering of crustal materials [8,9]. Improper treatment of industrial wastewater is also known for the release of different heavy metals such as zinc, copper, nickel, cadmium, lead and chromium etc. into the environment [9-11].

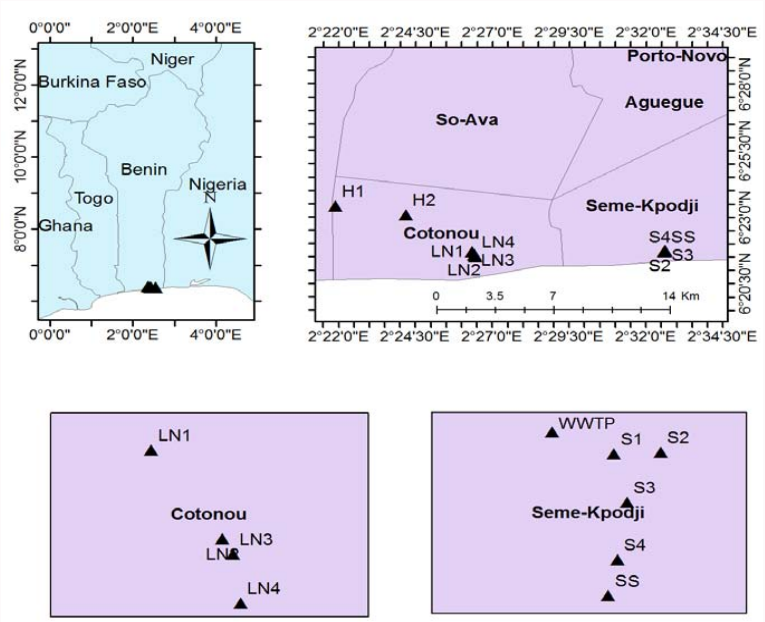


**Figure 1:** Maps show sampling area and locations: H1, H2, LN1, LN2, LN3 and LN4 are sediment samples from Lake Nokoué. S1, S2, S3 and S4 are treated sludge samples from 4 different discharge points (each from a treatment basin). SS is the soil sample collected by the sea side where the WWTP wastewater is discharged via direct connection.

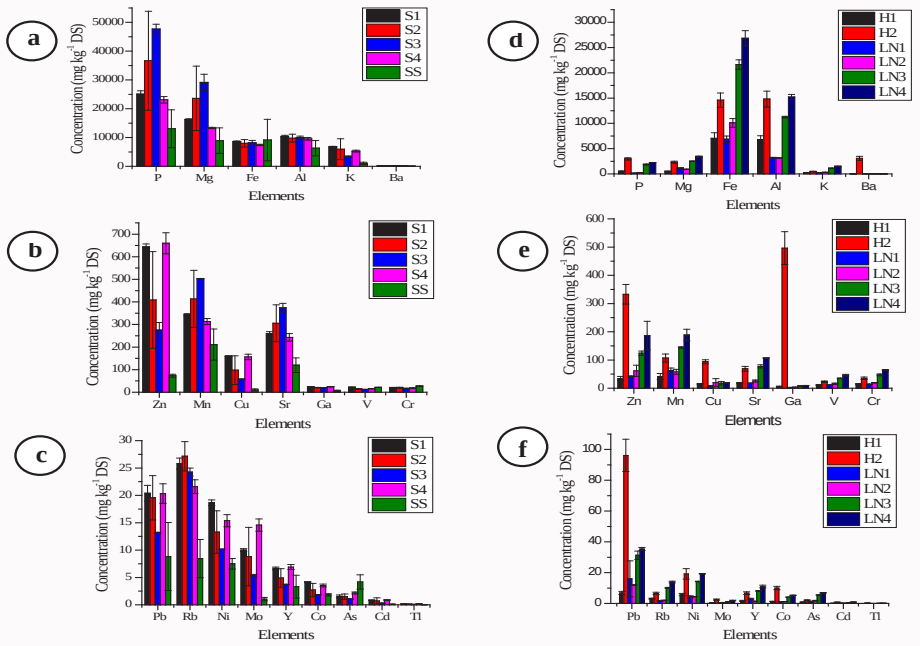


**Figure 2:** Concentrations of inorganic elements in sludge (a, b, c) and sediment (d, e, f) samples.

Regulations to limit or minimize the amount of pollutants before being discharged are getting more stringent and the need of better technology to meet the regulations has become a great challenge. Some developing countries are more vulnerable to environmental pollution as waste treatment technologies and management to encounter the pollution are less available. However, the pollution status in those countries is mostly unclear. It is the case of some countries in Africa where the management of wastes still remains a great challenge to decision makers. Wastewater treatment plants (WWTPs) do not exist in most of cases, and if it is available, the treatment capacity is often limited thus putting in doubt the treatment efficiency. In the Republic of Benin (West Africa), only one WWTP exists, and it is setup in the biggest and most populated town (Cotonou) of the country. The only existing stabilization treatment plant, managed by a private company SIBEAU, is overloaded. It was built to treat wastewater from 0.3 million inhabitants. Nowadays, the population has grown and the WWTP has to treat wastewater from over 1 million inhabitants [12]. The effluent quality is poor and does not meet any quality standards [13]. Ninety six percent of survey respondents recognized that the sanitation situation in the city was not good. It has to be improved by providing an appropriate and sustainable sanitation management system [13]. Despite all the mentioned observations, the wastewater

effluent from the WWTP is continuously discharged in the natural water reserves such as the Lake, sea; and sewage sludge is used as soil amendment mostly for garden. Some hospitals directly discharged their wastes in the natural reserves without good pre-treatment. To be noticed, the lake (e.g. Lake Nokoué) serves as the source of provision in Crustacea and fishes. Coastal inhabitants used to fish in this lake for their own and sell part of the products. Such situation poses environmental eco-toxicological risks and threatens human health as heavy metals can accumulate in the sediment and enter food chain through the uptake by aquatic organisms. In-depth investigations to assess the risks of pollution and suggest potential solution are necessary. Therefore, this study, which follow after our previous report with respect precious and rare earth elements in sediments and sewage sludge in the republic of Benin [14] aims to (i) evaluate the pollution status of sediment from Lake Nokoué and treated sewage sludge from the urban WWTP in the republic of Benin with respect 22 inorganic elements including 19 metals, 2 metalloids and phosphorus, and (ii) assess the eco-toxicological risks related to the pollutant contamination. To our knowledge, this is the first comprehensive investigation on broad range of inorganic pollutants in Lake's sediment and urban sewage sludge in the Republic of Benin.

## Materials and Methods

### Study area and sampling

A total of 11 samples were collected from different sources including sediments from Lake Nokoué (H1, H2, LN1, LN2, LN3 and LN4) and dewatered sludge from the urban WWTP (S1, S2, S3 and S4) and a sea soil sample (SS) in Cotonou town in the Republic of Benin (West Africa) as shown in Figure 1. Samples from H1 and H2 were nearby the discharging point of the effluent from hospitals (Hi) and SS sample was that of effluent discharging point of the WWTP by the sea side. The WWTP receives domestic wastewater and fecal sludge of over 1 million inhabitants. The lagoon system is adopted for the treatment. It includes pretreatment (screening and grit removal), primary treatment and secondary treatment. The facility consists of two series of three waste stabilization basin, which receive effluent from an anaerobic pond. The ponds were designed for the following purposes: (i) anaerobic ponds used for settling and degradation of organic matter, (ii) optional basins used for the removal of BOD and pathogens, and (iii) maturation basins, whose essential function is to effectively reduce the quantity of fecal bacteria. Sludge samples were collected after maturation [15]. The overall treatment process is described in the Figure S1 in the Supporting Information (SI).

### Samples collection and pretreatment

Sediments and sludge samples were collected in June 2016 in 50mL glass bottles. In the lake, samples were taken using a 2-meter cylindrical tube (≈2.5cm diameter) at 5-10 cm depth. Collected samples were packed and sent to China at the Institute of Urban Environment, Chinese Academy of Sciences. After freeze-drying, samples were grounded in the glass mortar and sieved through a <2mm mesh size, the pulverized samples were used for physicochemical characterization and inorganic elements determination.

### Physicochemical characterization

Physicochemical characteristics including pH, electrical conductivity (EC), organic matter content (OM), and elemental determination (total carbon, nitrogen and sulfur) were determined. The pH and EC were determined in a dissolved sediment/sludge sample in milliQ water at ratio 1:10 (m:v) [16]. The reading of pH

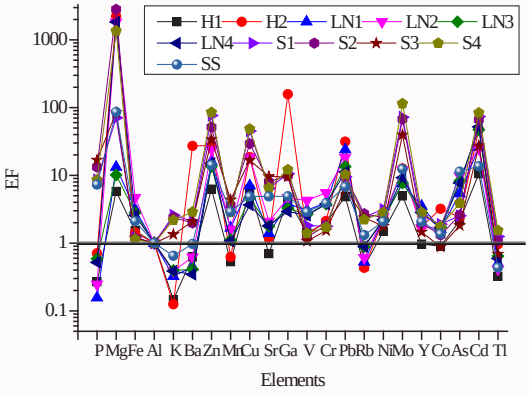


**Figure 3:** EF of each inorganic element.

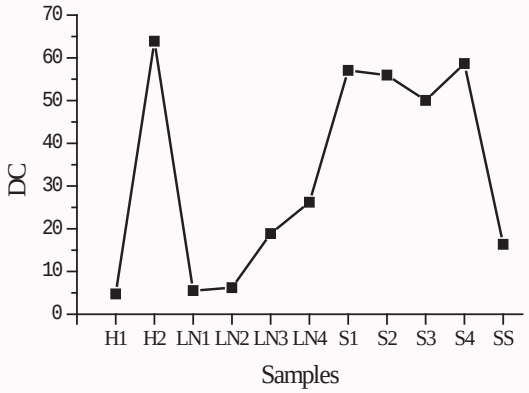


**Figure 4:** Degree of contamination in each sample.

and EC values were performed by a multi-parameter meter (HACH, HQ40d). OM was determined via weight loss by ignition at 500°C. Elemental analysis was performed by pyrolysis using macro elemental CNHS/O Analyzer (Vario MAX, Elementar, Germany).

### Vessel cleanup

Sample digestion was carried out in a polytetrafluoroethylene tube which was pre-cleaned by leaching with hot 10mL aqua regia (mixture of $HNO_3$: HCl at the ratio of 3:1) at 90°C for a minimum of 2h under microwave as recommended by the [17]. All volumetric wares were carefully acid washed in 10% nitric acid and then in 10% hydrochloric acid and finally rinsed with milliQ water. The acids were analytical grade (Merck KGaA, Darmstardt, Germany).

### Sample digestion

Zero point one gram of sludge/sediment sample was weighted in a polytetrafluoroethylene (PTFE) digestion tube followed by adding 12mL of freshly prepared aqua regia. The mixture was brought under microwave for hot digestion at 180°C at high pressure [17]. The running program is as follows: 10min for temperature to rise from ambient temperature to 180°C and then 45min residence time at 180°C. After complete digestion, samples were cooled, filtered and collected in a 50mL plastic centrifugation tube, and then diluted to 45 mL using milliQ water. Samples were stored at 4°C prior analysis. For quality control, an instrument blank, procedural blank and the certified reference material with known concentrations of elements (GBW07309, GSD-9, Inspection and Quarantine of the People's Republic of China) were applied for each batch and the recovery

ranged between 92.6% and 105%. All experiments were performed in triplicate.

### Inorganic elements detection

The quantification of inorganic elements in sludge/sediment samples was performed using inductively coupled plasma mass spectroscopy (ICP-MS, Agilent 7500CX). Stock solution (GNM-M27867-2013) of inorganic elements was purchased from National Center of Analysis and Testing for Nonferrous Metals and Electronic Materials (NCATN). The target elements include Ag, Al, As, Ba, Ca, Cd, Co, Cr, Cs, Cu, Fe, Ga, K, Mg, Mn, Mo, Na, Ni, P, Pb, Rb, Re, Sb, Sr, Tl, V and Zn. All the quantified data calculation was based on the sediment/sludge dry weight.

### Pollution indices

To evaluate the pollution state of the sediment and sludge and assess the eco-toxicological risks, parameters such as enrichment factor (EF), pollution index (PI) and the degree of contamination (DC) where evaluated [18,19].

**Enrichment factor (EF):** EF is a useful approach to determine the level and influence of anthropogenic sources on metal or element pollution in the environment using a normalizing element in order to assuage the variations produced by heterogeneous sediments [19,20]. Different elements (Al, Fe, K, Li, Sc, Ga, Zr and Ti) are often used as the normalizing element [21,22]. In the present study, EF of an element (Y) was calculated relative to the average composition of crustal abundance [23,24] using Al as the reference element according to the Equation 1. Al is selected as a normalizing element because not only it is one of the major elements in sediments, but it is also one of the abundant elements in earth's crust assuming that it has little anthropogenic impact.

$$EF=\frac{\left(\frac{Y}{Al}\right)Sample}{\left(\frac{Y}{Al}\right)Crustal} \quad \text{(Eq. 1)}$$

where (Y/Al) sample and (Y/Al) Crustal refer, respectively, to the ratios of average concentrations (mg $kg^{-1}$) of the target metal and Al in the studied sample and continental crust of earth [25]. According to [26], the EF values are interpreted as follows: (EF < 1)-no enrichment, (EF < 3)-minor enrichment, (3 ≤ EF < 5) moderate enrichment, (5 ≤ EF <10)-moderately severe enrichment, (10 ≤ EF < 25)-severe enrichment, (25≤ EF < 50) very severe enrichment, and (EF ≥ 50) extremely severe enrichment.

**Degree of contamination (DC):** DC is used to assess the extent of multi-metal or element contamination in the sediment, soil or sludge, which is different from the single-element factor. According to [27], DC could be calculated according to the Equation 2:

$$DC=\sum_{j-1}^{n}(PI)_j \quad \text{(Eq. 2)}$$

where *n* is the number of elements and (PI) is the pollution index calculated according to [27] via Equation 3.

$$PI=\frac{Cj\ sample}{Coj\ Crustal} \quad \text{(Eq. 3)}$$

where Cj is the concentration of a given element j in the studied samples (mg $kg^{-1}$), and Coj is its corresponding upper crustal concentration (mg $kg^{-1}$). According to [27], DC level was classified as follows: (DC ≤ 6) low degree of contamination, (6 < DC ≤ 12) moderate degree of contamination, (12 < DC ≤ 24) considerable degree of contamination, (24 < DC ≤ 24) very high degree of contamination.

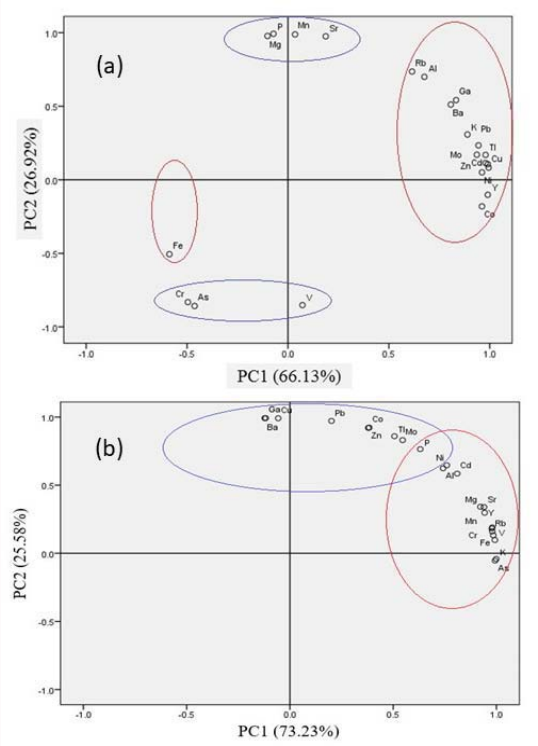


**Figure 5:** PCA plot of inorganic elements in the sludge (a) and sediment (b).

### Statistical analysis

Statistical analysis including correlation and principal component analysis (PCA) were performed by SPSS 20.0 and graphical plotting was performed by Origin software version 9.0.

## Results and Discussion

### Physicochemical characterization

Results of studied physicochemical parameters are summarized in Table 1. From Table 1, it can be seen that both sediment and sludge samples exhibited a near neutral pH. It can also be noticed that sludge samples possessed higher EC (3.00mS $cm^{-1}$ ≤ EC ≤ 4.1mS $cm^{-1}$) compared to sediment samples, which EC was lower than 3mS $cm^{-1}$. This could be explained by higher concentrations of charged substances in the sludge samples [28]. The sample collected by the sea side had the highest EC (5.95mS $cm^{-1}$). Furthermore, as it was expected, sludge samples (S1-4) were very rich in OM, carbon and nitrogen contents contrary to sediment samples (H1, H2, LN1, LN2, LN3 and LN4) and the sea soil sample. This could be explained by the fact that sludge is generally constituted of microorganisms and various organic matters contrary to sediment and sea soil which abound in sand.

### Inorganic elements in sludge and sediment samples

Results of quantitative analysis of inorganic elements in the sludge and sediment samples are shown in Figure 2. Data revealed broad range concentrations of the elements in the samples.

**Inorganic elements in sludge:** P, Mg, Fe, Al and K were predominantly detected in the sludge (S1-4) and in the sea soil (Figure

**Table 1**: Sediment and sludge physicochemical characterization.

| Sample | pH | EC (mS cm$^{-1}$) | C (%) | N (%) | S (%) | OM (%) |
|---|---|---|---|---|---|---|
| H1 | 6.9 | 2.23 | 0.634 | 0.04 | 0.21 | 1.8 |
| H2 | 6.8 | 2.71 | 8.81 | 0.51 | 0.37 | 14.8 |
| LN1 | 6.4 | 2.74 | 0.274 | 0.03 | 0.17 | 1.04 |
| LN2 | 7.2 | 2.81 | 0.28 | 0.04 | 0.13 | 1.35 |
| LN3 | 6.7 | 2.53 | 1.38 | 0.09 | 0.18 | 2.61 |
| LN4 | 7.3 | 2.02 | 1.63 | 0.08 | 0.17 | 3.8 |
| S1 | 7.1 | 4.04 | 32.6 | 3.2 | 1.1 | 66 |
| S2 | 7.5 | 3.27 | 29.7 | 3.01 | 0.91 | 65.8 |
| S3 | 6.9 | 3.73 | 26.2 | 2.68 | 0.48 | 62.3 |
| S4 | 7.8 | 3.15 | 37.2 | 3.55 | 1.15 | 68.2 |
| SS | 7.6 | 5.95 | 2.34 | 0.88 | 0.15 | 6.97 |

2a). Their concentrations ranged (13052 - 47700, 8917 - 29158, ≈ 10000, ≈ 12000 and 1042 - 6802) (mg kg$^{-1}$) for P, Mg, Fe, Al, K respectively. S2 and S3 exhibited the highest concentrations of P and Mg [S3 (47700mg kg$^{-1}$and 29157mg kg$^{-1}$) and S2 (36666mg kg$^{-1}$and 23620mg kg$^{-1}$)]. Our results are comparable to that reported by [29] for most of the element in the sewage sludge in China. P is known as the major nutrient in the synthetic fertilizer; as a consequence, the current sludge would be a good candidate for soil amendment [30,31]. However, the usability of a sewage sludge as soil amendment is dependent to the content of environmental hazard elements (e.g. heavy metals (Cd, Cr, Pb, Zn, Cu, Ni etc.), emerging contaminants etc.) in the sludge; as this would lead to their accumulation in soil and compromise food chain [32,33]. Ba, Zn, Mn and Sr concentrations were lower compared to the above mentioned elements in the sludge samples (Figure 2a and 2b). The concentrations ranged 50 – 760 mg kg$^{-1}$, 200 – 500 mg kg$^{-1}$, 100 – 400 mg kg$^{-1}$ and 40 - 210 mg kg$^{-1}$ for Zn, Mn, Sr and Ba, respectively. Cu concentration was low (10 – 150 mg kg$^{-1}$), elements including Cr, V, Ga, Pb, Rb, Ni, Mo, Y, Co, As, Cd and Tl were at very low concentrations (≤ 30mg kg$^{-1}$) (Figure 2b and 2c). The concentrations of Cr, V, Ga, Cd and Tl were quite close in all sludge samples. Summing up, the environment elements of great concerned such as As, Cd, Co, Cu, Cr, Ni, Pb and Zn, concentrations ranged 1.06 – 4.16, 0.30- 0.87, 1.84- 4.13, 57.28- 159.34, 17.14- 20.36, 10.15- 18.65, 13.15- 20.39 and 275.44 - 659.70 mg kg$^{-1}$, respectively in the sludge, while it were: 4.16, 0.01, 1.81, 10.47, 27.36, 7.49, 8.83 and 73.17 mg kg$^{-1}$, respectively in the sea soil sample. These values were considerably lower compared to those reported before in sludge samples in China [29,34-37], and that reported by [38] in sludge samples in U.S (Table 2). As a matter of fact, the detected concentrations values meet with U.S. Environmental Protection Agency recommendations which regulate their limit in sludge to 75, 85, 3000, 4300, 420, 75 and 7500 mg kg$^{-1}$ for As, Cd, Cr, Cu, Ni, Pb and Zn, respectively [39,40].

**Inorganic elements in sediment samples:** In sediment samples, the major detected inorganic elements (≥ 1000 mg kg$^{-1}$) remained the same as those in the sludge (Figure 2d). However, contrary to the sludge samples, Fe and Al were the elements with the highest concentrations in the sediment. Their concentrations ranged 6887-26833 and 3215–15228 mg kg$^{-1}$ for Fe and Al, respectively. Fe and Al concentrations in the sediment sampling site follow the following order: LN4 >, LN3 > H2. P, Mg, K and Ba, concentrations vary from 100 to 3500 mg kg$^{-1}$ in all sediment samples. The remaining elements' concentrations were generally much lower [from 0.2mg kg$^{-1}$ (for Tl) to 500mg kg$^{-1}$ (for Ga)] (Figure 2e and 2f). Metals of environmental great concern (e.g. As. Cd, Co, Cu, Cr, Ni, Pb and Zn) were < 500mg kg$^{-1}$ for Zn, Cu and Cr and < 25mg kg$^{-1}$ for As, Cd, Co, Ni and Pb in the sediment. Compared to others studies, our results with respect heavy metal concentrations were relatively higher than those reported by [48] in a river sediments in Nigeria, [41] in surface sediments in Iran, and those reported on surface sediments in China [42,43,45] (Table 2). The comparison of our results to the standard limits according to Consensus-Based Sediment Quality guide line (CBSOG) [44] (Table 2), showed that the concentrations of the elements did not fully meet with the recommendation. Anthropogenic sources such as the effluent from industrial facilities and sewage treatment plants, domestic wastewater and hospital waste discharge [46,49,50], would contribute to the higher concentrations of inorganic elements in the lake's sediments. However, more in-depth survey is needed to figure out which are the major sources contributing to the occurrence/accumulation of contaminants in the sediment.

## Correlation between inorganic elements and environmental factors

Pearson correlation between inorganic element concentrations and environmental factors including pH, EC, C, N, S and OM in both sediment and sludge samples has been performed (Table S1-2).

In the sewage sludge, the correlation between environmental factors and studied inorganic elements were noted. Indeed, EC, C, N, S and OM developed a strong correlation with most of the target inorganic elements in the sludge. For example, EC showed strong and significant correlation with Fe, Ba, Ga, Cr, and Rb ($0.77 \leq r^2 \leq 0.91$, $p< 0.05$), and marginal correlation with the rest of the inorganic elements ($0.32 \leq r^2 \leq 0.85$, $p> 0.05$). C, N, S and OM positively correlated with the majority of target inorganic elements (Table S1), while negatively correlated with Fe and As. pH, only showed marginally and negative correlation with elements such as P, Mg, Mn, Sr and As ($0.34 \leq r^2 \leq 0.46$, $p > 0.05$). Strong correlation among some target inorganic elements was also observed, which suggested the possible common sources of the elements in the sludge. However, several elements such as P, Fe, Mn, Sr and V, did not show correlation with most of other target inorganic elements.

In the sediment, EC developed marginally negative correlation with P, Al, Ni and Cd ($0.26 \leq r^2 \leq 0.57$, $p > 0.05$), and strong negative correlation with Mg, Fe, K, Mn, Sr, V, Cr, Rb, Y and As ($0.70 \leq r^2 \leq 0.84$, $p < 0.05$). C, N, S and OM developed significantly positive correlation with some of the target elements (Ba, Zn, Cu, Ga, Pb and Co) in the sediment (Table S2). The correlations among most of the target inorganic elements in the sediment were significant ($p < 0.05$). Particularly, P, Mg, Al, Sr and Cr showed correlation with almost all other target elements. This suggested that inorganic elements in the sediment could be originated from similar and common sources.

## Environmental risk assessment

The evaluation of pollution indices permitted us to better understand the pollution state of the lake's sediments and sewage sludge. The results of analysis are shown below.

**Enrichment factor (EF):** According to [25], EF< 1 indicates no enrichment and the given element would be originated from crustal materials or from natural weathering processes, and EF > 1 indicated that a significant portion of the element was originated from anthropogenic sources, suggesting the possibility of pollution. In the current study, EF was evaluated for each element using Al as the normalizing element. Results showed that by some inorganic

**Table 2:** Comparison of the detected concentrations of heavy metals with the recent reports (concentration in mg $kg^{-1}$).

| Matrix and country | As | Cd | Co | Cr | Cu | Ni | Pb | Zn | References |
|---|---|---|---|---|---|---|---|---|---|
| **Surface Sediment, Republic of Benin** | **0.85-6.85** | **0.078-0.867** | **1.08-10.0** | **19.5-64.1** | **7.72-93.8** | **4.33-19.3** | **6.76-96.2** | **34.5-333** | **Present study** |
| Surface Sediment, Nigeria | na | 0.550- 1.142 | na | 9.57-15.95 | na | 9.15-13.96 | 2.00-8.9 | 91.5-121.6 | [41] |
| Surface Sediment, PR China | 1.94-13.67 | 0.03-0.13 | na | Oct-85 | 1-39.5 | na | Nov-56 | 13-125 | [42] |
| Surface Sediment, PR China | 3.4–13.6 | 0.020–0.240 | na | 11.6–76.2 | 9.6–35.6 | 3.5–35.8 | 22.6–43.7 | 12.9–94.7 | [43] |
| Surface Sediment, PR China | 4.9-67.8 | 0.10-2.00 | na | 17.7-182.3 | 15.2-121.4 | 11.2-82.3 | 11.4-76.7 | 30-161 | [44] |
| Sediment, Iran | na | 0.17-2.1 | na | 57.8-141 | 19.9-47.9 | 69.8-196 | 17.6-94.6 | 45.9-107 | [45] |
| **CBSOG SQG *** | **<9.8** | **< 0.99** | **na** | **< 43** | **< 25** | **< 23** | **< 40** | **< 90** | **[46]** |
| **Sewage Sludge, Republic of Benin** | **1.06-4.17** | **0.094-0.873** | **1.82-3.54** | **17.1-27.4** | **10.5-157** | **7.49-15.4** | **8.83-20.3** | **73.2-497** | **Present study** |
| Urban Sewage Sludge China[a] | na | 64.1 | 73.4 | 604.1 | 1102.2 | 483.9 | na | 2060.3 | [38] |
| Industrial Sewage sludge China[b] | na | 172,300 | na | na | 237 | 22,225 | na | 1700 | [39] |
| Urban Sewage Sludge , China[c] | na | na | na | 293.7 | 181.7 | 114.8 | 40.3 | 1453.9 | [35] |
| Urban Sewage Sludge, US[d] | 10.19 | 3.62 | 3.5 | 57.84 | 435.7 | 27.8 | 24.49 | 620.1 | [39] |
| **USEPA, NRC** | **75** | **85** | **na** | **3000** | **4300** | **420** | **75** | **7500** | **[40,48]** |

**Note**:
*Consensus-Based Sediment Quality guide line (CBSOG SQG 2003).
[a]Municipale wastewater treatment plant receiving both domestic and industrial wastewater.
[b]Industrial wastewater treatment plant.
[c,d]Municipal wastewater treatment plants receiving only domestic wastewater.
na: not available.

elements were enriched in the sediment and sludge samples (Figure 3). Mg was the most enriched element in the sludge samples and sediment samples. The level of enrichment was extreme (EF > 50), except in H1, LN1 and LN3 where Mg was severely enriched (EF = 5.8, 13.2 and 10.1, respectively). Regarding P, the EFs were 8.5, 8.5 and 7.3, respectively in S1, S4, and SS, which indicated moderately severe enrichment. In S2 and S3, the EF values (13.3 and 16.8, respectively) showed that P enrichment was very severe. Fe, K, Ba, Mn, V, Cr, Rb, Ni, Y, Co and Tl showed minor enrichment in all samples (EF < 3), except Ba which was very severely enriched in H2 (EF = 27.2). Heavy metals Zn, Cd and Cu showed much higher enrichment in the sludge samples compared to sediment samples. Indeed, the EFs of Zn, Cd and Cu in sludge samples could reach very severe enrichment to extremely severe enrichment. Otherwise, Pb showed higher enrichment in the sediment samples compared to sludge samples. It showed severe to very severe enrichment in sediment samples. Meanwhile, Arsenic (As) exhibited moderately severe enrichment in the sediment samples (LN1, LN2, LN3 and LN4) and severe enrichment in SS sample (EF = 11.2). Contrary to sediment samples, showed minor enrichment in the studied sludge samples. In summary, the overall results of the studied sludge and sediment samples revealed that investigated elements were enriched in the sludge and that anthropogenic activities strongly influenced their occurrence in the Lake's sediment. We further the pollution risk assessment of the sludge and sediment by evaluating the degree of contamination (DC) discussed below.

**Degree of contamination (DC):** According to the results shown in Figure 4, DC values of sediment samples H1, LN1 and LN2 were around 6, and therefore, those sediment samples were at low degree of contamination according to [26]. DC values of LN3, LN4 and SS were between 12 and 24. According to [26], those three samples showed considerable degree of contamination. Finally, each of H2, S1, S2, S3 and S4 has a DC value greater than 24, indicating a high degree of contamination. As a consequence, necessary measures should be taken to (i) restore the lake, (ii) regulate urban and hospital wastewater and sewage sludge treatment and management, and

**Table 3:** PC analysis loadings of metals in sediments of Nokoué Lake and sewage sludge.

| Elements | Sewage sludge | | Sediment | |
|---|---|---|---|---|
| | PC1 | PC2 | PC1 | PC2 |
| Cu | **0.99** | 0.09 | -0.06 | **0.99** |
| Ni | **0.98** | 0.12 | **0.76** | **0.65** |
| Co | **0.98** | -0.12 | 0.38 | **0.92** |
| Y | **0.98** | -0.11 | **0.94** | 0.3 |
| Zn | **0.98** | 0.1 | 0.38 | **0.92** |
| Cd | **0.97** | 0.11 | **0.81** | **0.59** |
| Tl | **0.96** | 0.14 | **0.51** | **0.86** |
| Pb | **0.93** | 0.24 | 0.2 | **0.97** |
| K | **0.9** | 0.37 | **1** | -0.04 |
| Mo | **0.89** | 0.09 | **0.55** | **0.83** |
| Ga | **0.81** | **0.52** | -0.12 | **0.99** |
| Ba | **0.76** | 0.44 | -0.12 | **0.99** |
| Mn | 0.02 | **0.99** | **0.98** | 0.18 |
| Mg | -0.11 | **0.99** | **0.92** | 0.34 |
| P | -0.09 | **0.98** | **0.63** | **0.76** |
| Sr | 0.17 | **0.97** | **0.94** | 0.34 |
| As | -0.46 | **-0.89** | **0.99** | -0.05 |
| Rb | **0.61** | **0.77** | **0.98** | 0.16 |
| Cr | -0.45 | **-0.76** | **0.98** | 0.19 |
| Al | **0.67** | **0.73** | **0.74** | **0.62** |
| V | 0.15 | **-0.7** | **0.99** | 0.1 |
| Fe | **-0.5** | -0.33 | **0.98** | 0.13 |
| Initial Eigen value | 14.55 | 5.92 | 16.11 | 5.63 |
| %o of Variance | 66.13 | 26.92 | 73.23 | 25.88 |
| Cumulative % | 66.13 | 93.04 | 73.23 | 98.81 |

**Note**: Data in bold are the main contributors to PC.

finally (iii) avoid discharging untreated wastewater and sewage sludge into natural receptors such as the lake, sea, soil etc.

### Principal component analysis (PCA)

To better elucidate the potential sources of inorganic elements in the surface sediments of Lake Nokoué and sewage sludge, PCA was separately performed on sludge and sediment samples. PCA reduced the number of variables to two principal components (PCs) for both sludge and sediment (Table 3 and Figure 5).

In the sludge, 93.04% of the variance was explained (Table 3 and Figure 5a). PC1 explained 66.13% of the total variance and was largely dominated by Cu, Ni, Co, Y, Zn, Cd, Tl, Pb, K, Mo, Ga, Ba, Rb, Cr, Al and Fe. PC2, which explained 26.92% of the total variance, is dominated by Ga, Ba, Mn, Mg, P, Sr, As, Rb, Cr, and V. Al is known as a petrogenetic element and constitutes one of the earth crust's major components, andits concentration is rarely influenced by anthropogenic activities [45,51]. In addition, as aforementioned, Al developed a strong correlation with most of the elements loaded on PC1 ($0.25 \leq r^2 \leq 0.99$). This suggests that elements loaded on PC1 would have similar source to Al. However, EF analysis showed that most of those elements were enriched in the sludge, which also suggested the influence of anthropogenic sources on the occurrence of these elements in the sludge [43]. Mn, Mg, P, Sr, As, Cr and V were loaded on the second component (PC2). EF analysis revealed that those elements, especially Mg and P, were highly enriched in the sludge, which implied that anthropogenic activities were likely the major source responsible for their accumulation in the sludge.

In the sediment, 98.81% of total variance was explained. PC1 explained 73.23% of the total variance and loaded Ni, Y, Cd, Tl, K, Mo, Mn, Mg, Sr, As, Rb, Cr, Al, V and Fe. PC2explained 25.88% of the total variance and loaded Cu, Ga, Ba, Pb, Co, Zn, Mo and Tl. Al and Fe are considered as petrogenetic elements and their concentrations are not often influenced by human contamination in the aquatic sediment [51,52]. As a consequence, elements loaded in PC1 (Ni, Y, Cd, Tl, K, Mo, Mn, Mg, P, Sr, As, Rb, Cr, Al, V and Fe) are likely mainly originated from natural sources. Other elements such as Cu, Co, Mo, Zn, Pb, Ga and Ba were loaded in PC2, and they were moderately to severely enrich in the sediment. Their occurrence in the sediment is probably attributed to anthropogenic activities. It is important to notice that some elements such as Ni, P were partially loaded in both PC1 and PC2 (Table 3). It could be considered that both natural and anthropogenic sources contributed to their occurrence and accumulation in the sediment.

## Conclusion

The occurrence and risk assessment of 22 inorganic elements, including 19 metals, 2 metalloids and phosphorus, in sewage sludge and sediment samples were evaluated in this study. Elements were detected at broad range concentrations both in sediment and sludge samples. The evaluation of eco-toxicological risks based on two indices, EF and DC, permitted us to evaluate the pollution state of the sewage sludge and lake sediment. Results revealed that the sediment and sludge were under different pollution status. When considering EF index, which assesses the contamination level based on each of the element, the contamination was evident as some elements were enriched in both of the sludge and sediment, and anthropogenic activities could be responsible for such situation. Furthermore, when considering the DC index, which takes into account the overall elements, the pollution status of the sediment and sludge was between considerable and extreme degree of contamination. As a consequence, it urges to take adequate measures to restore the lake, improve wastewater treatment processes and avoid discharging untreated wastewater and sewage sludge in natural receptors and soil. However, more investigation is still needed to better understand the bioavailability of the inorganic elements and contamination levels in aquatic organisms.

## Acknowledgments

The authors would like toacknowledge the support from institute of Urban Environment, Chinese Academy of Sciences and from the Ministry of High Education and Scientific Research of the Republic of Benin.

## References

1. Turhanen PA, Vepsäläinen JJ & Peräniemi S. Advanced material and approach for metal ions removal from aqueous solutions. Sci Rep. 2005; 5: 8992.
2. Chipasa KB. Accumulation and fate of selected heavy metals in a biological wastewater treatment system, Waste Manage. 2003; 23: 135-143.
3. Ruilian Y, Xing Y, Yuanhui Z, Gongren H, Xianglin T. Heavy metal pollution in intertidal sediments from Quanzhou Bay, China. J Environ Sci. 2008; 20: 664-669.
4. Bhattacharya BD, Hwang JS, Tseng L, Sarkar SK, Rakshit D, Mitra S. Bioaccumulation of trace elements in dominant mesozooplankton group inhabiting the coastal regions of Indian Sundarban mangrove wetland. Mar Pollut Bull. 2014; 87: 345-351.
5. USEPA. Guidance for assessing chemical contaminant data for use in fish advisories. Washington, DC: Office of Water, Office of Science and Technology. 2000.
6. Lin YS, Caffrey JL, Lin JW, Bayliss D, Faramawi MF, Bateson TF, et al. Increased risk of cancer mortality associated with cadmium exposures in older Americans with low zinc intake. J Toxicol Environ Health-Part A. 2013; 76: 1-15.
7. Park RM, Bena JF, Stayner LT, et al. Hexavalent chromium and lung cancer in the chromate industry: A quantitative risk assessment. Risk Anal. 2004; 24: 1099-1108.
8. Amman AA, Michalke B, Schramel P. Speciation of heavy metals in environmental water by ion chromatography coupled to ICP-MS. Anal Biochem. 2002; 372: 448-452.
9. Fu F & Wang Q. Removal of heavy metal ions from wastewaters: A review. J Environ Manage. 2011; 92: 407-418.
10. Li XG, Ma XL, Sun J & Huang MR. Powerful reactive sorption of silver (I) and mercury (II) onto poly(o-phenylenediamine) microparticles. Langmuir. 2009; 25: 1675-1684.
11. Huang M-R, Li S & Li X-G. Longan Shell as Novel Biomacromolecular Sorbent for Highly Selective Removal of Lead and Mercury Ions. J Phys Chem B. 2010; 114: 3534-3542.
12. Suanon F, Tomètin ASL, Dimon B, Agani IC, Mama D & Azandegbe EC. Utilization of Sewage Sludge in Agricultural Soil as Fertilizer in the Republic of Benin (West Africa): What are the Risks of Heavy Metals Contamination and Spreading? Am J Environ Sci. 2016a; 12: 8-15.
13. Hounkpe PS, Adjovi CE, Crapper M, Awuah E. Wastewater Management in Third World Cities: Case Study of Cotonou, Benin. J Environ Protect. 2014; 5: 387-399.
14. Yessoufou A, Ifon BE, Suanon F, et al. Rare earth and precious elements in the urban sewage sludge and lake surface sediments under anthropogenic influence in the Republic of Benin. Environ Monit Assess. 2018; 189: 625.
15. Rap final Progeboue. Development de technologies alternatives

décentralisées de valorisation des boues résiduaires de lagunage par compostage et essais agronomiques. 2014.

16. Soil Survey Staff. Keys to soil taxonomy. 11th ed. USDA-NRCS. GPO, Washington, DC. 2010.
17. UEPA 3051A. Microwave assisted acid digestion of sediments, sludge, soils, and oils.
18. Cheng Z, Man YB, Nie XP, Wong MH. Trophic relationships and health risk assessments of trace metals in the aquaculture pond ecosystem of Pearl River Delta, China. Chemosphere. 2013; 90: 2142-2148.
19. Wang J, Liu R, Zhang P, Yu W, Shen Z, Feng C. Spatial variation: environmental assessment and source identification of heavy metals in sediments of the Yangtze River Estuary. Mar Pollut Bull. 2014; 87: 364-373.
20. Martin GD, George R, Shaiju P, Muraleedharan KR, Nair SM, Chandra mohana kumar N. Toxic metals enrichment in the surficial sediments of a Eutrophic Tropical Estuary (Cochin Backwaters, Southwest Coast of India). Sci World J. 2012: 17.
21. Amin B, Ismail A, Arshad A, Yap CK, Kamarudin MS. Anthropogenic impacts on heavy metal concentrations in the coastal sediments of Dumai, Indonesia. Environ Monit Assess. 2009; 148: 291-305.
22. Hasan AB, Kabir S, Reza AHMS, Zaman MN, Ahsan A, Rashid M. Enrichment factor and geo-accumulation index of trace metals in sediments of the ship breaking area of Sitakund Upazilla (Bhatiary-Kumira) Chittagong, Bangladesh. J Geochem Explor. 2013; 125: 130-137.
23. Taylor SR. Abundance of chemical elements in the continental crust: a new table. Geochim Cosmochim Acta. 1964; 28: 1273-1285.
24. Rudnick RL & Gao S. The composition of the continental Crust. 2003: 1-64. In The Crust (ed. RL Rudnick). Volume 3, Treatise on Geochemistry (eds. H.D. Holland and K.K.
25. Lide DR. Section 14, geophysics, astronomy, and acoustics; abundance of elements in the Earth's crust and in the sea, Boca Raton. In: CRC Handbook of Chemistry and Physics, 85th edn. CRC Press, Florida, USA. 2005.
26. Sakan SM, Djordjevic DS, Manojlovic DD, Polic PS. Assessment of heavy metal pollutants accumulation in the Tisza river sediments. J Environ Manage. 2009; 90: 3382-3390.
27. Hakanson L. An ecological risk index for aquatic pollution control. A sedimentological approach. Water research. 1980; 14: 975-1001.
28. Lee CS, Li XD, Shi WZ, Cheung SC, Thornton I. Metal contamination in urban, suburban and country park soils of Hong Kong: a study based on GIS and multivariate statistics. Sci Total Environ. 2006; 356: 45-61.
29. Wong JWC, Mak KF, Chna NW, Lam A, Fang M, Zhou LX, et al. Co-composting of soybean residues and leaves in Hong Kong. Bioresour Technol. 2001; 76: 99-106.
30. Suanon F, Sun Q, Yang X, Chi Q, Mulla IS, Mama D, et al. Assessment of the occurrence, spatiotemporal variations and geoaccumulationof fifty-two inorganic elements in sewage sludge: A sludge management revisit. Sci Report. 2017; 7: 5698.
31. Kvasauskas M & Baltrenas P. Research on anaerobically treated organic waste suitability for soil fertilization. J Environ Eng Landsc Manag. 2009; 17: 205-2011.
32. Peng G, Tian G, Liu J, Bao Q, Zang L. Removal of heavy metals from sewage sludge with a combination of bioleaching and electrokinetic remediation technology. Desalination. 2011; 271: 100-104.
33. Karvelas M, Katsoyiannis A, Samara C. Occurrence and fate of heavy metals in the wastewater treatment process. Chemosphere. 2003; 53: 1201-1210.
34. Wei YJ, Liu YS. Effects of sewage sludge compost application on crops and cropland in a 3-year field study. Chemosphere. 2005; 59: 1257-1265.
35. Dong B, Liu X, Dai L, Dai X. Changes of heavy metal speciation during high-solid anaerobic digestion of sewage sludge. Bioresour Technol. 2013; 131: 152-158.
36. Wu Q, Cui Y, Li Q, Sun J. Effective removal of heavy metals from industrial sludge with the aid of a biodegradable chelating ligand GLDA. J Hazard Mater. 2015; 283: 748-754.
37. Suanon F, Sun Q, Dimon B, Mama D, Yu C-P. Heavy metal removal from sludge with organic chelators: Comparative study of N, N-bis(carboxymethyl) glutamic acid and citric acid. J Environ Manage. 2016b; 166: 341-347.
38. Suanon F, Sun Q, Mama D, Li J, Dimon B, Yu C-P. Effect of nanoscale zero-valent iron and magnetite (Fe3O4) on the fate of metals during anaerobic digestion of sludge. Water Res. 2016c; 88: 897-903.
39. Westerhoff P, Lee S, Yang Y, Gordon G, Hristovski K, Halden R, et al. Characterization, Recovery Opportunities, and Valuation of Metals in Municipal Sludges from U.S. Wastewater Treatment Plants Nationwide. Environ Sci Technol. 2015; 49: 9479-9488.
40. USEPA. Process Design Manual for land application of sewage sludge and domestic septage. EPA/625/R-95/001, Office of Research and Development, Washington, DC. 2016c; 33.
41. Benson NU, Anake WU, Essien JP, Enyong P, Olajire AA. Distribution and risk assessment of trace metals in Leptodiusexarata, surface water and sediments from Douglas Creek, Qua Iboe estuary. J Taibah Univ Sci. 2016; 11: 434-449.
42. Zhao G, et al. Distribution and contamination of heavy metals in surface sediments of the Daya Bay and adjacent shelf, China. Mar Pollut Bull. 2016; 112: 420-426.
43. Liu J, Yin P, Chen B, Gao F, Song H, Li M. Distribution and contamination assessment of heavy metals in surface sediments of the Luanhe River Estuary, northwest of the Bohai Sea. Mar Pollut Bull. 2016; 109: 633-639.
44. Lin Q, Liu E, Zhang E, Li K, Shen J. Spatial distribution, contamination and ecological risk assessment of heavy metals in surface sediments of Erhai Lake, a large eutrophic plateau lake in southwest China. Catena. 2016; 145: 193-203.
45. PejmanaA, Bidhendia N, Ardestania MG, Saeedib M, Baghvanda A. A new index for assessing heavy metals contamination in sediments: A case study. Ecol Indicat. 2015; 58: 365-373.
46. Consensus-Based Sediment Quality guide line (CBSOG SQG 2003). 2003.
47. NRC. Biosolids applied to land: advancing standards and practices. National Research Council. National Academy Press, Washington, DC. 2002.
48. World Health Organization. Exposure to cadmium: a major public health concern. Preventing Disease through Healthy Environments. 2010; 27 (Geneva).
49. Abdullah M, Fasola M, Muhammad A, Malik SA, Boston N, Bokhari H, et al. Avian feathers as a non-destructive bio-monitoring tool of trace metals signatures: a case study from severely contaminated areas. Chemosphere. 2015; 119: 553-561.
50. Hanif N, Ali S, Akber M, Eqani S, Ali SM, Cincinelli A, et al. Geo-accumulation and enrichment of trace metals in sediments and their associated risks in the Chenab River, Pakistan. J Geochem Explor. 2016; 165: 62-70.
51. Yu X, Yan Y, Wang WX. The distribution and speciation of tracemetals in surface sediments from the Pearl River estuary and the Daya Bay, southern China. Mar Pollut Bull. 2010; 60: 1364-1371.
52. Zhao YY, Yan MC. Geochemistry of Sediments of the China Shelf Sea. Science Press, Beijing. 1994; 37-92.