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Assessment of zinc concentrations in surface sediment from urban and industrial sites of Umeda River, Japan

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ABSTRACT

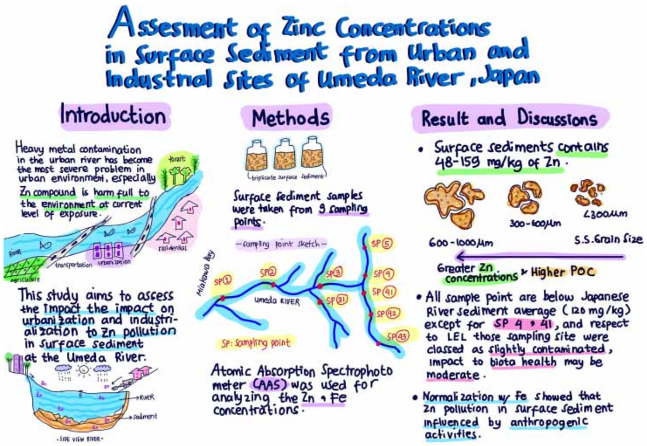
Heavy metal contamination in the urban river has become the most severe problem in the urban environment, especially the zinc (Zn) compound is harmful to the environment at current levels of exposure. This study aims to assess the impact of urbanization and industrialization on Zn pollution in surface sediment at the Umeda River. Triplicate surface sediment samples were collected at nine sample points. An atomic absorption spectrophotometer was used for analyzing the Zn and Fe concentrations. The surface sediments in the Umeda River contained 48–159 mg/kg of Zn. The results indicated that Zn concentrations were abundant in the ascending order of 600–1,000 μm, 300–600 μm, and smaller than 300 μm. The higher the grain size, the higher the particulate organic carbon which might contribute to the higher Zn and Fe concentrations. In addition, the sediment quality guidelines indicated that the Zn levels in the study area did not constitute a major threat to biota. Normalization with Fe showed that the Zn pollution in surface sediment had been influenced by anthropogenic activities such as industrialization and urbanization.

Key words: atomic absorption spectrophotometer, heavy metal, river water, surface sediment, Umeda River, zinc pollution

HIGHLIGHTS

- Unusual condition of Zn contents in the surface sediment compared with other studies.
- Represents the zinc studies in urban areas with some type of land use in the watershed.
- The first heavy metals research in the Umeda River, Aichi, Japan.

GRAPHICAL ABSTRACT



INTRODUCTION

Heavy metal contamination in the urban river has become the most serious problem in urban environments, and has attracted increased attention from researchers (Xu *et al.* 2017). Especially, zinc is a crucial element for organisms, but its toxicity to aquatic organisms at high concentrations has remained a concern. Based on Environmental Protection Agency (EPA) heavy metals guidelines for sediments, in particular for Zn, <90 mg/kg is categorized as not polluted, 90–200 mg/kg as moderately polluted, and >200 mg/kg categorized as heavily polluted. (Ogbeibu *et al.* 2014) In Japan, in 2003, Environmental Quality Standards (EQS) were established for the annual mean of Zn concentration in surface water, i.e., 0.03 mg/L, which is stricter than preceding regulations in order to protect aquatic life. While sediment standards have not yet been defined, the danger of heavy metals, unlike other pollutants, lies in their being non-degradable and their accumulation on the Earth's surface. Through the food chain, heavy metals in the soil may pile up in the body of a human as well as livestock, endangering human health directly or indirectly (Harikumar & Jisha 2010). The main anthropogenic sources of heavy metal contamination are mining and smelting activities, disposal of untreated and partially treated effluents, metal chelates from different industries and the indiscriminate use of heavy metal-containing fertilizers and pesticides in agricultural fields. (Ogunfowokan *et al.* 2013).

Sediments act as sinks and sources of contaminants in aquatic systems because of their variable physical and chemical properties (Evans *et al.* 2003). Analysis of pollutants in sediments is vital as they are adsorbed by materials in suspension and by fine-grained particles (Rainey *et al.* 2003). Pekey (2006) demonstrated that heavy metals tend to be trapped in aquatic environments and accumulate in sediments.

This study aimed to assess the level of Zn concentrations in surface sediments, to examine the distributions of Zn concentrations in different grain sizes of surface sediment, and to explore the effect of the natural and anthropogenic input on the Umeda River.

METHODS

The research method is presented on Figure 1.

Study area and sample collection

For this study, triplicate surface sediment samples of 250 g weight were taken at nine sampling points of the Umeda River. The description of sampling site locations on Umeda River and tributaries is presented in Table 1 and visualized in Figure 2. The sampling events were conducted every 2 months between July 2019 and January 2020.

Pre-treatment, digestion and analysis

For surface sediment, the samples were dried in an oven 40 °C within 3 days. After the sediment samples were dried, then the dried sediment samples were sieved with a mesh size of 600–1,000 μm, 300–600 μm, and <300 μm. About 1 g (dry weight) of each sample fraction was acid digested with addition of HCl according to EPA method 3050B. After heating up on a hot plate at 90 °C and cooling, each digest was filtered through Whatman No. 41 filter paper into a 50 mL volumetric flask and the volume was made to the mark with deionized water (EPA 1996). The sample was centrifuged at 2,300 rpm for 10 minutes. Once centrifuged, all the samples were decanted and kept immediately in bottles, ready to be analyzed.

The concentrations of Zn for sediment were determined by an atomic absorption spectrophotometer (AA-7000, Shimadzu Corporation, Japan) with four calibration standards ranging from 0 to 0.5 mg/L (detection limit: 0.005 mg/L). Every sample was analyzed three times, and standard solutions were analyzed every six samples for quality assurance purposes.

RESULTS AND DISCUSSION

Comparison of Zn concentration at surface sediment with sediment quality guidelines

To evaluate the contamination levels of Zn in the study areas, the Coastal Ocean Sediment Database, the lowest effect level (LEL) and the severe effect level (SEL), and the Canadian Sediment Quality Guidelines (CSeQGs), including the interim sediment quality guidelines (ISQGs) and the probable effect level (PEL), were applied as benchmarks (Figure 2). The Coastal Ocean Sediment Database values are indicative of metal contamination and are used to quantify the degradation of sediment quality in estuarine and marine ecosystems (Ruiz-Fernández *et al.* 2003). The New York State Department of Environmental Conservation (NYSDEC 1999) points out that if both the LEL and SEL criteria are exceeded, the metal concerned may severely impact biota health, whereas if only the LEL criterion is exceeded, impact on biota health may be moderate. The

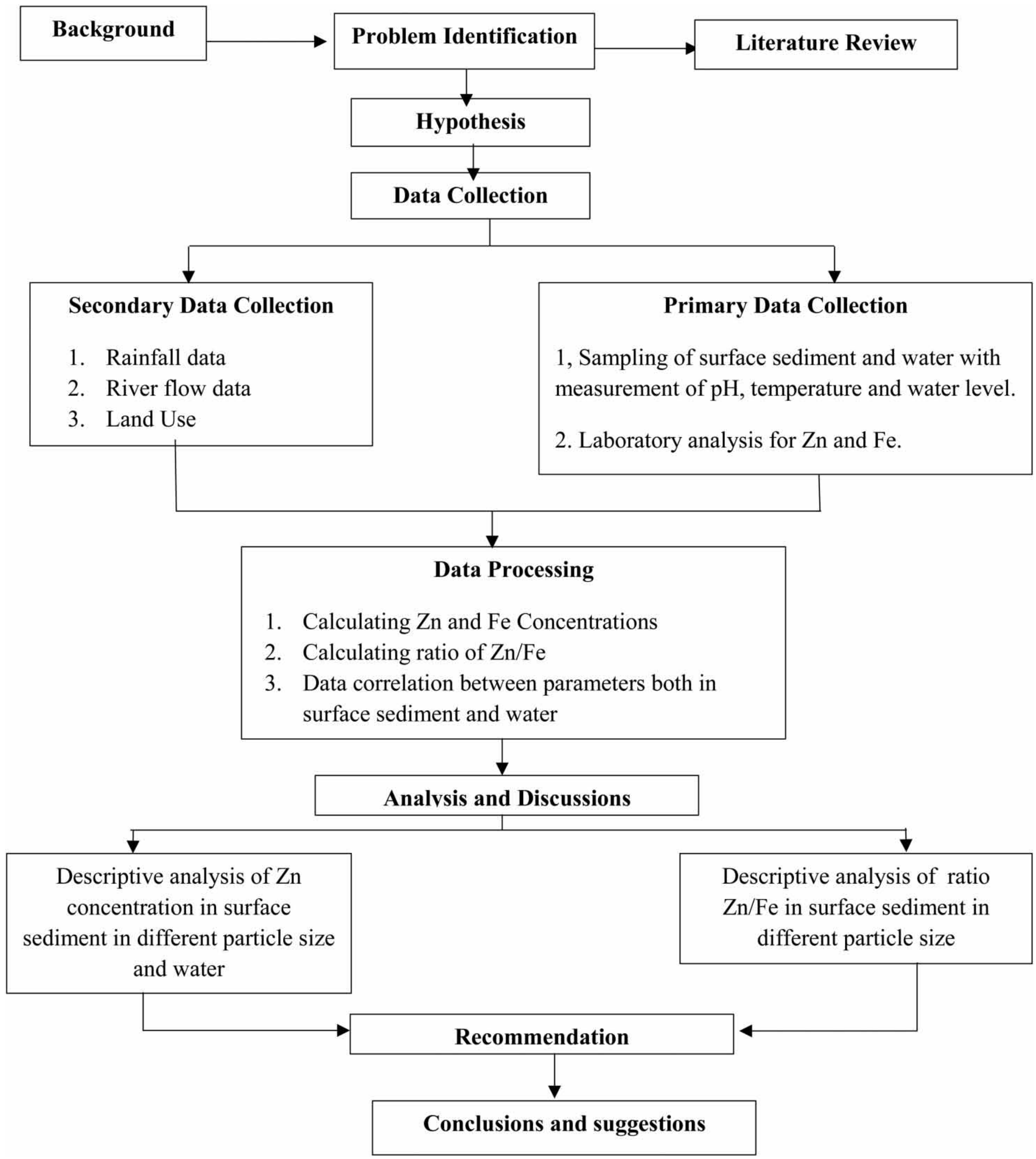


Figure 1 | Research flow diagram.

CSeQGs present values for individual chemicals or elements in both freshwater and marine (including estuarine) sediments for the protection of aquatic life, and were developed from available scientific information on the biological effects of chemicals associated with sediments (SAIC 2002). The CSeQGs present two numerical limits: (1) the lesser limit is termed the ISQG value, and (2) the greater limit is called the PEL (CSMWG 2003). Sediment chemical concentrations below the ISQG values are unlikely to be associated with adverse biological effects, whereas concentrations above the PEL are expected

Table 1 | Presents the site codes, locations and a brief description of the sampling sites

Sampling site	Description of sampling site location	Longitude	Latitude
1	Umeda River; Downstream	137°23′46.8″	34°42′44.0″
2	Urban area	137°25′32.6″	34°43′21.3″
3	Urban area	137°27′21.9″	34°43′16.8″
4	Urban area	137°28′57.2″	34°43′22.9″
5	Umeda River; Upstream	137°28′42.3″	34°44′02.4″
31	Ochiai River; Industrial area	137°27′22.8″	34°43′15.0″
41	Sakai River; Industrial area	137°28′55.2″	34°43′20.3″
42	Industrial area	137°29′19.3″	34°42′49.5″
43	Agriculture and urban area	137°29′33.9″	34°42′07.1″

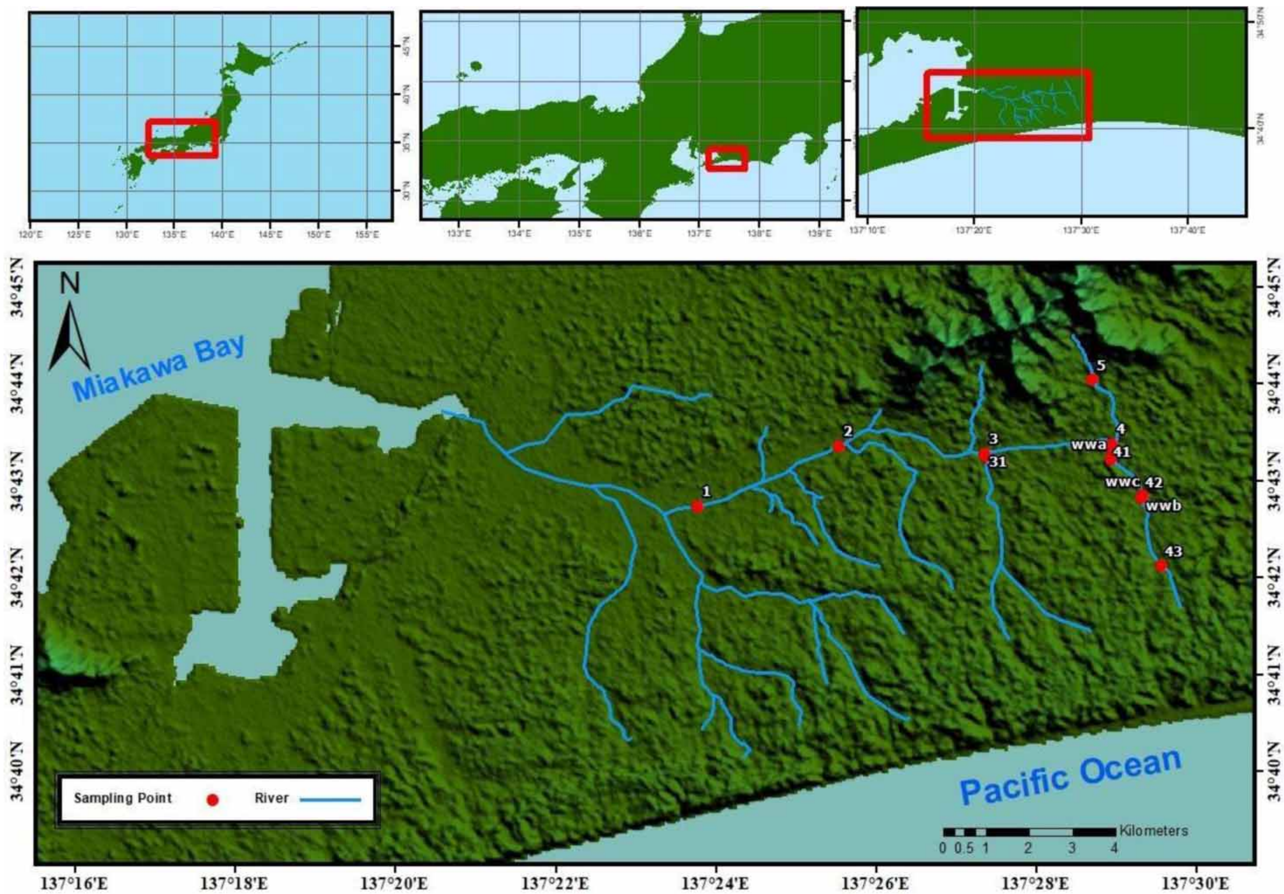


Figure 2 | The geographical location of the sampling sites in Umeda River.

to be frequently associated with adverse biological effects. Adverse effects are occasionally observed in sediments that contain metal concentrations between the two threshold values (CSMWG 2003).

The average concentrations of Zn in each site of the study area (Figure 3) compared with sediment quality criteria representing all the sampling sites were still below of sediment quality criteria except for sampling site 4 which was slightly higher than Japanese river sediment average. In addition, with respect to the LEL, Zn concentration in sampling sites 4 and 41 were classed as slightly contaminated, and the impact on biota health may be moderate. In addition, compared with the EPA heavy metal guideline for sediment all sampling sites were below 90 mg/kg meaning that these levels were classified as unpolluted,

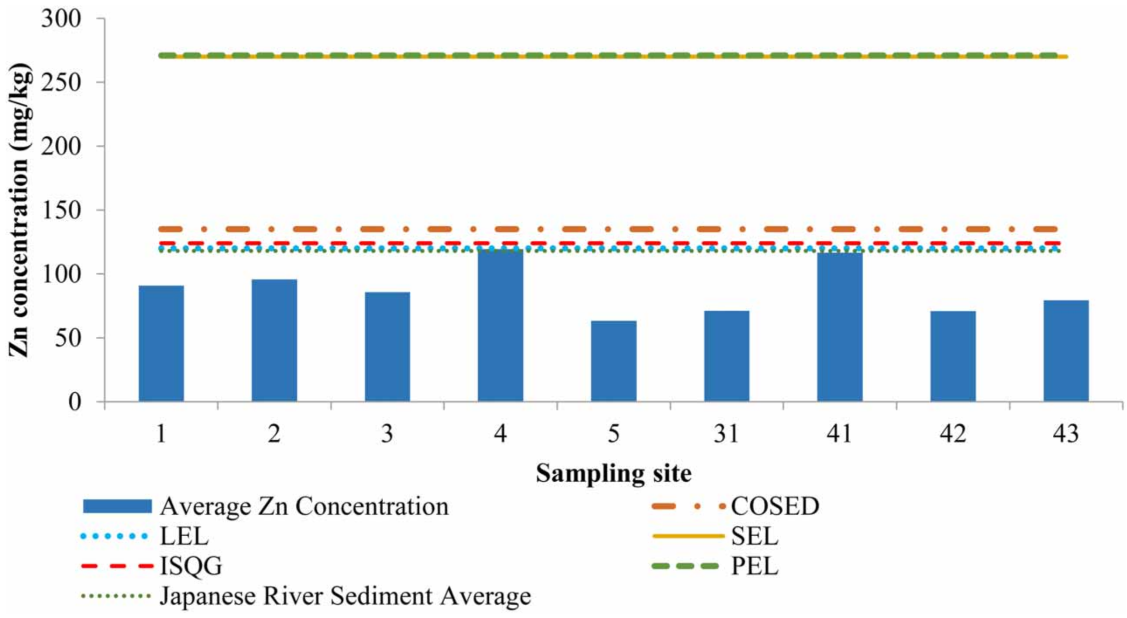


Figure 3 | Sediment quality criteria and average Zn concentration in surface sediment samples of Umeda River.

except for sampling sites 4 and 41 categorized as moderately polluted. Hence, those sites should be monitored frequently and need further investigation to identify the possible sources of Zn fluxes to the Umeda River.

Comparison of Zn concentration on sediment in study area with previous studies from other regions

In order to appreciate the level of trace heavy metal in the Umeda River relative to water bodies around the world, the average concentrations of Zn and Fe in surface sediments from the Umeda River were compared to those from Japan, Indonesia, Malaysia and China, and Africa (Table 2).

The average concentrations of Zn in surface sediments reported by Sin *et al.* (2001) for ShingMun River area were all greater than those concentrations from Umeda River, which was almost 15 times higher than in the Umeda River. Compared with other rivers in Japan, the concentration of Zn in Umeda River was slightly below that of Kumozu River (115 mg/kg), Hannai River (105 mg/kg), Nomi River (95 mg/kg), and Miya River (102 mg/kg), and was almost double that seen in River around 'Firefly Village' (160 mg/kg). In addition, the average concentration of Fe in the surface sediment of Jakarta

Table 2 | Average metal concentration of surface sediment (mg/kg) in the Umeda River and rivers compared to those from other studies

	Sediment (mg/kg)		
Area	**Zn**	**Fe**	**Reference**
Umeda River, Aichi, Japan	88	12,754	This study
Kumozu River, Matsusaka, Japan	115	–	GSJ AIST
Hannai River, Matsusaka, Japan	105	–	
Nomi River, Tokyo Japan	95	47,000	Sharmin *et al.* (2009)
Miya River, Ise, Japan	102	–	GSJ AIST
River around 'Firefly Village', Shikoku, Japan	160	40,794	Anazawa *et al.* (2004)
Kampala, Uganda, Africa	446.2	–	Sekabira *et al.* (2010)
Estuary of Brantas River and Solo River, Indonesia	81	–	Everaarts (1989)
Shing Mun River, Hongkong	1,299	–	Sin *et al.* (2001)
Jakarta Bay, Indonesia	307	49,931	Budiyanto & Lestari (2017)

Bay, Indonesia (49,931 mg/kg) were the highest compared with Umeda River and other studies, and was almost triple compared with rivers in Japan such as Nomi River (47,000 mg/kg), and the river around Firefly village (40,794 mg/kg).

Grain size of surface sediment

Grain size is the most fundamental physical property of sediment. Grain size distributions (weight % 600–1,000 μm, 300–600 μm, smaller than 300 μm) of the Umeda River surface sediments are illustrated in Figure 4. In the Umeda River, except for sample sites 5 and 31 both of which contained below than 40% of 300–600 μm, almost all samples were dominated by medium sand (300–600 μm), ranging approximately from 44% for sampling site 1 to 56% for sampling site 43. In addition, coarse sand (600–1,000 μm) values ranged from 23% for sampling site 3 to 46% for sampling site 31. In this same area, fine sand (smaller than 300 μm) was present in all the sampling sites, with the maximum value of 36% observed at sampling site 5, and a minimum value of 12% observed at sampling site 42.

Zn and Fe concentrations

All the samples collected along the Umeda River showed that Zn concentration was correlated with particle size (Figure 5). The results indicated that Zn concentrations were rich in the ascending order of 600–1,000 μm, 300–600 μm, and smaller than 300 μm fractions. The statistical analysis showed that the Zn concentration decreased with decreased grain size.

This phenomenon corresponded with the findings by Krumgalz (1989), profound enrichment of autumn trace metals' content and organic matter content in the coarse and medium fraction (larger than 0.250 mm) sometimes even more than in the fraction of clay and silt. A possible reason for anomalously high concentrations of trace metals and organic matter content in the medium and coarse fractions might be due to the formation of large particle sizes of surface sediment that was from the smaller particles enriched by contaminants. The formation of large particle size occurs during the generally accepted drying procedures, all of which were carried out without prewashing the studied sediments. Therefore, during any drying procedure (either lyophilization or heating), the small sediment particles will be cemented both by dissolved organic matter and by sea salt, present in the marine sediment, to form a large particle size. In this case, the total amounts of either trace metals or organic matter retained by each agglomerate particle will be much larger compared to the amounts which could be adsorbed only on the outer surface of such agglomerates.

In addition, this was supported by the findings from Singh *et al.* (1999) at these two sites that concentrations of Fe and Mn increased towards the finer fractions, but all other metals (Si, Al, Cu, Zn, Ni, Cr) showed similar to or higher concentrations towards the coarser size fractions. The increased concentration of metals in the coarser fractions at these sites may be attributed to inputs from the mining and other anthropogenic sources. Many workers have pointed out the fact that larger particles stay in a place longer, often in shallow oxygenated areas of streams (Singh *et al.* 1999) and therefore may have more time to

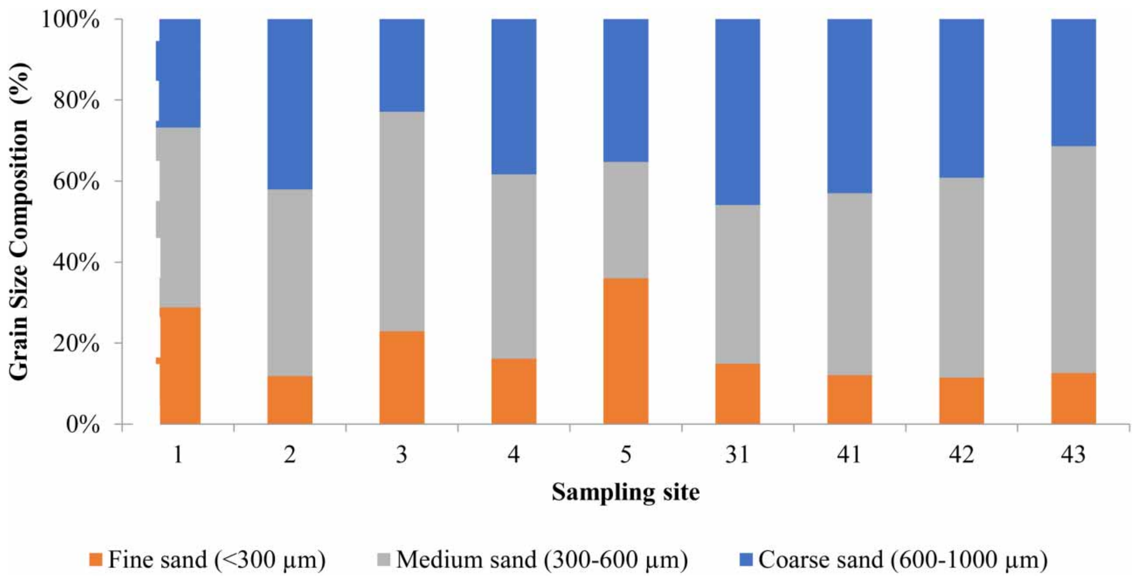


Figure 4 | Grain size of the sediments from Umeda River and its tributaries.

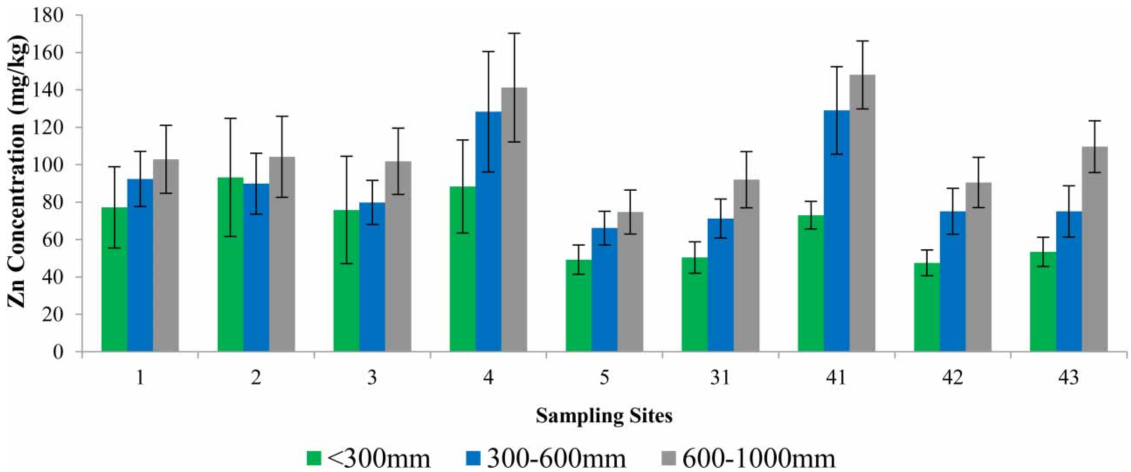


Figure 5 | Variation of Zn concentrations against particle size at different sampling site locations.

develop oxide coating and absorb more trace metals than smaller particles. Presence of heavy minerals or coarse fractions of mine and industrial waste may also increase metal concentration in the coarser fractions (Singh *et al.* 1999).

Liang *et al.* (2019) found that other factors such as the grain size and the organic matter (OM) content of sediment can also affect the concentration of heavy metals. For instance, sediment with a higher content of OM adsorbs more heavy metals. It was previously suggested that OM in sediment can adsorb heavy metals from the environment (Damak *et al.* 2019).

Figure 6 illustrates that the percentage of particulate organic carbon (POC) were rich in the coarser particle size of surface sediment. Organic matter has a high specific storage capacity for heavy metals (Elith & Garwood 2001). The result of this study similarly showed that an increase in POC in the coarser particle size was followed by an increase in the concentrations of Zn and Fe.

In this study, the land use of the sampling area was mostly dominated by urban, industrial, or agricultural use hence minerals coming from mining industries to pollute the sediments is not possible. Therefore, there was a possibility that anthropogenic activities such as industry and domestic use can affect the characteristics of sediment at the Umeda River. In addition, corresponding with Krumgalz (1989), is also possible that this phenomenon happened because the procedure during the drying of the sediment which the small particles will be cemented both by dissolved OM, caused the heavy metals on sediments to be not entirely digest and could be detected.

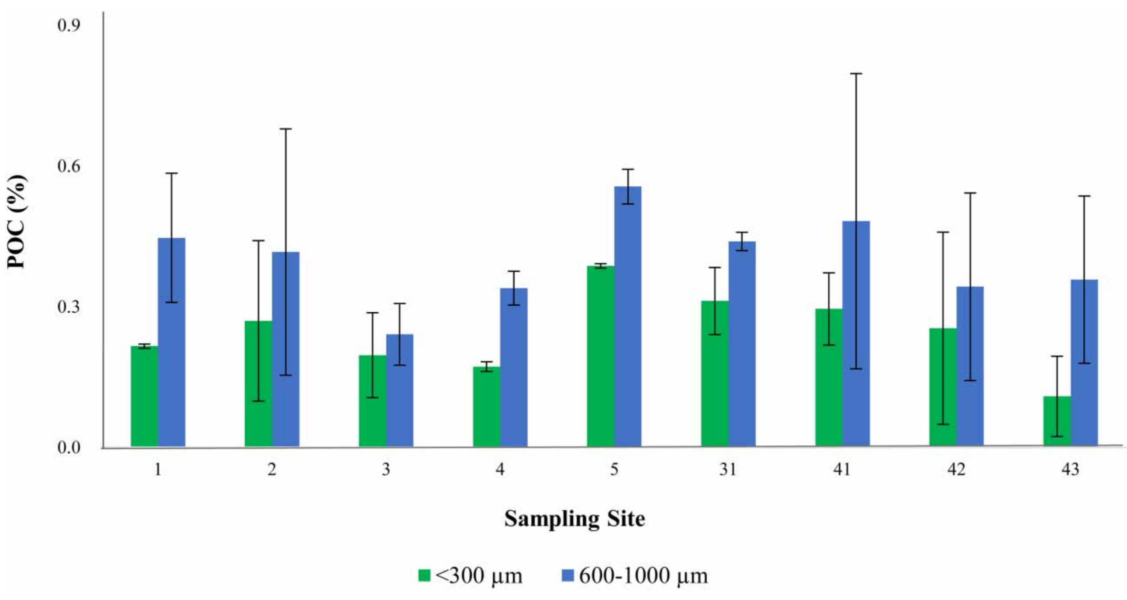


Figure 6 | Variation of organic carbon against particle size.

Normalization of Zn concentration

Based on a study from Sharmin *et al.* (2009) and Gamo (2007), we have also used Fe as a conservative tracer to differentiate natural from anthropogenic components and Japanese river sediment average, data have been considered as baseline values. Geochemical normalization can compensate for both the granulometric and mineralogical variability of base metal concentration in sediments (Liu *et al.* 2003). Consequently, in order to assess the possible anthropogenic impact, several authors (Guerra-Garcia & Garcia-Gomez 2005) have successfully used Fe to normalize the base metal contaminants. Fe is abundant in the Earth's crust and is scarcely influenced by anthropogenic inputs, especially compared with the naturally occurring high levels of this element (Villares *et al.* 2003; Guerra-Garcia & Garcia-Gomez 2005). The normalization of the concentrations using Fe as a conservative element confirmed that with little exception all the sampling sites were most polluted by Zn.

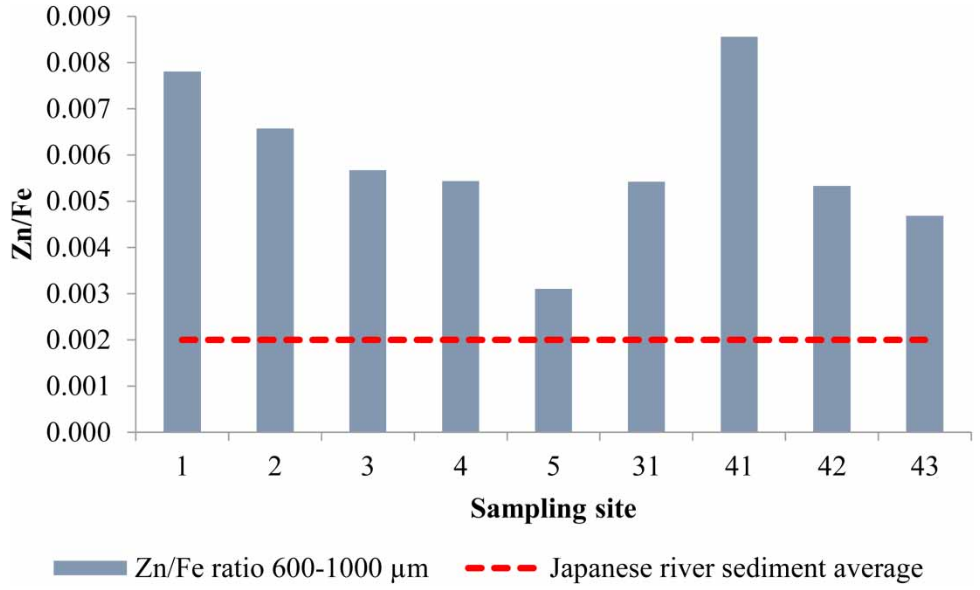


Figure 7 | Normalized ratios of Zn concentration in surface sediment of grain size 600–1,000 μm using Fe as conservative element. Japanese river sediment average (Gamo 2007) data have been considered as baseline values (broken lines).

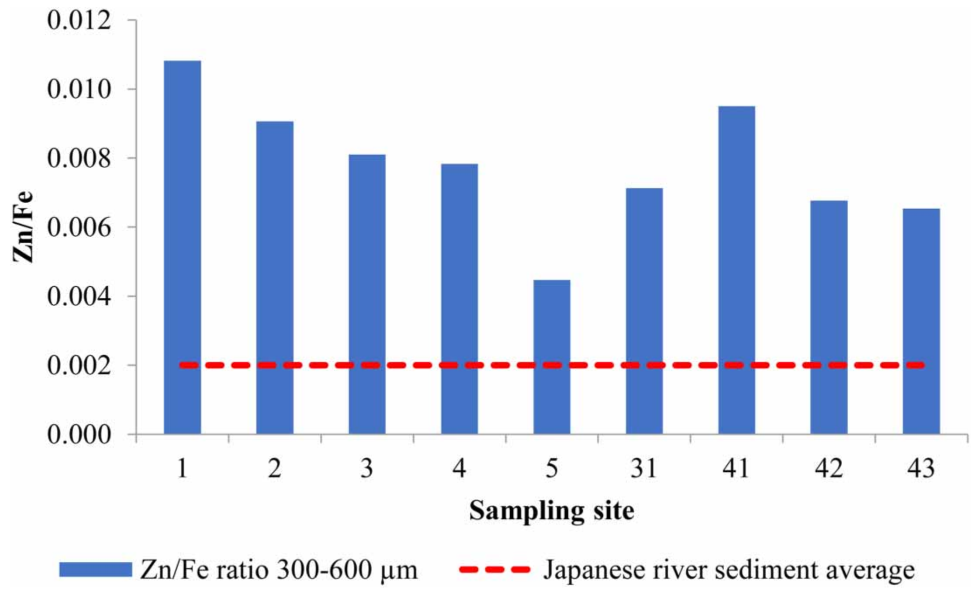


Figure 8 | Normalized ratios of Zn and Fe concentration in surface sediment on grain size 300–600 μm compared with Japanese river sediment average.

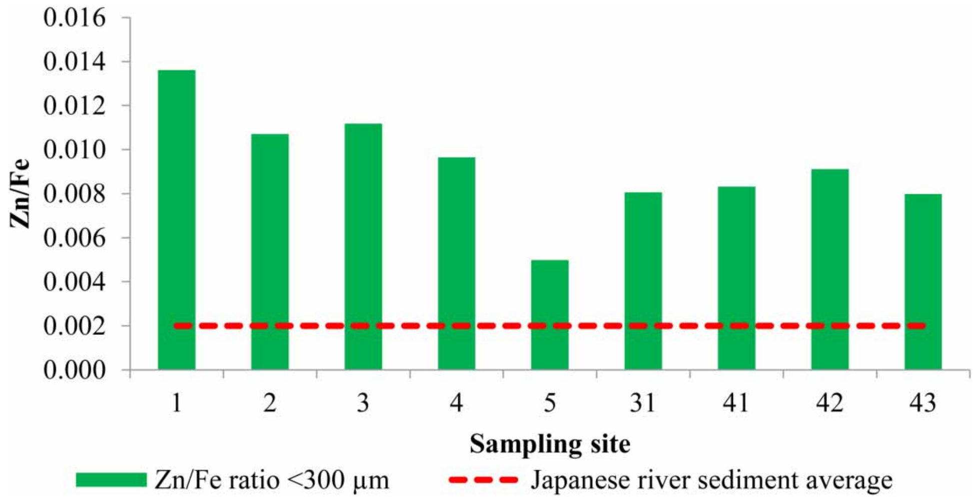


Figure 9 | Normalized ratios of Zn and Fe concentration in surface sediment on grain size smaller than 300 μm compared with Japanese river sediment average.

Normalizing with Fe, revealed that Zn enrichment in surface sediments at the sampling sites of Umeda River was not natural but anthropogenic (Figures 7–9). Contradiction with Figures 4, 7–9 showed that the ratio of Zn/Fe was increasing with the finer particle size of sediment, which means there was a very strong positive correlation between decreasing grain size and increasing ratio Zn/Fe. Additionally, it can be summarized that the concentrations of Zn and Fe were more available or that it was more attractive to catch Zn/Fe from anthropogenic sources in the finer grain size of surface sediment on Umeda River.

In addition, supported by the studies from Schiff (1999), inputs such as treated municipal wastewater discharges from deep ocean outfalls or urban runoff from large river and creek mouths contributed not only varying levels of pollutants but large quantities of silt and clay-sized particulates (smaller than 63 mm diameter). These inputs can significantly alter the proportions of fine-grained materials (Gorsline *et al.* 1990).

CONCLUSION

The result of the assessment of Zn concentration at Umeda River compared with the criteria of sediment quality standard was that it was unpolluted, except for sampling sites 4 and 41 which were classed as slightly contaminated, and the impact on biota health may be moderate. In addition, based on Zn/Fe, all sampling points indicated that Zn in sediment has slightly adverse effects due to anthropogenic activities. In addition, the results indicated that Zn concentrations were abundant in the ascending order of 600–1000 μm, 300–600 μm, and smaller than 300 μm that correlated with the higher POC (%) in the coarser grain size, which might contribute to the higher Zn and Fe concentrations. The ratios of Zn/Fe were increased with the finer particle size of sediment, it can be summarized that the finer grain size of surface sediment contained more Zn/Fe pollution from the anthropogenic sources in the Umeda River. In addition, this study showed that the downstream side had lower concentrations of Zn compared to the upstream side that could be markedly affected by industrialization and urbanization. According to this study, the Umeda River sediment has been affected by anthropogenic activities. Further investigation is necessary to identify the possible sources of Zn fluxes to the Umeda River and photographs of sediment fractions need to be taken for the possibility of Zn and Fe analysis in the laboratory.

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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