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ASSESSMENT OF HEAVY METALS POLLUTION IN SOFT SEDIMENT OF THE SEVERN ESTUARY AND INNER BRISTOL CHANNEL SYSTEM

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ABSTRACT : *For several decades, the Severn Estuary and Bristol Channel have been contaminated by heavy metals despite the many ecosystem services it provides. Therefore, this paper is devoted to the systematic assessment of heavy metals (Cd, Cr, Ni, Zn and Pb) pollution in soft sediments of the Severn Estuary and Inner Bristol Channel. An unpublished 2016 monitoring survey data of the Severn Estuary and Bristol Channel was obtained from the UK Environment Agency to assess seasonal variations, spatial distribution, exceedances, ecological risk, and potential sources of heavy metals. Seasonal variations between the concentration of heavy metals during summer and winter revealed that irrespective of seasons the concentration of heavy metals had the trend Zn > Cr > Pb > Ni > Cd. Concentrations of metals were higher in summer than winter, and there was a significant difference between metals concentration in summer and winter ($P < 0.05$). Spatial distribution of metals using the Empirical Bayesian Kriging (EBK) model revealed that irrespective of the seasons, concentrations of Cd, Cr, Ni, Zn and Pb in the distinct reaches of the estuary followed the trend Severn middle > Severn lower > Inner Bristol Channel > Severn upper. One-way ANOVA, which was followed by the Tukey's HSD test, showed a significant difference between the mean concentration of heavy metals in the distinct reaches of the study area at $p \leq 0.05$. Assessment of heavy metals using the USEPA Sediment Quality Guideline revealed that none of the reaches of the estuary was polluted with Cd. The Severn upper was not polluted with Pb. The Severn middle was moderately polluted by Ni but heavily polluted with Zn, Cr and Pb. Other reaches of the estuary were moderately polluted with Cr, Ni, Zn and Pb. Assessment of ecological risk of heavy metals using pollution indices such as geo-accumulation index (Igeo), contamination factor (CF), metal pollution index (MPI) and pollution load index (PLI) revealed that heavy metals such as Zn, Pb, Ni and Cr in the distinct reaches of the estuary and might pose an adverse effect on biota and ecosystem services of the Severn Estuary and Inner Bristol Channel. Identification of potential sources of metals using Pearson correlation, hierarchical cluster analysis (HCA) and principal component analysis (PCA) suggested that heavy metals all originated from anthropogenic sources.*

KEYWORDS*:* Heavy metal, Soft sediment, seasonal variations, spatial distribution, ecological risk, potential sources

INTRODUCTION

Over the past few decades, there has been increasing worldwide systematic monitoring of aquatic pollution by heavy metals (Salomons, 1993; Fernandes *et al*., 2008; Rahman *et al*., 2014), which have been demonstrated to pose significant challenges to human society both as a direct health risk and indirectly through undermining the health of aquatic ecosystems and their supportive capacities (Sakan *et al*., 2015; Ali and Khan, 2017).

Although there is no consensual definition of the term 'heavy metal', this study refers to heavy metals as metals and metalloids with a density of more than 5 g/cm3 (Banfalvi, 2011; Yirgu, 2011; Leong and Chang, 2020). Environmental scientists commonly use the term in the context of potential toxicity. Some common examples include arsenic (As), cadmium (Cd), chromium (Cr), nickel (Ni), copper (Cu), iron (Fe), lead (Pb), mercury (Hg), silver (Ag) and zinc (Zn) (Duruibe *et al*., 2007). Pollution of the aquatic ecosystem due to heavy metals is of global concern as heavy metal atoms are conservative (they do not degrade) so tend to persist and bioaccumulate once released into the environment (Ahmed *et al*., 2015a; Ahmed *et al*., 2015b; Islam *et al*., 2015; Vhahangwele and Khathutshelo, 2018).

The term 'sediment' is referred to as unconsolidated materials that form the upper layer of aquatic ecosystems (Margaret *et al*., 2000). These materials may include sand, fine silt, loose cobble, boulder, and partially or fully buried organic matter (Tiwari, 2020). Metals bind preferentially to soft sediments which are surficial sediments that make up the top 2cm layer of river sediment (Mccready *et al*., 2006; Lutgen *et al*., 2020). In an aquatic environment, sediments are considered as sinks for heavy metals (Xie *et al*., 2019). About 99% of the heavy metals load in an aquatic system has been reported to eventually precipitate onto the sediment (Peng *et al*., 2009; Jin *et al*., 2017). Therefore, concentrations of heavy metal in the sediment are often four or five orders of magnitude higher (by mass) than that found in the overlying water as sediment tends to accumulate metals. Also, concentrations in the overlying water may be transient, whereas sediment store episodic inputs (Yuan *et al*., 2011). Thus, the sediment is considered a suitable medium which can reflect the heavy metal pollution status of an aquatic ecosystem (Kalimur, 2011; Joksimović *et al*., 2020). Consequently, monitoring of the concentration of heavy metals in sediments can be an indicator to provide information on the heavy metal contamination of the entire aquatic ecosystem.

Heavy metals are mobilised in the sediment of aquatic ecosystems via two primary sources (Guo *et al*., 2018). The first is through natural processes, including atmospheric deposition, rock weathering and erosion, and hydrodynamic alteration (Li *et al*., 2020). The second is via anthropogenic activities, such as industrial wastewater discharge and agricultural fertiliser leaching (Nguyen *et al*., 2020). Due to the rapid development of agriculture, industrialisation, and urbanisation since the nineteenth century, anthropogenic activities have become the main source of heavy metal pollution in the sediment of aquatic ecosystems (Khan *et al*., 2008; Haiyan *et al*., 2013; Xu *et al*., 2018).

The Severn Estuary, one of the largest estuaries in the United Kingdom, contains essential habitats such as areas of mudflats, sandbanks and salt marshes that support aquatic organisms such as fish and serve as a significant resource for other organisms including wading birds (Rehfisch *et al*., 2003; Kuroki *et al*., 2008; Hooper and Austen, 2013; Zhao *et al*., 2018). The Severn Estuary and Inner Bristol Channel have also been recognised as a Special Protection Area (SPA) and a possible Special Area of Conservation (pSAC) (Langston *et al*., 2003).

Despite the many ecosystem services provided by the Severn Estuary and Inner Bristol Channel, for several decades, this ecosystem has been contaminated by heavy metals as reflected in several monitoring results published from the early 1970s (Butterworth *et al*., 1972; Chester and Stoner, 1975; Hamilton *et al*., 1979; Little and Smith, 1994). Although some authors have argued that heavy metal concentrations in the sediment of the Severn Estuary and Bristol Channel have begun to decline because of reduced industrial activity and improved environmental emission control, there remain areas of the estuary with locally elevated concentration levels that could have adverse effects on flora and fauna of the estuary (Duquesne *et al*., 2006; Langston *et al*., 2010).

In response to heavy metals enrichment of the Severn Estuary and Inner Bristol Channel, several studies have investigated the concentration of heavy metals in their sediments (Butterworth *et al*., 1972; Chester and Stoner, 1975; Hamilton *et al*., 1979; Thorne and Nickless, 1981; Little and Smith, 1994; Duquesne *et al*., 2006). However, these studies have only focused on reporting heavy metal concentration levels and establishing the metal concentrations associated with different types of sediment and particle sizes. No study has systematically investigated the spatial distributions of heavy metals in estuarine sediments, assessed the ecological risk posed by heavy metals, and apportioned the pollution sources of heavy metals in the sediment.

Hence, this study aims to (1) determine the spatial distribution of heavy metals in the sediments of the Severn Estuary and Inner Bristol Channel using Empirical Bayesian Kriging (EBK) interpolation; (2) check for exceedances of heavy metal concentration in the sediment of the Severn Estuary and Inner Bristol Channel by comparing monitored data with USEPA sediment quality guidelines (USEPA, 1999); (3) assess the ecological risk of metal contamination using pollution indices (4) determine the potential sources of heavy metals in the sediment of the Severn Estuary and Inner Bristol Channel using multivariate analysis.

METHODOLOGY

The study area

The study area of this research is the Severn Estuary and Inner Bristol Channel (Fig 1). The Severn Estuary is located between the south-west of England and south-east of Wales and stretches from Haw Bridge, North of Gloucester to a line between Lavernock Point (immediately south of Penarth n South Wales) and Brean Down (ABPmer and Atkins, 2010). The Inner Bristol Channel begins at the endpoint of the Severn Estuary and terminates at the point between Donats in South Wales and Minehead. The Severn Estuary and Bristol Channel together (Fig 2) form an extended and funnel-shaped area of sea that causes the tidal range to increase from a maximum of 7m at the outer reaches to about 14m at Avonmouth on springs and 6.5m on neaps (Binnie, 2016). Hence, the Severn Estuary is hyper-tidal and has one of the most extensive tidal range in the world (ABPmer and Atkins, 2010; Severn Estuary Partnership, 2011). The study area is also characterised by high-velocity currents and high turbidity (Manning *et al*., 2010).

The dispersive characteristics of the estuary have seen it formerly regarded as a receptor for the disposal of industrial effluents and sewage amounting to 0.2 × 106 m3 day -1 and 0.8 × 106 m3 day -1, respectively (Owens, 1984). The effect of considerable input of heavy metals into the estuary became evident after a comprehensive assessment was undertaken about 45 years ago (Abdullah *et al*., 1973; Abdullah and Royle, 1974). Recent studies have argued that heavy metal concentrations in the sediment of the Severn Estuary and Bristol Channel have begun to decline because of reduced industrial activity and improved environmental emission control. However, there remain areas of the estuary with locally elevated concentration levels that could have adverse effects on the biota of the estuary (Duquesne *et al*., 2006; Langston *et al*., 2010).

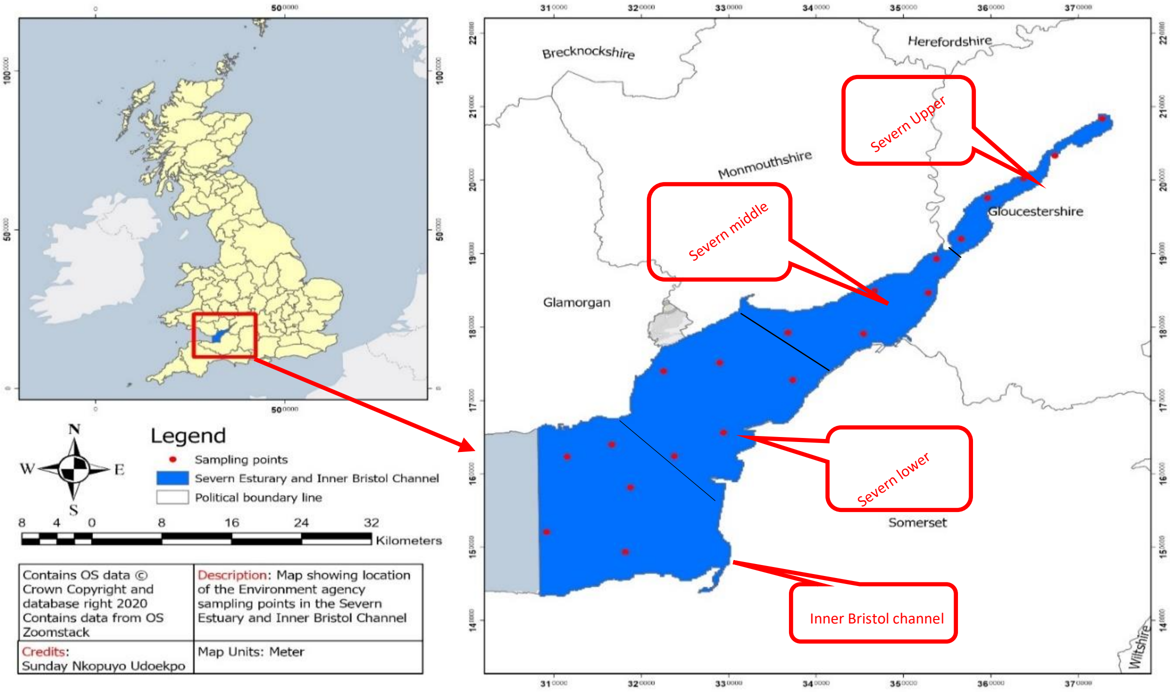

Figure 1: *Map of the study area showing location of sampling points. Lines within the estuary indicates the cut off between the distinct reaches of the Severn Estuary and Inner Bristol Channel*

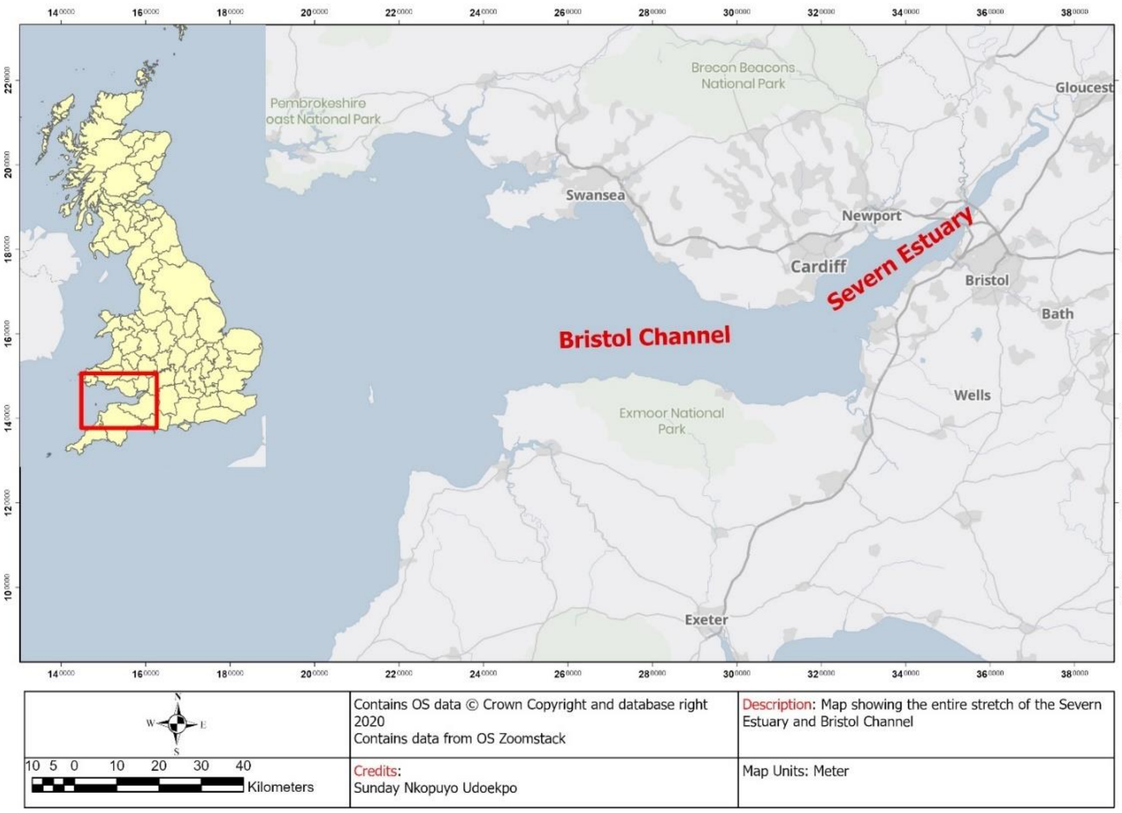

***Figure 2**: Map of the entire stretch of the Severn Estuary and Inner Bristol Channel*

Monitoring data of the Severn Estuary and Inner Bristol Channel

A Freedom of Information (FOI) request under the Freedom of Information Act 2000 (FOIA)/ Environmental Information Regulations 2004 (EIR) was made to the Environment Agency (EA) to provide the investigators with the recent available heavy metals monitoring data the agency holds on the Severn Estuary and Bristol Channel. The data obtained from the EA was the result of a 2016 monitoring survey on the Severn Estuary and Bristol Channel. The monitoring data which was sent by the Environment Agency in an Excel spreadsheet were filtered to remove certain information which was not relevant to the present study. The data were then reorganised in a format that would present heavy metal concentrations in both summer and winter seasons, and aid both spatial and statistical analysis of Cd, Cr, Ni, Zn and Pb, which are the heavy metals of concern. These metals have been chosen as the metals of concern in this study because they are most relevant metals discharged into the Severn Estuary and Inner Bristol Channel (Owen, 1984; Duquesne *et al*., 2006) and their concentrations are still regarded as high and so a cause for continuing concern (OSPAR Commission, 2000; Duquesne *et al*., 2006).

Spatial distribution of heavy metals

The Empirical Bayesian Kriging (EBK) model simulated in ArcGIS Pro version 2.6.2 software was used to model the spatial distribution of heavy metals in sediment of the Severn Estuary and Inner Bristol Channel because the model creates a spectrum of variograms which accounts for the error introduced by estimating a variogram model. The model also accounts for moderate non-stationarity by building local models on subsets of the input data (Krivoruchko, 2012; Gribov and Krivoruchko, 2020). A prediction surface and a prediction standard error surface, which are exported as rasters, using an extent and mask were the outputs of the EBK model. Figure 3 presents the procedure of the EBK interpolation used in this study.

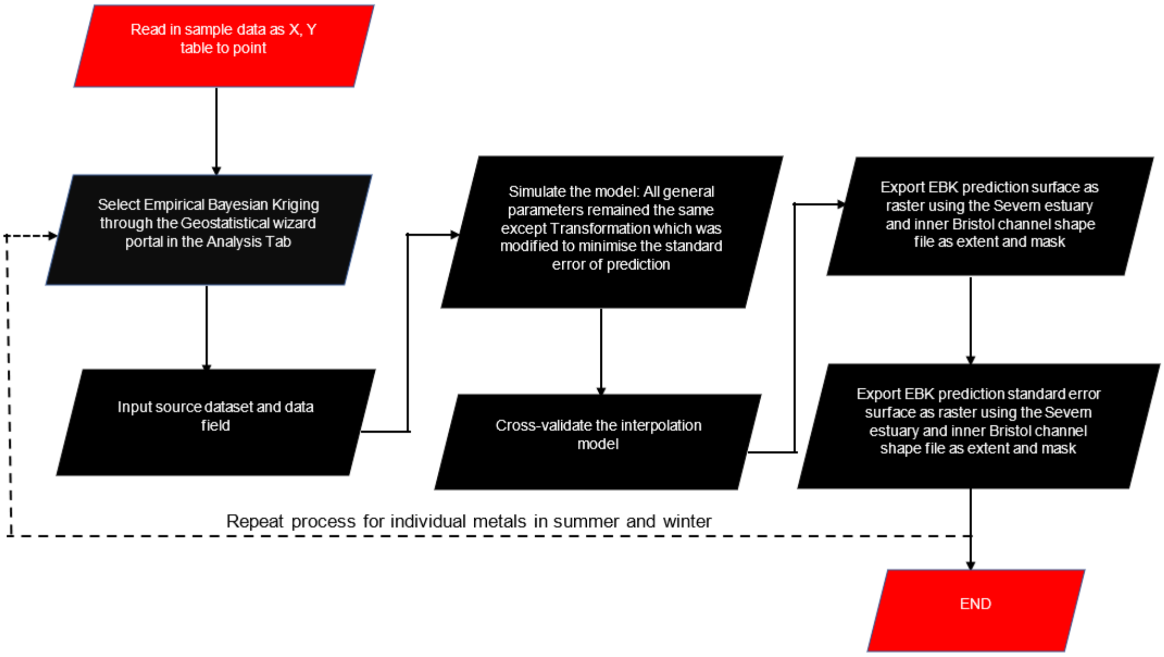

***Figure 3**: Procedures of Empirical Bayesian Kriging (EBK) used in this study*

Assessment of heavy metals using the USEPA sediment quality guideline

Monitored concentrations of Cd, Cr, Ni, Zn and Pb in the sediment of the Severn Estuary and Inner Bristol Channel were compared to the USEPA sediment quality guideline in Table 1 to determine the pollution status of the estuary.

Assessment of heavy metals in sediment using pollution indices

2.1.1 Geo-accumulation index (Igeo)

The geo-accumulation index proposed by Muller (1969) was used to determine metals contamination in sediment by comparing current concentrations with preindustrial levels.
The geo-accumulation index was calculated using the formula:

$$I_{geo} = \log_2\left[\frac{Cn}{(1.5\ Bn)}\right]$$

Where C_n is the measured concentration of the sediment for metal (n), B_n is the geochemical background value of metal (n) and factor 1.5 is the possible variations of background data due to lithogenic impacts. In the absence of background values of metals for the Severn Estuary and Inner Bristol Channel, the crustal abundance given by Wedepohl, (1995) was adopted as the background/preindustrial values of the Severn Estuary and Inner Bristol Channel in this research. Table 2 presents the classification scale of the geo-accumulation index proposed by Muller (1969).

Even though the Geo-accumulation index is widely used, this mathematical model still possesses some inherent limitations. Firstly, given the inhomogeneous geological conditions of sediments in most cases, geochemical background information is often represented in the form of uncertain intervals instead of concrete values (Matschullat *et al*., 2000; Štrbac *et al*., 2018), and the geo-accumulation index cannot deal with such uncertainty. Secondly, sediments usually contain multiple heavy metal species (Ma *et al*., 2016; Gloaguen and Passe, 2017; Ke *et al*., 2017). However, the geo-accumulation index only quantifies the pollution degree of a single heavy metal and cannot perform a comprehensive pollution evaluation of multiple heavy metals.

Table 1: *USEPA sediment quality guideline*

	Heavy metals (mg/kg)				
	Cd	Cr	Ni	Zn	Pb
Not polluted	-	<25	<20	<90	<40
Moderately polluted	-	25 – 75	20 – 50	90 – 200	40 – 60
Heavily polluted	>6	>75	>50	>200	>60

Adapted from USEPA, (1999)

Table 2: *Classification scale of the geo-accumulation index*

Igeo Value	**Class**	**Sediment Quality**
≤ 0	0	Unpolluted
0 – 1	1	From unpolluted to moderately polluted
1 – 2	2	Moderately polluted
2 – 3	3	From moderately to strongly polluted
3 – 4	4	Strongly polluted
4 – 5	5	From strongly to extremely polluted
> 6	6	Extremely polluted

Contamination factor (CF)

The contamination factor (CF) proposed by Hakanson (1980) was used to assess the degree of contamination of sediment by metal.

The contamination factor (CF) was calculated using the formula:

$$Cf = \frac{C_{metal}}{C_{background}}$$

Where, C_{metal} is the concentration of the given metal in sediment, and $C_{background}$ is the crustal abundance given by Wedepohl, (1995). Table 3 presents the classification scheme used to describe the contamination factor.

Table 3: *Classification scheme used to describe the contamination factor*

CF	**Description**
CF < 1	Low degree of contamination
1 – 3	Moderate degree of contamination
3 – 6	Considerable degree of contamination
CF > 6	Very high degree of contamination

Adapted from Hakanson (1980).

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Metal pollution index
The Metal pollution index (MPI) was calculated according to Usero *et al*. (1997) to assess the overall degree of river sediment metal contamination.
The Metal pollution index (MPI) was calculated using the formula:
$MPI = (M_1 \times M_2 \times M_3 \times \ldots \ldots \ldots \ldots \ldots \times M_n)^{(1/n)}$
Where M_n, is the concentration of metal n expressed in mg/kg of dry weight. This model is simple but does not compare the contaminant concentration with any baseline. No sediment threshold classification from unpolluted to highly polluted. Although it has Geometric average advantages when compared with other indices since it shows concentration differences (Kalimur, 2011).
The pollution load index (PLI)
For each contaminant, the PLI was calculated based on the formula proposed by Wilson and Jeffrey (1987):

$$PLI = antilog \log_{10}(1 - \frac{C - B}{T - B})$$

Where B, is the baseline value, T is the threshold, and C is the concentration associated with degradation or changes in the quality of the aquatic system. Baseline values for each contaminant proposed by Wilson and Jeffrey (1987) were used except the baseline value of Cd. The reason is that, in some sample locations, Cd concentrations were lower than the baseline values proposed by Wilson and Jeffrey (1987). Therefore, the minimum concentration of Cd in this study was used as the baseline value of Cd which would eliminate any error in the index calculation that would have resulted from the use of Wilson and Jeffrey (1987) baseline value. Wilson and Jeffrey (1987) defined T for the different contaminants, and C is the concentration of the pollutant. For each place, the PLI calculation considers any (or n) contaminants:
$PLI = (PLI_1 \times PLI_2 \times PLI_3 \times \ldots \ldots \ldots \times PLI_n)^{(1/n)}$
PLI varies from 10 (unpolluted) to 0 (highly polluted).

This index allows a comparison between several estuarine systems. It has been applied successfully in European aquatic systems (Wilson, 2003). However, the values of baseline and threshold are not defined locally for each coastal zone investigated (Caeiro *et al*., 2005).
In sum, these pollution indices employed considered the enrichment, pollution status of a single element and combined effects of heavy metals. Therefore, it is essential to determine the values of several combined indices for a comprehensive understanding of heavy metal pollution in sediments.

Statistical analysis
Firstly, descriptive statistics, including mean and standard error of mean (SEM) were calculated to give a summary of the dataset in this study (Kaliyadan and Kulkarni, 2019). The data were subjected to one-way analysis of variance (ANOVA) to test for significant differences at $p \leq 0.05$ between the means of the distinct reaches of the study area in summer and winter. Where significant differences were recorded, a posthoc analysis called the Tukey's HSD (Honest Significant Difference) was used to compare means of the distinct reaches of the study area (Shi *et al*., 2019). A two-sample t-test was used to test for significant differences at $p \leq 0.05$ between the concentration of heavy metals in summer and winter (Bastami *et al*., 2014). Finally, multivariate analysis, including Pearson's correlation, cluster analysis (CA) and principal component analysis (PCA), was conducted to determine potential sources of heavy

metals (Bhuyan *et al*., 2019). All statistical analysis was conducted using the R statistical programming language version 4.0.3.

RESULTS AND DISCUSSION

Seasonal variations of heavy metal concentration in sediments

The seasonal variations between heavy metals concentration in the different reaches of the Severn Estuary and Inner Bristol Channel are presented in Table 4 Results of the two-sample t-test, which shows whether there is a significant difference at $p \leq 0.05$ between the concentration of heavy metals in summer and winter are contained in Table 5. The concentration of heavy metals had the trend Zn > Cr > Pb > Ni > Cd irrespective of the season. Furthermore, it was revealed that summer sediment concentrations of all five metals, Cd, Cr, Ni, Zn and Pb, were higher than winter concentrations. The two-sample t-test confirmed that at $p < 0.05$, there was a statistically significant difference between the concentration of heavy metals in summer and winter.

Although there is a statistically significant difference between metals concentration in summer and winter, a critical view of the heavy metal concentration figures in summer and winter reveals that the figures are not wildly apart. Therefore, the reason for the significantly higher concentrations recorded in summer might be attributed to the fact that the summer season is marked with increased temperatures, decreased pH, and low dissolved oxygen which might cause an increased concentration of heavy metals in sediments. Meanwhile, during winter, low temperatures, increased dissolved oxygen and high pH might be responsible for the lower concentrations compared to the summer season. Albeit some authors have suggested that high pH during winter might be a responsible for reduced heavy metal concentration in sediments, the relationships that exist between sediment pH and heavy metals are complex and therefore requires further study (Li and Zhang, 2010; Haiyan *et al*., 2013; Baxa *et al*., 2020). Similarly, Sarasiab *et al*. (2014) also reported higher concentrations of heavy metals in soft sediments from the Arvand River in the Persian Gulf in the summer season. The author attributed high summer concentrations to variation in temperature, salinity, pH, and summer discharge.

Table 4: *Seasonal variations between heavy metals in the Severn Estuary and Inner Bristol Channel*

Heavy metals (mg/kg)	season	Reaches of the Severn Estuary and Inner Bristol Channel			
		Severn upper n = 5	Severn middle n = 5	Severn lower n = 5	Inner Bristol Channel n = 5
Cd	Summer	0.13 ± 0.03	0.50 ± 0.05	0.35 ± 0.04	0.20 ± 0.03
	Winter	0.09 ± 0.02	0.38 ± 0.06	0.27 ± 0.03	0.13 ± 0.03
Cr	Summer	50.26 ± 4.70	88.45 ± 5.21	67.67 ± 8.95	57.87 ± 6.26
	Winter	43.44 ± 5.51	78.43 ± 3.16	60.77 ± 8.11	53.37 ± 5.91
Ni	Summer	25.30 ± 2.27	38.31 ± 1.00	33.57 ± 2.41	30.92 ± 2.05
	Winter	21.64 ± 2.49	35.29 ± 1.63	30.83 ± 2.38	25.98 ± 1.35
Zn	Summer	133.68 ± 15.84	211.69 ± 9.58	196.27 ± 6.15	149.90 ± 10.13
	Winter	122.55 ± 14.82	199.84 ±11.98	190.33 ± 6.15	141.21 ± 9.34
Pb	Summer	39.33 ± 5.36	67.53 ± 3.81	57.23 ± 2.93	42.34 ± 3.45
	Winter	34.66 ± 5.15	60.19 ± 2.81	53.46 ± 3.20	37.82 ± 3.97

Values are Mean ± Standard Error of mean (SEM).

Table 5: *Results of the two-sample t-test for seasonal variations of the heavy metals*

Season	Heavy metals (mg/kg)				
	Cd	Cr	Ni	Zn	Pb
Summer	0.30 ± 0.04	66.06 ± 4.43	32.02 ± 1.42	172.88 ± 8.92	51.61 ± 3.20
Winter	0.22 ± 0.03	59.00 ± 4.00	28.44 ± 1.50	163.48 ± 9.01	46.53 ± 3.02
P value (significance level)	$3.802e^{-09}$	$5.998e^{-06}$	$1.324e^{-07}$	$1.876e^{-07}$	$1.491e^{-08}$

Values are Mean ± Standard Error of Mean (SEM)

Spatial distribution of heavy metals

The EBK model was used to predict the spatial distribution patterns of heavy metals in sediment of the Severn Estuary and Inner Bristol Channel. The model outputs the actual prediction surface and a prediction standard error surface that measures the uncertainties in the model prediction. The actual prediction maps of heavy metals distribution patterns in summer and winter are shown in Fig 4 to Fig 13, while maps of the prediction standard error are

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presented in the supplementary data. The result of the one-way ANOVA for each heavy metal concentration in summer and winter seasons showed a significant difference between the mean concentration in the distinct reaches of the study area at $p \leq 0.05$. Since statistically significant differences were recorded at $p \leq 0.05$, the Tukey's test was conducted to compare mean concentrations of heavy metals in the distinct reaches in the study area during summer and winter. The result of the Tukey's test for each heavy metal is presented alongside its corresponding spatial distribution map in Fig 4 to Fig 13. From the prediction maps of heavy metals distribution patterns in summer and winter, one can observe that irrespective of season, concentrations of Cd, Cr, Ni, Zn and Pb in the distinct reaches followed the trend Severn middle > Severn lower > Inner Bristol Channel > Severn upper.

Heavy metals concentration was generally low in Severn upper during the summer and winter seasons. In Severn middle, metals concentration peaked and then decreased from the Severn lower to the Inner Bristol Channel. The spatial trend of heavy metals concentration in the distinct reaches of the Severn estuary and Inner Bristol Channel is a significant finding from this study as it indicated a point-source type of pollution. Heavy metal concentration hotspots were noticed in the Severn middle where a more significant number of industrial sites on the Severn Estuary are located (Duquesne *et al*., 2006). Therefore, this study suggests that, although there have been reduced industrial activities and improved environmental emission control in the Severn Estuary and Inner Bristol Channel (Duquesne *et al*., 2006), the high concentration of heavy metals observed in the Severn middle area might be attributed to the activities of the numerous process industries, coal mines and sewage treatment works (STWs) which had operated or are still operating in the area. Butterworth *et al*. (1972) and Duquesne *et al*. (2006) recorded a similar spatial variation of heavy metals where the authors found the highest metal concentration in the Avonmouth and Portishead area which are located in the Severn middle area in this study.

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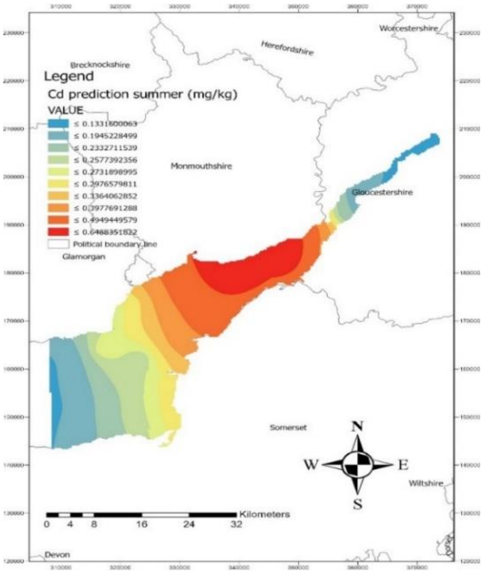

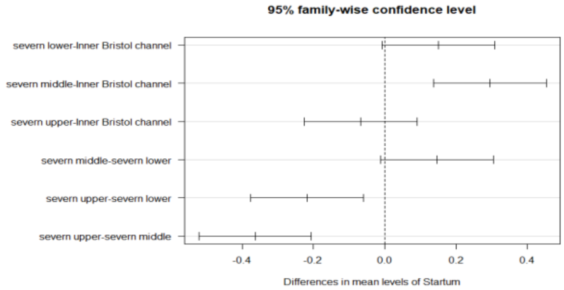

Key: *Any confidence intervals that do not contain 0 indicates that there is a statistically significant difference in the groups.*

Figure 4: Spatial distribution of Cd in soft sediments during summer(a). Pair-wise comparisons from Tukey's HSD for the mean concentrations of Cd in the distinct reaches of the study area during summer (b).

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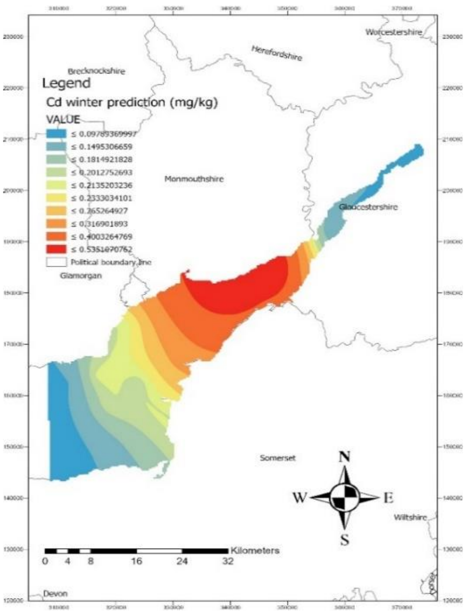

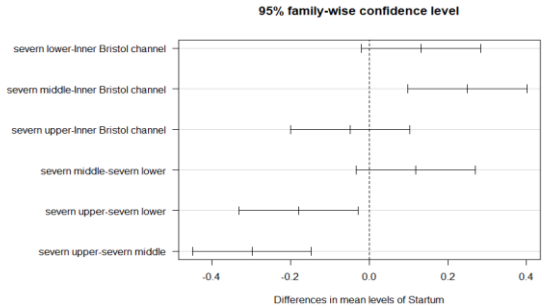

Key: *Any confidence intervals that do not contain 0 indicates that there is a statistically significant difference in the groups.*

Figure 5: Spatial distribution of Cd in soft sediments during winter (a). Pair-wise comparisons from Tukey's HSD for the mean concentrations of Cd in the distinct reaches of the study area during winter (b).

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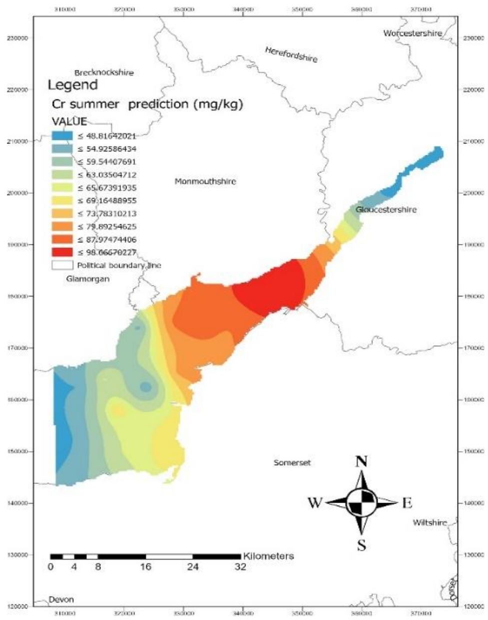

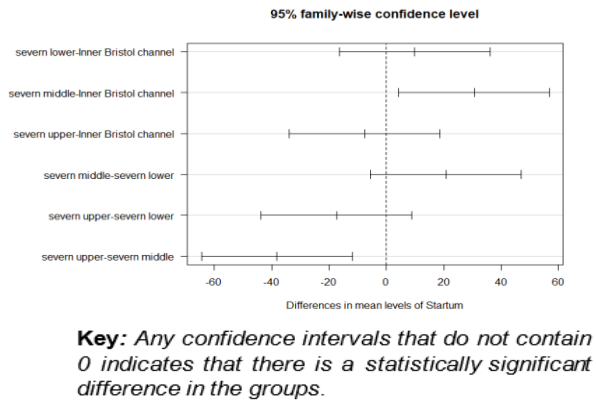

Figure 6: Spatial distribution of Cr in soft sediments during summer (a). Pair-wise comparisons from Tukey's HSD for the mean concentrations of Cr in the distinct reaches of the study area during summer (b).

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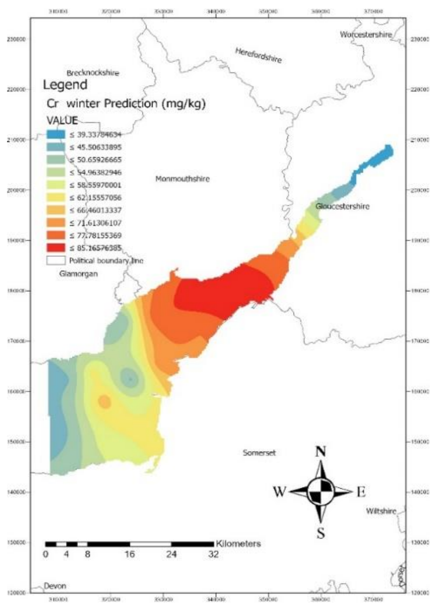

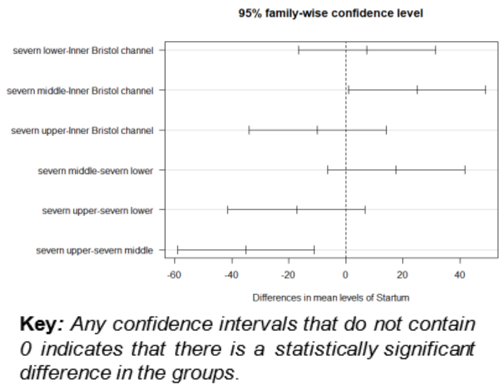

Figure 7: Spatial distribution of Cr in soft sediments during winter (a). Pair-wise comparisons from Tukey's HSD for the mean concentrations of Cr in the distinct reaches of the study area during winter (b).

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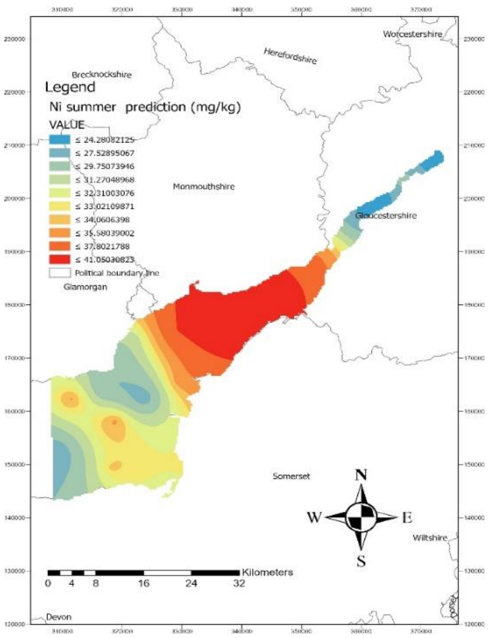

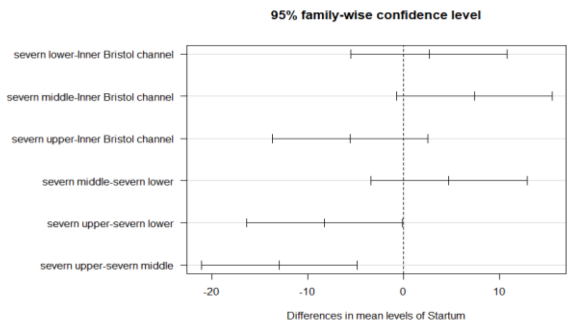

Key: *Any confidence intervals that do not contain 0 indicates that there is a statistically significant difference in the groups.*

Figure 8: Spatial distribution of Ni in soft sediments during summer (a). Pair-wise comparisons from Tukey's HSD for the mean concentrations of Ni in the distinct reaches of the study area during summer (b).

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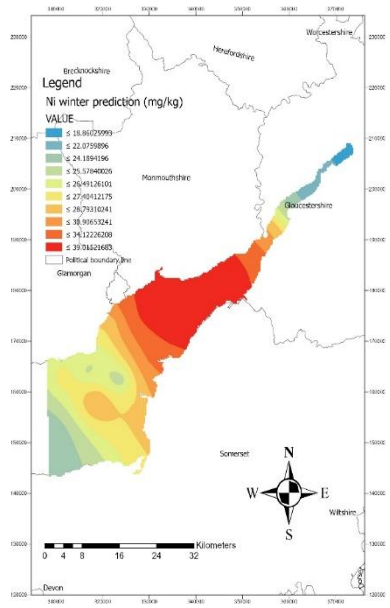

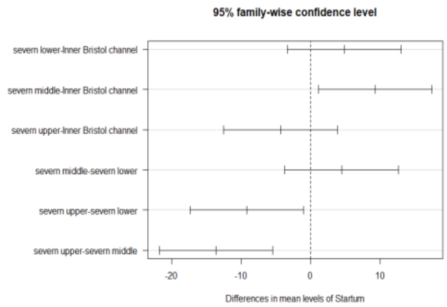

Key: *Any confidence intervals that do not contain 0 indicates that there is a statistically significant difference in the groups.*

Figure 9: Spatial distribution of Ni in soft sediments during winter (a). Pair-wise comparisons from Tukey's HSD for the mean concentrations of Ni in the distinct reaches of the study area during winter (b).

A **B**

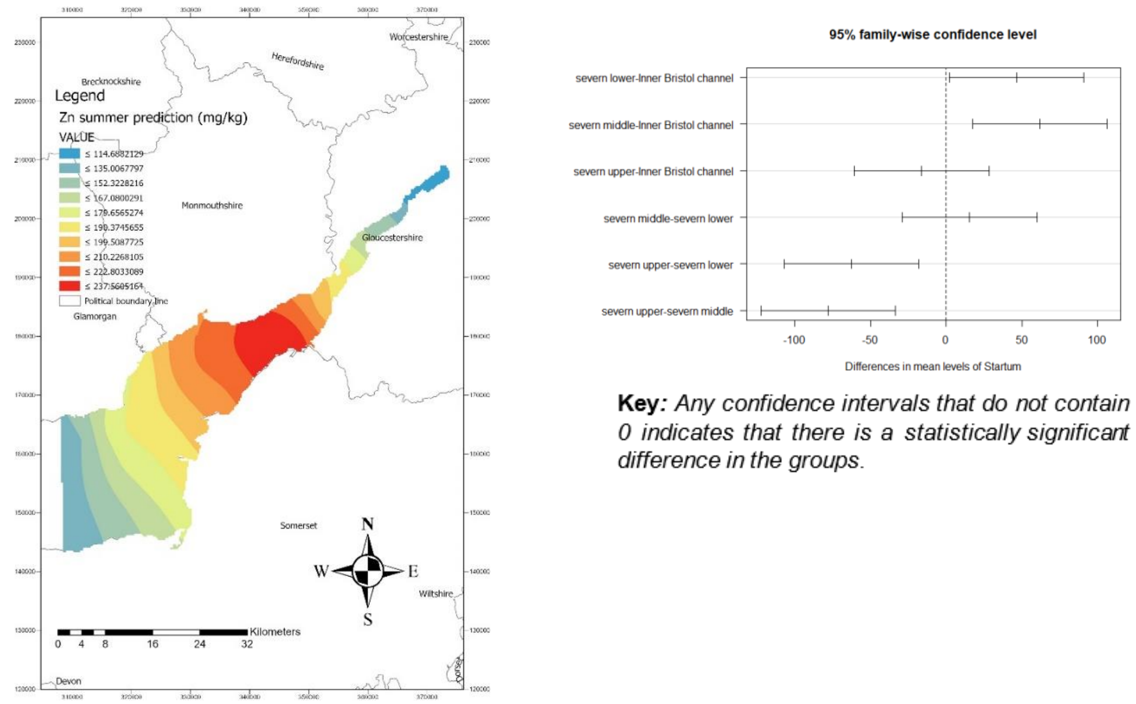

Figure 10: Spatial distribution of Zn in soft sediments during summer (a). Pair-wise comparisons from Tukey's HSD for the mean concentrations of Zn in the distinct reaches of the study area during summer (b).

A **B**

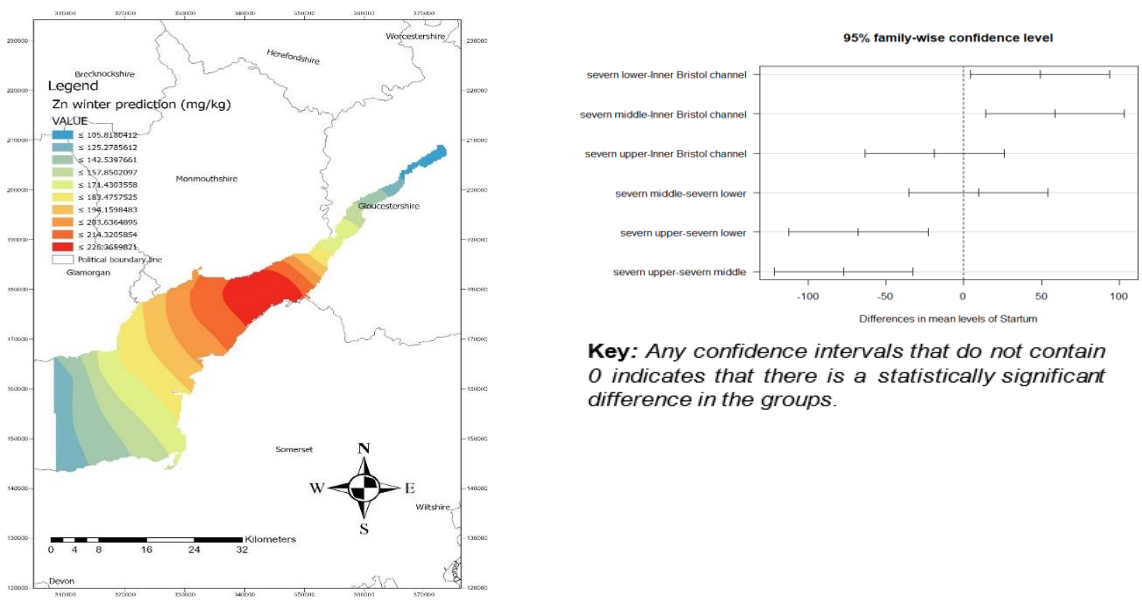

Figure 11: Spatial distribution of Zn in soft sediments during winter (a). Pair-wise comparisons from Tukey's HSD for the mean concentrations of Zn in the distinct reaches of the study area during winter (b).

A **B**

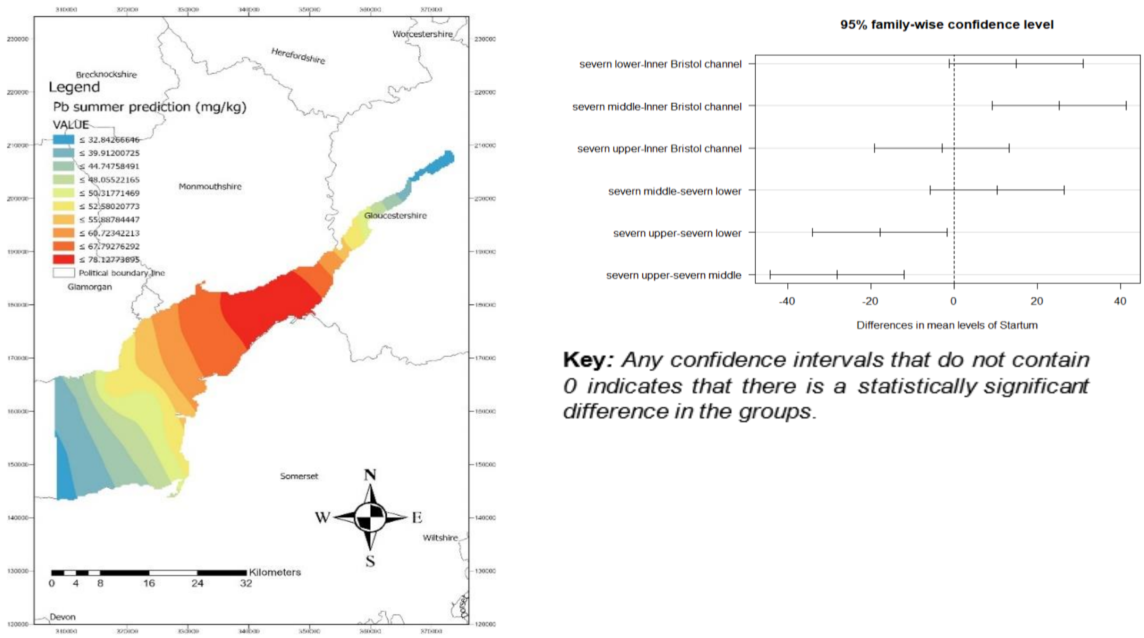

Figure 12: Spatial distribution of Pb in soft sediments during summer (a). Pair-wise comparisons from Tukey's HSD for the mean concentrations of Pb in the distinct reaches of the study area during summer (b).

A **B**

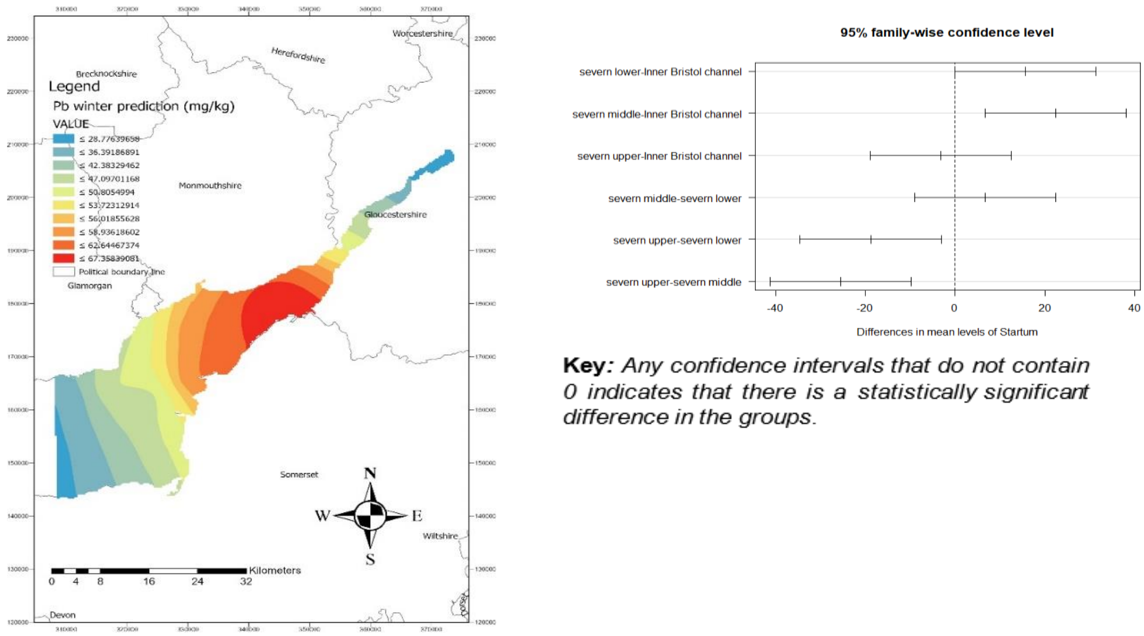

Figure 13: Spatial distribution of Pb in soft sediments during winter (a). Pair-wise comparisons from Tukey's HSD for the mean concentrations of Pb in the distinct reaches of the study area during winter (b).

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Comparison between USEPA sediment quality guidelines and monitored data

Average heavy metal concentrations between summer and winter seasons in the distinct reaches of the Severn Estuary and Inner Bristol Channel were compared with the USEPA sediment quality guideline in the absence of a formal sediment quality guideline in the UK. Th result of the comparison is presented in Table 6. These results confirm in no small extent the result of the spatial distribution of pollutants where local elevations of metal concentration were observed in the Severn middle area. Although the USPEA SQG is not a definitive indicator of toxicity (Hubner *et al*., 2009), it has a high predictive ability and is a useful tool for identifying areas with potentially adverse biological effects (Pazi, 2011; Awosusi and Adisa, 2020).

Table 6: *Comparison between USEPA sediment quality guideline and monitored concentrations*

Heavy metals (mg/kg)	Cd	Cr	Ni	Zn	Pb
USEPA Sediment quality Guideline					
Not polluted	-	<25	<20	<90	<40
Moderately polluted	-	25 – 75	20 – 50	90 – 200	40 – 60
Heavily polluted	>6	>75	>50	>200	>60
Monitored concentrations					
Severn upper	0.11	46.85	23.47	128.12	37.00
Severn middle	0.44	83.44	36.80	205.76	63.86
Severn lower	0.31	64.22	32.20	193.30	55.35
Inner Bristol channel	0.17	55.62	28.45	145.55	40.08
Results					
Severn upper	Not polluted	Moderately polluted	Moderately polluted	Moderately polluted	Not polluted
Severn middle	Not polluted	Heavily polluted	Moderately polluted	Heavily polluted	Heavily polluted
Severn lower	Not polluted	Moderately polluted	Moderately polluted	Moderately polluted	Moderately polluted
Inner Bristol channel	Not polluted	Moderately polluted	Moderately polluted	Moderately polluted	Moderately polluted

Assessment of heavy metals in sediment using pollution indices

Geo-accumulation index (Igeo)

The geo-accumulation index (Igeo) at the different reaches of the Severn Estuary and Inner Bristol Channel is shown in Table 7. Figure 14 attempts to describe the result presented in Table 7, according to Muller (1969). It was observed that in Severn upper, Igeo scores for Cd, Cr and Ni in sediments were >0, which means that the Severn upper is *not polluted* with Cd, Cr and Ni. However, Igeo scores for Zn and Pb lay between 0 and 1, which implies that the Severn upper is *from unpolluted to moderately polluted* with Zn and Pb. In Severn middle, Igeo

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scores for Cr and Ni lay between 0 and 1, meaning that the sediments in the Severn middle are *from unpolluted to moderately polluted* with Cr and Ni. Meanwhile, Igeo scores for Cd, Zn, and Pb lay between 1 and 2, which suggests that the Severn middle is *moderately polluted* with Cd, Zn, and Pb. The Igeo scores for Cr and Ni in Severn lower lay between 0 and 1, which means that the Severn lower is *from unpolluted to moderately polluted* with Cr and Ni. Meanwhile, Igeo scores for Cd, Zn and Pb lay between 1 and 2 meaning that sediments of the Severn lower are *moderately polluted* with Cd, Zn and Pb. In the Inner Bristol Channel, Igeo scores of all metals (Cd, Cr, Ni, Zn and Pb) lay between 0 and1 which means that sediments of the Inner Bristol Channel are from *unpolluted to moderately polluted* with Cd, Cr, Ni, Zn, and Pb.

Table 7: *Geo-accumulation index (Igeo) at the different reaches of the study area*

	Reaches of the Severn Estuary and Inner Bristol Channel			
Heavy metal	Severn upper	Severn middle	Severn lower	Inner Bristol channel
Cd	-0.45	1.56	1.04	0.16
Cr	-0.16	0.67	0.29	0.08
Ni	-0.25	0.40	0.21	0.03
Zn	0.72	1.40	1.31	0.90
Pb	0.54	1.32	1.12	0.65

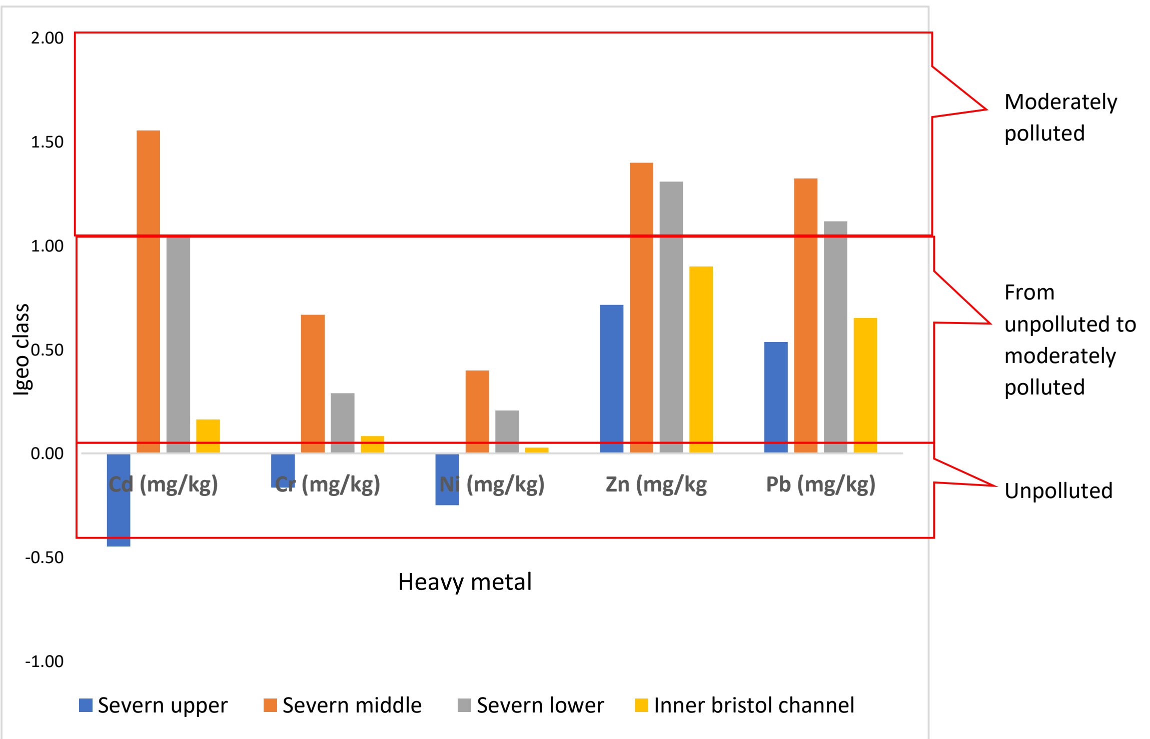

Figure 14: Spatial variation of Igeo index in the study area

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Contamination factor (CF)

The contamination factor (CF) at the different reaches of the Severn Estuary and Inner Bristol Channel is shown in Table 8. Figure 15 describes the result presented in Table 8, according to Hakanson (1980). The CF of Cd, Cr, Ni, Zn, and Pb in Severn upper lay between 1 – 3. These values imply that the sediments in the Severn upper are classified as having a *moderate degree of contamination*. In Severn middle, CF of Cr and Ni lay between 1 – 3 which means that the sediments of the Severn middle are classified as having a *moderate degree of contamination* concerning concentrations of Cr and Ni while CF of Cd, Zn and Pb lay between 3 – 6 implying a *considerable degree of contamination* by Cd, Zn and Pb. The CF of Cr and Ni in Severn lower lay between 1 – 3 which indicates that the sediment of the Severn lower has a *moderate degree of contamination* with regards to Cr and Ni while the CF of Cd, Zn and Pb lay between 3 – 6 which means that the sediment of the Severn lower has a *considerable degree of contamination* with regards to Cd, Zn and Pb. In the Inner Bristol Channel, the CF of Cd, Cr, Ni, Zn, and Pb all lay between 1 – 3. Therefore, it means that the sediment of the Inner Bristol Channel can be said to have a *moderate degree of contamination*.

Table 8: *Contamination factor (CF) at the different reaches of the study area*

	Reaches of the Severn Estuary and Inner Bristol Channel			
Heavy metal	Severn upper	Severn middle	Severn lower	Inner Bristol channel
Cd	1.10	4.41	3.09	1.68
Cr	1.34	2.38	1.83	1.59
Ni	1.26	1.98	1.73	1.53
Zn	2.46	3.96	3.72	2.80
Pb	2.18	3.76	3.26	2.36

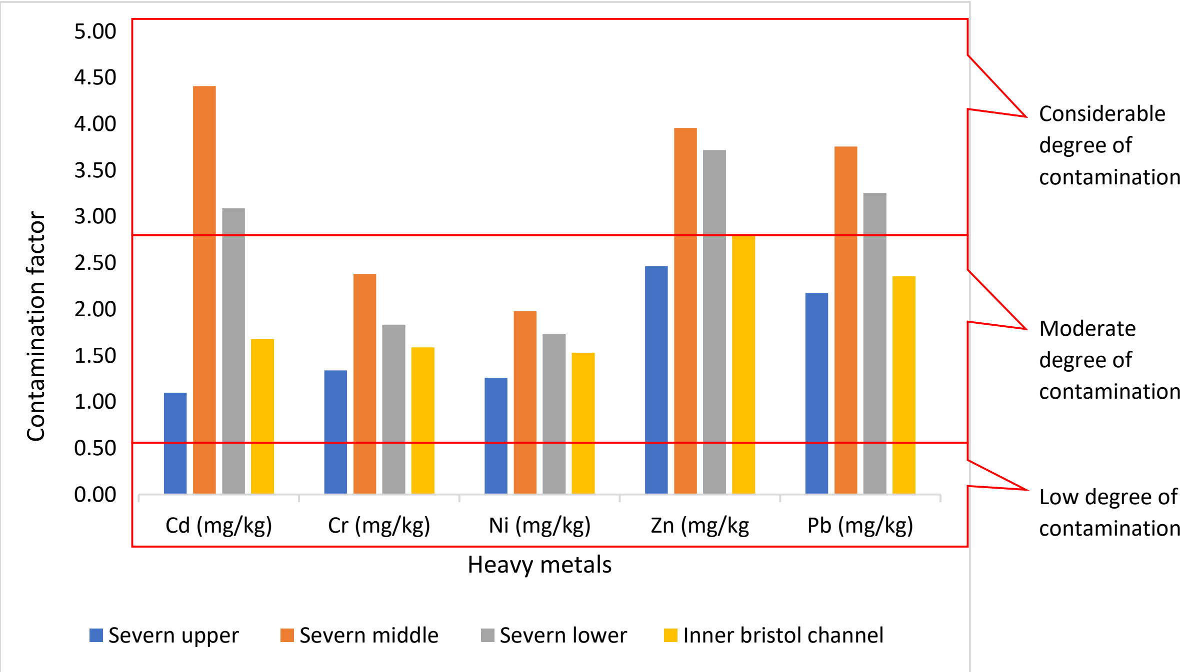

Figure 15: Spatial variation of CF in the study area

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Metal pollution index (MPI)
The MPI at the different reaches of the Severn Estuary and Inner Bristol Channel is presented in Table 9. The MPI considered the combined effects of heavy metals in each of the reaches of the estuary. It is useful in presenting a clear picture of the overall degree of metal contamination in each section of the Severn Estuary and Inner Bristol. From the MPI result presented in Table 9, it is evident that the Severn upper having an MPI of 14.18 is the least contaminated followed by the Inner Bristol channel (17.30), Severn lower (23.28) and the Severn middle having an MPI of 28.19 is the most contaminated site. Therefore, heavy metal contamination in the distinct reaches of the Severn Estuary and Inner Bristol Channel is ranked as Severn middle > Severn lower > Inner Bristol channel > Severn upper. This result again is similar to the findings in the spatial distribution of heavy metals where the same trend of heavy metals contamination in the distinct reaches was observed.

Table 9: *Metal Pollution Index (MPI) at the different reaches of the study area*

Reaches of the Severn Estuary and Inner Bristol Channel	Metal pollution index
Severn upper	14.18
Severn middle	28.19
Severn lower	23.28
Inner Bristol channel	17.30

The pollution load index (PLI)
The PLI at the different reaches of the Severn Estuary and Inner Bristol Channel is presented in Table 10. The PLI focused on the enrichment of heavy metals in each of the reaches of the estuary. PLI values of 2.52, 0.76, 1.13 and 1.91 were recorded for Severn upper, Severn middle, Severn lower and Inner Bristol Channel, respectively. According to Wilson and Jeffrey (1987), PLI varies from 10 (unpolluted) to 0 (highly polluted) and the values of PLI in the research were within 0 and 10. Therefore, each of the reaches of the Severn Estuary and Bristol Channel can be described to be moderately polluted. However, in order of magnitude of pollution load, the Severn middle had the highest pollution load followed by Severn lower, Inner Bristol Channel and Severn upper, which had the least pollution load.

Table 10*: Pollution Load Index (PLI) at the different reaches of the study area*

Reaches of the Severn Estuary and Inner Bristol Channel	Pollution load index
Severn upper	2.52
Severn middle	0.76
Severn lower	1.13
Inner Bristol channel	1.91

In sum, both the USEPA SQGs and the pollution indices employed in this research considered the pollution status of a single element, combined effects of heavy metals and heavy metals enrichment in the distinct reaches of the Severn Estuary and Inner Bristol Channel. Therefore, it is essential to determine the values of several combined indices for a comprehensive understanding of heavy metal pollution in sediments. Furthermore, form the findings of the assessment using the USEPA SQGs and pollution indices, it is evident that concentration heavy metals such as Zn, Pb, Ni and Cr are above thresholds in the distinct reaches of the estuary and might pose an adverse effect on biota and ecosystem services of the Severn Estuary and Inner Bristol Channel.

Identification of potential sources of heavy metals

The relationships that exist between heavy metals can suggest the sources of heavy metals in sediments of aquatic ecosystems (Tchounwou *et al*., 2012). Multivariate analysis (Pearson correlation, Hierarchical cluster analysis (HCA) and Principal component analysis (PCA)) were used to suggest potential sources of heavy metals by determining heavy metal relationships. The correlation matrix presented in Table 11 revealed that all of the five heavy metals showed strong correlations between themselves which were statistically significant at $p < 0.05$. This result suggests that Cd, Cr, Ni, Zn and Pb might have originated from the same sources which could be anthropogenic—thereby overriding any other assumption that natural sources such as mineralisation of metal-rich rocks might be responsible for increased levels of heavy metals concentration in the Severn Estuary and Inner Bristol Channel (Bagheri *et al*., 2011; Sakan *et al*., 2015).

Since the Pearson correlation has already suggested all heavy metals to originate from anthropogenic sources, the dendrogram presented in Figure 16 which shows the hierarchical clusters of heavy metals and the PCA loading plot in Figure 18 further explains what anthropogenic sources are responsible for the presence of the heavy metals in the sediment of the Severn Estuary and Inner Bristol Channel. Langston *et al*. (2003) suggest that the operations of process industries at Severnside, sewage treatment, coal mining in the Rhondda valley and agricultural run-off may cause pollution by heavy metals in the sediments of the Severn Estuary and Inner Bristol Channel. The patterns of HCA and PCA clustering would help understand which of these processes is responsible for the presence of the Cd, Cr, Ni, Zn and Pb in sediments.

The HCA revealed two major clusters. Cluster one consists of Zn while cluster two had two sub-clusters including Cr and Pb in the first cluster and Cd and Ni in the second. This result reveals that Zn may have originated from coal mining and several manufacturing processes involving metals which have taken place in recent years (Dong *et al*., 2012). Cr, Pb, Cd and Ni may have originated from agricultural run-offs, sewage treatment and discharge of effluents from process industries (Bagheri *et al*., 2011).

The scree plot in Figure 17 helped to determine the number of principal components based on the percentage of explained variances. It was observed that the scree plot was steep between dimension 1 and 2 before levelling out. This observation means that the first two principal components (PCs) are the most significant. A similar result was observed in the HCA where two clusters were also observed. The two PCs explain 95.6% of the total variance, as shown in the PCA loading plot for heavy metals in Figure 18.

Furthermore, the first PC, including Zn, Pb and Cd explains 91.6% of the total variance while the second PC, including Cr and Ni, explains 4% of the total variance. The first PC can be considered to reveal in addition to coal mining and several manufacturing processes, run-offs from roads due to the wearing of brake lining and tyres, oil spills and emission from the exhaust

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of vehicles as sources of Cd, Zn and Pb (Napier et al., 2008; Dong *et al*., 2012; Ozonzeadi, 2014; Hanfi *et al*., 2020). The second PC suggests discharge of effluents from process industries and sewage treatment as the sources of Cr and Ni in the sediments of the Severn Estuary and Inner Bristol Channel (Bagheri *et al*., 2011; Aliu *et al*., 2020).

Table 11*: Correlation matrix showing the coefficients of correlation between metal concentrations in the sediment of the study area*

	Cd	Cr	Ni	Zn	Pb
Cd	1				
Cr	0.9^{***}	1			
Ni	0.86^{***}	0.88^{***}	1		
Zn	0.91^{***}	0.87^{***}	0.85^{***}	1	
Pb	0.93^{***}	0.91^{***}	0.86^{***}	0.98^{***}	1

Values with superscript *** indicate a statistically significant correlation at $p<0.05$.

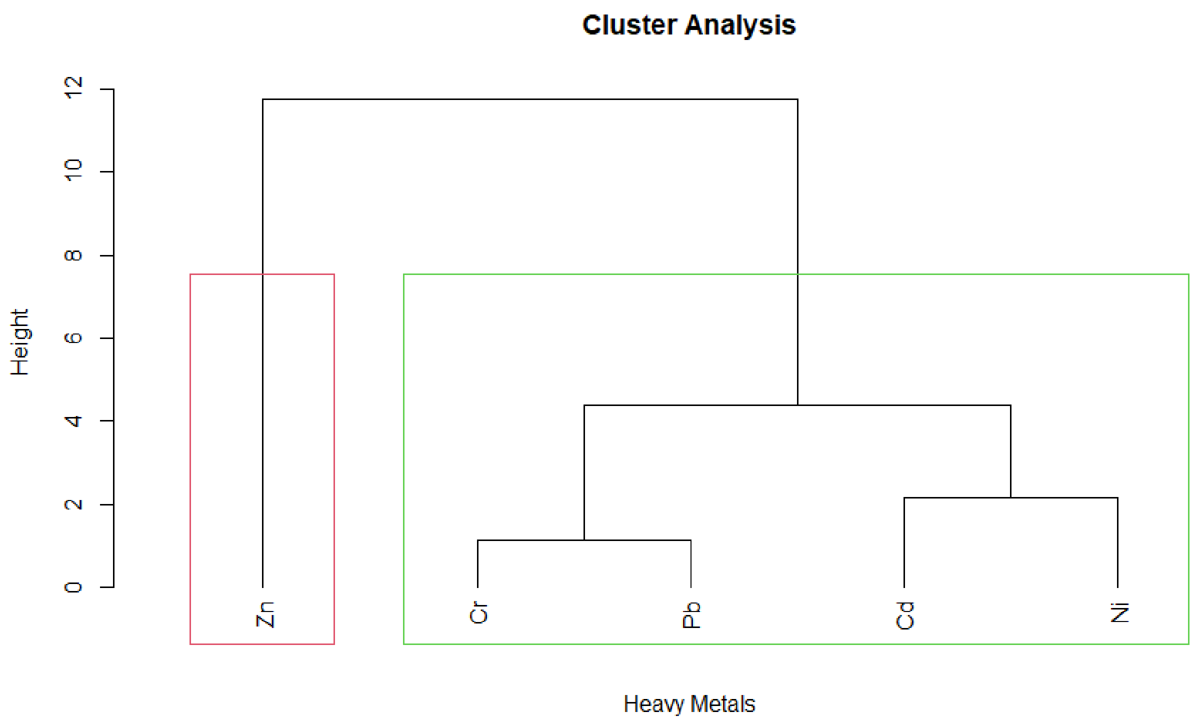

Figure 16: *Dendrogram showing heavy metal clusters in the sediment of the Severn Estuary and Inner Bristol Channel*

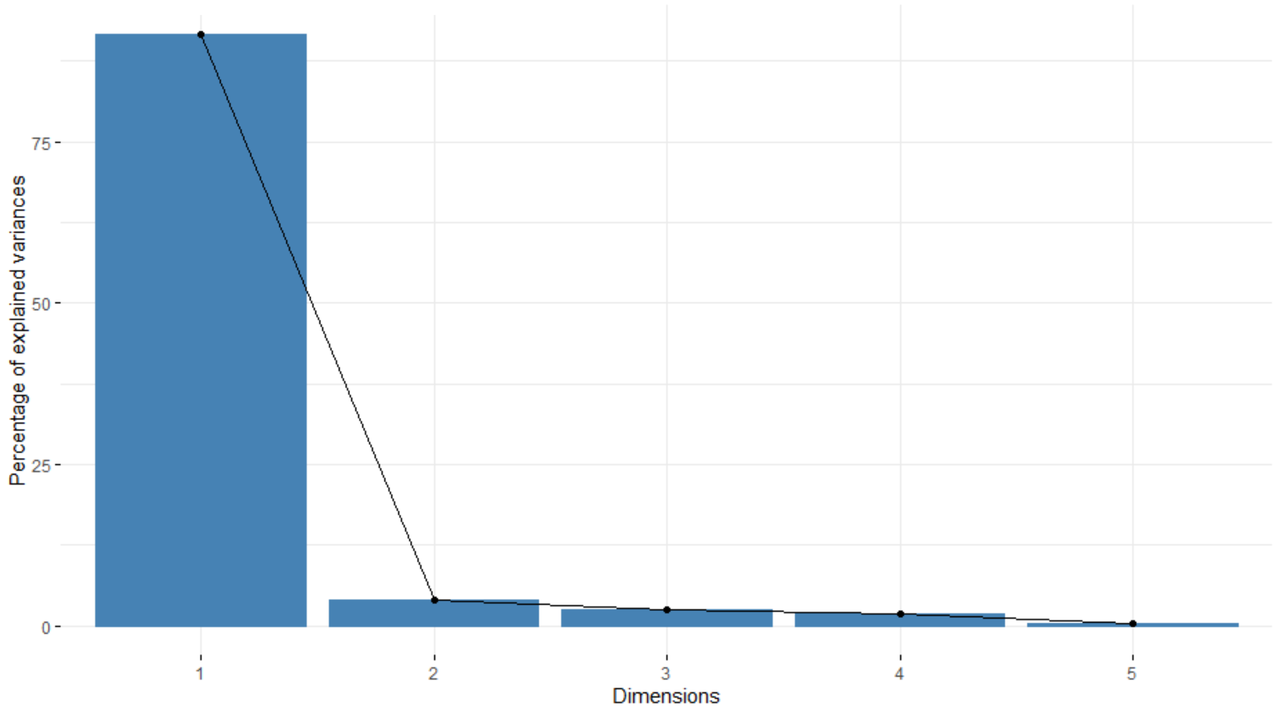

Figure 17: *Scree plot of the percentage of variance explained by each principal component*

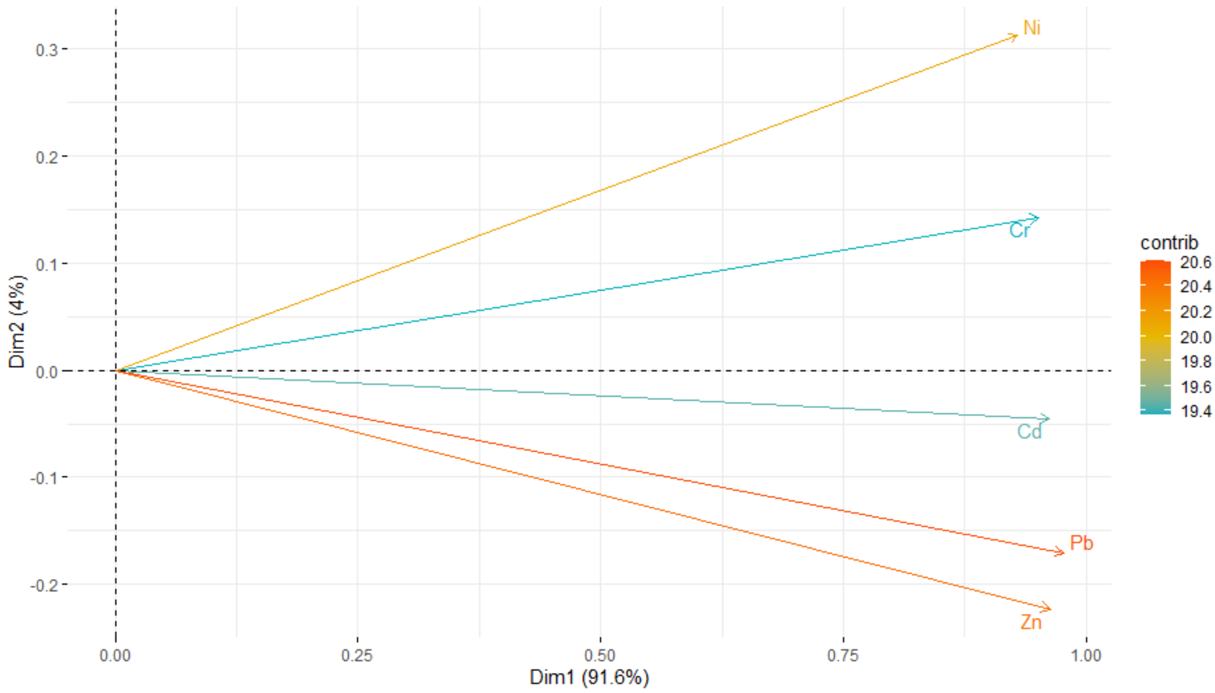

Figure 18: *PCA loading plot for heavy metals*

CONCLUSIONS

This study was devoted to the systematic assessment of heavy metals (Cd, Cr, Ni, Zn and Pb) contamination in soft sediments of the Severn Estuary and Inner Bristol Channel. The assessment considered the seasonal variations between selected heavy metals, the spatial distribution of selected heavy metals, exceedances of selected heavy metals concentration, ecological risk of selected metal contamination and the potential sources of selected heavy metals in the sediment of the Severn Estuary and Inner Bristol Channel. Seasonal variations between the concentration of heavy metals during summer and winter revealed that irrespective of seasons the concentration of heavy metals had the trend Zn > Cr > Pb > Ni > Cd. Concentrations of metals were higher in summer than winter, and there was a significant difference between metals concentration in summer and winter ($P < 0.05$). The assessment of the spatial distribution of metals using the EBK model revealed that irrespective of the seasons, concentrations of Cd, Cr, Ni, Zn and Pb in the distinct reaches of the estuary followed the trend Severn middle > Severn lower > Inner Bristol Channel > Severn upper.

Assessment of heavy metals using the USEPA SQG revealed that none of the reaches of the estuary was polluted with Cd. The Severn upper was not polluted with Pb. The Severn middle was moderately polluted by Ni but heavily polluted with Zn, Cr and Pb. Other reaches of the estuary were moderately polluted with Cr, Ni, Zn and Pb. Assessment of ecological risk of heavy metals using pollution indices such as Igeo, CF, MPI and PLI revealed that heavy metals such as Zn, Pb, Ni and Cr in the distinct reaches of the estuary and might pose an adverse effect on biota and ecosystem services of the Severn Estuary and Inner Bristol Channel. Identification of potential sources of metals using Pearson correlation, HCA and PCA revealed that Cd, Cr, Ni, Zn and Pb all originated from anthropogenic sources including coal mining, manufacturing processes involving metals, agricultural run-offs, run-offs from roads due to the wearing of brake lining and tyres, oil spills and emission from the exhaust of vehicles and sewage treatment. Based on the findings of this research, the remediation of polluted sediments, especially in the Severn middle area is necessary. Additionally, industries involving the emission of heavy metals on the Severn Estuary and Inner Bristol Channel should be strictly monitored with the sole aim of making sure they are compliant with relevant environmental policies.

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Highlights

- Sediment heavy metals concentrations in summer were significantly higher than in winter.
- Sediment heavy metals such as Zn, Cr, Ni and Pb occurs at concentrations that may pose adverse effects on biota and ecosystem services of the Severn Estuary and Inner Bristol Channel.
- The Severn middle is the most polluted reach of the Severn Estuary and Inner Bristol Channel.

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- Sources of heavy metals in the Severn Estuary and Inner Bristol Channel are mostly anthropogenic.

Conflict of interest

The authors declare that there is no conflict of interest.

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