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Assessment of Metal Concentrations and Variability Using Censored Data Analysis in Hu'u Rivers, Sumbawa, Indonesia

Karizma Fahlevy[1]†, Agustinus Sembiring[2], Rias Kaessari Magenta[2], Adi Suroso[2] and Charles P. H. Simanjuntak[3]
[1]Nabat, Abu Dhabi Emirate, United Arab Emirates
[2]PT Hirax Environmental Indonesia, Jakarta 12950, Indonesia
[3]Department of Aquatic Resources Management, Faculty of Fisheries and Marine Sciences, IPB University, Bogor 16880, Indonesia
†Corresponding author: Karizma Fahlevy; karizma.fahlevy@nabat.ai

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ABSTRACT

Water quality monitoring is essential for managing watersheds; however, values below detection limits are often mishandled, which can hinder interpretation and lead to these measurements being ignored or discarded. This study aimed to analyze metal concentrations and their variability in freshwater rivers, focusing on datasets with a high proportion of censored values, where many measurements fell below the detection limits. This study was conducted in Hu'u, Sumbawa, Indonesia, from 2019 to 2023. Thirteen dissolved metals were analyzed: As, Ba, B, Cd, Cr(VI), Co, Cu, Fe, Pb, Mn, Hg, Se, and Zn. Significant differences between watersheds were observed for six metals, including As, Ba, B, Fe, Mn, and Zn ($p < 0.05$), whereas no significant seasonal effects were detected for any parameter. The findings indicate that the watersheds are likely impacted by nearby agricultural and domestic activities, as evidenced by the higher metal concentrations near settlements and cornfields. Conversely, more remote watersheds with limited agricultural exposure exhibited lower pollutant concentrations. This study practically enhances the representation of censored data concentrations with multiple detection limits, not only by visualizing censored values but also by illustrating data variability.

1. INTRODUCTION

Heavy metals are naturally present in aquatic environments and are mainly introduced into rivers through natural rock weathering, soil erosion, or the dissolution of water-soluble salts (Garbarino et al. 1995; Kennish 1998; Khethi et al. 2013; Jeong et al. 2023). Metals belong to a broader category of substances, some of which are essential for human health (Mertz 1993). According to Bosch et al. (2016), metals can be categorized as essential, non-essential, or toxic. Essential elements include iron (Fe), copper (Cu), zinc (Zn), and selenium (Se), which have specific functions in regulating body metabolism but can be harmful when exceeding certain levels (Bosch et al. 2016, Pasco et al. 2024). Conversely, non-essential elements are typically recognized to have no specific function in living organisms. Toxic elements, such as arsenic (As), chromium (Cr), nickel (Ni), cadmium (Cd), mercury (Hg), and lead (Pb), are detrimental when present in high concentrations (Bosch et al. 2016). Despite this, naturally discharged metals into river systems without anthropogenic intervention are generally considered harmless (Kennish 1998).

Indonesia has approximately 17,000 watersheds that are vital for managing the country's water resources (Ditjen PDASRH 2021). In addition to their role in natural cycles, watersheds provide water for agriculture, industry, and households and support traditional livelihoods (Duan et al. 2016, Bons et al. 2016, Rozana et al. 2020, Narendra et al. 2021). However, the quality of water in these watersheds is increasingly at risk due to the rising concentration of heavy metals, which can be exacerbated by untreated industrial and agricultural runoff. Concerns regarding heavy metal contamination in river waters are mounting due to their toxic properties, persistent effects, and potential for accumulation in aquatic organisms (Saturday et al. 2023, Supangat et al. 2023; Ahamad et al. 2024). This poses a significant risk to aquatic ecosystems, leading to bioaccumulation throughout the food chain. Such bioaccumulation presents a potential threat to human health, making it crucial to accurately assess the distribution of heavy metal concentrations in river ecosystems (Srivastava et al. 2020; Mofijur et al. 2021; Zhou et al. 2022).

Over the last decade, water quality monitoring has become key to evaluating watersheds and prioritizing management strategies (Supangat et al. 2023). It provides timely information necessary to detect and address threats to water quality. A common issue encountered in water quality monitoring is the presence of data below the detection limit (DL) of the analysis, often arising from variations in laboratory methods or instrumentation and interferences in the matrix of the water samples (Shoari & Dube 2018; Hites 2019). This issue creates difficulties in managing the data, commonly known as "censored data" (Hewett & Ganser 2007). Such values are difficult to interpret, which can result in bias and misinterpretation of the results (Verbovšek, 2011). Methods to compute summary statistics for censored data include the Kaplan-Meier survival method, maximum likelihood estimation, and regression on order statistics (Helsel 2012). However, these methods are seldom employed because of the need for specific statistical techniques and software (Verbovšek 2011). This added complexity has led to the adoption of more traditional approaches for handling these data, such as substituting censored values with one-half of the detection limit derived from the analytical method. This simplistic approach is not recommended for environmental data, as it may result in both overestimation and underestimation. Further criticized this traditional method, considering it a form of data fabrication (Helsel 2006). In Indonesia, many water quality studies report various parameters, but censored data are often inconsistently managed and simply reported as "<DL" (Suyatna et al. 2017, Novita et al. 2020, Surbakti et al. 2021, Prayoga et al. 2022) due to the end users' lack of experience with data handling and interpretation of analytical results (Sulistiyowati & Uyun 2021). While such results indicate that the parameter falls below the applicable guidelines or regulations, assessing variability over space and time remains challenging, limiting insights into the original data. Addressing this issue is critical for developing effective and sustainable water management strategies.

Throughout human history, freshwater has emerged as one of the most vital resources worldwide. Simultaneously, monitoring heavy metal concentrations in river ecosystems is crucial for global environmental assessments (Khan et al. 2022, Manfreda et al. 2024). While numerous studies have investigated the spatial and temporal variations in heavy metal concentrations in freshwater rivers, detailed analyses of censored data have not been extensively addressed (Li et al. 2015; Anyanwu et al. 2024; William et al. 2024). For example, McGrory et al. (2020) analyzed heavy metal concentrations and biological and physical data in groundwater using multiple detection limits and clustering analysis in Ireland. In New Zealand, Gadd et al. (2024) examined trends in metal concentrations using censored data, whereas Whitehead (2019) focused on biological and nutrient parameters. In France, Laroche et al. (2022) conducted a study on pesticide analyses. However, comparable studies analyzing heavy metals in river ecosystems are scarce. To our knowledge, no studies in this region have addressed censored values. Furthermore, continuous, focused, and long-term monitoring of metals in freshwater, particularly when using data with over 80% censored values and multiple detection limits across different watersheds and seasons, remains limited on a global scale. To fill these gaps, this study utilized both censored (up to 80%) and uncensored data with multiple detection limits (DLs) collected over five years of monitoring. By focusing on the specific challenges posed by high levels of censored data, this study offers insights into future studies and policy frameworks in Indonesia and beyond, into the underrepresented issue of censored data interpretation in environmental monitoring.

A case study was conducted in the freshwater rivers of the Hu'u District in Dompu Regency, West Nusa Tenggara Province (NTB), Indonesia. The Hu'u District plays a significant role in Indonesia's agricultural sector, with rice and corn as its primary commodities (Paryono et al. 2023). Residents often utilize river water for irrigating their crops, and the use of chemical pesticides and fertilizers poses a potential risk of mobilizing heavy metal concentrations into rivers (Arifin et al. 2019). To develop effective and sustainable water management strategies, it is important to assess the variability in heavy metal concentrations (Kumar et al. 2024). Currently, no studies have examined heavy metal variability across watersheds and seasons using censored data analysis in the country. The novelty of this study is highlighted through the following specific objectives:

1. Determining heavy metal concentrations in freshwater rivers in Hu'u,

2. Comparing the heavy metal concentrations with the permissible limits.
3. Assessing the variability of heavy metal concentrations across watersheds and seasons using a censored data analysis.

Univariate and multivariate statistical analyses were used to examine the patterns across sampling locations by considering all metal parameters. The focus parameters include one metalloid, arsenic (As), and twelve heavy metals: barium (Ba), boron (B), cadmium (Cd), chromium hexavalent (Cr(VI)), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), mercury (Hg), selenium (Se), and zinc (Zn), as outlined in the Government of Indonesia's (GoI) regulations on environmental protection and river management. The aspects of this study are particularly important, as there are currently few studies demonstrating how both censored and uncensored data can be integrated to interpret variability in heavy metal concentrations.

2. MATERIALS AND METHODS

2.1. Sampling Location

The study was conducted in the Hu'u District, West Sumbawa, West Nusa Tenggara (NTB), Indonesia. A total

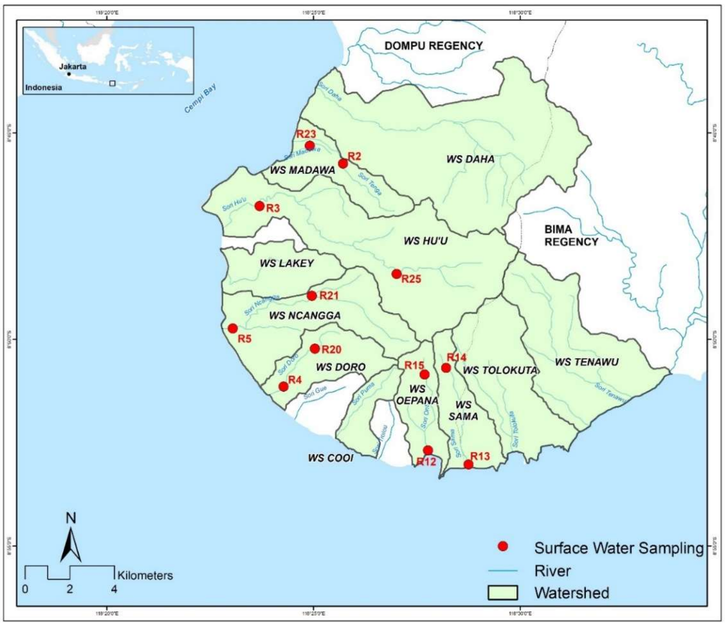

Fig. 1: Map of surface water sampling locations in Hu'u District, Sumbawa, Indonesia.

Table 1: Surface water sampling locations in Hu'u, Sumbawa, Indonesia.

No.	Watershed	River	Landuse	ID	Latitude	Longitude
1	Madawa	Tenga	Cornfield	R2	118° 25' 42.767" E	8° 45' 44.334" S
2		Madawa	Cornfield, low-density settlement	R23	118° 24' 54.540" E	8° 45' 18.122" S
3	Hu'u	Hu'u	Cornfield, low-density settlement	R3	118° 23' 41.793" E	8° 46' 45.954" S
4			Wildwood	R25	118° 27' 00.587" E	8° 48' 24.627" S
5	Ncangga	Ncangga	Corn-Paddy field, low-density settlement	R5	118° 23' 02.891" E	8° 49' 43.839" S
6			Wildwood	R21	118° 24' 57.724" E	8° 48' 56.412" S
7	Doro	Doro	Cornfield	R4	118° 24' 16.556" E	8° 51' 08.302" S
8			Wildwood	R20	118° 25' 01.945" E	8° 50' 13.221" S
9	Oepana	Onto	Wildwood	R12	118° 27' 46.066" E	8° 52' 40.827" S
10			Wildwood	R15	118° 27' 41.375" E	8° 50' 50.753" S
11	Sama	Sama	Wildwood	R13	118° 28' 45.262" E	8° 53' 01.492" S
12			Wildwood	R14	118° 28' 12.528" E	8° 50' 41.158" S

of 12 locations in freshwater rivers across six watersheds (Hu'u, Doro, Ncangga, Onto, Sama, and Madawa) were deliberately selected based on accessibility, proximity to infrastructure, land use, and their positions along the river, including both upstream and downstream areas (Fig. 1, Table 1). Land adjacent to the rivers is primarily used for irrigated and rain-fed agriculture, such as corn and paddy fields, as well as residential areas comprising small village settlements and forested regions (wildwood, Table 1). Most residents of Hu'u are farmers, and their agricultural activities, particularly irrigation, may impact water quality.

2.2. Sample Collection

Samples were collected twice annually to represent the wet and dry seasons from 2019 to 2023. Wet season sampling was conducted from January to March or from November to December, and dry season sampling occurred from May to September. Water samples were obtained by immersing 1 L high-density polyethylene (HDPE) bottles just below the water surface. The samples were subsequently passed through a 0.45 μm Millipore filter and poured into 150 mL labeled high-density polyethylene (HDPE) bottles. The filtered samples were preserved using acid, stored in a cooler at 4°C, and transported to accredited laboratories in Jakarta for further analysis. Thirteen dissolved metals were measured: arsenic (As), barium (Ba), boron (B), cadmium (Cd), chromium (VI) (Cr(VI)), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), mercury (Hg), selenium (Se), and zinc (Zn). The results were compared with the Indonesian and FAO water quality standards. The Indonesian standard is stipulated in the Government of Indonesia Regulation No. 22/2021, Appendix VI concerning Water Quality Management and Control of Water Pollution: Class II and Class III (Government of Indonesia 2021). Class II water is intended for use in recreational facilities, freshwater fish farming, livestock, irrigation, or other activities requiring comparable water quality. Class III water is suitable for freshwater fish farming, livestock, irrigation, and other uses with similar water quality requirements. The FAO standard used in this study was the recommended concentration of trace elements in irrigation water (Ayers & Westcot 1985). Table 2 summarizes the parameters, including their abbreviations, detection limits, and analytical methods.

2.3. Data Handling

At certain locations, owing to the episodic nature of the rivers, sampling could not be conducted during the dry season in some years. Consequently, some observations were missing when the rivers were dry; therefore, these points were omitted from the data table, leaving datasets of 107 and 108 observations. In addition, many samples contained metal concentrations below the detection limit (DLs). Non-detectable values, often referred to as censored data, occur when the concentration in a water body falls below the detection limits, either due to methodological or instrumental limitations (Brown 2008). The presence of censored data renders the use of defined distributions for continuous data unsuitable (Ogden 2010, Helsel 2012). To address this issue, the empirical cumulative distribution function (ECDF) was calculated for each metal parameter using the nonparametric Kaplan-Meier method (She 1997, Taylor et al. 2002, Smith 2011). This method falls under the likelihood-based approach for estimating the overall survival function when the data are incomplete or censored (Lira et al. 2020; Sun & Wang 2024). The Kaplan-Meier method was selected instead of regression on order statistics (ROS) and maximum likelihood estimation

Table 2: Parameters, methods, detection limits and permissible standards. Detection limits show the range for the different analytical laboratories.

Parameters	Method	Units	Detection Limit [DL]	GoI (2021)	FAO
Arsenic, As	APHA 3125 B	$mg.L^{-1}$	0.0005 - 0.002	0.05	0.1
Barium, Ba	APHA 3125 B	$mg.L^{-1}$	0.01	NR	NA
Boron, B	APHA 3125 B	$mg.L^{-1}$	0.01 - 0.02	1	NA
Cadmium, Cd	APHA 3125 B	$mg.L^{-1}$	0.0001 - 0.003	0.01	0.01
Chromium Hexavalent, Cr(VI)	APHA 3500-Cr B	$mg.L^{-1}$	0.0001 - 0.01	0.05	NA
Cobalt, Co	APHA 3125 B	$mg.L^{-1}$	0.001 - 0.01	0.2	0.05
Copper, Cu	APHA 3125 B	$mg.L^{-1}$	0.001 - 0.01	0.02	0.2
Iron, Fe	APHA 3125 B	$mg.L^{-1}$	0.005 - 0.02	NR	5.0
Lead, Pb	APHA 3125 B	$mg.L^{-1}$	0.001 - 0.02	0.03	5.0
Manganese, Mn	APHA 3125 B	$mg.L^{-1}$	0.001 - 0.02	NR	0.2
Mercury, Hg	APHA 3112 B	$mg.L^{-1}$	0.00005 - 0.0008	0.002	NA
Selenium, Se	APHA 3125 B	$mg.L^{-1}$	0.00006 - 0.001	0.05	0.02
Zinc, Zn	APHA 3125 B	$mg.L^{-1}$	0.005 - 0.01	0.05	2.0

(MLE) because it does not require any assumptions about the underlying data distribution, making it more robust for datasets with multiple detection limits, relatively small sample sizes, and a significant proportion of censored values (Hwang et al. 2023).

2.4. Statistical Analysis

All statistical analyses were conducted using R software v.4.4.2 (R Core Team 2024), at a significance level of p <0.05 (95% confidence level). The data were structured according to metal parameters, watersheds, and seasons. The next step was to calculate the ECDF value using the Kaplan-Meier method from the EnvStats package (Millard 2013). We included a 95% upper confidence limit (UCL95) in the Kaplan-Meier estimates because of multiple detection limits in the datasets, obtained from a percentile bootstrap estimate using the "enparCensored" function. A bootstrap repetition of 5,000 was performed to ensure the stability of the estimates from one iteration to another (Singh et al. 2006).

Environmental data (e.g., air, water, and soil) frequently exhibit a right-skewed distribution owing to a lower bound of zero and less frequent high values (Helsel 2012). Applying logarithmic or square root transformations can help achieve a more symmetric distribution, making it more suitable for statistical methods that assume normality (Helsel 2012). The "gofTestCensored" function from the EnvStats package was used to normalize the distribution (Millard 2013). This function adapts the Shapiro-Francia test to assess the lognormality of data containing multiple censored values (Millard 2013). The analysis requires datasets containing a minimum of two non-missing, uncensored, and distinct values. Accordingly, cadmium (Cd), Cr (VI), lead (Pb), mercury (Hg), and selenium (Se) were excluded from the analysis. Moreover, because most measurements were below the detection limit, the summary statistics reflect only the possible range of metal concentrations. In some cases, the minimum value could not be reported because the lowest uncensored value was lower than one or more censored observations. Reporting a numeric minimum in such cases would be misleading because the censored data may indicate a higher or lower value than the lowest observed uncensored measurement. Similarly, a maximum value may not be reported if some censored values exceed the highest uncensored observations.

To account for instances of single (one value censored) or double censorship (both values censored), the non-parametric Kendall's tau (τ) correlation coefficient was applied using the "cenken" function in the NADA package (McGrory et al. 2020, Travis et al. 2023). This analysis specifically assessed the strength of the relationships for parameters not limited by sample size constraints (Helsel 2012). The correlation coefficient values were categorized as follows: $|\tau| = 0$ indicates no relationship, $|\tau| < 0.3$ represents a weak relationship, $0.3 \leq |\tau| \leq 0.5$ indicates a moderate relationship, and $|\tau| \geq 0.5$ signifies a strong relationship (McGrory et al. 2020). Significant relationships among the metal variables are indicated by * for $p < 0.05$ and ** for $p < 0.001$.

Considering the sample size limitations, tests for comparing specific parameters across watersheds and/or seasons were excluded (Table 3). Several metals, including arsenic (As), barium (Ba), boron (B), manganese (Mn), and zinc (Zn), did not meet the assumptions required for a parametric statistical analysis. Consequently, the nonparametric Peto and Peto tests were employed to calculate the differences in the empirical cumulative distribution function (ECDF) between groups, which is appropriate for censored datasets with two or more groups (Helsel 2012). For datasets with more than two groups, the analysis was followed by a nonparametric multiple comparison test using the Benjamini-Hochberg p-value adjustment method. This analysis was performed using the "cen1way" function in the NADA2 package in R (Julan & Helsel 2021). Other metals, including cobalt (Co), copper (Cu), and iron (Fe), which followed a log-normal distribution, were tested using the "cenanova" function, followed by Tukey's multiple comparison tests in the NADA2 package (Julan & Helsel 2021). ANOVA results for Co and Cu showed significant differences ($p < 0.05$); however, post hoc tests detected no significant differences ($p > 0.05$),

Table 3: Parameters that were skipped in the statistical test.

Metal	% Censored	Number of samples	Skipped test
Arsenic, As	63.89	108	-
Barium, Ba	0.93	108	-
Boron, B	29.63	108	-
Cadmium, Cd	96.3	108	All factor
Chromium Hexavalent, Cr(VI)	100	108	All factor
Cobalt, Co	90.74	108	Watershed
Copper, Cu	77.57	107	Watershed
Iron, Fe	19.63	107	-
Lead, Pb	100	108	All factor
Manganese, Mn	39.81	108	-
Mercury, Hg	99.07	107	All factor
Selenium, Se	98.15	108	All factor
Zinc, Zn	70.09	107	-

% censored: percentage of data with concentration below the detection limit.

likely because of the high proportion of censored data. To address this limitation, both parameters were reanalyzed using the cenpermanova function in the NADA2 package. This permutation-based approach evaluates differences in group means for censored data, providing reliable mean estimates and *p*-values, as no values are modeled below zero (Julan & Helsel 2021, Kittlaus et al. 2024). The significant parameters were visualized using boxplots generated with the "cboxplot" function from the NADA2 package (Julan & Helsel 2021), which creates plots specifically for left-censored data with one or more detection limits.

Hierarchical clustering analysis (HCA) was used to examine the similarity between variables, utilizing Ward's linkage method on the significant parameters (Zhao et al. 2011). Clustering was conducted using data from all years and seasons combined, as seasonal separation was not possible because of the large number of censored values. Hierarchical cluster analysis (HCA) followed the approach of McGrory et al. (2020), clustering both watersheds and parameters. The distances between watersheds were calculated from the absolute differences in the estimated Kaplan–Meier empirical cumulative distribution function (ECDF, $F(v_k)$) values. We used the Manhattan distance to calculate the overall distance between watersheds i and j across all p parameters as follows:

$$d_{ij} = \sum_{k=1\ldots p} d_{kij} = \sum_{k=1\ldots p} \left| F(v_{ki}) - F(v_{kj}) \right| \quad \ldots(1)$$

The distance matrix for parameters x and y was constructed using Kendall's tau correlation coefficient, which was calculated through the "cenken" function of NADA (McGrory et al. 2020, Simmonet-Laprade et al. 2019), represented as:

$$dxy = \tau xy \quad \ldots(2)$$

The clustering results are shown in a heatmap, depicting the dendrograms of both parameters and watersheds.

3. RESULTS

3.1. Concentrations of Dissolved Metals

Of the metals analyzed, chromium (Cr(VI)) and lead (Pb) were below the detection limits (<DLs) in all samples. The concentrations of cadmium (Cd), cobalt (Co), mercury (Hg), and selenium (Se) were below the detection limit in 96%, 91%, 99%, and 98% of the samples, respectively (Table 3). For other metals, the ranges and proportions of censored data were as follows: arsenic (As: <0.0005–0.004 mg.L^{-1}, 64% censored), barium (Ba: <0.01–0.34 mg.L^{-1}, 1% censored), boron (B: <0.01–0.16 mg.L^{-1}, 30% censored), copper (Cu: <0.001–0.03 mg.L^{-1}, 78% censored), and zinc (Zn: <0.005–0.15 mg.L^{-1}, 70% censored, Tables 3 and 4). The highest dissolved metal concentrations were recorded for iron (Fe: <0.005–1.13 mg.L^{-1}) and manganese (Mn: <0.001–1.08 mg.L^{-1}) (Supplementary Table A1).

At all sampling locations, the concentrations of the 13 metals were below Indonesia's Class II and III water quality standards, except for copper (Cu) at site R25 in the Hu'u watershed during the 2019 wet season (Supplementary Tables A1 and A2). Although Indonesian standards do not specify limits for manganese (Mn), the FAO guidelines for irrigation water indicate that Mn concentrations in the Doro, Hu'u, Madawa, Ncangga, and Sama watersheds exceeded the maximum level in 2019, 2020, and 2022 (Supplementary Table A2). Compared to heavy metal concentrations in other countries (Table 4), the overall results of the present study were generally lower, although the maximum concentrations of barium (Ba) and manganese (Mn) were higher. Correlation analysis showed weak positive or negative relationships among the metals ($r < 0.3$). However, significant correlations were observed for several metals (Fig. 2), notably barium (Ba), arsenic (As), boron (B), cobalt (Co), iron (Fe), manganese (Mn), and zinc (Zn).

3.2. Concentrations Variability of Dissolved Metals

The Peto–Peto test (Table 5) revealed differences in ECDF-based concentration patterns across watersheds for arsenic (As), barium (Ba), boron (B), manganese (Mn), and zinc (Zn). Concentrations were higher for As in Madawa, Ba in Madawa and Ncangga, B in Madawa, Mn in Hu'u, and Zn in Doro, Hu'u, and Oepana (Table 5, Fig. 3). ANOVA results showed higher iron (Fe) concentrations in Doro, Hu'u, Ncangga, and Sama (Table 5, Fig. 3), whereas the permutational test revealed no significant differences in cobalt (Co) and copper (Cu) across watersheds (Table 5). Seasonal comparisons across all sampling locations and years revealed no significant differences between the wet and dry seasons (Table 6).

3.3. Multivariate Analysis

Hierarchical clustering identified three separate groups for both parameters and watersheds (Fig. 4). Among the parameter clusters, the first included zinc (Zn), iron (Fe), and manganese (Mn); the second contained arsenic (As), and the third comprised barium (Ba) and boron (B) (Fig. 4). Watershed clustering grouped Ncangga, Doro, and Sama in the first cluster, Oepana and Madawa in the second, and Hu'u in the third (Fig. 4).

4. DISCUSSION

4.1. Dissolved Metal Variability

Dissolved metals are more easily absorbed by aquatic

Table 4: Reported metal concentrations ($mg.L^{-1}$) in freshwater rivers from the literature.

Country	River	Parameter [$mg.L^{-1}$]													
		As	Ba	B	Cd	Cr(VI)	Co	Cu	Fe	Pb	Mn	Hg	Se	Zn	References
Bangladesh	Barnoi [n = 5]	0.030 - 0.075			0.002 - 0.005			0.021 - 0.062		0.020 - 0.088				0.023 - 0.118	Haque et al. (2024)
China	Yangtze [n=110]	0.00042 - 0.00852			0.00001 - 0.00013			0.00011 - 0.00013	0.00011 - 0.00013	0.0008 - 0.00097				0.0024 - 0.0446	Jin et al. (2022)
India	Yamuna [n = 7]				0.0017 - 0.4330			0.0184 - 17.6424	0.8785 - 53.9401	0.0068 - 1.1121				0.0154 - 28.5209	Bhardwaj and Garg (2017)
Kazakhstan	Syr Darya [n = 42]						0.008 - 0.105	0.002 - 0.034	0.125 - 8.180	0 - 0.023	0.014 - 0.691			0.027 - 0.169	Zhang et al. (2020)
Kazakhstan	Esil River Basin [n = 78]	0.00252 - 0.0043			0.00008 - 0.000032	0.00282 - 0.0034				0.00098 - 0.00172					Bahetnur and Lee (2023)
Malaysia	Linggi [n = 15]	0.00022 - 0.00131			0.00001 - 0.00261			0.00006 - 0.00306	0.00683 - 0.17966	0.00003 - 0.00027	0.00259 - 0.04069		0 - 0.00006	0.00006 - 0.00635	Razak et al. (2021)
Mexico	Guadalajara River Basin [n = 25]	0 - 0.011	0.06 - 0.085	0.023 - 0.415	<0.002 - 0.003			0 - 0.009	0.158 - 1.174	0 - 0.023	0.002 - 0.544			0.002 - 0.128	Gonzaez-Díaz et al. (2025)
Nigeria	Ossiomo [n = 4]								0.19 - 0.57		0.01 - 0.32			0.09 - 1.29	Anani and Olomukoro (2021)
Thailand	Chao Phraya [n = 20]				0.000029 – 0.000193					0.000190 – 0.004428	0.000190 – 0.004428	0.000001 – 0.000007		0.000102 – 0.001047	Asokbunyarat and Sirivithayapakorn (2020)
Vietnam	[DOI]-Cho Dem-Ben Luc [n = 7]							0.026 - 0.057	1.01 - 2.38	0.008 - 0.08	0.14 - 0.28			0.026 - 0.048	Thieu et al. (2022)
Present study [n=12]		<0.0005 - <0.01	<0.01 - 0.34	0.16*	<0.0001**	<0.0001 - 0.01	<0.001 - 0.021	<0.001 - 0.03	<0.005 - 1.13	<0.001 - <0.02	<0.001 - 1.08	<0.00005 - 0.002	<0.00006 - 0.0011	<0.005 - 0.15	

*: The minimum value is not reported because the uncensored data is lower than the value of the censored data.
**: The maximum value is not reported because the censored data is higher than the maximum value of the uncensored data.

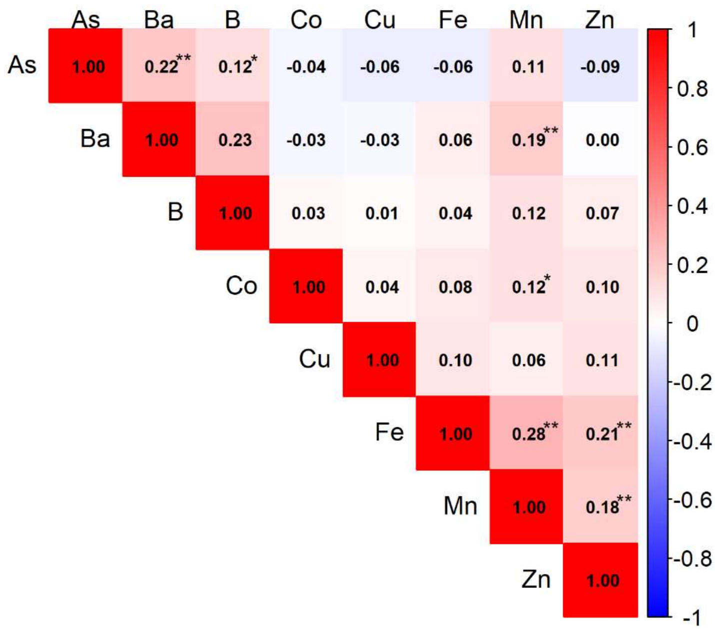

Fig. 2: The correlation matrix of Kendall's tau illustrates strong positive correlation (1.0) in red, indicates no correlation (0.0) without color, and represents strong negative correlation (-1.0) in blue. *=p-value<0.05, **= p-value<0.01.

Table 5: The concentration ranges (min-max) of metal parameters at all watersheds.

Watershed	Parameter [mg.L^{-1}]												
	As	Ba	B	Cd	Cr(VI)	Co	Cu	Fe	Pb	Mn	Hg	Se	Zn
Doro	<0.0005 - 0.0013[a]	0.05 - 0.1[a]	<0.01 - 0.03[a]	<0.0001 - <0.001	<0.002 - <0.01	<0.001 - <0.01	<0.001 - 0.02	<0.02 - 0.5[a]	<0.001 - <0.02	<0.001 - 0.049[a]	<0.00005 - <0.0008	<0.00006 - <0.001	<0.005 - 0.07[ab]
Hu'u	<0.0005 - 0.01[ab]	0.05 - 0.1[a]	<0.01 - 0.14[a]	<0.0001*	<0.002 - <0.01	<0.001 - 0.021	<0.001 - 0.03	<0.02 - 1.13[a]	<0.001 - <0.02	<0.01 - 1.08[b]	<0.00005 - <0.0008	<0.0005 - 0.0011	<0.005 - 0.15[a]
Madawa	<0.001 - 0.004[b]	<0.01 - 0.34[b]	<0.02 - <0.02[b]	<0.0001 - <0.003	<0.001 - <0.01	<0.001 - <0.01	<0.001*	<0.005 - 0.24[b]	<0.001 - <0.02	0.418**[a]	<0.00005 - <0.0008	<0.0005 - <0.001	<0.005 - <0.01[ab]
Ncangga	<0.0005 - <0.001 [ab]	0.07 - 0.17[b]	<0.01 - 0.15[a]	<0.0001*	<0.002 - <0.01	<0.001 - <0.01	<0.001*	0.383**[a]	<0.001 - <0.02	0.03**[a]	<0.00005 - <0.0008	<0.0005*	<0.005 - 0.06[b]
Oepana	<0.0005 - 0.009[a]	0.02 - 0.07[c]	<0.01 - 0.12[a]	<0.0001 - <0.001	<0.0001 - <0.01	<0.001 - <0.01	<0.001 - <0.01	<0.005 - 0.138[b]	<0.001 - <0.02	<0.001*[c]	<0.00005 - 0.002	<0.0005 - <0.001	<0.005 - 0.05[b]
Sama	<0.0005*[a]	0.03 - 0.09[c]	<0.01 - 0.09[a]	<0.0001*	<0.002 - <0.01	<0.001 - <0.01	<0.001 - 0.02	1.07**[ab]	<0.001 - <0.02	0.31**[a]	<0.00005 - <0.0008	<0.0005 - <0.001	<0.005 - <0.01[a]
GoI	0.05	NR	1	0.01	0.05	0.2	0.02	NR	0.03	NR	0.002	0.05	0.05
FAO	0.1	NR	NR	0.01	NR	0.05	0.2	5	5	0.2	NR	0.02	2

Similar letters indicate no statistical difference based on the Peto-Peto test (adjust = Benjamini-Hochberg) and ANOVA test.
*The maximum value is not reported because the censored data is higher than the maximum value of the uncensored data.
**The minimum value is not reported because the uncensored data is lower than the value of the censored data.

organisms (Zahran et al. 2015, Tolkou et al. 2023). In this study, 13 dissolved metals were analyzed in the river water of Hu'u: arsenic (As), barium (Ba), boron (B), cadmium (Cd), hexavalent chromium (Cr(VI)), cobalt (Co), copper (Cu), iron (Fe), lead (Pb), manganese (Mn), mercury (Hg), selenium (Se), and zinc (Zn). Although all metals were detected, most concentrations were below the analytical detection limits, with cadmium (Cd), chromium (VI), mercury (Hg), lead (Pb), and selenium (Se) being undetected in 96–100% of the samples (Table 3). These metals may be regarded as safe since their detection thresholds were lower than the river water quality standards set by the Government of Indonesia (2021, Class II and III) and the Food and Agriculture Organization for irrigation water. Low levels of these metals may reflect limited inputs from pollution sources (Mokarram et al. 2020) and natural deposition processes (Botutihe et al. 2021).

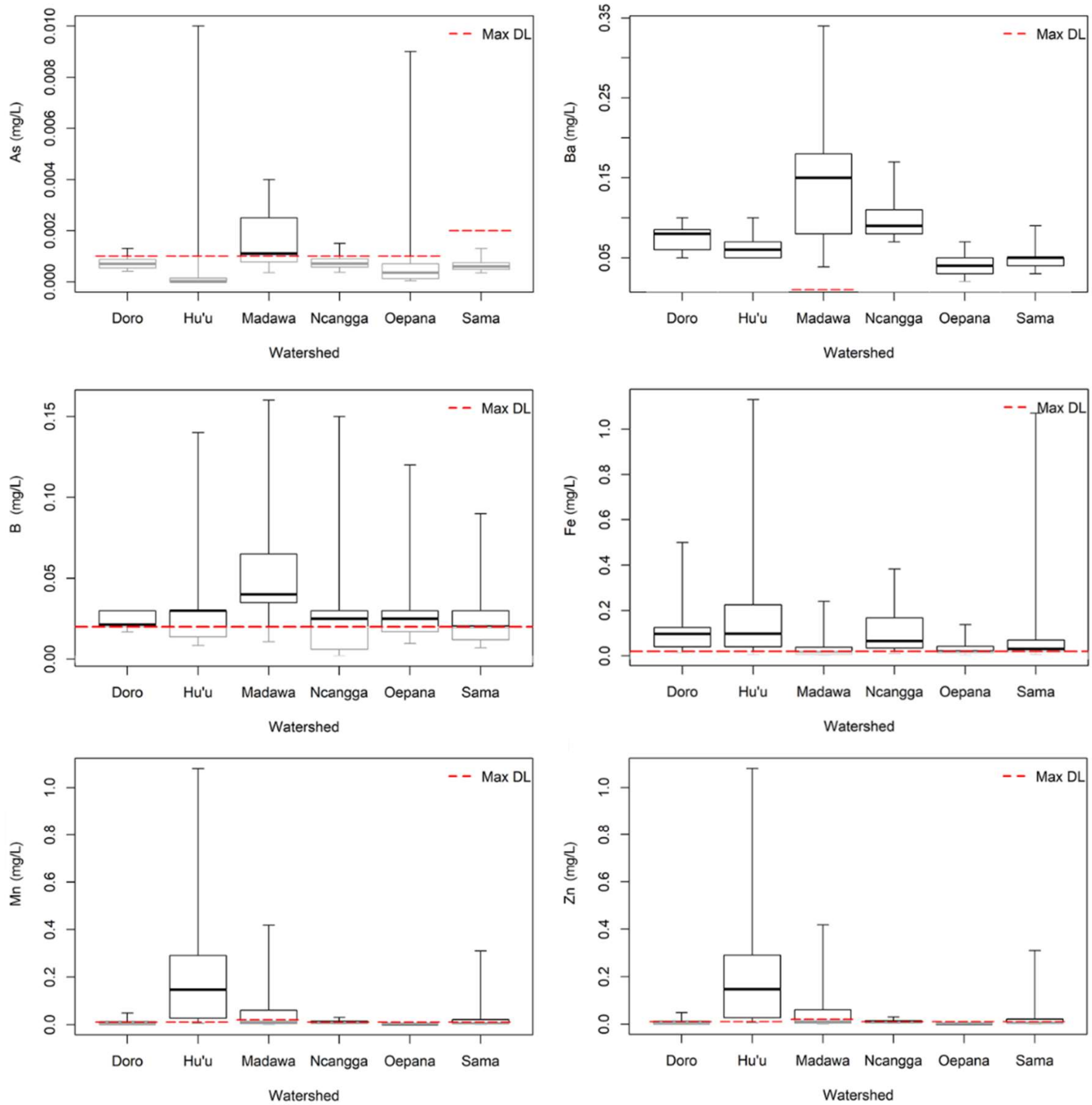

Fig. 3: Variability of metal concentrations for six parameters across the watersheds, selected based on statistical significance.

Table 6: The concentration ranges (min-max) of metal parameters at all seasons.

Season	Parameter [mg.L-1]											
	As	Ba	B	Cd	Cr(VI)	Co	Cu	Fe	Pb	Mn	Hg	Se	Zn
Dry	<0.0005 - 0.01	<0.01 - 0.21	<0.01 - 0.16	<0.0001*	<0.001 - <0.01	<0.001 - 0.021	<0.001 - 0.015	<0.005 - 0.917	<0.001 - <0.02	<0.001 - 1.08	<0.00005 - 0.002	<0.00006 - 0.0011	<0.005 - <0.01
Wet	<0.0005 - <0.001	0.02 - 0.34	0.07**	<0.0001 - <0.002	<0.0001 - <0.01	<0.001 - 0.02	<0.001 – 0.03	<0.005 - 1.13	<0.001 - <0.02	<0.001 - 0.94	<0.00005 - <0.0008	<0.0005 - <0.001	<0.005 - <0.01
GoI	0.05	NR	1	0.01	0.05	0.2	0.02	NR	0.03	NR	0.002	0.05	0.05
FAO	0.1	NR	NR	0.01	NR	0.05	0.2	5	5	0.2	NR	0.02	2

*The maximum value is not reported because the censored data is higher than the maximum value of the uncensored data.
**The minimum value is not reported because the uncensored data is lower than the value of the censored data.

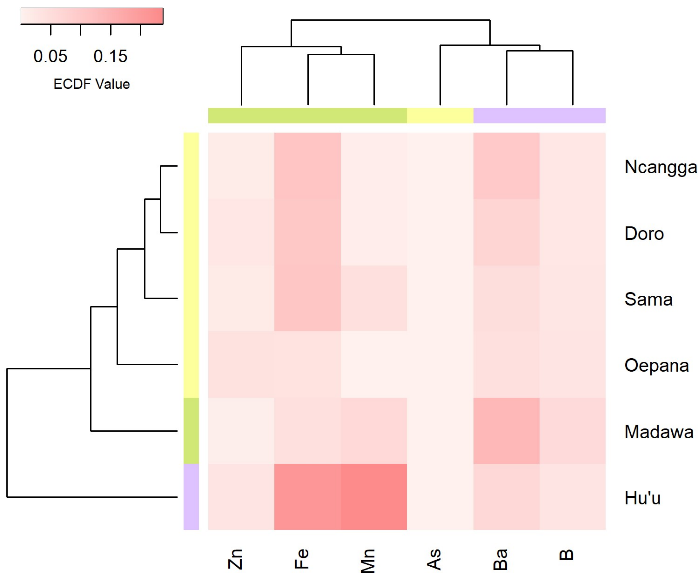

Fig. 4: Heatmap of hierarchical clustering based on watersheds (y-axis) and significant parameters (x-axis), clustered by Kendall's tau for parameters and ECDF interval distances for watersheds.

The latest assessment of the concentrations of 13 metals in the Hu'u district freshwater rivers found that they were below the Government of Indonesia (GoI) Class II and III water quality standards (WSP 2023, Supplementary Table A3), consistent with the findings of this study in 2023. The results from 2019 to 2023 showed that only copper (Cu) exceeded the GoI standards, occurring during the wet season of 2019 in Hu'u, while manganese (Mn) exceeded the FAO standard during both wet and dry seasons in 2019, 2020, and 2022 (Table 6, Supplementary Tables A1 and A2). Despite most parameters falling below the standard, continuous monitoring of heavy metals in surface water is crucial for sustainable management (Hartiningsih et al. 2024). As the human population continues to grow, water resources may be adversely affected by increasing metal concentrations if they are not properly monitored and managed (Mokarram et al. 2020).

No significant differences were observed in the dissolved metal concentrations between the wet and dry seasons (Table 6). Similar observations have been reported for some metal parameters in countries such as Nigeria and Bangladesh, where seasonal variation was also not significant (Ogungbile et al. 2020, Islam et al. 2021, Adano et al. 2023). The primary sources of metals may remain consistent throughout the year, contributing to stable concentrations regardless of seasonal changes (Adano et al. 2023). Although higher runoff during the wet season could increase the mobilization of metal-rich sediments and raise concentrations in surface waters through soil leaching (Oluyemi et al. 2008, Valencia-Avellan et al. 2017), frequent mild rainfall likely reduced concentrations through dilution (Huang et al. 2020). The predominance of such rainfall throughout the year may explain the absence of marked differences between the wet and dry seasons.

Significant variations in metal concentrations were detected for six parameters: arsenic (As), barium (Ba), boron (B), iron (Fe), manganese (Mn), and zinc (Zn) across watersheds (Table 5). The maximum concentrations of barium (Ba) and manganese (Mn) in this study exceeded values previously reported in the literature (Bhardwaj & Garg 2017, Zhang et al. 2020, Asokbunyarat & Sirivithayapakorn 2020, Razak et al. 2021, Anani & Olomukoro 2021, Jin et al. 2022, Thieu et al. 2022, Bahetnur & Lee 2023, Haque et al. 2024, González-Díaz et al. 2025). The dissolved concentrations of As, Ba, and B were particularly elevated in the Madawa watershed (Table 5). These metals may originate from shared sources, as indicated by their low but significant positive correlation (Fig. 2). Such patterns are likely linked to agricultural practices and residential waste inputs in Madawa, where large areas adjacent to the river are dominated by cornfields (Table A1). The use of As, Ba, and B in pesticides, fungicides, and fertilizers for crops has been recorded in many studies and may be a possible source of these metals in this case (Castro & Brighenti 2007, Coupe & Capel 2015, Upadhyay et al. 2018, Nejad & Etesami 2020, Sundaresan et al. 2023).

Although the use of chemical pesticides persists in Indonesia (Prajawahyudo et al. 2022, Ismindarto et al. 2024), considerable efforts have been made to reduce their application owing to environmental concerns (Maryono &

Irham 2001). Initiatives promoting the use of bio-organic pesticides have been organized by the local government in the NTB region since 2015 (BPTP NTB 2019) and could be expanded to further diminish the reliance on chemical pesticides in the Hu'u District. Although these metals were found at higher concentrations in Madawa, their levels remained within water quality standards, suggesting that their occurrence may be linked to residues from historical use in the area. Elevated Ba concentrations were also detected in Ncangga, although they were still lower than those in Madawa (Table 5 and Fig. 3). This pattern may be related to the predominance of wildwood areas in the upstream region, combined with cornfield activities in the lower parts of the watershed (Table 1).

Dissolved Fe, Mn, and Zn concentrations were higher in the Hu'u District watersheds (Table 5 and Fig. 3), likely reflecting similar sources, as indicated by their significant but weak positive correlations (Fig. 2). The Hu'u District watershed experiences substantial riverine activity from upstream to downstream and features mixed agricultural land use, including corn and paddy fields near settlements (Table 1). Watersheds with more intensive land use generally showed higher metal concentrations, as shown in Fig. 4. As previously noted, the Fe, Mn, and Zn levels are closely linked to agricultural practices and domestic waste. Although elevated Mn and Zn concentrations were also detected in Madawa, they were lower than those in the Hu'u District, likely reflecting the more limited agricultural activity, which is primarily restricted to cornfields in Madawa. Overall, the Hu'u watersheds showed the greatest influence from agriculture and settlements, whereas watersheds dominated by wildwood and natural areas, such as Ncangga, Doro, Sama, and Oepana, formed closer clusters (Fig. 4).

Geological formations and local mineralization may also contribute to the presence of soluble metals such As, Ba, B, Fe, Mn, and Zn in this area. Sumbawa Island is geologically active, experiencing tectonic plate movements, and can be classified as a seismic region (Intan 2016). Tectonic activity in the Hu'u District has formed geological structures that influence the landscape, hydrology, and mineralization (Intan, 2016). Petrographic analysis of the Hu'u District has identified several rock types, including alluvial deposits, reef limestone, volcanic breccia, tuff, and andesite (Sofyan & Fadhlan 2004, Intan 2016). These rocks are associated with various metal-bearing minerals. For instance, Fe-Mn concentrations have been linked to volcanic breccia in mineralized volcanic systems (Berdnikov et al. 2021), and higher Zn concentrations have been found in alluvial sediments of rivers (Sajn et al. 2008). However, further in-depth studies are required to fully evaluate this factor.

4.2. Strengths and Limitations

Censored data are common in environmental studies, requiring methods that properly handle observations below the detection limits (Wood et al. 2011). The Kaplan–Meier estimator is widely used for this purpose because it accommodates censored values without substitution and is available in most statistical software packages (Smith 2011, Helsel 2012). However, its reliability decreases with small samples or high censoring, and it cannot be applied to datasets composed entirely of censored values (Helsel 2012, Zhao et al. 2011). Although its asymptotic properties are well established (Breslow & Crowley 1974), the approximation performs poorly in small datasets (Borgan & Liestøl 1990). To improve inference under these conditions, alternative approaches such as data transformations (e.g., log, logit, log-minus-log, or arcsine square-root) are recommended because they can yield more robust results in small or heavily censored datasets (Nagashima et al. 2021). Several recommendations and limitations are outlined based on the findings of this study.

i) Carefully define the objectives and deliverables during the planning stage, considering the high proportion of censored data and the presence of multiple detection limits (McGrory et al. 2020). For example, implementing stricter sampling protocols and maintaining consistent analytical methods, including the use of blank and replicate samples, could help reduce variability in detection limits and improve the likelihood of measuring actual concentrations,

ii) Testing for differences between years, as well as interactions among year, location, and season, was not possible due to dataset size constraints, as no two samples collected within the same year contained uncensored values, limiting the ability to assess the effect of year on both season and location,

iii) Consistent long-term monitoring with stable sample sizes will be necessary to observe trends in metal concentrations or other environmental elements over time (Gadd et al. 2024) to better understand the complexity of temporal variability,

iv) To gain deeper insight into the sources of heavy metals and enhance interpretative capacity, future studies should incorporate additional parameters, including in situ field measurements, nutrient sampling and analysis, and biological assessments.

5. CONCLUSIONS

Analysis of dissolved metals in freshwater rivers showed that a large proportion of the data fell below detection limits. The key findings of this study are as follows:

i) Of the 13 dissolved metals examined, cadmium (Cd), chromium (Cr (VI)), mercury (Hg), lead (Pb), and selenium (Se) were rarely detected, with over 90% of values censored,

ii) From 2019 to 2023, dissolved metal concentrations generally remained below the water quality standards set by the Government of Indonesia (GoI) for Classes II and III, except for copper (Cu), which exceeded the limit in the Hu'u River during the wet season of 2019. Manganese (Mn) concentrations also surpassed the FAO standard in five watersheds during 2019, 2020, and 2022,

iii) Censored data analysis revealed no significant seasonal effects on dissolved metal concentrations, as wet- and dry-season samples showed no statistical differences,

iv) Watershed characteristics and surrounding land use significantly influenced six parameters: arsenic (As), barium (Ba), boron (B), iron (Fe), manganese (Mn), and zinc (Zn). Higher concentrations of these metals are likely associated with agricultural practices and domestic discharges.

In general, using censored data analysis based on Kaplan-Meier estimates yields more valuable insights for presenting results on heavy metal concentrations in water. Although this approach is not yet fully optimized, especially when censored data exceeds 80%, it remains a valuable tool for supporting water quality assessment and management.

6. DATA AVAILABILITY

The supplementary data is available at https://doi.org/10.6084/m9.figshare.32915180

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