



Spatiotemporal variations, health risk assessment, and sources of potentially toxic elements in potamic water of the Anday Stream Basin (Türkiye), Black Sea Region

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Abstract Monitoring and protecting freshwater habitats are paramount for a sustainable water management perspective. This study investigated potentially toxic elements (PTEs) in the potamic water of the Anday Stream Basin (Türkiye), Black Sea Region, for a hydrological year (from May 2020 to April 2021). Among PTEs, the highest average values were recorded for sodium (Na) at 41.3 mg/L and the lowest for mercury (Hg) at 0.009 µg/L and noted under quality guidelines. The stream was found to be at the level of “Low Heavy Metal Pollution” and “Low Contamination” based on the ecotoxicological risk indices. The highest calculated hazard quotient (HQ) value of 1.21E-02 for Cd was noted in the children via the

dermal pathway and the lowest of 6.91E-06 for Fe in adults via the ingestion pathway. Results revealed a higher hazard index (HI) value of 1.50E-02 for Cd to children and the lowest of 1.98E-05 for Fe to adults. As a result of applying agricultural risk indices, the stream showed sodium adsorption ratio values less than 6 and was found to be “Excellent” for agriculture. However, the sodium percentage values were less than 20 and found “Permissible” and the magnesium hazard > 50 and noted as “Unsuitable” for agriculture. Statistical analysis revealed that natural factors mainly attributed to PTE contamination of the Anday Stream Basin.

Keywords Health index · Ecotoxicological risk assessment · Sodium absorption ratio · Statistical analyses

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Introduction

Potentially toxic elements (PTEs) are regularly discharged into the environment and considered hazardous for aquatic habitats due to their toxicity, persistence, and bioaccumulation (Carvalho, 2022; Jafarzadeh et al., 2022; Kumar et al., 2023). Few PTEs, such as zinc (Zn), iron (Fe), and copper (Cu), are essential for all living beings at low levels (Sepúlveda et al., 2023). Lowering essential elements than the required amounts may cause deficiency effects such as hair loss, poor healing of wounds, and anemia. However, essential elements may cause significant toxic effects at higher

intakes (Haq et al., 2023; Qasemi et al., 2023; Sajjadi et al., 2022). The concentrations of PTEs above the recommended limits may cause a significant public health threat (Ahmed et al., 2022; Jiang et al., 2022; Muhammad, 2023). Long-lasting exposure to PTEs may have a severe toxic effect when using such contaminated water for cooking and as potable water. This exposure has been associated with different diseases, including malformation, kidney loss, cancer, and even death due to elevated levels of PTEs (Aziz et al., 2023; Hasan et al., 2021). It may pose a substantial health threat to residents sourcing from both natural (Liu et al., 2023; Zhang et al., 2021; Zhu et al., 2023) and anthropic factors (Abualhajja, 2023). Therefore, PTE monitoring in the aquatic environment has an exceptional place for the sustainability of these significant ecosystems (Tokatli & Ustaoglu, 2020; Tokatli et al., 2023b).

Recently, several evaluation techniques have been widely used to assess the multiple and synergistic effects of water quality (Jafarzade et al., 2023; Kiani et al., 2021; Moghaddam et al., 2022; Shams et al., 2022) for better resource management (Shang et al., 2023; Wen et al., 2024; Yin, 2023b; Yin, 2023c). Spatiotemporal investigations of PTEs using ecotoxicological indices may supply valuable data to determine the contamination factors and probable health risks of such toxicants (Lin et al., 2022; Qin et al., 2023; Ustaoglu et al., 2020; Yin et al., 2023a). Heavy Metal Evaluation Index (HEI) and Heavy Metal Pollution Index (HPI) are two of the widely used inorganic pollution risk assessment methods developed from the aspect of water quality for consumption (Chettri et al., 2022; Muhammad et al., 2022). The spatiotemporal assessment of PTEs can also provide helpful information on freshwater habitats where resource managers and regulatory bodies may decide to utilize the alternatives (Egbueri & Agbasi, 2022; Reza & Singh, 2010). Therefore, monitoring of the freshwater habitat is essential tool used to assess the impacts of these contaminants on aquatic system (Abd El Tawaba et al., 2022; Hakuzimana & Masasi, 2020; Nasreen & Ashraf, 2020; Olusola, 2020).

Multivariate statistical techniques are very beneficial and robust methods in evaluating the multiple impacts of PTEs on water ecosystems. Factor analysis (FA), cluster analysis (CA), and Pearson Correlation Index (PCI) are among the most popular multivariate statistical techniques (Bodrud-Doza et al., 2016; Hasan et al., 2021). Among them, PCI is employed to determine the associations, FA can aid in elucidating

the variance of a vast data set with a small data set of independent parameters, and CA shows the relationships among contaminants (Tokatli et al., 2023b; Varol et al., 2022a; Venkatramanan et al., 2014).

The contaminations of PTEs in freshwater (Tokatli et al., 2021) and sediments (Şimşek et al., 2022; Varol et al., 2022b) along with the Black Sea Region have been reported in few sporadic studies. However, these studies primarily focused on the PTE contamination of the Black Sea Region sediments. PTE contaminations of freshwater in the region have been rarely studied. Other limitations of previous studies were one-time sampling techniques and single evaluation indices. A study reported that seasons play a significant role in determining the freshwater contamination levels in the Black Sea Region (Tokatli et al., 2023a). However, no earlier study has assessed the spatiotemporal variation of the PTE accumulations in this rare potamic water of unpolluted habitat (Anday Stream Basin), Türkiye Black Sea Region. This study hypothesizes that Anday Stream has very high unpolluted water quality due to the protection of rare aquatic habitats in the study region. The main targets of this study were (1) to determine the spatiotemporal variations of PTEs in water of the Anday Stream Basin (Türkiye) from May 2020 to April 2021, (2) to define the quality of Anday Stream Basin in terms of irrigation water supply using agroecological risk indices, (3) to evaluate the quality of Anday Stream Basin water in terms of PTE contamination levels using ecotoxicological indices, (4) to identify the potentially non-carcinogenic risks of PTEs in potamic water of Anday Stream Basin using health risk assessment indices in Black Sea Region, Türkiye, and (5) to assess the water quality of Anday Stream Basin using a multi-statistical perspective technique for sources apportionment.

Material and method

Description of the basin and collection of freshwater samples

Anday Stream Basin is a rare potamic habitat far away from any point or non-point contamination sources and anthropogenic stress. Anday Stream Basin has limited settlement in its watershed and even agricultural activities, one of the most critical pollution pressures on many fluvial ecosystems, and does not pose a significant problem for this potamic habitat due to quite dense forest

vegetation. The stream flows south-north from the borders of Küre District from Kızılkaya Hill, at an altitude of approximately 1500 m in the west part of the Black Sea Region, to the Black Sea via the Küre Stream within the borders of İnebolu District. Anday Stream Basin dominates a typical Black Sea climate. The most fundamental characteristic of this climate is abundant rainfall in all seasons, which causes the basin to be covered with rich natural vegetation and forests (Turkes, 2020).

In this study, potamic sampling was performed monthly from May 2020 to April 2021 (once each month) from ten selected stations on the Anday Stream Basin of Türkiye and processed according to the flow diagram (Fig. 1). Sampling stations were selected on the Anday Stream Basin (S1-S3, S6-S10) and the most essential branch of the creek (S4-S5) (Fig. 2). In this way a total of 120 water samples were collected from the ten known stations of Fig. 1. Before sampling, the sampler was washed three times with the respective water of the sampling point. Water samples were taken 10 cm below the surface using a deep-water sampler in disposable, sterile, 1000-ml pre-cleaned polyethylene bottles. Water samples were then placed in a 4 °C portable vehicle cooler and transferred immediately.

Water analysis

Potamic water samples were filtered and measured for chloride (Cl), sodium (Na), potassium (K), calcium (Ca), and magnesium (Mg) using an analytical ultraviolet and

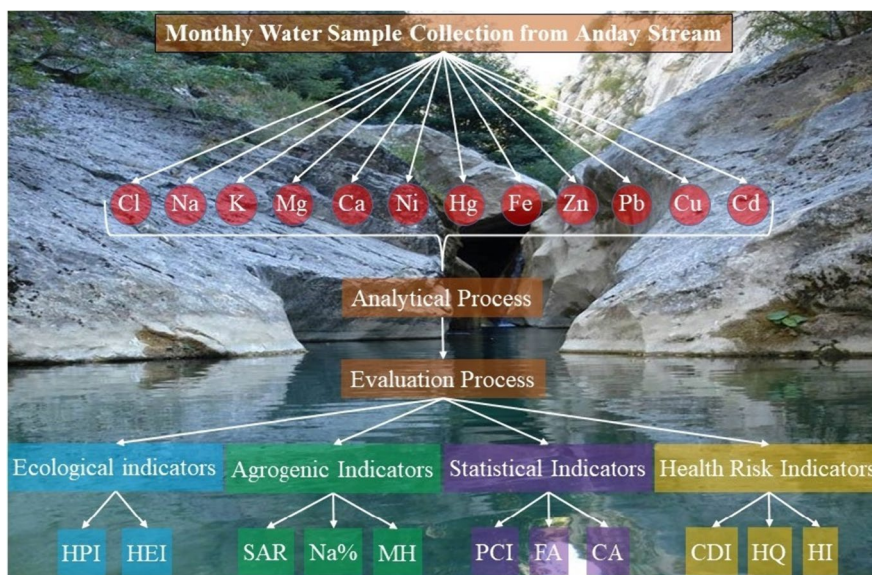
visible light (UV–VIS) absorption spectrophotometer (UV Pharmaspec 1700 – Shimadzu). Iron (Fe), lead (Pb), mercury (Hg), copper (Cu), cadmium (Cd), nickel (Ni), and zinc (Zn) levels were determined using an ICP-OES (Spectro, SpectroBlue). For the analysis of PTEs, the pH of 1 L potamic water was adjusted to 2 by adding nitric acid (HNO₃) into each sample. Then, the potamic water samples were filtered (0.45-µm pore-spaced filter) on the same sampling day. Finally, the contamination of PTEs was measured in the filtered samples. The whole data of investigated PTEs were determined as the averages of triplicate readings. All the analyses were conducted in the Central Research Laboratory of Kastamonu University (accredited by the Accreditation Agency of Türkiye). The accreditation content includes all the analyses performed in the current study (APHA, 1998, 2017).

The standard stock solution (SSS) (Merck, Germany) supplied for ICP-OES was used to prepare the calibration standards. Each instrument was daily calibrated with standards. The water sample for each parameter was measured three times, and the mean values were used for result interpretation. The recovery rates of the determined PTEs by ICP-OES for the SSS analysis are in the supplementary information (Table S1).

Ecotoxicological risk indices

The HEI and HPI, which are among the best inorganic pollution assessment risk indices, describe the overall trend evaluation of PTE contamination in water

Fig. 1 Stages of the current research and an image from Anday Stream Basin



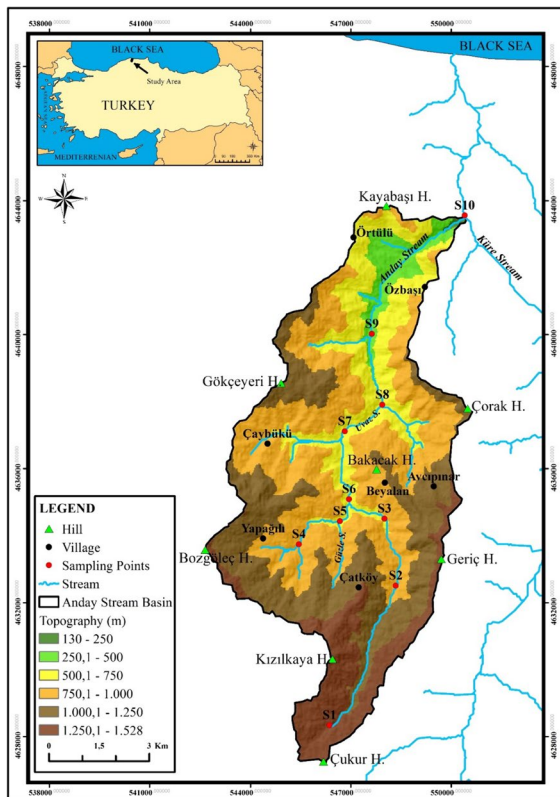


Fig. 2 Sampling points of the Anday Stream Basin

(Bodrud-Doza et al., 2016; Chorol & Gupta, 2023; Reza & Singh, 2010). The methods of calculating applied ecotoxicological risk indices are given in supplementary information (Table S2).

Agrogenic risk indices

Magnesium harness (MH), sodium percept (Na%), and sodium adsorption ratio (SAR), which are among the best agrogenic risk assessment tools, were used to gauge the water quality of Anday Stream Basin for irrigation purposes (Haq & Muhammad, 2023; Muhammad, 2023). The methods of calculating applied agrogenic indices are given in supplementary information (Table S3). In calculating irrigation water indices, mg/L values have been converted to meq/L.

Probabilistic assessment of health risks

The PTEs are taken from water to the human body through skin contact or ingestion. Non-carcinogenic health risks of PTEs from skin contact and oral

intake may be calculated as an approximation by some experimental models. In this study, a widely used non-carcinogenic health risk assessment technique suggested by USEPA (2004) was preferred and applied to the data. The applied indices including CDI obtained from dermal absorption (CDI_{dermal}) and digestion (CDI_{ingestion}), HQ obtained from digestion (HQ_{ingestion}) and dermal absorption (HQ_{dermal}), and HI are shown in the supplementary information (Table S4).

Statistical analysis

The PCI was implemented to data using the “Origin-Pro Lab 2021” package statistical software, the CA was implemented to data by using the “PAST” package statistical software, and the FA was implemented to data by the “SPSS 23” package statistical software.

Results

Water characteristics

Spatiotemporal distributions of investigated PTEs contamination in the water of Anday Stream Basin were summarized as matrix plot diagrams (Fig. 3). The values of Cl ranged 4.30 to 10.7 mg/L, Na 24.9 to 75.5 mg/L, K 2.82 to 9.77 mg/L, Mg 16.7 to 20.6 mg/L, Ca 16.6 to 21.0 mg/L, Fe 0.005 to 0.027 mg/L, Pb 0.50 to 2.90 µg/L, Cu 5.00 to 38.0 µg/L, Cd 0.40 to 2.10 µg/L, Hg 0.004 to 0.024 µg/L, Ni 4.00 to 16.0 µg/L, and Zn 5.00 to 28.0 µg/L, respectively (Fig. 3). Among water characteristics, the highest average value of 41.3 mg/L was noted for Na in the Anday Stream Basin, while the lowest of 0.009 µg/L for Hg.

Ecotoxicological risk indices

The data of spatiotemporal HPI and HEI scores are shown as spider diagrams (Fig. 4). The HPI and HEI values were recorded as below the limit values (<100 for HPI; <10 for HEI), and the water quality characteristics of Anday Stream Basin in terms of PTEs contamination were found to be as “Low Heavy Metal Pollution” for applied HPI and “Low Contamination” for applied HEI.

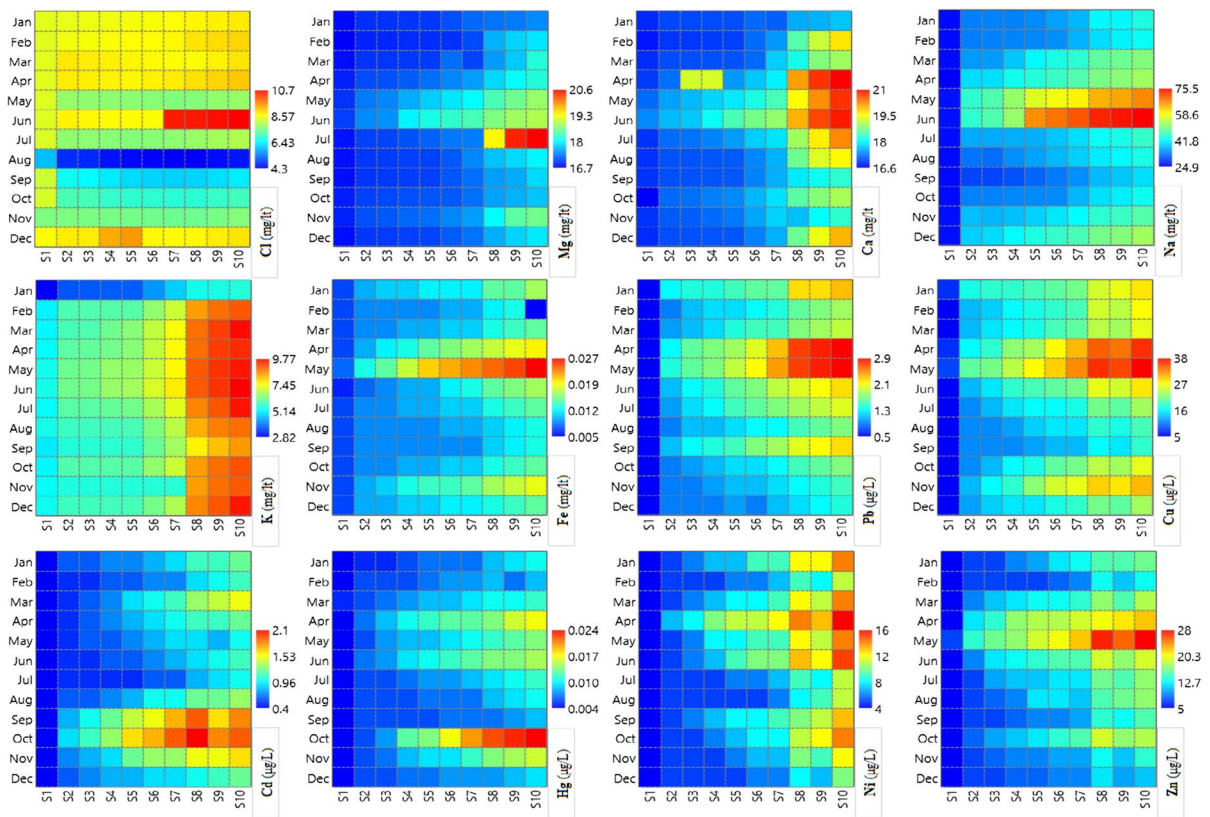
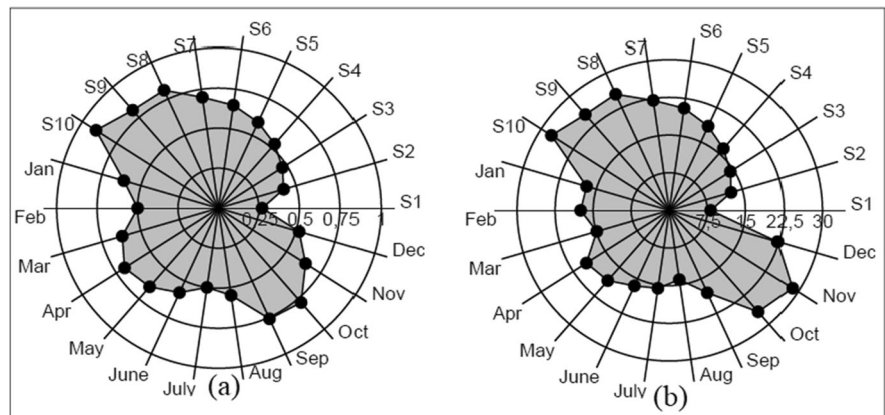


Fig. 3 Potentially toxic elements in the water of Anday Stream Basin

Fig. 4 Spatial–temporal variations of HEI (a) and HPI (b) scores of Anday Stream Basin



Agrogenic risk indices

The results of spatiotemporal MH, Na%, and SAR scores are given as spider diagrams (Fig. 5). As a result of agricultural water quality evaluation indices, the Anday Stream Basin was found to be “Unsuitable” in terms of MH (> 50), “Permissible” in terms of Na% (< 20), and “Excellent” in terms of SAR (< 6).

Probabilistic assessment of health risks

The PTEs from water may be taken to the body through skin contact and ingestion (Chen et al., 2023; Omeka & Egbueri, 2023; Turdiyeva & Lee, 2023). In the current investigation, the non-carcinogenic health risk effects of Fe, Zn, Ni, Cd, Cu, Hg, and Pb from oral intake and skin contact were calculated, and the

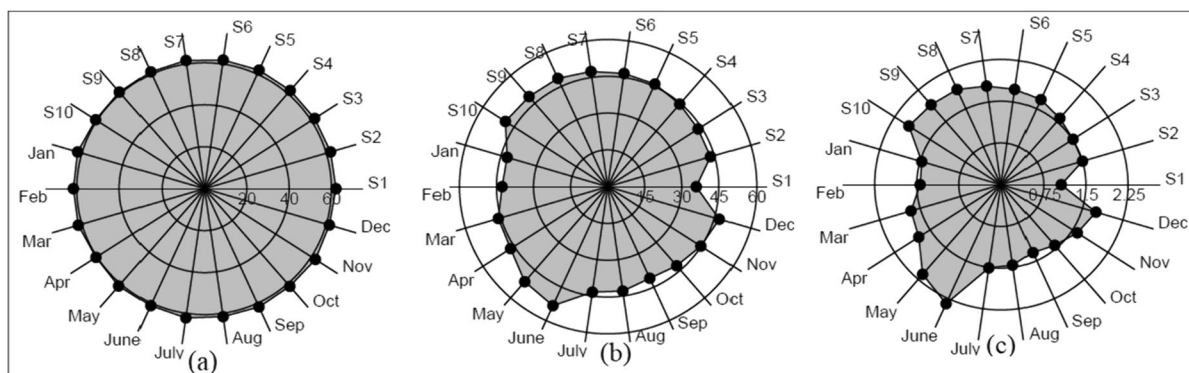


Fig. 5 Spatial–temporal variations of MH (a), %Na (b), SAR (c) scores of Anday Stream Basin

results are shown in Table 1. Among PTEs, the highest HI values were noted for the Cd, while the lowest for Fe. None of the PTE HI values exceeded the limit value of 1 for HQ and HI.

Statistical evaluation

The PCI was used to identify the significant relations among PTEs in the water of the Anday Stream Basin ($n=120$). The recorded relations among PTEs with the correlation coefficients were shown (Fig. 6). According to the results of PCI, all the investigated PTEs showed significant positive correlations (at the $p<0.05$ and $p<0.01$ significance levels).

The FA was used to identify the influential variables—factors on the water quality of Anday Stream Basin ($n=120$). The Kaiser–Meyer–Olkin value was relatively high in the current application and recorded as 0.88, which means that the sampling adequacy was sufficient. As a result of FA, two factors (eigenvalues >1) explained 80.36% of the total variance. The

scree plot, rotated component plot, and rotated component matrix were shown (Fig. 7). Component 1 was named “Natural Factor (F1)” and clarified 55.85% of the total variance. This factor was interrelated to the parameters of Na, Zn, Fe, Ca, Pb, Cu, Mg, Ni, and K. Ni and K were moderate (0.50–0.75), and Na, Zn, Fe, Ca, Pb, Cu, and Mg were strongly (>0.75) positively associated with this factor (Fig. 7). Component 2 was named “Anthropogenic Factor (F2)” and clarified 24.51% of the total variance. This factor was interrelated to the parameters of Cd and Hg. The Hg was moderate (0.50–0.75), and Cd was strongly (>0.75) positively associated with this factor (Fig. 7).

The CA was used to present the groups—clusters of similar water quality characteristics among the sampling locations on the Anday Stream Basin ($n=120$). The tree diagram of CA was shown (Fig. 8). As a result of CA, three clusters were formed. Cluster 1 (C1) associated to the S1 station, Cluster 2 (C2) associated to the S2, S3, S4, S5, S6, and S7 stations, and Cluster 3 (C3) associated to the S8, S9, and S10 stations.

Table 1 Health risk assessment indices values of water in the Anday Stream Basin

Kp		RfD _{ing}	RfD _{derm}	ABS _g	HQ _{ing}		HQ _{derm}		HI	
		μg/kg/day			Ad	Ch	Ad	Ch	Ad	Ch
Fe	1.00E-03	700	140	0.014	6.91E-06	7.74E-06	1.29E-05	2.85E-05	1.98E-05	3.62E-05
Ni	2.00E-04	20	0.8	0.04	4.47E-04	5.01E-04	2.92E-04	6.46E-04	7.39E-04	1.15E-03
Cu	1.00E-03	40	8	0.57	7.56E-03	8.47E-03	3.46E-04	7.66E-04	7.91E-03	9.23E-03
Zn	6.00E-04	300	60	0.2	2.31E-04	2.59E-04	1.81E-05	4.01E-05	2.50E-04	2.99E-04
Cd	1.00E-03	0.5	0.025	0.05	2.61E-03	2.93E-03	5.46E-03	1.21E-02	8.07E-03	1.50E-02
Hg	1.00E-03	0.3	0.021	0.07	6.09E-05	6.82E-05	6.49E-05	1.44E-04	1.26E-04	2.12E-04
Pb	1.00E-04	1.4	0.42	0.117	1.03E-03	1.15E-03	1.53E-05	3.38E-05	1.04E-03	1.18E-03

Ad, adults; Ch, children

Fig. 6 PCI coefficients of PTEs ($p < 0.05$ are boxed) in the Anday Stream Basin

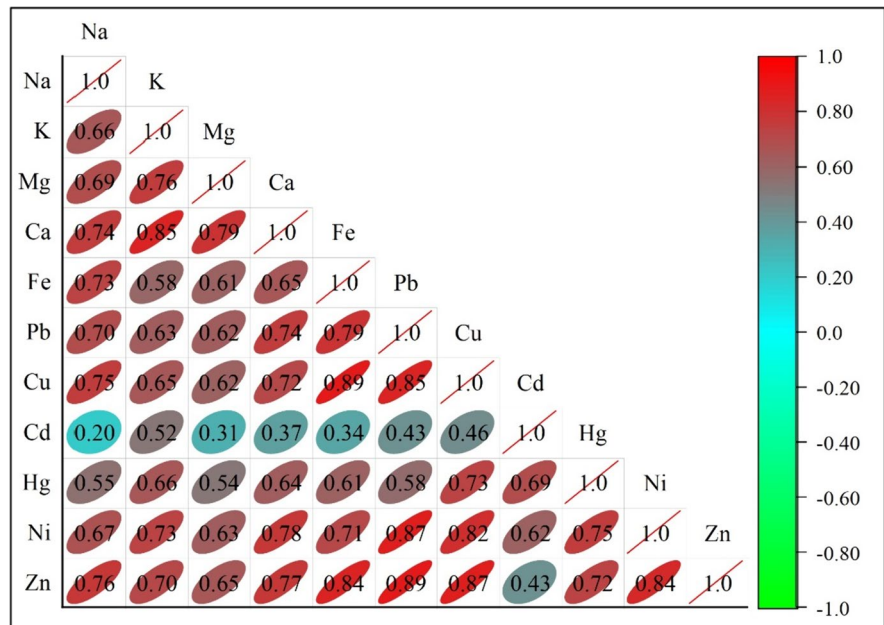
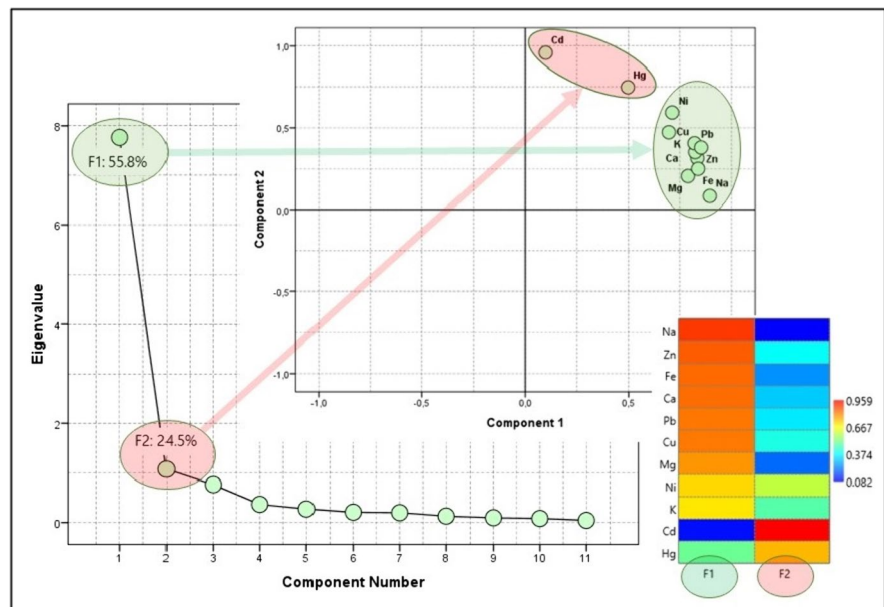


Fig. 7 Rotated component matrix (right), rotated component plot (middle), and scree plot (left) for potentially toxic elements in the water of the Anday Stream Basin

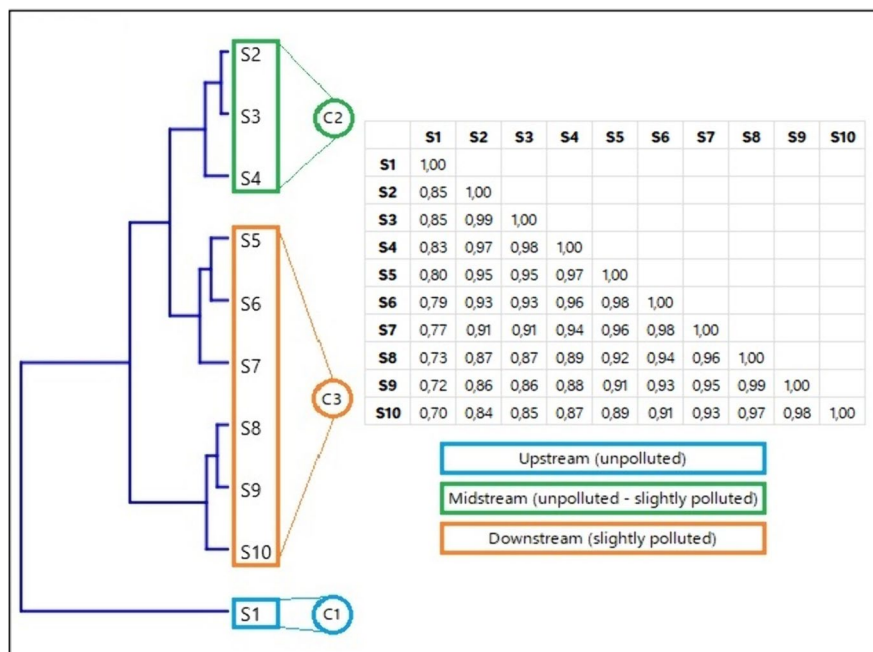


Discussion

Results revealed that water characteristics gradually increase from the upstream to the downstream. The values of water characteristics for all sampling points and periods in the Anday Stream Basin were noted under the drinking water guidelines (EC, 2007; WHO, 2017). Moreover, the water of Anday Stream

Basin showed first-class water quality characteristics in terms of almost all the determined PTEs (except for the Cu contents) during the investigated monthly sampling hydrological year (WQCR, 2015). The Cu values of water in Anday Stream Basin were found to be relatively higher (second-class water quality) at almost all the stations starting from the middle basin and in almost the entire basin in April and

Fig. 8 CA diagram for potentially toxic elements in the water of the Anday Stream Basin



May, when the heaviest precipitation was noted in the region (GDM, 2023).

Copper is an essential element at low levels of intake for all organisms, while it may cause toxic effects at higher levels. Although it exists naturally in soil, rock, and water, it may be released into the environment through anthropogenic sources, primarily factories, domestic wastewater, fertilizer use, and wood production (ATSDR, 2017). The area of Kastamonu Province constitutes about 4% of the total area of forests in Türkiye, and the region plays a vital role in manufacturing forest-originated products (OGM, 2006; Tokatli et al., 2021). The detected higher Cu content at the mid-downstream basin of Anday Stream Basin is thought to have originated mainly from the intensive wood production and the agro-genic applications conducted around the watershed. In addition, the main reason for the increase in Cu content in the entire basin in April and May could be the surface runoff from wood production and agricultural lands due to heavy rainfall during this season. The water quality results of this study were similar to a study reported in the vicinity of the Şehriban Stream which was found to be first class, except for the Cu concentration, which was quite risky and higher than the other investigated PTEs (Tokatli et al., 2021). The values of these parameters were noted much less than those reported for stream and river water by Ali and Muhammad (2022) and Haq et al. (2023) in Pakistan. This variation in water characteristics

was attributed to geological mineralization and ongoing surface activities in the basin.

Based on the current ecotoxicological and health risk assessment results, Anday Stream Basin showed relatively high water quality, and no non-carcinogenic risk was determined for the system. The HPI and HEI scores of Anday Stream Basin were much lower than those reported by Muhammad et al. (2022) for Kunhar River Basin, Pakistan. The children showed higher HQ and HI values among the human population than adults. Children's higher HQ and HI values were attributed to their higher vulnerability to toxicants. Higher values of children compared to adults were noted, consistent with a previous study on PTEs reported by Amin et al. (2021). The dermal route showed higher HQ and HI values than others. The Cd showed higher HQ and HI values than other PTEs. Higher HQ and HI values of Cd were attributed to its higher toxicity and low RfD value (Muhammad, 2023; Muhammad et al., 2022).

Nevertheless, as a result of agro-genic risk indices, the stream's water quality was recorded as "unsuitable" in terms of MH. A higher level of Mg than Ca in irrigation water may cause more alkaline agricultural soil characteristics, which may cause a decline in the harvest-yielding limit of the soil. Consequently, MH values greater than 50 in the irrigation water are stated as harmful for agro-genic purposes (Rahman et al., 2022). It was reported that Ca levels were higher than Mg in the

stagnant water bodies of the Black Sea Region (Mutlu et al., 2021). However, Mg concentrations were higher than Ca in the waters of potamic habitats, as in the Anday Stream Basin, which was more affected by the surface runoff due to the heavy precipitation regime of the region (Demir et al., 2021). This study reported that water of Anday Stream for agricultural activities seems quite risky in terms of MR values, while that of Na% (<20 , permissible) and SAR (<6 , excellent) values.

The observed high positive correlations among the parameters reflect that these contaminants may be released from the same source to the freshwater habitats through similar pathways (Tokatlı et al., 2023c; Yüksel et al., 2022). This indicates that the investigated PTEs are entering the water of the Anday Stream Basin from similar natural sources like wood preservatives, soil and rock weathering, and fertilizer.

The result of FA, the first factor, named the “natural factor,” is thought to be most affected by intensive precipitation and rock structure. In contrast, the second factor, the “anthropogenic factor,” is thought to be most affected by rural settlements and agricultural applications performed in the region (Fig. 7). Most PTEs, including Na, Zn, Fe, Ca, Pb, Cu, Mg, Ni, and K, are naturally occurring elements in the environment and can enter the water ecosystems due to the mineral disintegrations in soil and rocks through exposure to wind and water (Tokatlı et al., 2021). Cd may reach very high levels in phosphate rock used as a fertilizer for agricultural purposes, while it is a widely distributed element in the earth’s crust (ATSDR, 2017). Many studies have reported that Cd may attain relatively high levels in surface and groundwater having intense agricultural activities in the vicinity (Emiroğlu et al., 2013). It has also been stated that about 80% of the Hg released into the environment due to human activities comes from burning fossil fuels and 15% from fertilizers and fungicides used in agrogenic applications (ATSDR, 1999).

Potentially toxic elements in potamic habitats that exceed standard values may have a long-term negative chronic health effect on aquatic organisms and humans (Köse et al., 2020). Although anthropogenic factors are the main reasons for the increase of PTEs in aquatic habitats, natural factors like rock structure and erosion–washout may also be significant contamination sources (Wang et al., 2017; Wetzel, 2001). In the Black Sea Region of Türkiye, where the average precipitation rates are quite high, natural contamination factors may be more effective on the environment (Tokatlı et al.,

2021). The current research showed that the levels of PTEs are slightly increased in the water of the Anday Stream Basin, and the quality is decreased in the rainy season. Surface runoff may carry several anthropogenic or natural contaminants. In addition to the agricultural and wood production activities in the region, the rock structure of the Anday Stream Basin and the erosion due to heavy rainfall are thought to be among the primary sources of PTEs contamination in the region.

It is known that SDG 6, one of 17 SDGs set by the United Nations General Assembly in 2015, is about “clean water and sanitation for all.” The outcome-oriented targets of “Goal 6” include improving freshwater quality and restoring water-related ecosystems (UN, 2017; UN (2018)). The current study focused on the freshwater quality monitoring of Anday Stream Basin is a potamic habitat in the world. This study used various ecological, agrogenic, statistical, and health risk assessment indicators, to draw a comprehensive and complete picture of the Anday Stream Basin and provided recommendations to protect and ensure the sustainability of this significant aquatic habitat that will make significant contributions to the achieving the targets planned for “Sustainable Development Goal 6.” This study was limited to the water quality of Anday Stream Basin. Further studies need to be carried out on the sediments, bedrock geology, flora, and fauna of the Anday Stream Basin.

Conclusion

This study concluded that water characteristic parameters of the Anday Stream Basin were noted within the drinking water guidelines. The HPI and HEI results were noted as less than 100 and 10, respectively. Based on that Anday Stream Basin was classified as having “Low Heavy Metal Pollution” and “Low Contamination.” The HI values of Anday Stream Basin water were noted as less than 1 for the studied PTEs. This study noted higher HI values for Cd and observed that children were more vulnerable than adults. Statistical analyses showed that the natural and anthropic factors contributed to the water contamination of the Anday Stream Basin. This study recommends regular monitoring of future studies on the sediments, flora, and fauna of the Anday Stream Basin for the conservation, management, policy development, and fulfilling the Sustainable Development Goal.

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Author contribution C. Tokatli, designed this study, Supervision, and edited the manuscript. E. Mutlu, data curation, and edited the manuscript. F. Ustaoglu, A.R.T. Islam, and S. Muhammad performed data analyses and drafted and edited the manuscript.

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Data availability The data supporting this study's findings are available from the corresponding author upon reasonable request. No datasets were generated or analyzed during the current study.

Code availability Not applicable.

Declarations

Competing interests The authors declare no competing interests.

Ethics approval Not applicable.

Consent to participate All authors reviewed and approved the final manuscript.

Consent for publication All authors are approved for this publication.

Conflict of interest The authors declare no competing interests.

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