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Original Article

Impacts of a Garbage Disposal Facility on the Water Quality of Çavuşlu Stream in Giresun, Turkey: A Health Risk Assessment Study by a Validated ICP-MS Assay

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ABSTRACT

Environmental concerns have been raised because of a garbage disposal facility (GDF) constructed near Çavuşlu Stream in Giresun, Turkey. This article proposes a fully validated ICP-MS technique to investigate the impacts of the GDF on the water quality of the stream and provide a human health risk estimation. Water sampling was carried out at four different stations and in tap water since the stream is the primary source of drinking water for the inhabitants of the town. Quantification of selected toxic metals (As, Pb, Cd, Hg, Sb, Al, and Ni) in conjunction with the essential and other elements (Se, Cu, Fe, Mg, Mn, Zn, and Co) was performed by the use of the previously validated ICP-MS method. Once water quality index (WQI), heavy metal pollution index (HPI), and heavy metal evaluation index (HEI) were computed, the health risk assessment was studied according to the US EPA's method. Although the stations (2, 3, 4, and tap water) showed excellent water quality, station-1, which is the closest one to the facility, was classified as poor water quality. Lifetime cancer risk (LCR) was only significant both for adults and children in station-1. In addition, low risk regarding non-carcinogenic health hazards was found for children. The results indicate that the facility decreases the water quality of this stream and is possibly responsible for LCR. In conclusion, the ecological environment and human health should be protected by further monitoring the effect of the GDF on the ecological system.

Keywords: ICP-MS, water quality, health risk assessment, toxic metals, industrial pollution

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INTRODUCTION

Ecological pollution is an increasing difficulty all over the world as a result of industrialization, which influences each lifeform (Mutlu et al., 2016; Aydın et al., 2021). In particular, water pollution is one of the most significant issues, since the accessibility of high-quality drinking water is necessary for environmental and human health (Taş & Şişman, 2020). Nowadays, many water supplies are at risk because of uncontrolled industrialization and urbanization (Küçükosmanoglu & Filazi, 2020; Egbueri & Mgbenu, 2020). Hence, pollution is a global complication that affects surface water like rivers and streams (Taş & Kolören, 2017; Hadi et al., 2019). Continuing the release of heavy metals into surface waters can cause various chemical, physical, and biological problems (Ustaoğlu et al., 2020a). Contamination may originate from anthropogenic and geogenic sources (Ustaoğlu et al., 2017; Ustaoğlu & Tepe, 2018; Yuksel & Arica, 2018). Pollution in drinking water sources is associated with organic and inorganic impurities involving heavy metals and chemical ions (Egbueri & Mgbenu, 2020). In water ecosystems, excess amounts of metals with high persistence in nature are responsible for toxicity, as they can accumulate in aquatic organisms (Mutlu & Kurnaz, 2018; Ali et al., 2019).

Exposure to arsenic generally occurs through drinking water supplies polluted by natural, geological, and anthropogenic sources of inorganic arsenic. Today, the limit recommended by the World Health Organization (WHO) is 10 µg/L (WHO, 2011). Yet, it is likely that millions of individuals are using drinking water with an arsenic concentration above this safety standard. The relationship between the consumption of arsenic-contaminated drinking water and health disorders has been studied all around the world. Hence, arsenic exposure has been linked with certain cancer types such as liver, kidney, bladder, lung, and skin cancers, as well as other medical disorders like adverse effects in pregnancy, neurological disorders, and cardiovascular complaints (Ustaoğlu & Aydın, 2020; Thakur et al., 2020; Zhang et al., 2018; Yüksel et al., 2018). New technics are used to reduce arsenic concentration to a safe limit in the water treatment process (Zhu et al., 2018). Even so, previous studies proposed the possible toxicity of even low-level arsenic exposure because of its life-long accumulation capability in organisms (Roh et al., 2017; Tsuji et al., 2015).

Lead is a toxic heavy metal, and studies regarding lead exposure in drinking water have been well documented over the past few decades as the number of lead pollution cases has risen (Ding-Quan et al., 2020). It may distress nearly every body system, but chiefly, the hematologic, gastrointestinal, and nervous systems are affected. Furthermore, children are extra vulnerable to medical disorders due to lead exposure (Bozalan et al., 2019, Yüksel et al., 2016), as it damages children's behavioral and mental health (Redmon et al., 2020; Dórea, 2019).

Investigating the mercury levels in surface water such as rivers and lakes, as well as tap and bottled water, is important in water quality assessment because it is a toxic element having no biological or physiological function in humans. However, it is responsible for different sorts of health problems, such as neuropathological degradation, kidney deficiency, renal system failure, and leukemia (Marinho et al., 2020, Yüksel et al., 2017a).

According to the US EPA, cadmium is another toxic element that has been classified as Group B1 (probable human carcinogen). Cadmium pollution in drinking water occurs because of industrial debris and agricultural fertilizers. Specific examples of health disorders that have possibly been linked with cadmium exposure are renal failure, liver injury, muscle cramps, diarrhea, nausea, and vomiting (ATSDR, 2012; Cai et al., 2019). The permissible limit for cadmium in drinking water is 5.0 µg/L, according to the US EPA, EU, and TSE. However, the WHO has established this limit as 3.0 µg/L (WHO, 2011) since cadmium levels in uncontaminated drinking water are usually below 1.0 µg/L.

Certain metals are fundamental for aquatic life and other living organisms. The essential metals may be classified in two groups: micronutrients (Cu, Cr, Co, Fe, Se, Mn, Mo, and Zn) and macronutrients (Mg, Ca, Na, S, and P). However, elevated concentrations of these metals may exert toxicity by distressing reproduction, biotransformation, and growth in living organisms, including human beings (Gheorghe et al., 2017; Stankovic et al., 2014).

Origin characterization of metals dissolved in river water, as well as computing the proportional participation, is required to ensure environmental safety of aquatic ecosystems (Tepe & Aydın 2017; Tokatlı et al., 2019; Tokatlı et al., 2020). It is therefore beneficial to employ principal component analysis (PCA), hierarchical cluster analysis (HCA), and Pearson's correlation coefficient (PCC) analysis to assess the source and spread of metals in river waters (Köse et al., 2014; Çiçek et al., 2019; Ustaoğlu, 2020). There are critical activations to monitor the water quality of rivers. For instance, source identification of pollution, determining water quality status, and controlling water pollution are employed for effective water management (Taş et al., 2019; Varol, 2020). Since high-quality freshwater sources have rapidly deteriorated, water quality assessment in Turkey has become a significant issue in recent years. Water quality index (WQI), heavy metal pollution index (HPI), heavy metal evaluation index (HEI), hazard quotient (HQ), hazard index (HI), and carcinogenic risk (CR) are techniques that are widely used in water quality assessment (Ustaoğlu & Tepe, 2019). Therefore, they play an essential role in water resources management.

Çavuşlu Stream, which flows from northeastern Turkey into the Black Sea, is one of the principal watercourses in the region, as the water needs of the town of Çavuşlu are met by caisson wells in the basin. Çavuşlu Stream, also known as Yalakoda Stream, may be exposed to contamination through industrial, domestic, and medical waste from the GDF located near the stream. No environmental studies of water quality and health risk from the Çavuşlu Stream's water have been performed. Since the GDF is suspected to be the origin of water pollution in the town of Çavuşlu, environmental concerns have been rising day by day. For this reason, the primary objective of this research is to investigate the impact of the GDF on the water quality of Çavuşlu Stream and the human health risk. The complementary tools used in this study are to validate an ICP-MS assay to determine metal levels in stream water in the scope of ISO/IEC 17025 certification, and to address the probable origins of metal pollution through statistical tests such as PCC, PCA, and HCA.

MATERIAL AND METHODS

Research items and sample preparation

Sample collection was performed in early May 2020. Sample raw volume was 300 mL. Triple sampling was conducted at four different stations, starting from the closest point to the GDF near Çavuşlu Stream in Görele, Giresun, Turkey (Figure 1). In addition, tap water samples from three different houses in the town were utilized in this study. After 10-mL amounts of the samples were filtered through the Acrodisc® Minispike PTFE membrane with a pore size 0.45 µm (Merck, Germany), they were mixed with the same amounts of 8% (v/v) nitric acid. Calibration standards, at the concentrations of 1.0, 5.0, 10.0, 25.0, 50.0, and 100.0 µg/L, were produced by diluting a multi-element calibration mother solution (VHG LABS, Manchester, NH, USA) with an appropriate amount of 4% (v/v) nitric acid. To avoid any sort of cross-contamination, all glassware (Analitik Kimya, Istanbul, Turkey) was stored in 10.0% (v/v) nitric acid for 24 hours before the analysis.

Instrumentation

The metal levels in the water samples were quantified utilizing an ICP-MS (7700x ICP-MS, Agilent Corporation, USA). The generation of ultrapure water with a resistivity of 18MΩ cm was provided

Figure 1. Map of Çavuşlu Town, located in Görele, Giresun, Turkey (The garbage disposal facility and sampling stations are labeled).

for the sample preparation step employing Direct-Q8 (Merck-Millipore, Germany). The operating parameters of the instrument were set as follows: The water specimens were injected (60 s, 0.3 rps) by means of a Meinhard® nebulizer as well as a chilled spray chamber. The autosampler was promptly established to rest in the sampling stand for the specified time, and no flow injection valve was employed. As for the argon gas plasma conditions, reflected power and forward power were 7 W and 1300 W, respectively. The gas flow rates of plasma, auxiliary, and nebulizerwere set at 16.0, 1.0, and 1.0 L/min, respectively. Next, nickel interface cones were utilized. The peak jumping mode was employed when running the instrument. The autosampler pump was cleaned between injections in three steps, as follows: i) washing with ultrapure water for 30 seconds, ii) rinsing with 2% (v/v) nitric acid for 50 seconds, and iii) concluding the cleaning by using ultrapure water for 50 seconds.

Standard solutions and reagents

To plot the calibration graphs, VHG LABS (Manchester, NH, USA) multi-element standards solutions at the concentration of 10 mg/L of each element were employed. The internal standard multi-element stock solution to control the quantification stability of the instrument was obtained from Agilent® (USA). Nitric acid (HNO_3, 65% v/v) was purchased from Merck (Darmstadt, Germany) to prepare calibration standards and sample solutions. The certified reference material (CRM), ERM®-CA713 (Sigma-Aldrich, Germany), was utilized to test the validation of the assay. Finally, argon gas with an analytical purity (99.999%) was obtained from a local supplier in Turkey.

Assay optimization

As was well documented in our previous paper (Yüksel & Arica, 2018), quantification of trace element levels in natural water subjects is challenging with the matrix elements (Na, Mg, Ca, K, and Cl) in water. Thus, the water specimens were diluted by the amount of 10 mL 8% (v:v) nitric acid to weaken the matrix effects. The most favorable signal intensity throughout multi-element quantification at very low concentrations was achieved, employing the three elements ^{7}Li at low mass, ^{89}Y at medium mass, and ^{205}Tl at high mass.

Validation

Having taken into account the validation guide of the ISO/IEC 17025 standard (Gisbert Albaga et al., 2017), CRM ERM-CA713 Waste Water (Sigma-Aldrich, Taufkirchen, Germany) was analyzed 11 times to validate the assay based on accuracy, precision, recovery, and limit of detection. To improve the validation study, an in-house secondary reference standard solution at the concentration of 100 µg/L was produced by diluting the multi-element calibration mother solution with an appropriate amount of 4% (v:v) nitric acid. As described in previous papers, precision was calculated in terms of the coefficient of variation, while accuracy was expressed by relative error (Yüksel et al. 2020; Arica et al. 2018; Horwitz, 1982). The results of the validation study are given in Table 1, demonstrating that the assay is accurate and precise.

Statistical analysis

The use of different statistical approaches evaluated elemental quantifications in water samples. The PCC analysis was employed to assess the association between metals and their probable origin. Next, HCA was utilized to interpret the correlation among metals. Finally, PCA was employed to decrease data sets and uncover novel factors. SPSS® software version 22.0 was used throughout the statistical analysis.

RESULTS AND DISCUSSION

Quantification of metal levels

Metal levels in aquatic specimens can be measured by various techniques. However, ICP-MS is one of the most widely used assays because of its multi-element analysis capability (Yüksel & Arica, 2018). Most environmental research does not provide sufficient information regarding the accuracy of the method utilized. The ICP-MS method was validated with respect to accuracy and precision before the determination of metal levels in water samples, which improved the significance of this study. In this research, certified reference material and in-house secondary reference standard solutions were employed to perform the validation procedure. As a result, relative error, the variation of coefficient, and recovery were calculated between 0.5-2.7%, 0.9-4.0%, and 97.3-102.4%, confirming that the ICP-MS assay was accurate and precise. Quantified metal concentrations versus WHO, EU, US EPA, and TSE standards are shown in Table 2. Aluminum, iron, and manganese in station-1 exceeded all limits.

Aluminum levels in natural waters may alter dramatically contingent upon numerous mineralogical and physicochemical circumstances. For instance, aluminum levels typically vary between 1.0 to 50 µg/L in water with a neutral pH value, while they may be as high as 500–1000 u/L in increased acidity or organic content

Table 1. Validation Study for the ICP-MS method (Values are given in µg/L).

Metals	Reference Material	Certified Value	Measured Value	RE %	CV %	R %	LOD
Al	In-House Reference	100.0±0.3	98.4±2.1	1.6	2.1	98.4	1.58
Sb	In-House Reference	100.0±0.1	99.5±1.6	0.5	1.6	99.5	0.01
As	ERM-CA713	10.8±0.3	11.0±0.2	1.9	1.8	101.9	0.02
Cu	ERM-CA713	101±7	100.2±0.9	0.8	0.9	99.2	0.02
Hg	ERM-CA713	1.84±0.1	1.79±0.05	2.7	2.8	97.3	0.01
Zn	In-House Reference	100.0±0.4	101.7±2.2	1.7	2.2	101.7	0.38
Fe	ERM-CA713	445±27	451.4±6.7	1.4	1.5	101.4	5.50
Cd	ERM-CA713	5.09±0.2	4.97±0.2	2.4	4.0	97.6	0.02
Pb	ERM-CA713	49.7±1.7	50.2±1.1	1.0	2.2	101.0	0.02
Mn	ERM-CA713	95±4	96.3±2.2	1.4	2.3	101.4	0.20
Ni	ERM-CA713	50.3±1.4	49.4±1.9	1.8	3.9	98.2	0.03
Se	ERM-CA713	4.9±1.1	5.0±0.2	2.0	4.0	102.0	0.10
Ca	In-House Reference	100.0±0.5	100.5±1.9	0.5	1.9	100.5	12.89
Mg	In-House Reference	100.0±0.2	97.9±3.1	2.1	3.2	97.9	0.05
Co	In-House Reference	100.0±0.5	102.4±2.2	2.4	2.2	102.4	0.01

RE, CV, R, and LOD refer to relative error, variation of coefficient, recovery, and limit of detection, respectively.

Table 2. Measured metal concentration versus WHO, EU, US EPA, and TSE standards. Values are given in µg/L.

Metals	Station-1	Station-2	Station-3	Station-4	Tap Water	WHO, 2011	EU	USEPA	TSE
Aluminum	**1168.33±15.22**	51.34±1.20	**215.33±2.5**	4.32±0.3	16.80± 0.45	200	200	200	200
Antimony	0.70±0.01	0.02±0.00	0.01±0.00	<LOD	0.01±0.00	20	5	6	5
Arsenic	2.86±0.02	0.79±0.01	0.14±0.00	0.49±0.01	0.23±0.01	10	10	10	10
Copper	11.78±0.13	9.69±0.13	1.88±0.01	9.05±0.01	2.12±0.02	2000	2000	1300	2000
Mercury	0.01±0.00	0.02±0.00	<LOD	<LOD	0.01±0.00	6	1	2	1
Zinc	148.7±1.25	22.39±0.33	64.66±0.16	122.44±1.82	9.76±0.18	3000	…	5000	…
Iron	**1273.40±1.25**	85.42±3.45	88.4±3.1	176.52±6.14	2.98±0.13	200	200	300	200
Cadmium	1.53±0.04	1.57±0.03	0.03±0.00	1.36±0.07	0.63±0.04	3	5	5	5
Lead	1.78±0.05	0.28±0.01	1.36±0.13	0.92±0.04	1.28±0.05	10	10	15	10
Manganese	**150.72±1.10**	**60.27±0.45**	3.68±0.06	2.51±0.03	1.64±0.02	50	50	50	50
Nickel	10.27±0.26	5.31±0.07	1.88±0.01	7.19±0.12	2.68±0.04	20	20	…	20
Selenium	1.57±0.05	3.25±0.17	2.04±0.14	<LOD	0.62±0.04	10	10	50	10
Calcium	21958.93±229.6	18305±141	4048.41±82.65	26336±256	12252.68±5.5	75000	…	…	…
Magnesium	6826.06±62.20	5060.10±15.90	1728±34	13694±39.88	5362.75±38	50000	…	…	…
Cobalt	5.12±0.01	6.27±0.00	3.3±0.14	5.97±0.01	3.46±0.01	50	…	…	…

(Yavuz et al., 2013). Therefore, elevated aluminum concentration in station-1 may be linked with rich organic content that has originated from the GDF.

Iron is an essential element for humans in terms of cellular biochemical processes, as long as its amount is at trace levels. Nevertheless, iron can also become toxic when its concentration is elevated (Yüksel et al., 2017b). Iron concentrations in natural waters are varied between 500 to 50000 µg/L, while its levels in drinking water are generally below 300 µg/L (WHO, 2011). Raised iron levels in station-1 compared to other stations and tap water may be related to high metallic content in the GDF.

Manganese is a naturally rich essential element necessary for many integral biological processes in humans. Manganese concentration in drinking water is usually lower than 100 µg/L while it may be more than 1000 µg/L in freshwater. Drinking water with manganese levels of more than 100 µg/L results in an unpleasant flavor. Although manganese toxicity seldom happens, its level in drinking water should be quantified to avoid toxic exposures (Evans & Masullo, 2020; WHO, 2011).

Other measured essential metals and toxic metals were not found to be above the maximum contaminant limits. However, toxic metals like arsenic, lead, mercury, and cadmium can have toxic effects even at very low concentrations, as was described in the introduction section.

Water quality index (WQI)

WQI is one of the best classifying methods computed by taking into account the collective effect of different water quality factors on overall water quality. Therefore, it provides an inclusive and actual perception of the water quality.Initially, WQI was established by Horton (1965) in the USA, and today this approach is broadly benefitted through water quality examiners (Kükrer & Mutlu, 2019; Ustaoğlu et al., 2020b; Tokatli & Ustaoğlu 2020). Hence, WQI, in this study, was computed with the formula below (1).

$$WQI = \sum \left[W_i \times \left(\frac{C_i}{S_i} \right) \times 100 \right] \qquad 1$$

$W_i = w_i / \Sigma w_i$ refers to relative weight. Taking into consideration the comparatively critical impacts of heavy metals on public health, the W_i values are designated by minimum and maximum magnitudes of 1 and 5, respectively. C_i represents the microelement level quantified in the water where Si expresses the reference values reported by WHO (2011) in respect of drinking water. Concerning WQI, water quality is assessed in five categories: WQI ≥ 300, undrinkable; 200 ≤ WQI <300, very poor; 100 ≤ WQI<200, poor; 50 ≤ WQI <100, good; WQI <50, excellent (Xiao et al., 2019). Assigned weight (AS) and weight relative (WR) are presented in Table 3. Hence, Station-1, which is the closest one to the GDF, showed poor water quality (Table 4).

Table 3. Relative weight of each heavy metal.

Metals	Assigned Weight (AW)	Weight Relative (RW)
Al	4	0.073
Sb	4	0.073
As	5	0.091
Cu	2	0.036
Hg	5	0.091
Zn	3	0.055
Fe	4	0.073
Cd	5	0.091
Pb	5	0.091
Mn	5	0.091
Ni	5	0.091
Se	2	0.036
Ca	2	0.036
Mg	2	0.036
Co	2	0.036
	55	1.0

Table 4. List of calculated WQI, HPI, and HEI in all stations along with tap water.

	Station-1	Station-2	Station-3	Station-4	Tap Water
WQI	**131.42**	28.43	15.45	18.68	7.02
HPI	38.71	29.07	5.66	20.16	10.60
HEI	**16.91**	3.40	2.14	2.08	0.76

Heavy metal pollution index (HPI)

Since HPI is a valuation approach considering the united impact of every single heavy metal on total water quality, it has been employed by most scientists to expansively evaluate the overall water quality. HPI, in this study, was calculated with formulas (2 and 3), proposed as follows (Mohan et al., 1996):

$$HPI = \frac{\sum_{i=1}^{n}(Q_i W_i)}{\sum_{i=1}^{n} W_i} \qquad 2$$

$$Q_i = \sum_{i=1}^{n} \frac{M_i}{S_i} \times 100 \qquad 3$$

In formulas 2 and 3, Wi expresses the unit weight of the ith factor, Qi refers to the sub-index of the trace-toxic metal, Si states the reference values of the factor, Mi represents the screened values of toxic metals, and n stands for the number of factors taken into account. When HPI<100, it is, therefore, a low level of heavy metal pollution that is possibly not responsible for severe health effects (Saleh et al., 2018). Apparently, in this study, all stations, along with the tap water, showed HPI<100, which is a low level of heavy metal contamination (Table 4).

Heavy metal evaluation index (HEI)

Similar to HPI, HEI defines the general trend in the examination of water quality with reference to heavy metal pollution in water. Thereby, it may simply be employed to interpret the pollution degree in water (Edet & Offiong, 2002). In this study, HEI was computed based on the formula below (4).

$$HEI = \sum_{i=1}^{n} \frac{H_C}{H_{MAC}} \qquad 4$$

In this formula, Hc refers to the value detected for every single factor, where Hmac expresses the magnitude of maximum admissible concentration (MAC) for all variables (WHO, 2011). With reference to MAC, the elevated levels of the metal lead to further unpleasant water quality (Goher et al. 2014). As a general rule, when the level of individual metal exceeds the MAC value (HEI > 10), the water is not advisable for consumption. The water quality diminishes because of other impacts when metal levels are not exceeding but in the vicinity of the MAC values. Therefore, HEI is checked out by three classifications as follows: 20 < HEI means high contamination, 10 < HEI < 20 means medium contamination, and HEI < 10 means low contamination (Saleh et al., 2018). In our study, only station-1 showed medium contamination since it is suspected that the leaking from the GDF contaminates Çavuşlu Stream, whereas other stations and tap water showed low contamination (Table 4). The reason why other stations and tap water have low contamination is possibly the dilution of toxic metals with fresh water in the stream.

Health risk assessment: Hazard quotient, Hazard index, and cancer risk

Although current health risk assessment approaches, along with the mathematical patterns, may vary in different countries and organizations, the principle employed for this assessment remains the same. Throughout this research, the health risk assessment procedure proposed by the US EPA (2004) was applied. With regard to the trace elements in water, the health risk assessment process was performed by taking into consideration the amount of ingestion along with dermal absorption. To calculate the average daily dose (ADD) as a result of dermal absorption (ADD dermal) and direct digestion (ADD ingestion), the following formulas (5 and 6) offered by the US EPA (2004) were enforced:

$$ADD_{ingestion} = \frac{C_{Water} \times IR \times ABS_g \times EF \times ED}{BW \times AT} \qquad 5$$

$$ADD_{dermal} = \frac{C_{Water} \times SA \times K_p \times ET \times EF \times ED \times CF}{BW \times AT} \qquad 6$$

In these formulas, $ADD_{ingestion}$ means average daily dose by ingestion and ADD_{dermal} expresses average daily dose by ingestion in the unit of μg/kg/day. In addition, C_{water} refers to the concentration of the metals in surface water in the unit of μg/L. IR is ingestion rate (L/day), which, in our paper, is 2.0 for adult and 0.64 for children. As a parameter, ABSg refers to the unitless gastrointestinal absorption factor. EF is exposure frequency, which is set in our paper at 365 days/year. Next, ED represents exposure length in years, which is set at 70 for adults and 6 for children. BW displays average body weight in the unit of kg, and it was 70 for adults and 20 for children. AT is another parameter expressing the averaging time in days, and it is set in our paper at 25550 and 2190 for adults and children, respectively. SA is a parameter in the formula that stands for exposed skin area in the unit of cm^2, and it is 18,000 and 6600 for adults and children, respectively. Where Kp is the dermal permeability coefficient in water in the unit of cm/h, ET represents the exposure time throughout shower and bathing, which is 0.6 h/day in this paper. Finally, CF is the unit conversion factor, which is 1 L/1000 cm^3 (Xiao et al., 2019). Values of metals along with toxicological parameters employed for health risk assessment are illustrated in Table 5.

With regard to heavy metals, the non-carcinogenic risk by means of ingestion and dermal absorption was computed, and assessment was performed both for children and adults. As well described in a previous paper (Das et al., 2018), the risk hazard quotient formula (HQ) divides the average daily dose (ADD) by reference dose (RfD), while hazard index (HI) refers to the overall quantity of HQs as well as probable non-carcinogenic effects originatingfrom whole heavy metals.

Table 5. Hazard quotient and cancer risk for each element of the Çavuşlu Stream.

	HQ_{ing}		HQ_{derm}		HI		LCR(Ingestion+Dermal)	
Station-1	**Adult**	**Child**	**Adult**	**Child**	**Adult**	**Child**	**Adult**	**Child**
Al	3.04E-02	3.41E-02	8.35E-04	1.85E-03	3.12E-02	3.59E-02		
Sb	4.79E-02	5.37E-02	1.25E-02	2.77E-02	6.05E-02	8.14E-02		
As	2.48E-01	2.78E-01	1.44E-03	3.18E-03	2.50E-01	2.81E-01	**1.13E-04**	**1.28E-04**
Cu	4.60E-03	5.15E-03	2.11E-04	4.66E-04	4.81E-03	5.62E-03		
Hg	6.39E-05	7.16E-05	6.81E-05	1.51E-04	1.32E-04	2.22E-04		
Zn	2.72E-03	3.04E-03	2.13E-04	4.71E-04	2.93E-03	3.51E-03		
Fe	6.98E-04	7.81E-04	1.30E-03	2.88E-03	2.00E-03	3.66E-03		
Cd	4.19E-03	4.69E-03	8.75E-03	1.94E-02	1.29E-02	2.41E-02		
Pb	6.41E-04	7.18E-04	9.53E-06	2.11E-05	6.51E-04	7.39E-04		
Mn	1.03E-02	1.16E-02	2.25E-02	4.97E-02	3.28E-02	6.12E-02		
Ni	5.63E-04	6.30E-04	3.67E-04	8.12E-04	9.30E-04	1.44E-03		
Co	4.68E-01	5.24E-01	4.88E-03	1.08E-02	4.72E-01	5.34E-01		
				HI_{total}	8.71E-01	**1.03E+00**		
Station-2								
Al	1.34E-03	1.50E-03	3.67E-05	8.12E-05	1.37E-03	1.58E-03		
Sb	1.37E-03	1.53E-03	3.58E-04	7.91E-04	1.73E-03	2.33E-03		
As	6.85E-02	7.68E-02	3.96E-04	8.77E-04	6.89E-02	7.76E-02	3.13E-05	3.55E-05
Cu	3.78E-03	4.24E-03	1.73E-04	3.83E-04	3.96E-03	4.62E-03		
Hg	1.28E-04	1.43E-04	1.36E-04	3.01E-04	2.64E-04	4.45E-04		
Zn	4.09E-04	4.58E-04	3.20E-05	7.09E-05	4.41E-04	5.29E-04		
Fe	4.68E-05	5.24E-05	8.73E-05	1.93E-04	1.34E-04	2.45E-04		
Cd	4.30E-03	4.82E-03	8.98E-03	1.99E-02	1.33E-02	2.47E-02		
Pb	4.08E-03	4.56E-03	6.06E-05	1.34E-04	4.14E-03	4.70E-03		
Mn	4.13E-03	4.62E-03	8.98E-03	1.99E-02	1.31E-02	2.45E-02		
Ni	2.91E-04	3.26E-04	1.90E-04	4.20E-04	4.81E-04	7.46E-04		
Co	5.73E-01	6.41E-01	5.98E-03	1.32E-02	5.79E-01	6.55E-01		
				HI_{total}	6.86E-01	7.97E-01		
Station-3								
Al	5.60E-03	6.28E-03	1.54E-04	3.41E-04	5.76E-03	6.62E-03		
Sb	6.85E-04	7.67E-04	1.79E-04	3.96E-04	8.64E-04	1.16E-03		
As	1.21E-02	1.36E-02	7.03E-05	1.55E-04	1.22E-02	1.38E-02	5.54E-06	6.28E-06
Cu	7.34E-04	8.22E-04	3.36E-05	7.44E-05	7.68E-04	8.96E-04		
Hg	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00		
Zn	1.18E-03	1.32E-03	9.25E-05	2.05E-04	1.27E-03	1.53E-03		
Fe	4.84E-05	5.43E-05	9.03E-05	2.00E-04	1.39E-04	2.54E-04		
Cd	8.22E-05	9.21E-05	1.72E-04	3.80E-04	2.54E-04	4.72E-04		
Pb	3.11E-03	3.49E-03	4.63E-05	1.02E-04	3.16E-03	3.59E-03		
Mn	2.52E-04	2.82E-04	5.48E-04	1.21E-03	8.00E-04	1.50E-03		
Ni	1.03E-04	1.15E-04	6.72E-05	1.49E-04	1.70E-04	2.64E-04		
Co	3.01E-01	3.38E-01	3.15E-03	6.96E-03	3.05E-01	3.44E-01		
				HI_{total}	3.30E-01	3.75E-01		
Station-4								
Al	1.12E-04	1.26E-04	3.09E-06	6.84E-06	1.16E-04	1.33E-04		
Sb	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00		
As	4.25E-02	4.76E-02	2.46E-04	5.44E-04	4.28E-02	4.82E-02	1.94E-05	2.20E-05
Cu	3.53E-03	3.96E-03	1.62E-04	3.58E-04	3.70E-03	4.32E-03		
Hg	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00		

Table 5. Continue.

Station-4								
Zn	2.24E-03	2.50E-03	1.75E-04	3.87E-04	2.41E-03	2.89E-03		
Fe	9.67E-05	1.08E-04	1.80E-04	3.99E-04	2.77E-04	5.07E-04		
Cd	3.73E-03	4.17E-03	7.78E-03	1.72E-02	1.15E-02	2.14E-02		
Pb	2.11E-03	2.36E-03	3.13E-05	6.93E-05	2.14E-03	2.43E-03		
Mn	1.72E-04	1.93E-04	3.74E-04	8.27E-04	5.46E-04	1.02E-03		
Ni	3.94E-04	4.41E-04	2.57E-04	5.69E-04	6.51E-04	1.01E-03		
Co	5.45E-01	6.11E-01	5.69E-03	1.26E-02	5.51E-01	6.23E-01		
				HI_{total}	6.15E-01	7.05E-01		
Tap Water								
Al	4.37E-04	4.90E-04	1.20E-05	2.66E-05	4.49E-04	5.16E-04		
Sb	6.85E-04	7.67E-04	1.79E-04	3.96E-04	8.64E-04	1.16E-03		
As	2.00E-02	2.23E-02	1.15E-04	2.55E-04	2.01E-02	2.26E-02	9.10E-06	1.03E-05
Cu	8.28E-04	9.27E-04	3.79E-05	8.39E-05	8.66E-04	1.01E-03		
Hg	6.39E-05	7.16E-05	6.81E-05	1.51E-04	1.32E-04	2.22E-04		
Zn	1.78E-04	2.00E-04	1.40E-05	3.09E-05	1.92E-04	2.31E-04		
Fe	1.63E-06	1.83E-06	3.04E-06	6.74E-06	4.68E-06	8.56E-06		
Cd	1.73E-03	1.93E-03	3.60E-03	7.97E-03	5.33E-03	9.91E-03		
Pb	2.93E-03	3.28E-03	4.36E-05	9.64E-05	2.97E-03	3.38E-03		
Mn	1.12E-04	1.26E-04	2.44E-04	5.41E-04	3.57E-04	6.66E-04		
Ni	1.47E-04	1.64E-04	9.58E-05	2.12E-04	2.43E-04	3.76E-04		
Co	3.16E-01	3.54E-01	3.30E-03	7.30E-03	3.19E-01	3.61E-01		
				HI_{total}	3.51E-01	4.01E-01		

HQ and HI were computed based on the following formulas (7 and 8) (US EPA, 2004):

$$HQ = \frac{ADD_{ingestion}/ADD_{dermal}}{RfD_{ingestion}/RfD_{dermal}} \qquad 7$$

$$HI = \sum (ADD_{ingestion} + ADD_{dermal}) \qquad 8$$

Medical disorders in humans caused by heavy metals may be observed when HI, HQ>1. In contrast, no adverse health effect is observed if HI, HQ <1. Wu & Sun (2016) report that HI (hazard index) is evaluated in 5 categories: $HI_{total} > 4$ means extreme risk, $3 < HI_{total} < 4$ means high risk, 2 < HItotal < 3 means medium risk, $1 < HI_{total} < 2$ means low risk, and $HI_{total} < 1$ means no risk. Therefore, in this paper, low risk in association with non-carcinogenic health hazards was found for children, while no risk was calculated for adults (Table 5).

Lifetime cancer risk (LCR) may be defined as conveying possible risk because of exposure to a carcinogen throughout life, and it was calculated using formula (9) (US EPA, 2004).

$$LCR = ADD \times CSF \qquad 9$$

In the present investigation, LCR was assessed due to arsenic content, since it is one of the most carcinogenic toxic metals analyzed. Based on previous articles (Gao et al. 2019; Saha et al. 2017), Cancer Slope Factor (CSF) values were utilized as 0.0015 and 0.00366 μg/kg/day for ingestion and dermal exposure, respectively. As reported by the US EPA (2004), the tolerable or acceptable carcinogenic risk ranges from 10^{-6} to 10^{-4}. From another point of view, adverse health effect is very likely by the time LCR ≥ 10^{-4}. As can be seen in Table 5, Station-1 has LCR, while other stations and tap water do not show LCR. The reason why station-1 shows poor water quality and LCR may be due to toxic materials leaking from the GDF.

Source identification

The origin identification was based on hierarchical cluster analysis (HCA) and principal component analysis (PCA) to classify clusters of water quality variables and sampling sites of similar contamination features, as described by Mishra et al. (2017). Therefore, PCA was conducted to associate the contribution source as well as providing consistent evidence through the correlation of metals (Table 6). As evidenced in Figure 2, the component plot in rotated space indicated that metals are linked with three different sources. The outcome from PCA was supported by HCA, signifying sample grouping within the dataset by three clusters (Figure 3). Since sampling stations are in the vicinity of GDF and an agricultural region where pesticides are applied, one of the most significant reasons for toxic metal pollution can be explained as anthropogenic. In addition, it is possible that geogenic contamination has an effect on water quality, as Çavuşlu Stream is also fed from groundwaters.

Pearson's correlation (PC) matrix was employed to observe whether or not metal levels in the stream water are interconnected with one another (Ustaoğlu & Islam, 2020; Mutlu, 2019). As proposed by Ali et al. (2016), strong interrelation among certain metals in riv-

Table 6. PCA Component values of metals analyzed.

	Components		
Variable	**PCA1**	**PCA2**	**PCA3**
Al	*.999*	-.007	.040
Sb	*.987*	.118	.038
Fe	*.982*	.187	-.035
As	*.940*	.308	.131
Mn	*.891*	.285	.352
Pb	*-.829*	-.147	.528
Ni	*.715*	.689	-.117
Zn	*.700*	.333	-.532
Co	.023	*.956*	.188
Ca	.228	*.940*	-.231
Cd	.299	*.925*	.208
Cu	.511	*.833*	.148
Mg	-.055	*.784*	-.615
Se	.060	-.077	*.918*
Hg	.046	.325	*.871*
Eigenvalues	8.564	3.073	2.835
% of variance	57.095	20.487	18.897
Cumulative %	57.095	77.582	96.479

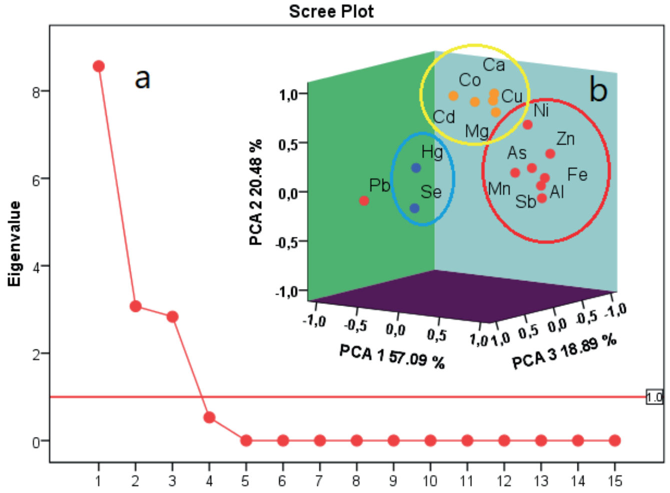

Figure 2. PCA of measured parameters by (a) scree plot of the characteristic roots (Eigen values) and (b) component plot in rotated space.).

Table 7. Correlations of metals.

	Correlations														
	Al	**Sb**	**As**	**Cu**	**Hg**	**Zn**	**Fe**	**Cd**	**Pb**	**Mn**	**Ni**	**Se**	**Ca**	**Mg**	**Co**
Al	1														
Sb	_.986**_	1													
As	_.941*_	_.976**_	1												
Cu	.513	.595	.746	1											
Hg	.074	.153	.282	.369	1										
Zn	.680	.677	.667	.608	-.448	1									
Fe	_.978**_	_.992**_	_.977**_	.650	.082	.762	1								
Cd	.299	.423	.600	_.938*_	.535	.370	.461	1							
Pb	-.804	-.827	-.763	-.451	.332	-.870	-.861	-.287	1						
Mn	_.902*_	_.930*_	_.973**_	.741	.451	.521	_.916*_	.606	-.597	1					
Ni	.705	.781	.867	_.925*_	.149	.797	.834	.825	-.753	.792	1				
Se	.102	.048	.127	.160	.637	-.340	.006	.096	.490	.344	-.111	1			
Ca	.210	.338	.480	.849	.155	.557	.409	_.901*_	-.462	.393	.836	-.312	1		
Mg	-.086	.021	.113	.523	-.259	.526	.115	.588	-.402	-.041	.571	-.654	.873	1	
Co	.027	.121	.325	.870	.393	.311	.191	_.906*_	-.035	.353	.657	.187	.835	.616	1

**. Correlation is significant at the 0.01 level (2-tailed). *. Correlation is significant at the 0.05 level (2-tailed).

er water can be evidence of the same origin of the contamination. The results of the PC matrix are illustrated in Table 7. Mn has a positive correlation with Al, Sb, As, and Fe, proposing that these metals in the stream water have similar sources. Furthermore, these findings are consistent with the outcome of PCA and HCA.

CONCLUSION

For the first time, this paper has outlined the impact of the GDF on the water quality of the Çavuşlu Stream, located in Görele, Giresun, Turkey. We also developed a simple revalidation procedure employing in-house and certified reference materials, which resulted in further accurate results after instrumental analysis by ICP-MS. Station-1, in the vicinity of GDF, was classified as having poor water quality. Furthermore, LCR was significant only at station-1 both for adults and children, and non-carcinogenic health hazard was estimated only at station-1 for children. Source identification by PCA and HCA indicated that metals in the Çavuşlu Stream might primarily originate from anthropogenic and geogenic sources. Overall, the results point to the GDF decreasing the water

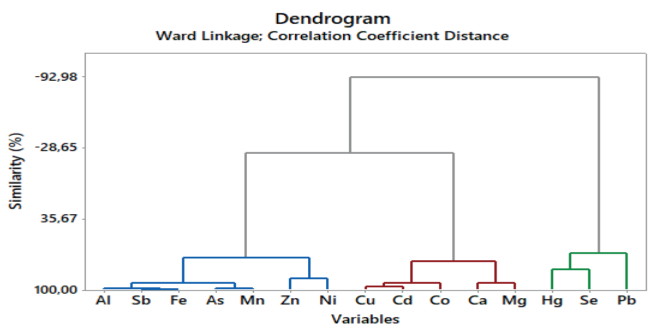

Figure 3. Hierarchical cluster analysis.

quality of the stream and possibly being responsible for LCR. Hence, we propose that the GDF should be moved somewhere else, not in the proximity of water resources and towns. In addition, as a temporary solution, new caisson wells should be constructed on the upper side of the stream where water quality is not directly affected by the GDF. However, the ecological environment and human health will be at risk as long as the activity of the GDF lasts. Environmental monitoring to assess the effect of the GDF on the ecological system should be maintained. Therefore, sediment and fish samples will be investigated to assess ecotoxicological risk in the next phase of this research project.

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