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Original Article

Temporal and spatial fluctuations of some heavy metals and silica in Tigris River water, downstream of Belad city, Iraq

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Abstract: This study was designed to assess the concentrations of six elements of zinc, cadmium, lead, nickel, aluminum, and total silica (SiO_2) in Tigris River water downstream of Belad city and evaluate their temporal and spatial fluctuations. For this purpose, water samples were collected monthly, and four sampling sites were chosen and sampled during 2022. According to the results, the average concentrations of all metals in the water were within the permissible range by Iraqi river standards. Only aluminum slightly crossed the allowed levels during the study. In addition, the values of metals varied spatially and increased longitudinally. Generally, the concentrations of metals in the Tigris before Baghdad's Province entrance were parallel with many global rivers and increased steadily toward the downstream. Elevated Al concentrations are discussed regarding the river region's features and the sedimentation method.

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Introduction

Heavy metals include necessary and unnecessary trace metals that vary according to their characteristics, abundance (chemical speciation), and concentration and may be toxic to organisms (Marcovecchio et al., 2013). Sources of heavy metals in aquatic systems include wastes from mining, agriculture, homes, and industries, as well as metals that dissolve in natural deposits (Marcovecchio et al., 2013; Spellman, 2014). In the riverine water, heavy metals like lead, zinc, cadmium, and nickel are present in both dissolved and particulate forms (Marcovecchio et al., 2013). In water, metals can be divided as either nontoxic or toxic. Toxic metals are dangerous in small concentrations, whereas others are defined as nontoxic metals (Marcovecchio et al., 2013).

Some heavy metals, such as zinc, iron, cobalt, manganese, copper, strontium, molybdenum, and vanadium, are necessary in trace quantities by living organisms. However, the organism may suffer if the concentrations of essential metals increase. Nonessential heavy metals of particular concern to aquatic systems are chromium, lead, mercury, antimony, cadmium, and arsenic (Marcovecchio et al., 2013; Spellman, 2014). For most plants, silica is an unnecessary trace element; however, most animals need it. Silica is not necessary for most plants but is required for most animals. There is no minimum concentration level (MCL) for silica in drinking water standards by the US EPA. There is a more explanation of the presence and structure of silica in aquatic environments (Baird et al., 2017). The average silica concentration in ground and surface water is 14 $mg.l^{-1}$ (Baird et al., 2017). Aluminum is nonessential for plants and animals. According to (Law No. 25, 1967) of the Iraqi rivers maintenance rules, the aluminum level should not exceed 0.10 $mg.l^{-1}$. While for irrigation, water should not exceed 5 $mg.l^{-1}$ (Baird et al., 2017).

Many works done to assess the concentrations of different metals in the Tigris River within Baghdad City (Sabri et al., 1993; Hashim and Rabee, 2014; Al-Ani et al., 2014; Al-Obaidy et al., 2016; Rasheed et al., 2017; Abed et al., 2019; Aljanabi et al., 2022; Oleiwi Al-Dabbas, 2022; Al-Bahathy et al., 2023). However, no investigations were performed in the

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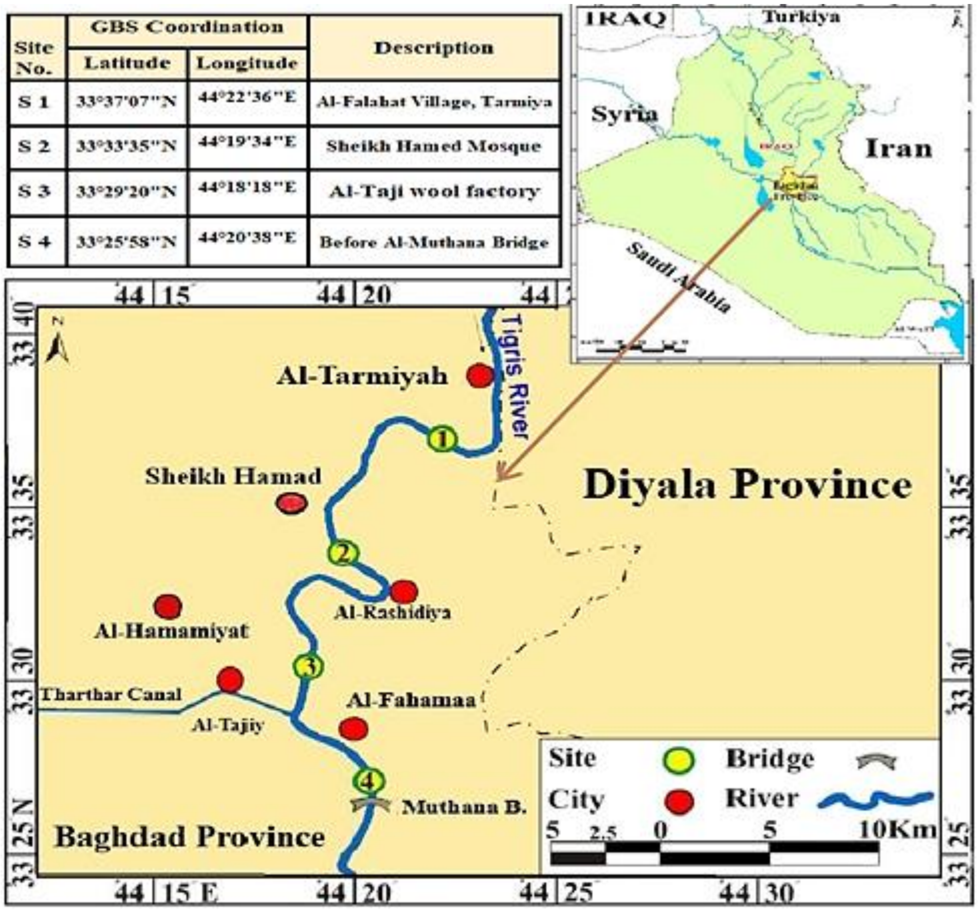

Site No.	GBS Coordination		Description
	Latitude	Longitude	
S 1	33°37'07"N	44°22'36"E	Al-Falahat Village, Tarmiya
S 2	33°33'35"N	44°19'34"E	Sheikh Hamed Mosque
S 3	33°29'20"N	44°18'18"E	Al-Taji wool factory
S 4	33°25'58"N	44°20'38"E	Before Al-Muthana Bridge

Figure 1. Study area and sampling sites in 2022.

Tigris River water downstream of Belad City; therefore, this study aimed to measure the concentrations of total silica with five toxic elements in Tigris River water 30 km before the entrance of Baghdad Province.

Materials and Methods

Study area: Tigris River is the major water resource for the Baghdad Province and splits into two parts, the eastern part known as "Risafa" and the western part known as "Karkh"(Ali et al., 2012; Majeed et al., 2022a, 2023). From Tarmiyah Town to the Diyala River confluence in southern Baghdad City, the river travels approximately 50 km (Nama, 2015; Majeed et al., 2022a). This river is a dynamic system that reacts to various physicochemical characteristics (Majeed et al., 2022b, c). Based on population distribution, confluence area, and agricultural activities on the Tigris River drainage, four sampling sites were selected, covering a stretch of about 25 km downstream of the Borders of Belad City (Fig. 1). Subsurface river water samples were collected between January and December 2022. The first station was near Falahat Village in Al-Tarmiya (33°37'07"N, 44°22'36"E). The second site was near the Sheikh Hamed mosque (33°33'35"N, 44°19'34"E). The third station is at 33°29'20"N and 44°18'18"E, next to Al-Taji wool-industries. The fourth station was 250 m ahead of the Muthana Bridge area (Fig. 1).

Sampling and sample preparation: Samples were collected in 1-liter screw-capped polyethylene bottles previously cleaned. After returning to the laboratory, the samples were refrigerated at 4°C. High-purity, concentrated nitric acid was used to digest the water samples, and the results were obtained fast (Marcovecchio et al., 2013; Baird et al., 2017).

Analytical techniques for assessing the metals: Using an atomic absorption spectrophotometric (AAS) method described in standard procedures, the cadmium, lead, nickel, and zinc contents in water

Table 1. The concentration levels of the analyzed heavy metals in Tigris water. 1st line shows the lowest and highest levels ($mg.l^{-1}$), and 2nd line the average and standard error.

Sites Metals	1	2	3	4	Standard Value
Pb^{2+}	0.0058-0.1416 0.0391±0.0102	0.018-0.075 0.0455±0.0058	0.0141-0.11 0.04±0.0076	0.014-0.12 0.0415±0.0085	0.0500
Zn^{2+}	0.008-0.096 0.033±0.007	0.015-0.075 0.032±0.005	0.11-0.089 0.039±0.009	0.014-0.1442 0.042±0.01	0.5000
Cd^{2+}	0.001-0.0041 0.0018±0.0003	0.0013-0.0046 0.0025±0.00032	0.001-0.004 0.0025±0.00032	0.00071-0.0042 0.00241±0.00032	0.00500
Ni^{2+}	0.0012-0.03 0.011±0.00275	0.0036-0.053 0.0162±0.0044	0.0036-0.0823 0.018±0.0068	0.0036-0.0411 0.0134±0.0036	0.1000
Al^{3+}	0.031-0.44 0.222±0.0449	0.0311-0.5 0.236±0.0436	0.025-0.54 0.244±0.0397	0.073-0.49 0.241±0.0464	0.1000
Silica	4.23-14.3 8.59±0.82	5.4-10.9 7.625±0.447	6.2-10.1 7.825±0.401	6.26-11.9 7.59±0.454	14

Standard value-based Iraqi river's-maintained standards (Law No. 25, 1967). Only Silica was taken from Baird et al. (2017).

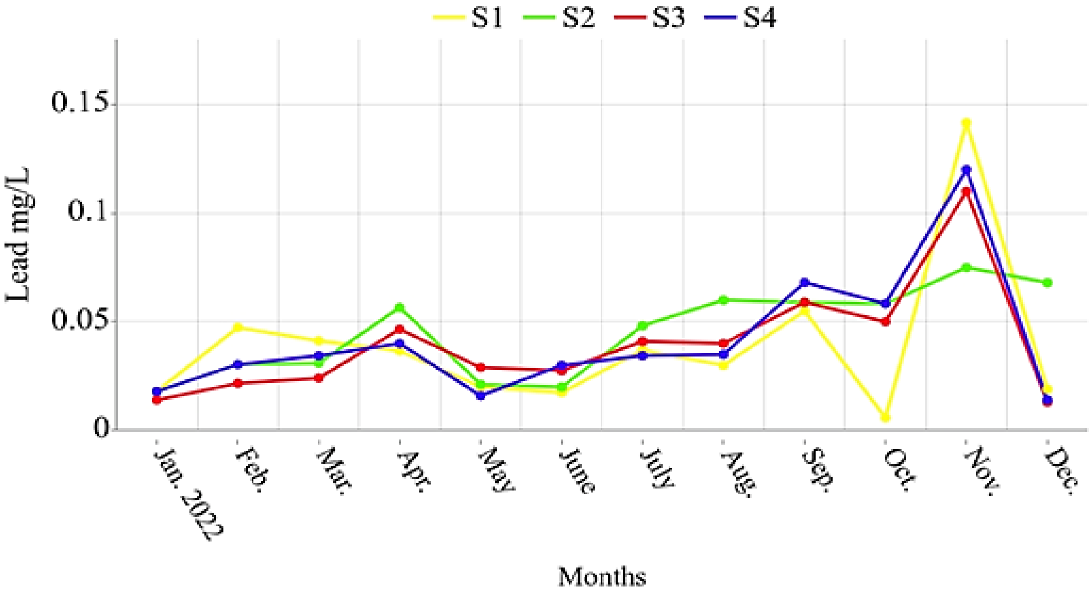

Figure 2. Variations of lead value in the studied stations of the Tigris water during 2022.

samples were ascertained (Chen and Teo, 2001; Baird et al., 2017). This technique is appropriate for determining very low amounts of metals. The total silica (SiO_2) was determined using the ammonium molybdate technique (Baird et al., 2017). The colorimetric method of Eriochrome cyanine R is used to find the lowest concentrations of aluminum ions at a wavelength of 535 nanometers (Baird et al., 2017; Majeed and Ibraheem, 2024). Calculate each metal ion's concentration in micrograms per liter ($mg.l^{-1}$) using the appropriate calibration curve. The average of three replicates served as the basis for each determination. Before the samples were tested, a suitable drift blank was taken (Marcovecchio et al., 2013).

Results and Discussions

Table 1 and Figure 2 show the Pb concentration in the Tigris River water. Our results corresponded with the findings of Hashim and Rabee (2014) recorded 0.035-0.86 $mg.l^{-1}$ in the Diyala River and 0.015-0.28 $mg.l^{-1}$ in the Tigris River. Hamdan (2020) reported that the water of Shatt Al-Arab contained Pb in a range of 0.0002 to 0.002706 $mg.l^{-1}$. Oleiwi and Al-Dabbas (2022) showed that the maximum values of Pb increased relatively along the Tigris River from 0.006 $mg.l^{-1}$ within Baghdad City to 0.006 $mg.l^{-1}$ in Al-Azizziyah City. In contrast, Al-Hussaini et al. (2018)

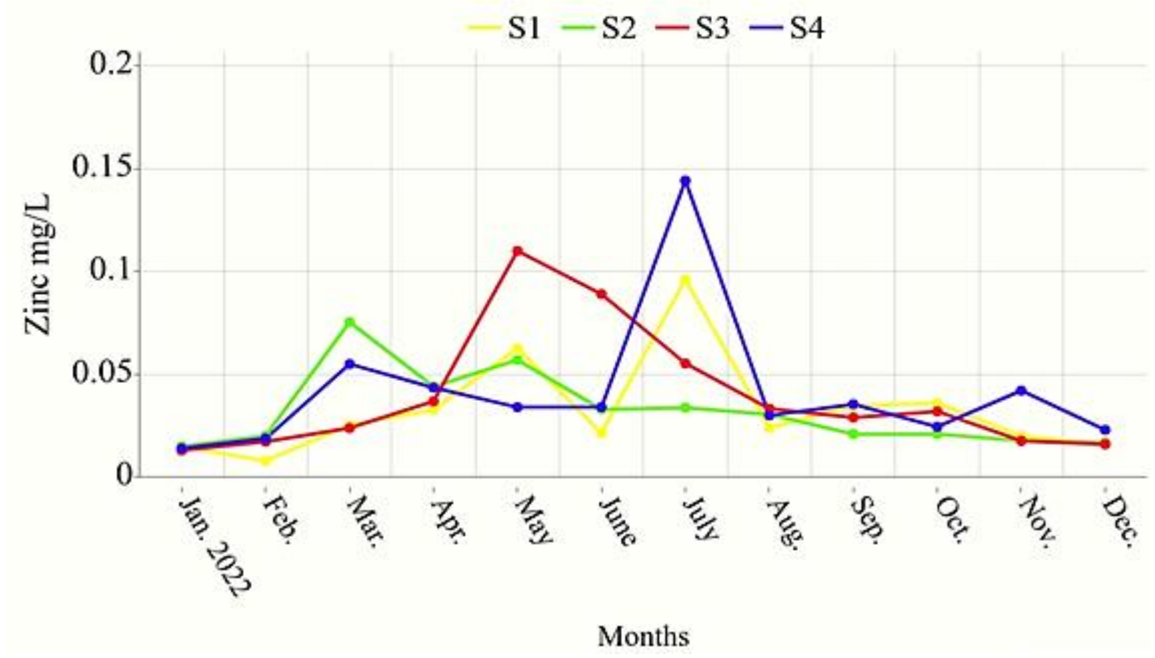

Figure 3. Variations of Zinc value in the studied stations of the Tigris water during 2022.

recorded high concentrations of Pb in the Diyala River, reaching 1.8202 mg.l^{-1}, attributing to the impact of Al-Rustimiyah wastewater treatment plants. Singh et al. (2005) found that Pb concentrations in the Gomti River ranged from 0.0158-0.0276 mg.l^{-1}. Zhaoyong et al. (2015) obtained the same results, showing that the minimum and maximum concentrations of lead ions were 0.000001 and 0.001227 mg.l^{-1}, respectively, in the surface water of China's Mountains in the Balikun-Yiwu section. Cao et al. (2018) found that the concentrations of Pb ranged from 0.00002-0.00041 mg L^{-1} in the Hai River water. Jiang et al. (2019) showed that the mean concentrations of lead ions in the Xiangjiang River water remained below the standard value, ranging between 0.00181 and 0.00135 mg.l^{-1} between 2005 and 2016, respectively. In contrast, Eneji et al. (2012) recorded a high concentration of 0.207 mg.l^{-1} of Pb along the Benue River, attributed that to solid wastes of industrial activities like lead batteries, Polyvinyl chloride plastics, paints alloys, explosions, and gunpowder. Myvizhi and Aruna Devi (2020) reported a high concentration of lead ions in the Cauvery River, which is related to the release of wastewater from the leather and chemical industries directly into the river. Jaiswal et al. (2022) found that the average values of Pb in the Yamuna River were within permissible limits, but in certain cities such as Delhi, Mohana, and Agra, the values exceeded the slandered limits due to the discharge of effluents containing lead into the river.

Based on our results, the lowest Pb levels were found during the winter season (Fig. 2). This may be linked to the rainwater. The metals' value in the river decreases when there is more rainfall. This finding, in agreement with Hashim and Rabee (2014), showed that the lowest Pb was during November and December. The maximum values were recorded during the autumn season, especially in November. Li and Zhang (2010) found that the peak values of Pb in the Han River in November related to the diluting effect in flood season. The lowest mean concentration was recorded at site 1 as 0.0391 mg.l^{-1}, and the highest at site 2 as 0.0455 mg.l^{-1} (Table 1). The lowest concentration at Site 1 may be related to the absence of anthropogenic activities. Based on Iraqi standards (Number 25, 1967), the mean values of Pb did not exceed the permission limit of 0.05 mg.l^{-1} (Table 1). Conversely, Aljanabi et al. (2022) found that Pb levels in the Tigris River in Baghdad Province were higher than the permission limits. This may be related to differences in sampling sites.

The Zn content of Tigris River water is shown in

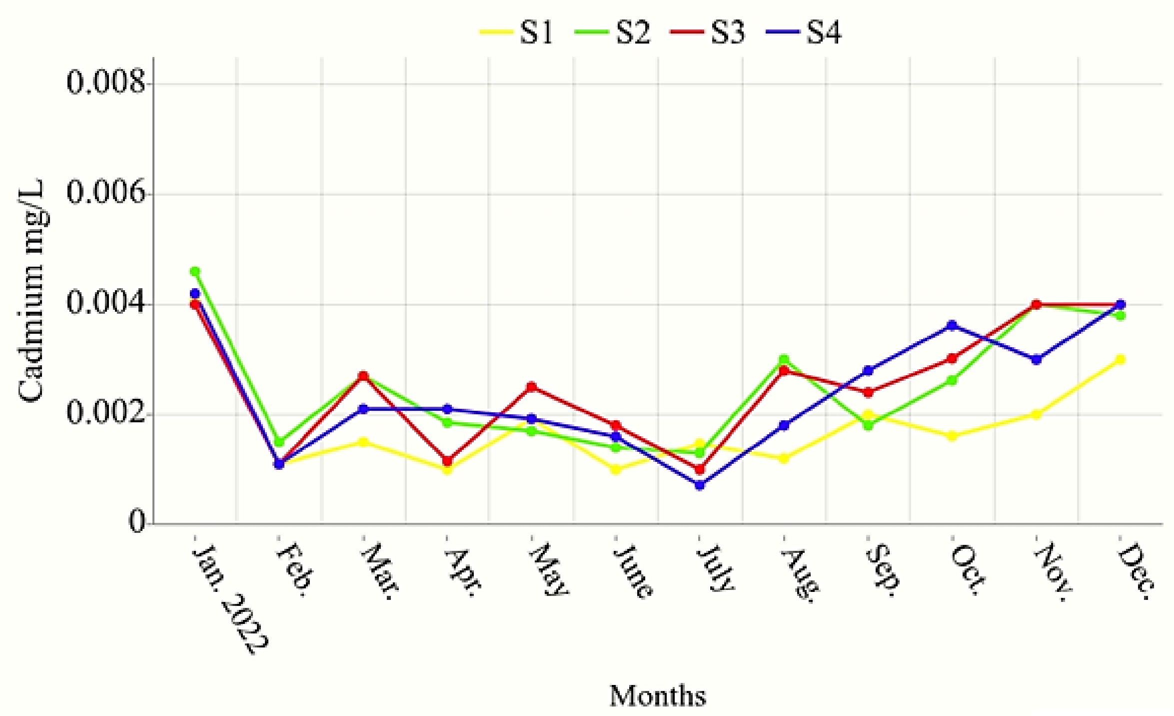

Figure 4. Variations of Cadmium value in the studied stations of the Tigris water during 2022.

Table 1 and Figure 3. The values of Zn in Tigris River water did not cross the permission limits of the Iraqi river's standards i.e., 0.5 mg.l^{-1} (Law No. 25, 1967), and global standard of 0.02 mg.l^{-1} (Baird et al., 2017). Our results were supported by previous studies such as Hashim and Rabee (2014), who found that the Zn in Tigris water varied from 0.02 to 0.36 mg.l^{-1} while in Diyala River, varied between 0.028-1.11 mg.l^{-1}. Hamdan (2020) reported that Zn ranged from 0.0032 to 0.010063 mg.l^{-1} in Shatt Al-Arab River's water. Oleiwi and Al-Dabbas (2022) reported the same results, showing that the maximum Zn value in Tigris water within the Baghdad region was 0.05 mg.l^{-1}, while in Al-Azizziyah City, it was recorded as 0.06 mg.l^{-1}.

Eneji et al. (2012) showed that the mean value of Zn was 0.0787 mg.l^{-1} in Benue River. Zhaoyong et al. (2015) found the same results, showing that the Zn varied between 0.000001-0.001268 mg.l^{-1} in the surface water of China's Mountains in the Balikun-Yiwu section. Cao et al. (2018) found that the concentrations of Zn ranged between 0.00001 and 0.00107 mg.l^{-1} in the Hai River water. Jiang et al. (2019) demonstrated that the local government's remediation and preventive strategies regulated the mean concentrations of zinc ions in the Xiangjiang River water, falling from 0.04967 mg.l^{-1} in 2005 to 0.0134 mg.l^{-1} in 2016. Similarly, another study in the Dankran River found very small concentrations of zinc, ranging between 0.0000618 and 0.000142 mg.l^{-1} (Afriyie et al., 2022). Jaiswal et al. (2022) also found that the mean concentrations of Zn in the Yamuna River ranged between 0.04527 mg.l^{-1} in the monsoon and 0.19202 mg.l^{-1} in the non-monsoon, within allowable limits of Indian standards.

Regarding temporal variation, zinc levels were highest in the summer (Fig. 3), which may be related to fertilizer effluent. Reza and Singh (2010) found that the highest value of Zn in the summer season was attributed to the remaining zinc sulfate in fertilizers. Salman et al. (2014) also showed that Zn increased in summer in the Al-Hilla River. The lowest values were in the winter season, and this may be a return to the diluting effect of the rainfall, which consequently decreased the concentration. For spatial variations in the current study, the lowest mean concentration recorded was 0.032 mg.l^{-1} in site 1, whereas the highest was 0.042 mg.l^{-1} in site 4 (Table 1). The lowest concentration at site one may be related to the absence of anthropogenic activities in this region.

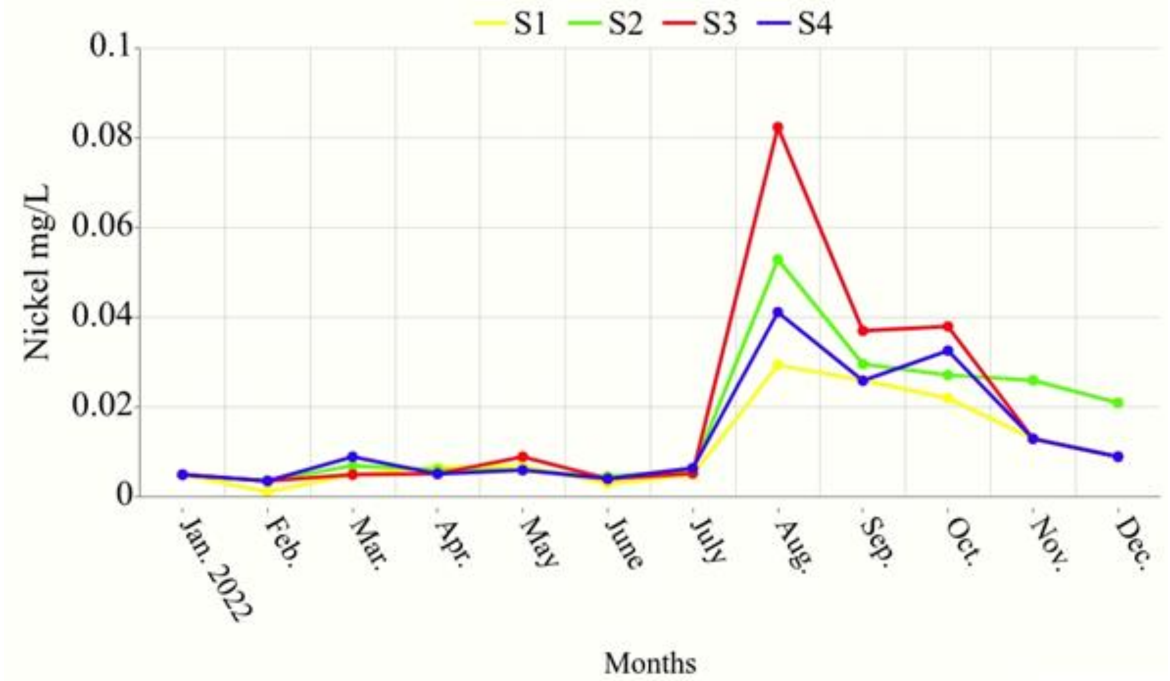

Figure 5. Variations of Nickel in the studied stations of the Tigris water during 2022.

Table 1 and Figure 4 summarize the Cd concentrations, ranging from 0.001 mg.l^{-1} at site 1 in April and June to 0.0042 mg.l^{-1} at site 4 in July and January. The results supported by the findings of Hashim and Rabee (2014) found that the amounts of Cd in the Tigris and Diyala Rivers were different, ranging from 0.001 to 0.0092 mg.l^{-1} in the Tigris River and between 0.0025 and 0.28 mg.l^{-1} in the Diyala River. Also, Hashim and Rabee (2014) recorded high concentrations of Cd in the Diyala River, reaching 0.0928 mg.l^{-1}, pointing to the effect of the absence of heavy metal treatment units in Al-Rustimiyah treatment plants for wastewater. Hamdan (2020) found that Cd in Shatt Al-Arab Rive water ranged between 0.000144 to 0.00555 mg.l^{-1}. Oleiwi and Al-Dabbas (2022) showed that the maximum value of Cd in the Tigris River was 0.002 mg.l^{-1} within Baghdad and Al-Azizziyah City.

Singh et al. (2005) found that Cd concentrations in the Gomti River ranged between 0.0001-0.0005 mg.l^{-1}. Eneji et al. (2012) recorded a high concentration of Cd in the Benue River, and the concentration was 0.052 mg.l^{-1} on average, attributed to industrial wastewater and poorly managed municipal solid waste. Zhaoyong et al. (2015) obtained the same results, indicating that Cd concentrations ranged from 0.000001 to 0.001132 mg.l^{-1} in the surface water of China's Mountains in the Urumqi-Akesu section. Cao et al. (2018) observed that the concentrations of Cd in the Hai River's water ranged from 0.00001 to 0.00015 mg.l^{-1}. Jiang et al. (2019) also demonstrated that the average Cd concentrations in the Xiangjiang River water remained below the standard value, between 0.00208 and 0.00022 mg.l^{-1} in 2005 and 2016, respectively. Whereas in the Nigerian Benue River, Myvizhi and Aruna Devi (2020) reported high concentrations of Cd in the Cauvery River due to wastewater from power stations, steel, cement, and dyeing industries discharged directly into the Cauvery River.

Seasonally, the maximum value of Cd was recorded at 0.0046 mg.l^{-1} in winter, especially in January, due to higher rainfall, which reduced metal concentrations. The minimum value was 0.000714 mg.l^{-1} during the summer, especially in July (Fig. 4). This may be related to decreasing water due to increasing evaporation. In contrast, Reza and Singh (2010) showed that the highest values of Cd in Brahmani River water in summer reached 0.004 mg.l^{-1}, related to coal-combustion and domestic purposes.

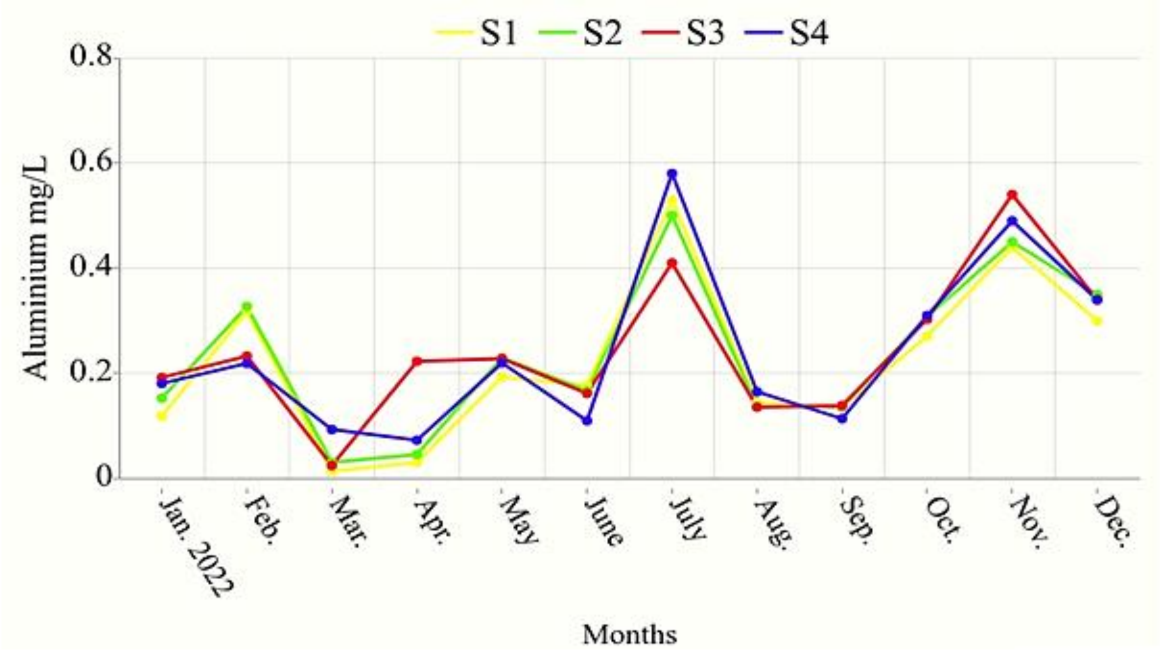

Figure 6. Variations of Aluminium in the studied stations of the Tigris water during 2022.

Furthermore, because of the high evaporation rate and agricultural activities, Li and Zhang (2010) found that the summer months had the highest cadmium ion concentrations in the upper stream of the Han River.

Spatially, the least and greatest mean were 0.0018 and 0.00254 mg.l^{-1} at sites 1 and 3, respectively (Table 1). The absence of industrial activities and human inputs close to the riverbanks may contribute. Based on Iraqi river-maintained standards (Law 25/1967), the concentrations of Cd were beyond the limits in all sites within 2022 (Table 1).

Over a yearly period, the values of nickel metal in four sites are shown in Table 1 and Figure 5. Our findings were compatible with the results of Sabri et al. (1993), which showed that the levels of Ni were within the Iraqi standard for river water. Hamdan (2020) found Ni in Shatt Al-Arab Rive water ranged from 0.0003 to 0.0301 mg.l^{-1}. Singh et al. (2005) found that Ni concentrations in the Gomti River ranged between 0.0066-0.011 mg.l^{-1}. Zhaoyong et al. (2015) obtained the same results, showing that the low and high levels of Ni were 0.000002-0.000985 mg.l^{-1}, consequently, in China's Mountains water in the Zhaosu-Tekesi Section. Cao et al. (2018) found that the values of Ni varied between 0.00103-0.00994 mg.l^{-1} in the Hai River water. Conversely, Al-Hussaini et al. (2018) recorded high concentrations of Ni in the Diyala River, reaching 0.6003 mg.l^{-1}, pointing to the effect of the absence of heavy metal treatment units in Al-Rustimiyah treatment plants for wastewater.

Seasonally, the peak values of Ni appeared in the summer season, especially in August, and decreased in winter, especially in February (Fig. 5). The increase in summer may be a return to high evaporation and intense agriculture activities. This finding, supported by Li and Zhang (2010), showed that Ni in the upper Han River increased in August. The decrease in winter may be related to the high amount of rain, which dilutes the river metals. Spatially, site 1 had the lowest average reading of Ni, recorded at 0.011 mg. l^{-1}. At site 3, the largest average reading was 0.018 mg.l^{-1} (Table 1). This could be attributed to the fact that the river stretches in site 1 is away from anthropogenic and industrial activities. Generally, the levels of Ni in every site during the analysis period did not exceed the limits of 0.1 mg.l^{-1} (Law 25/1967; Baird et al., 2017).

The values of Al in the Tigris River are presented in Table 1 and Figure 6. According to Iraqi river maintenance standards (Law 25/1967), Al concentrations slightly exceeded the limits in all sites (Table 1). These results are consistent with the findings of Al-Obaidi et al. (2010) observed that Al

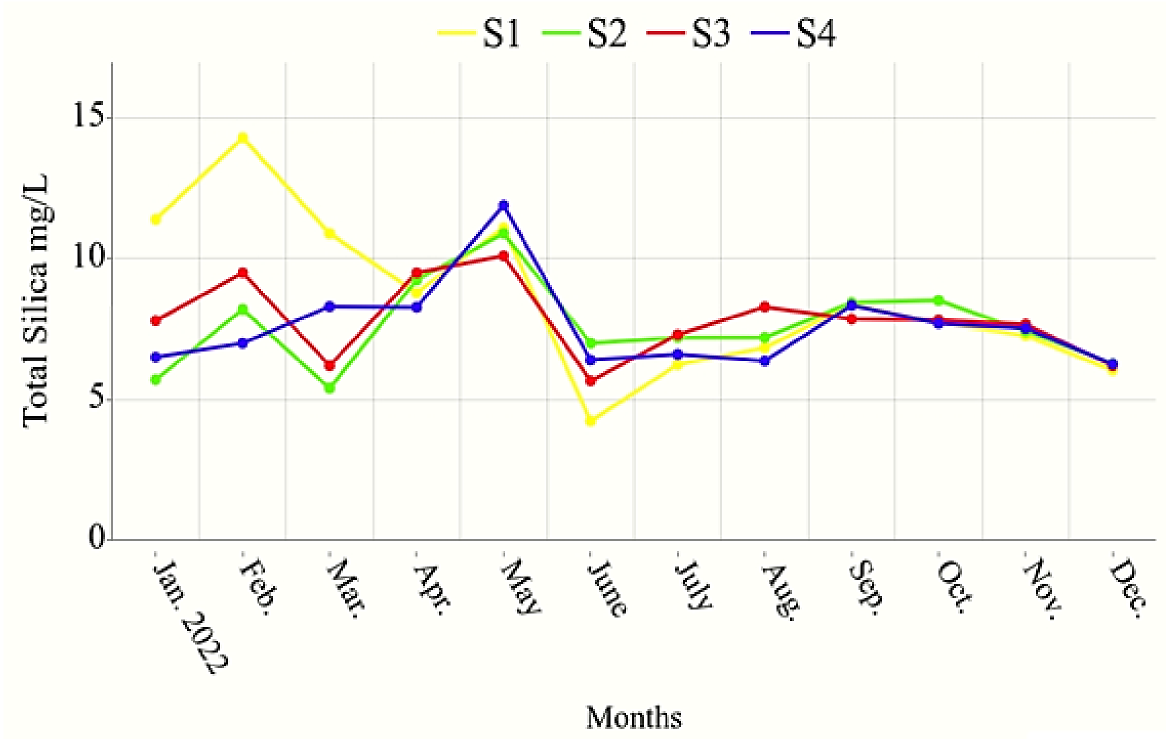

Figure 7. Variations of total silica in the studied stations of the Tigris water during 2022.

concentration in the Tigris River within Mosul City varied between 0.007 and 0.037 mg.l^{-1}. Abed et al. (2019) found that Al concentrations in Tigris River water within Baghdad City ranged between 0.1 and 0.4 mg.l^{-1}. Gibbs (1972) showed that the concentration of dissolved aluminum in Amazon River water ranged between 0.02 and 0.06 mg.l^{-1}. In China, Wang et al. (2013) found that Al concentrations in the Weihe River ranged between 0.065 to 0.928 mg.l^{-1} in Mongolia. Wang et al. (2013) found that Al concentrations in the Selenga River and its tributaries varied from 0.01 to 0.1 mg.l^{-1}. In the USA, Brown and Bruland (2009) found that aluminum concentrations in the Columbia River were 0.00216 mg.l^{-1}. Also, the concentration of aluminum decreased with increasing salinity. Our results showed that aluminum concentrations varied seasonally. Autumn had the highest values (Fig. 6). This result agrees with Senze et al. (2021), who showed that the highest concentrations of aluminum metal in the Nysa Szalona River occurred in the autumn season. The lowest values were observed in the summer.

For spatial variations, the mean values of Al for all sites were similar (Table 1). The highest average value reached 0.244 mg.l^{-1} at site 3, while the lowest average value recorded was 0.222 mg.l^{-1} at site 1. Senze et al. (2021) showed the same mean values for the Nysa Szalona and Strzegomka Rivers. Table 1 and Figure 7 show the values of total silica in Tigris water during 2022. In the first site, the value varied from 4.23 to 14.3 mg.l^{-1} in June and February. The second site showed 5.4 mg.l^{-1} as the lowest in March and 10.9 mg.l^{-1} as the highest in May. The lowest and highest measurements in the third site were 6.2 and 10.1 mg.l^{-1} in March and May, respectively. The fourth site fluctuated between 6.26 mg.l^{-1} in December and 11.9 mg.l^{-1} in May. Additionally, total silica concentrations in each site were lower than standard silica limits in surface water, i.e., less than 14 mg.l^{-1} (Baird et al., 2017). These findings are consistent with Gibbs's (1972) showing that the dissolved silica in Amazon River water ranged between 9.1 and 12.42 mg.l^{-1}. Abed et al. (2019) reported a low amount of silica, 0.6-0.72 mg.l^{-1}, in Tigris water within Baghdad city. The measurements varied spatially; site 1 had the lowest average value, 4.23 mg.l^{-1}, and site 4 had the greatest average value, 11.9 mg.l^{-1} (Table 1). This could be due to the physical process of sedimentation and the geography of the river areas. Regarding temporal variation, the summer and spring seasons had the lowest and highest silica values, respectively (Fig. 7).

Conclusions

The current study demonstrated that the average concentrations of lead, zinc, cadmium, nickel, and total silica in surface water were within the global average and the standards maintained by Iraqi rivers. The only metal that marginally went Aluminum exceeded the allowed limits, which will require further analysis. Concentrations varied spatially. The highest values for all metals were recorded in the first site as the lowest values, while in site 4, the concentrations increased longitudinally. The concentrations also varied seasonally.

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