Assessment of war impact on concentrations of pollutants and heavy metals and their seasonal variations in water and sediments of the Tigris River in Mosul / Iraq

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Abstract

War-related contamination of water and sediment of the Tigris River within the urban area of Mosul leads to seasonally independent exceedances of the WHO limit values for Cd, Pb, Cr, and Ni up to a factor of 100 in water and sediments. Furthermore, exceedances consistently occur for conductivity, PO_4^{3-}, and SO_4^{2-}, as well as sporadically for salinity and COD in water samples, and consistently for salinity in sediment samples, highlighting the direct impact of war (amunition, ignition of sulfur fields) as well as indirect effects (destroyed wastewater infrastructure). Conflict-related emissions from the former conflict zone (S5-S7) are highlighted by the sudden increases in load from S4 to S5, although partially masked by discharge of highly polluted water from the Khosr River (between S3 and S4). Due to sorption to sediments and presumed wind-borne discharge of highly polluted particles into the Tigris River, sediments at S10 on the southern edge of Mosul showed the highest pollutant loads. Significant statistical differences were observed by T-test analyses for E.C., TDS, salinity, COD, PO_4^{3-}, NO_3^-, SO_4^{2-}, Cd, Pb, Zn, Cr, and Ni for water samples as well as salinity, Cd, Pb, Zn, and Cr for the sediment samples for seasonal comparison. Since the percentage difference of water samples at S4-S7 is smaller than upstream and downstream, contaminant input is not limited to rainwater, but also via year-round infiltration of highly polluted wastewater from the surrounding valleys or suburban areas as well as presumably polluted groundwater or windblown particulate input.

1. Introduction

Over the centuries, wars have always brought disasters to humans and the environment, but the environment remains the silent victim (Reuveny et al. 2010). Such environmental damage and destruction can be divided into three categories:

- Environmental damage caused by pre-war activities to extract resources for direct warfare or to finance the conflicts, e.g., oil, sulfur, mineral resources, or timber (Snyder 2020). Low-tech processing methods are often used by competing armed groups to minimize investment costs and the need for skilled labor. As a result of low-tech processing, heavy metals such as mercury, lead, or cadmium can be released into the environment and pollute water bodies (RECEU 2015).
- Environmental damage from direct acts of war, such as the destruction of agricultural land and specialty crops as well as the fate of destroyed weapons and munitions remnants in the environment (Snyder 2020).
- Anarchic conditions or the proclamation of martial law result in a reduced control and monitoring function of state environmental authorities, leaving private companies operating in conflict zones subject to minimal environmental oversight (Bazyan 2012).

In terms of population and area, Mosul is the second largest city in Iraq after Baghdad. The Tigris River is one of the most important rivers in the region, as it is the main source of water for drinking, industry, agriculture, entertainment, and public purposes in the city (Khecher et al. 2016).

ISIS held the city from June 2014 to July 2017. The liberation battles, which lasted from October 2016 to July 2017, resulted in more than 40,000 casualties and the complete destruction of 9518 buildings (of which approximately 80% were residential), according to UN reports. The resulting debris in Mosul is estimated at 11 million tons. This mass is equivalent to three times the Great Pyramid of Giza or four times the Eiffel Tower (UN environment 2017, UNEP 2018).

The most severe pollution in the urban area, in addition to the direct use of weapons and resulting explosions, was caused by the burning of both 18 oil wells by ISIS in Qarrayah and their nine-month fire (soot) and 50,000 tons of pure sulfur in the Mishraq complex (SO_2, SO_3) (UNEP and OCHA 2016). The release of these contaminants had a direct impact on water, groundwater, soil and air and led to an accumulation of these contaminants over time. The consequences are clear today in the quality of water and sediment of the Tigris River (Khecher et al. 2016, Plebani and Magri 2017), especially in direct comparison with pre-war studies.

In the pre-war years, the water quality of the Tigris River met the limits set by Iraq and the World Health Organization (WHO) for drinking water and domestic use, although a wide variety of tributaries and activities resulted in the discharge of pollutants. For example, the drainage of many valleys to Lake Mosul, the discharges of the tributaries of the Al-Mur Valley, the Northern Aljazeera Irrigation Project, and the sulfur springs of the Badoosh Dam Project led to increased eutrophication and discharge of sulfur compounds. Furthermore, discharges of hazardous industrial wastes (e.g., cement industry), solid buffalo wastes (min. 10 tons dry matter/day) in the Badoosh area, and car maintenance, car washing, livestock washing, and livestock treatment facilities located on both sides of the river bank resulted in degradation of the water quality of the Tigris River in terms of the parameters of hardness, salinity, nutrients, organic loads, and algal growth (Jankeer and Mustafa 2017).

In 2012, a study by Al-Masri and Fadhel (Al-Masri and Fadhel 2014) investigated the variations in pollutant concentrations in the Tigris River within the Mosul urban area. The water qualities determined were on average equivalent to that of clean water, with 17% of the test samples classified as polluted. Although the river experienced pollutant discharges from many sources such as households, industry, and agriculture,

the dilution and self-purification potential of the river was sufficiently high, especially at high flow rates, to sustain good water quality in the Tigris. The WQI average was 83% (good water quality) (Al-Masri and Fadhel 2014). A later study by the authors addressed the water quality of the Tigris River upstream and downstream of Mosul city to elucidate the impact of wastewater discharges in Mosul during the period from 2002 to 2012. The detected nutrient and salinity loads were harmless to the growth and reproduction of all aquatic hydrophyte species. However, the organic loads as well as rotting of aquatic plants enhanced by eutrophication resulted in increased odor, change in color and taste, and significant increase in salinity (Fadhel 2020).

Farhan et al (Farhan et al. 2020) conducted an assessment of the water quality of the Tigris River in the urban area of Mosul for drinking water and domestic use using the CCME WQI based on field work from 2008 to 2014. Based on the selected ten parameters, namely pH, turbidity, and concentrations of calcium, dissolved oxygen, dissolved solids, nitrate, chloride, phosphate, and sulfate, the Tigris showed water quality during the study period ranging from 66.3–93.7, or moderate to good, respectively.

A study conducted in 2011–2012 of heavy metal concentrations in water and sediments within the Mosul urban area showed that heavy metal concentrations in both compartments were within the normal concentration ranges of Iraq, with concentrations in sediments always higher than in water. The concentrations in water followed the descending order of Zn < Cu < Pb < Cd, while in sediment a concentration order of Cu < Pb < Zn < Cd was observed. The results revealed a significant increase in concentration of these heavy metals in both aqueous phase and sediments in the area of Al-Busaif south of Mosul compared with Al-Mushirfa area in the northwestern part of Mosul as a control (Al-Sarraj et al. 2019).

The importance of heavy metal pollution, besides acute toxicity, lies particularly in its ability to bioaccumulate in living organisms (Aoyama et al. 1978). Even though heavy metals and other contaminants lead to a deterioration in water quality and thus in the health status of the population, no post-war studies were conducted so far looking at the impact of war on emissions of these contaminants.

Therefore, the aim of this study was to quantify the effect of heavy metal inputs resulting from the war on the water and sediment of the Tigris River within the urban area of Mosul, considering seasonal variations (test series winter and spring vs. summer and fall) and comparing both measurement points upstream and downstream of Mosul, and to compare them with existing global limits. The comparison of the measured values was made considering the percentage difference as well as the T-test approach.

2. Materials and Methods

2.1. Investigation area

The Tigris river basin is located in the city of Mosul between northwest Mosul at Al-Quba and southeast Mosul at Hammam al-Aleel. Within the investigation area with sampling points according to Table 1, the Tigris has an annual discharge of 250–300 m³/s (Meshhadani and Jassim 2012).

2.2. Sampling procedure

30 water and sediment samples each were taken at ten points on the Tigris (see Table 1) in triplicate during the two test series (series 1: January to March 2022; series 2: July to September 2022) using clean polyethylene bottles with a capacity of 250 ml were taken, resulting in a total sample volume of 120 samples. The sample temperatures during sampling were 0–10°C during the first test series, while they were between 35–45°C in the second test series. Each sample was directly stored in a refrigerated container at -4°C after sampling, transported directly to the laboratory, and further analyzed for 13 parameters: pH, electric conductivity (E.C.), % salinity, total dissolved solids (TDS), chemical oxygen demand (COD), PO_4^{3-}, NO_3^-, SO_4^{2-}, Pb, Zn, Cd, Cr, and Ni. The collection and analysis of the samples in the environmental laboratory was carried out according to standard methods (Lipps et al. 2017).

Water samples were taken according to DIN EN ISO 5667-6. During sampling from the river bank, special attention was paid to ensure that sampling was done from the homogeneous layer of the water body without picking up surface films, dead water, or swirling bottom sediments. Sampling was done at mid-height between the riverbed and the surface.

Sediment samples were taken according to DIN ISO 5667-12 from the upper river sediment layer at a sediment depth of 0–30 cm with a clean plastic scoop and placed in plastic bags. Sampling took place at the same 10 locations along the river as sampling for water samples.

2.3. Sediment sample preparation

Water samples were directly analyzed according to standard methods after membrane filtration. The sediment samples were dried, and their heavy metal concentrations were determined according to the method described in Jackson (Jackson 1958), in which 0.5 g of dry soil was placed in a glass flask and 5 ml of the digestion solution consisting of concentrated sulfuric acid (H_2SO_4), concentrated nitric acid (HNO_3)

and perchloric acid ($HClO_4$) were added in the ratio (3:1:1). The samples were then heated on a heater at 90°C in a hood for two hours. After the samples had cooled down, they were diluted to 25 ml by adding distilled water and the respective heavy metal concentrations were quantified in mg/kg dry weight of the soil using an atomic absorption spectrometer.

2.4. Field measurement parameters

Temperature, pH value, E.C., TDS, and % salinity of water samples were directly analyzed on-site using an Oumefar 5 in 1 digital water quality monitor analyzer of type UPC 886108495111.

2.5. Laboratory measurement parameters

Parameters of section 2.4. were measured for sediment samples by mixing the sample with distilled water (about 10–15 g of sample in 25 ml of distilled water). After 2 to 3 minutes, until the soil is well loosened, pH, E.C., and salinity of the solution were measured electrometrically with the Oumefar 5 in 1 analzyer. All following parameters were analyzed according to standard methods.

Phosphate PO_4^{3-}(in mg/L) was photometrically analyzed without digestion at 690 nm as phosphorus molybdenum blue using ammonium molybdate solution according to DIN EN 1189.

Nitrate NO_3^-(in mg/L) was also photometrically analyzed at 324 nm after acidification with HCl of the pre-diluted samples using colorimetric reaction of nitrate with 2,6-dimethylphenole to 4-nitro-2,6-dimethylphenole according to DIN 38405-D9-2.

Sulfate SO_4^{2-}(in mg/L) was quantified by precipitation from acetic acid solution with barium chloride crystals as barium sulfate according to DIN 38405-D5. The intensity of the turbidity of the final suspension was measured at 420 nm using a turbidimeter.

The COD (in mg O_2/L) was determined according to DIN 38409-H41. Abundant potassium dichromate was titrated with iron(II) ammonium sulfate and ferroin as an indicator until the color changed from blue-green to red-brown.

Heavy metals (Cd, Cr, Ni, Pb, Zn) were analyzed after acidic digestion according to APHA (Clesceri et al. 1998) and were analyzed by a Phoenix-986AAS atomic absorption spectrometer. Absorbance levels were converted into concentrations in ppm by calibrations for each compound.

3. Result and discussion

3.1 Water samples

3.1.1 pH value

The pH analysis at all sampling points revealed pH values between 6.84 and 7.53 (test series 1) and between 6.83 and 7.48 (test series 2), which were within the WHO guidelines (WHO 1996). Along the flow route of the Tigris River, pH decreased to 6.8-6.9 within the city passage compared to unpolluted conditions (test site 1; 7.48-7.53) due to the discharge of organic pollutant loads and biodegradation of these contaminants (Al-Saffawi 2018) effecting partial acidification due to oxygen-limiting conditions. Due to a combination of higher water temperature and thus lower oxygen solubility, higher biodegradation activity, and lower water flow velocity due to low river levels during dry months, oxygen limitation is more prominent in summer months. However, the drop did not exceed 0.69 pH units because the carbonate hardness of the river water resulted in a high buffering capacity. If this buffering capacity was not present, a significant fluctuation in pH would be expected with implications for aquatic life (Ali et al. 2019, Kevat et al. 2016).

3.1.2 Electric conductivity, total dissolved solids and salinity

Electrical conductivity of surface waters is highly regional and can range from 50 to 1500 µS/cm (EPA 2012). Studies of US inland fresh waters indicate that streams supporting good mixed fisheries have a range between 150 and 500 µS/cm (EPA 2012). Conductivity outside this range may indicate non-suitable water for certain species of fishes or bugs. High conductivity up to 10,000 µS/cm in inland surface water strongly indicates water contamination by industry or crises (ENR). According to the WHO, a mid-range conductivity (200 to 1000 µS/cm) is the regular level for most major rivers (ENR), but suggests a limit of 400 µS/cm for drinking water (Meride and Ayenew 2016). Measurements in the Tigris River were within WHO target values for rivers in both test series. As these values were also within the range of electrical conductivities of North American streams, this parameter is not an exclusion criterion for the use of Tigris water. However, it can be seen over the flow length that the conductivity increases over all 10 measuring points from originally 405 µS/cm (test series 1) and 258.3 µS/cm (test series 2) as mean value to 750 µS/cm (test series 1) and 593 µS/cm (test series 2). Thus, there is a discharge of saline wastewater or contaminated surface runoff through the urban area. Also, infiltration of saline groundwater cannot be excluded, since hydrogeologically the Lazzaga area is part of the Tigris hydrogelogical basin in which water flow converges towards the River Tigris (Jassim S. Z. et al. 1999, Alsam

et al. 1990, Araim 1991). Correspondingly, the detected TDS curve correlates with the electrical conductivity, which also increases continuously between the measuring points S1 to S6 (series 1: 405 µS/cm to 750 µS/cm; series 2: 258 µS/cm to 521 µS/cm; mean values in each case), drops by approx. 10 % to S7 in both series and then remains nearly unchanged. The TDS concentrations remain below the WHO specifications, whereby high TDS values are not fundamentally harmful to humans. However, the consumption of water with high TDS values can have an increased negative effect on people with kidney and heart diseases in particular. In general, this water may also increase laxative but also constipating effect (Meride and Ayenew 2016, Sasikaran et al. 2012).

The same effect was observed for the salt content, which increased from 0.39% (series 1) or 0.31% (series 2) at reference point S1 to measuring point S5 or S7 to a maximum of 1.0% (series 1) or 0.66% (series 2) and dropped by 25-30 % to S10. The WHO limit value is just met at S7 during wintertime. Comparing both series an inconsistent picture is drawn for these three parameters. Due to the lack of precipitation during the summer period and thus lower levels in the Tigris, an increase in electrical conductivity, TDS value and salinity due to anthropogenic discharges would be expected. However, direct comparison of Tables 2 and 3 shows that electrical conductivity outside the urban area remained almost unchanged, while within the urban area remained around 30 % lower than during wintertime (series 1).

In case of the TDS values, there was an increase by a factor of 1.85 in both series over the city passage, with summer levels also being around 35-40% lower. The situation is similar with the salinity, which is also about 30% lower in summer than in wintertime, but rises by 120 % (winter) or 25% (summer) over the city passage. It temporarily increases by a factor of 2-3 within the urban area. The most likely explanation for this scenario is that due to the lack of precipitation in summer and thus the lack of surface runoff, a significantly lower pollutant load is flushed into the Tigris. In addition to anthropogenic discharges, the increase in contaminants in the urban area is due to the infiltration of contaminated groundwater into the Tigris (Jassim S. Z. et al. 1999, Alsam et al. 1990, Araim 1991).

3.1.3 Chemical oxygen demand

The thesis of increased infiltration of contaminated groundwater can be confirmed using COD data. In both series, an increase in the background COD (about 25-30 mg O_2/L) by a factor of 2-3 to 89 mg O_2/L (series 1) and 58 mg O_2/L (series 2), respectively, occurred across the urban area. Within the city, even higher COD values of 98.4 mg O_2/L (S5, Series 1) and 74.33 mg O_2/L (S5, Series 2) were temporarily achieved, detectable at nearly identical levels at S10 during the winter period, while a decrease of about 35% due to biodegradation processes was observed during the summer period. Increased direct or indirect discharge of sanitary wastewater and thus increased organic loads into the river due to war-related damages to the sewage system were previously described (Jankeer and Mustafa 2017, Mustafa 2002).

3.1.4 Nitrate, phosphate, and sulfate

There are a variety of potential emission sources for nitrate, phosphate, and sulfate. For example, these anions can be released during the exploration of minerals, agricultural use of fertilizer, industrial effluents, general leakages in the destroyed municipal sewage network, but also through direct use of weapons and ammunition. In the latter case, nitrate is directly used in munitions as ammonium nitrate.

Conflict-related phosphate emissions in the urban area of Mosul are particularly known from the use of phosphorus bombs during the liberation in the summer of 2017 (Aljazeera 2016). However, this type of bomb was also used by U.S. forces during the second Gulf War (Reuters et al. 2005). Sulfate emissions can result from the combustion of antimony sulphides when munitions are fired (Mariussen et al. 2021, Jovanovic et al. 2018). However, in the case of Mosul, the burning of the Mishraq industrial complex (SO_2, SO_3) (UNEP and OCHA 2016, BBC 2016) 45 km south of Mosul in October 2016 in particular, where the resulting gases entered the river through wet deposition into the soil and surface drainage (UNEP 2018), and the sulfur springs extending from Mosul in the north to Qayara in the south for 60 km along the Tigris River, where the River Tigris is also a zone of discharge (Jassim S. Z. et al. 1999), played a substantial role. Accordingly, the analyses northwestern measuring point, showed in some cases significant exceedances of the WHO limits. Thus, phosphate concentrations increase by a factor of 1.8-1.9 through the urban passage in both series, regardless of the season. In some cases, detected phosphate levels of 0.81 mg P/L (series 2) and 1.20 mg P/L (series 1) exceeded WHO limit by a factor of 2-3 within the urban region.

Sulfate levels exceeded limits from S2 to S10 in R1 series, and from S5 to S10 in R2 series, at the highest value of 495 mg/l (S6 in R1). Again, a key aspect of lower sulfate levels, but also other anion levels, was the lack of surface drainage in combination with groundwater infiltration to Tigris. However, biodegradation represents a second, possibly even dominant factor, where both sulfate and nitrate may be used as electron acceptors instead of oxygen. This aspect could be easily verified due to the severe odor emissions of H_2S gas near the valleys. Direct drainage into the river was described previously (Al-Saffawi 2018, Al-Sarraj 2020). In addition to above factors, the detected sulfate levels were further increased by dissolution of gypsum rock due to impoundment of Tigris water in Mosul Lake (Fadhel 2020).

In the case of nitrate, the course over the river flow showed only a small increase at S3, which is mainly due to agriculture upstream, but remains at an almost constant level over the course of the river. As microbiological H_2S formation occurred, and thus inevitably

microbiological nitrate respiration as a sink of nitrate, a continuous discharge of nitrate over the urban area took place. The discharge was caused by defects in the sewage system and infiltration processes as consequence. Nitrate concentration was clearly below the WHO limit value at all locations, irrespective of the season.

3.1.5 Heavy metals

Due to international contracts prohibiting the use of expanding bullets, as well as the higher cost of hollow point ammunition, full metal jacket bullets are the most used cartridges by soldiers as well as militias (Deese 2021). They consist of a soft core, usually lead, and a harder jacket metal alloy such as gilding metal (CuZn), CuNi, or steel encasing the core (Mariussen et al. 2021). The purpose of this design is to provide improved trajectory as well as greater penetration of the projectiles into soft tissue (Coget et al. 2021, Deese 2021). Supplementary galvanic cadmium coatings are applied to improve some properties like corrosion resistance, improved tribological properties, and chemical stability (Jovanovic et al. 2018). Chromatic layers are applied as a supplementary passivation (Jovanovic et al. 2018). Therefore, the use of weapons in conflict zones leads to an increase in heavy metal concentrations.

Concentrations of Cd, Pb, Cr, and Ni exceeded WHO limits even before entering the city (S1), while increasing further especially through the Old Town passage as a direct conflict zone (S5 - S7) in both series. With maximum concentrations (in ppm) of 0.47 Cd, 0.89 Pb, 0.39 Cr, and 0.98 Ni (series 1) and 0.40 Cd, 0.63 Pb, 0.32 Cr, and 0.73 Ni (series 2), the WHO limits were exceeded here in part by a factor of 100. For Zn, a comparable concentration pattern occurred over the urban passage, but with maximum values of 2.03 ppm (Series 1) and 1.52 ppm (Series 2), the limits were not exceeded.

3.1.6 Comparison of water samples in conflict zone vs. urban zone

The pollution of the Tigris River can be divided into a total of four zones over its flow length in the urban area. Zone 1 (S1) as a reference before entering the densely populated urban area illustrates the background pollution of the river. Zone 2 (S2-S4) essentially represents contamination from agricultural drainage. Since the zone is otherwise characterized by recreation areas, the influence of municipal drains is still low here. Between sampling points S4 and S5, the Al-Khosr valley drains into the Tigris, which carries a heavy pollutant load on its flow route from the northeastern valleys through densely populated areas (Al-Safawi 2007) and therefore leads to a strong increase in the parameters E.C., TDS, salinity, COD, nitrate, phosphate, and sulfate at S5.

The military operations to liberate Mosul focused on the Old City of Mosul, where sampling points S5-S7 (Zone 3) are located. Therefore, within this zone, an increase in heavy metal concentrations is expected and has been observed as an indicator. Destruction of the drainage infrastructure also leads to a further increase of the above parameters. Zone 4, represented by the S8-S10, again represents an area with strong agricultural use and residential development outside the direct conflict zone, so that the discharge situation here is limited to agricultural and municipal parameters and the heavy metals decrease in concentration due to (bio)chemical processes such as adsorption, precipitation, uptake in tissues as well as dilution effects.

The differences in concentration between summer and winter periods caused by the lack of surface drains as well as existing groundwater infiltration have already been pointed out. In addition, the role of entrained sediments should be mentioned, which will be further discussed in section 3.2.

3.1.7 Statistical analysis

An adequate approach for statistical analysis of deviations in analytical data and their significance is the t-test. The results for E.C, TDS, COD, nitrate, phosphate, sulfate, Cd, Pb, Zn, Cr, as well as Ni showed levels below the critical value ($P < 0.05$), which is why they are considered as statistically significant differences. Thus, based on the null hypothesis that the sample mean, and population mean are statistically different at the 0.05 significance level due to the time lag between the two analytical series, a significant statistical difference results between these two series ($P<0.05$, see Table 4).

The percentage difference of heavy metal analyses between the two series at all sites showed that the values of Cd, Pb, Zn, Cr and Ni in S1, S2, S3, S8, S9, S10 were higher than the values in S4, S5, S6, S7, which is due to the fact that the pollutants at these sites are discharged throughout the year via the drains from the valleys and are not only discharged via rainwater. Rather, surface drains from rainwater additionally enter the Tigris in addition to existing drains, which significantly increases the percent of pollution in the river at these sites. The lowest percent differences between both runs were seen at S5 and S6 for Cd (12.5 % vs. 20.4 %), Pb (38.9 % vs. 22.6 %), Ni (24.9 % vs. 23.4 %), Zn (24.9 % vs. 28.2 %), and Cr (22.0 % vs. 12.6 %) as shown in Table 5 and Figure 3. Differences at other locations were higher in this case, since the main pollution source of the Tigris River is the winter rainwater drain, therefore the difference in heavy metal concentrations between both series is even more evident. The highest values were obtained for Cd at S1 (83.3%) > Pb at S10 (69.7%) > Cr at S3 (51.4%) > Ni at S3 (50.0%) > Zn at S2 (45.2%) as shown in Table 5.

Figure 4 shows that the ranking in the percent differences in site contaminants between upstream S1 and downstream S10 in both runs was as follows: Cd>Pb>Cr>Ni>Zn> COD>TDS>$SO_4^{2-}>PO_4^{3-}>NO_3^-$>E.C>pH. Except for pH, the percentage increase of all parameters between upstream and downstream was higher in series 1 (winter, spring) than in series 2 (summer, fall). The pH was lower in series 1 than in series 2, because the increased biodegradation of organics in summer combined with oxygen limitation led to an enhanced decrease in pH.

3.2 Sediment samples

3.2.1 Chemical parameters and heavy metals

Sediments are an important sink for pollutants, but they can also lead to contamination of the water body through desorptive processes. In particular, if the sediment is primarily of mineral nature, ions (e.g. heavy metals) may be adsorbed (Miranda et al. 2022). However, interaction strength of heavy metals and geochemical fractions of sediments strongly depends on exchangeable, reducible, oxidizable, and residual fractions of the sediment (Buyang et al. 2019, Keshavarzifard et al. 2019). The exchangeable and, therefore, weakly-bound geochemical sediment fractions include fixation of heavy metals to the solid surface by weak electrostatic interactions (Brady et al. 2014, Jayarathne et al. 2019). Hence, changes in environmental parameters like pH, E.C., or salinity can promote desorption of heavy metals by breakdown of these weak electrostatic interactions (Chon et al. 2012, Keshavarzifard et al. 2019, Xia et al. 2020). If the sediment is primarily of organic nature, organic contaminants (e.g., pharmaceuticals, flame retardants) may be adsorbed (Dobslaw et al. 2021, Xu 2021). In addition to the complexity of the sediment composition, the interpretation of occurring distribution equilibria between sediment phase and liquid phase is particularly complicated by the downstream transport of sediment or small-particle and thus suspended silt fractions. Hence, it is to be expected that even outside the direct conflict zone or the urban area increased pollutant concentrations may occur in the water phase as well as in the river sediment due to sediment transport and subsequent desorption.

In fact, the sediment analysis confirmed continuous increase of heavy metal concentrations over the flow distance, with the highest concentrations occurring at the southern urban border (S10). A similar result was already presented by Al-Sarraj et al (Al-Sarraj et al. 2019). With detected heavy metal concentrations as mean of 6.23 ppm Cd, 45.33 ppm Pb, 21.33 ppm Zn, 81.33 ppm Cr, 64.00 ppm Ni (Series 1) and 9,200 ppm Cd, 62,800 ppm Pb, 30,000 ppm Zn, 123,600 ppm Cr, 79,000 ppm Ni (Series 2), respectively, WHO limits were permanently exceeded at all locations except Zn. In the case of S10, the limit values for Cd, Pb, Cr and Ni were exceeded by a factor of 3.1, 1.3, 3.3 and 3.2 (series 1) and by a factor of 4.6, 1.8, 4.9 and 4.0 (series 2). For zinc, the limits were met despite an increase in concentration over the urban passage (see Tables 6 and 7). However, due to the interaction between dissolved heavy metals in the water phase and sorbed heavy metals at the solid phase, severe local variations in concentration, in contrast to the water phase, were not detectable.

In the case of the parameters pH, E.C., and salinity, no significant change occurred for the pH value during the winter series. During the summer series, however, a moderate decrease of the pH value by 0.77 units in absolute terms was observed over the urban area. This decrease is related to the increased degradation of organic pollutants induced by higher temperatures with partial oxidation of the components. The pH drop turns out to be comparatively small, since both the morphological sediment composition and the existing lime-carbonic acid equilibrium in the water phase as well as the sediment phase lead to a pH buffering (Zarraq 2012). This pH buffering prevents enhanced pH-induced mobilization of heavy metal ions. This effect has already been described for Cr, Ni, and Zn (Miranda et al. 2022). The mobility of heavy metals is further influenced by E.C. as well as salinity, with enhanced mobilization at increasing E.C. and decreasing salinity, respectively. Both parameters increased by a factor of 1.6 - 2.4 over the urban passage, and just exceeded the WHO limits by a factor of 6.4 for E.C. and by a factor of 3.3 for salinity in the summer measurement, while in the winter period only salinity exceeded the WHO limit by a factor of 1.8.

3.2.2 Comparison of sediment samples in conflict zone vs. urban zone

The direct comparison of these parameters between both series showed, with exception of the pH value (see above) and the E.C. (almost unchanged), a significant increase of 1.6-1.8 (salinity), 1.2-1.5 (Cd), 1.2-1.4 (Pb), 1.4 (Zn), 1.2-1.5 (Cr) as well as 1.1-1.2 (Ni) in the summer period (series 2, see tables 6 and 7). This increase during dry weather is probably related to a combination of low flow velocity of the Tigris during the dry weather phase and thus also an increased deposition of small particles, which either infiltrated directly into the Tigris from the conflict zones or were lifted up by wind and settled down above the water surface, as well as an increased particle-bound heavy metal input during the precipitation phase in winter and spring (series 1, see Figure 8). These particles, some even forming conglomerates, are slowly transported downstream as sediment mixtures, resulting in the high concentrations detected throughout the entire sampling stretch of the river (see Figure 5).

3.2.3 Statistical analysis

The statistical T-test analysis shows that the T-test values for salinity, Cd, Pb, Zn and Cr were below the critical value of P <0.05. This means that there is a significant statistical difference between series 1 and series 2 (P<0.05). Conversely, the numerical values of Ni and E.C. were greater than 0.05 (P>0.05), which means that there is no significant statistical difference between series 1 and series 2 for these two parameters. This indicates different pollution sources or input sources of nickel to the sediments (see Table 9).

The percent difference in sediment analysis data between the locations of both series indicated that there is no regular variation between sites, as the rates of variation between both series were different in all sites depending on the specific element. This is due to differences in the river´s flow rate and the movement of sediments in the river, as well as the different sources of pollution. The highest percent difference in heavy metals was as follows: Cr in S1 (36.8 %) > Cd in S10 (32.2 %) > Pb in S2 (26.6 %) > Zn in S7 (23.6 %) > Ni in S1 (23 %) (see Table 8).

There was a general increase in contaminant concentrations between upstream S1 and downstream S10 in both test series, with the highest percentage increase as follows: Cr> Cd >Pb>Zn > E.C > salinity. In the case of pH, an enhanced decrease was observed during summer tests due to present biodegradation.

4. Conclusions

The impact of the war was evident according to high contamination levels in water and sediment samples from the Tigris River within the urban area of Mosul. With exception of Zn, heavy metal loads exceeded WHO limits seasonally independent both in water and sediment. Further exceedances occurred in the water samples for conductivity, phosphate, sulfate, and in individual samples for salinity and COD, regardless of season and location. Salinity was exceeded almost consistently in sediments, while in contrast, pH, TDS, salinity, COD, nitrate and Zn were (almost) within limits for all water samples and pH, E.C. and Zn met limits for all sediment samples. The very high loads of heavy metals, but also the exceedances for sulfate, indicated direct effects of war (amunition, ignition of sulfur fields). The increased loads of phosphate, but also of COD and TDS within the urban area indicated indirect war effects in form of destroyed wastewater infrastructure. The direct impact of war within the old city of Mosul as a former conflict zone (S5, S6, S7) is particularly evident in the form of the sudden increase in pollution from S4 to S5. Already between S3 and S4 a clear increase in load occurred due to discharge of the Khosr river. Highest load in sediments occurred at S10, which can be attributed to the transport and deposition of polluted sediment.

Seasonal comparison showed a decrease in pollutant concentrations in the water samples during the dry season (summer/fall) compared to the wet season (winter/spring), while the pollutant concentration in the sediments increased in contrast during the summer. This effect is caused in the water body by the lack of infiltration of highly contaminated surface drains into the Tigris River and simultaneous existing infiltration of lower contaminated groundwater during the dry season. Pollutant increase in sediments over the flow distance may be caused by deposition of wind-borne, highly contaminated particles and their deposition in the sediment, or by low water flow supporting enhanced deposition of particles transported during the dry season.

Apart from pH, the T-test analysis for both series indicated a significant statistical difference between both series for all other parameters in the water samples (P <0.05). For the sediment samples, corresponding significant statistical differences were identified for salinity, Cd, Pb, Zn, as well as Cr. Since the percent difference in water samples at S4-S7 is smaller than upstream and downstream, the contaminants are not only introduced via rainwater, but also via year-round infiltration of highly polluted wastewater from the surrounding valleys draining into the river or suburban areas.

Declarations

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Ethics declarations

Ethics approval

Not applicable.

Consent to participate

All authors reviewed and approved the final manuscript.

Consent for publication

All authors approved this article for publication.

Competing Interests

The authors have no relevant financial or non-financial interests to disclose.

Author Contributions

Zena Fakhri Altahaan: Sampling, material preparation, data collection and analysis, literature research.

Daniel Dobslaw: Study conception, sampling plan, literature screening, writing.

Authorship Principles

All authors agree with the content of the manuscript and its submission in the present form. The University of Stuttgart as responsible organization showed consent according to the present work.

Availability of data and materials

Associated analytical data can be provided on request.

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Tables

Table 1: Coordinates and description of all study sites on the Tigris river in Mosul

Site No.	Location names	Latitude N	Longitude E	Describtion
S1	Al-Kuba	36,398501“N	43,074622“E	Residential Area
S2	Alrashidia	36,396060“N	43,114278“E	Agricultural Area
S3	Third bridge	36,363435“N	43,114976“E	Recreation Area
S4	Fifth bridge	36,354850“N	43,12616“E	Recreation Area
S5	Old bridge	36,345667“N	43,136953“E	Commercial +Residential
S6	Aljumhoria bridge	36,340904“N	43,145025“E	Residential Area
S7	Fourth bridge	36,332283“N	43,152415“E	Residential Area
S8	Jarimjah	36,298735“N	43,176358“E	Agricultural Area
S9	Albosaef	36,277681“N	43,163921“E	Agricultural Area
S10	Hammam al-Aleel	36,160292“N	43,263505“E	Residential Area

Table 2: Mean and standard deviation values for the concentration of pollutants and heavy metals for all study sites in the river water (series 1) WHO limits presented as reference (WHO 1996).

S1 Water		PH	E.c µS/cm	TDS mg/l	Sal. %	COD mg/l	PO4 mg/l	NO3 mg/l	SO4 mg/l	Cd ppm	Pb ppm	Zn ppm	Cr Ppm	Ni ppm
WHO std. limits		6.5-8.5	1400	1000	1%	100	0.4	50	250	0.005	0.01	5	0.05	0.02
S1	Mean	7.53	444.3	405	0.31	29.00	0.28	2.62	154.33	0.04	0.15	0.79	0.10	0.27
	±Sd	0.31	29.3	21.39	0.09	3.06	0.02	0.62	28.10	0.03	0.04	0.19	0.03	0.02
S2	Mean	7.51	544	488.6	0.59	43.3	0.55	3.00	419.6	0.18	0.48	0.66	0.19	0.28
	±Sd	0.26	12.10	52.32	0.10	5.51	0.11	0.58	6.24	0.12	0.06	0.20	0.06	0.01
S3	Mean	6.84	693	611.3	0.75	44.67	0.72	4.02	409.3	0.28	0.65	0.70	0.28	0.47
	±Sd	0.16	22.11	34.59	0.06	4.51	0.05	0.60	24.21	0.11	0.05	0.04	0.03	0.01
S4	Mean	6.87	702	711.	0.97	96.3	0.88	3.74	458	0.32	0.72	1.42	0.30	0.70
	±Sd	0.09	19.55	34.59	0.04	7.77	0.06	0.10	6.56	0.11	0.03	0.37	0.07	0.01
S5	Mean	6.85	815	734.6	0.86	98.4	1.20	3.44	485	0.45	0.89	2.03	0.32	0.98
	±Sd	0.22	17.62	48.17	0.07	8.08	0.08	0.06	32.19	0.08	0.03	0.43	0.07	0.02
S6	Mean	6.86	884	787	0.90	95.2	0.91	2.82	495	0.47	0.82	1.81	0.37	0.94
	±Sd	0.21	39.04	63.52	0.06	5.29	0.11	0.45	26.85	0.08	0.02	0.29	0.04	0.08
S7	Mean	6.94	829	717	1.0	90.6	0.94	4.03	434	0.43	0.80	1.60	0.39	0.92
	±Sd	0.15	53.59	45.65	0.07	9.61	0.06	0.50	130.66	0.07	0.03	0.24	0.03	0.07
S8	Mean	7.04	711	737	0.78	82.3	0.75	2.86	444.6	0.41	0.85	1.38	0.32	0.90
	±Sd	0.22	26.69	44.84	0.03	6.56	0.11	0.73	26.91	0.06	0.02	0.09	0.02	0.08
S9	Mean	7.20	660	748.3	0.69	92	0.59	3.04	450.3	0.38	0.82	1.52	0.33	0.85
	±Sd	0.23	36.01	71.15	0.09	5.03	0.03	0.45	62.66	0.04	0.04	0.17	0.06	0.02
S10	Mean	7.13	616.00	750	0.68	89	0.52	3.07	445.33	0.37	0.74	1.81	0.31	0.81
	±Sd	0.25	28.21	28.75	0.04	5.00	0.06	0.78	51.59	0.08	0.76	1.51	0.31	0.81

Table 3: Mean and standard deviation values for the concentration of pollutants and heavy metals for all study sites in the river water (series 2) WHO limits presented as reference (WHO 1996).

S2 Water		PH	E.c µS/cm	TDS mg/l	Sal. %	COD mg/l	PO4 mg/l	NO3 mg/l	SO4 mg/l	Cd ppm	Pb ppm	Zn ppm	Cr ppm	Ni ppm
WHO std. limits		6.5-8.5	1000	1000	1%	100	0.4	50	250	0.005	0.01	5	0.05	0.02
S1	Mean	7.48	488.33	258.33	0.39	24.67	0.32	1.82	121.67	0.03	0.06	0.63	0.08	0.21
	±Sd	0.306	29.297	21.385	0.090	3.055	0.020	0.616	28.095	0.031	0.045	0.188	0.032	0.023
S2	Mean	7.30	524.33	281.33	0.34	28.67	0.31	2.00	191.00	0.08	0.27	0.56	0.12	0.13
	±Sd	0.265	12.097	52.320	0.104	5.508	0.111	0.578	6.245	0.118	0.062	0.204	0.057	0.011
S3	Mean	7.16	558.00	347.67	0.51	32.67	0.38	2.02	204.33	0.11	0.46	0.68	0.14	0.23
	±Sd	0.158	22.113	34.588	0.064	4.509	0.051	0.602	24.214	0.110	0.052	0.042	0.029	0.011
S4	Mean	7.09	498.33	367.33	0.47	51.67	0.51	1.45	211.00	0.22	0.48	0.84	0.24	0.48
	±Sd	0.090	19.553	34.588	0.038	7.767	0.064	0.100	6.557	0.11	0.034	0.372	0.066	0.013
S5	Mean	6.98	593.67	435.67	0.68	74.33	0.70	1.73	315.33	0.40	0.54	1.52	0.30	0.73
	±Sd	0.220	17.616	48.170	0.068	8.083	0.080	0.056	32.192	0.080	0.031	0.428	0.065	0.018
S6	Mean	6.81	596.00	520.67	0.52	63.00	0.74	1.82	289.00	0.38	0.63	1.30	0.32	0.72
	±Sd	0.209	39.038	63.516	0.061	5.292	0.109	0.452	26.851	0.080	0.022	0.288	0.040	0.082
S7	Mean	6.99	471.33	484.67	0.60	65.30	0.81	2.44	434.00	0.34	0.50	1.03	0.26	0.53
	±Sd	0.148	53.594	45.654	0.070	9.609	0.063	0.503	130.664	0.068	0.035	0.242	0.028	0.075
S8	Mean	7.02	412.67	502.67	0.66	59.00	0.56	2.19	293.00	0.31	0.40	1.09	0.20	0.60
	±Sd	0.222	26.690	44.837	0.025	6.557	0.106	0.730	26.907	0.065	0.021	0.094	0.019	0.077
S9	Mean	7.09	583.00	482.6	0.49	50.33	0.59	3.04	318.33	0.23	0.35	1.07	0.21	0.56
	±Sd	0.228	36.014	71.150	0.093	5.033	0.030	0.450	62.660	0.043	0.042	0.167	0.059	0.016
S10	Mean	6.833	593	476.333	0.507	58	0.522	2.071	320.667	0.187	0.231	1.308	0.237	0.456
	±Sd	0.254	28.213	28.746	0.040	5.0	0.055	0.776	51.588	0.080	0.025	0.214	0.032	0.047

Table 4: T-test Value comparing first and second series for all sites in case of river water

Parameters	T-test	Df*	P	Status p<0.05
EC	3.366479	18	0.001719	1
TDS	5.047889	18	4.19E-05	1
Salinity%	2.483954	18	0.011532	1
COD	2.492992	18	0.011318	1
PO4	1.923167	18	0.035212	1
NO3	4.874697	18	6.09E-05	1
SO4	3.614803	18	0.00099	1
Cd	1.891262	18	0.0373972	1
Pb	3.402881	18	0.0015856	1
Zn	2.000151	18	0.0304019	1
Cr	2.291138	18	0.0171182	1
Ni	2.249799	18	0.0186051	1

*Df : degree of Freedom

Table 5: The percentage deviation of concentrations between first and second series in all water sites

Sites	Cd %	Pb %	Zn %	Cr %	Ni %
S1	83.4	58.1	35.5	39.0	22.5
S2	53.7	44.7	45.1	35.8	53
S3	61.5	29.7	28.7	51.4	50
S4	30.9	32.9	41.1	20.3	31.6
S5	12.5	39	25	22.1	24.9
S6	20.4	22.7	28.2	12.6	23.5
S7	21	37.7	35.6	33.1	42.2
S8	25.1	52.9	27.1	36.6	33.1
S9	39.6	56.8	29.7	35.3	34.1
S10	40.7	69.8	31.9	22.9	43.7

Table 6: Average and standard deviation values for pollutant and heavy metal concentrations in sediment samples of all sites studied (series 1)

S1		PH	Ec	Sal.%	Cd	Pb	Zn	Cr	Ni
WHO std. limits		6.5-8.5	1000	1%	2	35	90	25	20
S1	Mean	7.44	710	0.86	2.78	25	12	28.20	27
	±Sd	0.31	50	0.07	0.26	2.33	1.00	6.01	2.00
S2	Mean	7.78	680	0.88	2.87	24.70	10.33	36.00	32.67
	±Sd	0.43	20	0.28	0.32	1.12	1.53	7.94	6.51
S3	Mean	7.57	750	1.34	3.50	29.80	14.67	41.67	32.33
	±Sd	0.29	40	0.17	0.96	0.35	3.51	4.16	7.02
S4	Mean	7.80	800	1.50	4.44	33.70	11.67	40.67	37.00
	±Sd	0.10	40	0.00	0.30	0.30	2.07	8.50	2.65
S5	Mean	7.55	1150	1.17	5.63	35.40	16.50	73.67	49.00
	±Sd	0.19	240	0.12	0.83	1.26	3.97	3.51	6.24
S6	Mean	7.53	1640	1.44	5.48	38.20	18.67	80.00	52.00
	±Sd	0.40	710	0.23	0.30	1.53	3.06	1.00	4.36
S7	Mean	7.64	50	1.34	5.87	38.67	19.33	78.67	57.00
	±Sd	0.49	680	0.13	0.35	3.06	3.51	4.51	6.56
S8	Mean	7.93	20	1.03	6.30	40.00	17.83	77.67	59.67
	±Sd	0.05	750	0.12	0.60	1.00	5.11	6.51	4.16
S9	Mean	7.86	40	1.10	5.47	44.33	20.67	80.33	63.67
	±Sd	0.40	800	0.64	0.21	4.51	6.11	3.06	8.50
S10	Mean	7.17	40	1.82	6.23	45.33	21.33	81.33	64.00
	±Sd	0.32	1150	1.05	0.55	2.50	2.50	7.50	8.14

Table 7: Average and standard deviation values for pollutant and heavy metal concentrations in sediment samples of all sites studied (series 2)

S2		PH	Ec	Sal.%	Cd	Pb	Zn	Cr	Ni
WHO std. limits		6.5 -8.5	1400	1%	2	35	90	25	20
S1	Mean	7.62	611	1.387	3.3	31.3	10.16	32.0	30
	±Sd	0.19	88	0.528	0.6	7.87	0.76	9.5	3.0
S2	Mean	7.79	777	1.617	3.3	33.66	12.6	50.3	37.6
	±Sd	0.39	110	0.458	0.8	4.51	1.87	11.4	5.6
S3	Mean	7.27	807	1.343	4.28	35.6	15.1	50.6	39.3
	±Sd	0.26	90	0.151	0.7	3.7	2.9	10.4	6.8
S4	Mean	7.80	920	1.667	5.8	40	17.0	42.66	46.0
	±Sd	0.08	42	0.204	0.8	2.3	3.8	12.3	5.8
S5	Mean	7.35	1146	1.517	6.9	43.1	20.3	79.0	58.3
	±Sd	0.19	215	0.264	0.78	2.7	3.52	11.1	6.9
S6	Mean	7.28	1105	1.650	7.37	46.33	22.0	89.0	58.3
	±Sd	0.30	297	0.310	0.88	4.40	3.5	5.0	9.22
S7	Mean	7.13	1267	1.377	7.79	47.6	25.33	92.3	64.80
	±Sd	0.41	367.5	0.6194	0.87	4.41	9.83	9.35	0.41
S8	Mean	7.03	1062	1.600	7.76	51.6	21.6	97.0	67.90
	±Sd	0.53	36	0.431	0.46	7.67	4.7	11.7	3.34
S9	Mean	6.97	1365	1.857	7.86	49.6	24.3	96.0	71.2
	±Sd	0.55	143	0.478	1.20	6.06	5.9	13.1	6.43
S10	Mean	6.86	1445	3.292	9.20	62.8	30.	123.60	79.0
	±Sd	0.45	112	0.340	0.86	4.44	2.8	16.1	5.6

Table 8: The percentage deviation of concentrations between first and second series in all sediment sites

Sites	Cd %	Pb %	Zn %	Cr %	Ni %
S1	16.60	20.12	14.2	36.88	23
S2	13.39	26.63	18.42	28.48	15.93
S3	18.29	16.45	3.08	17.76	17.80
S4	23.51	15.75	31.37	32.97	19.57
S5	18.75	17.99	18.85	18.15	16.00
S6	25.76	17.55	15.15	10.11	10.81
S7	24.69	18.88	23.68	14.80	12.04
S8	18.85	22.58	17.69	19.93	12.13
S9	30.49	10.74	15.07	16.32	10.58
S10	32.25	13.38	14.67	21.29	17.95

Table 9: T-test values of first and second series of sediment samples of all sites

Parameter	T-test	Df*	P	Status p<0.05
E.C	-0.03433	18	0.486495	0
Salinity	-2.36597	18	0.014703	1
Cd	-2.02299	18	0.02909	1
Pb	-1.79197	18	0.04498	1
Zn	-1.81791	18	0.04392	1
Cr	-1.61075	18	0.04339	1
Ni	-1.17174	18	0.12829	0

*Df : degree of Freedom

Figures

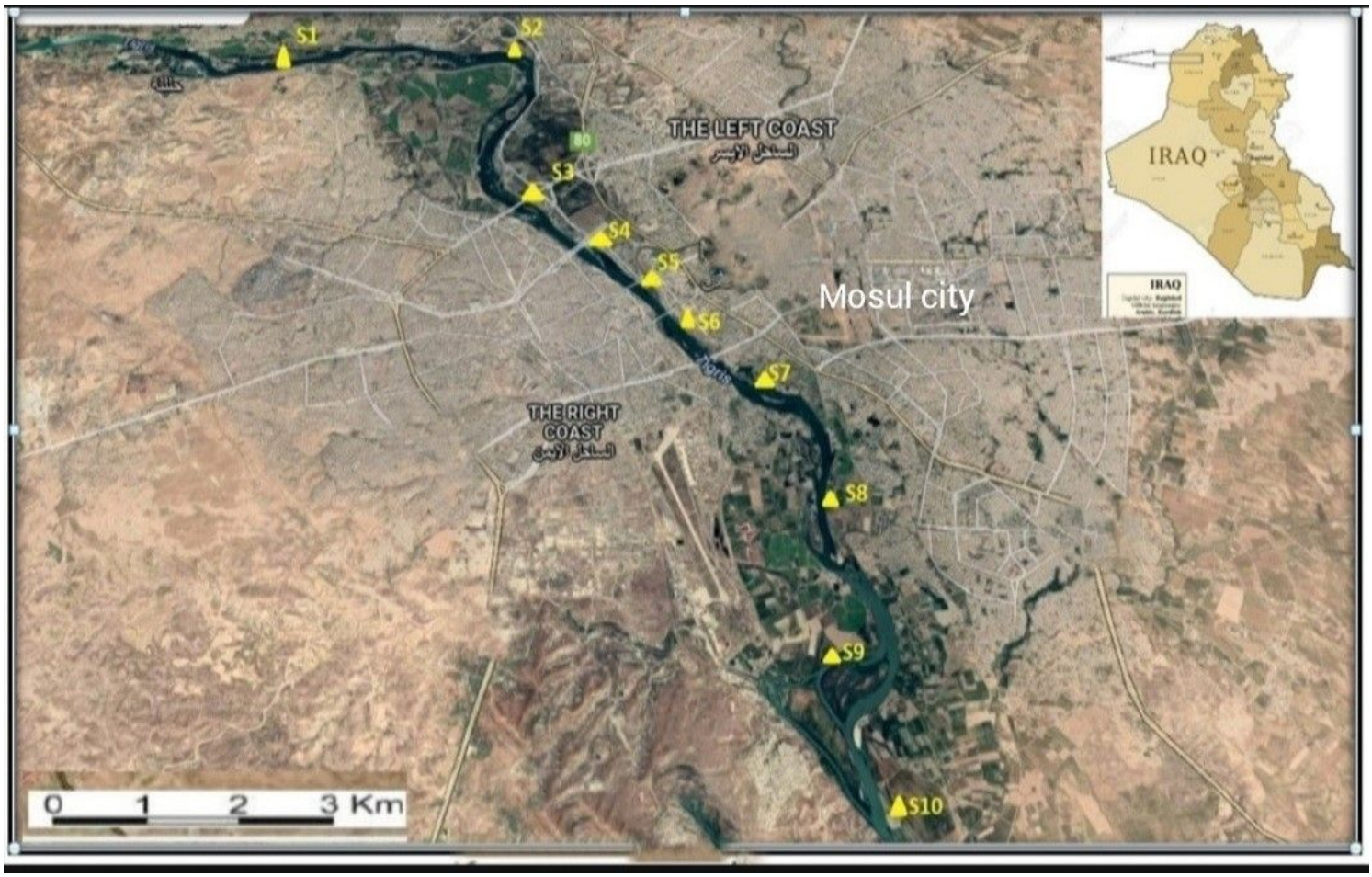

Figure 1

Map of Mosul city with highlighted sampling locations

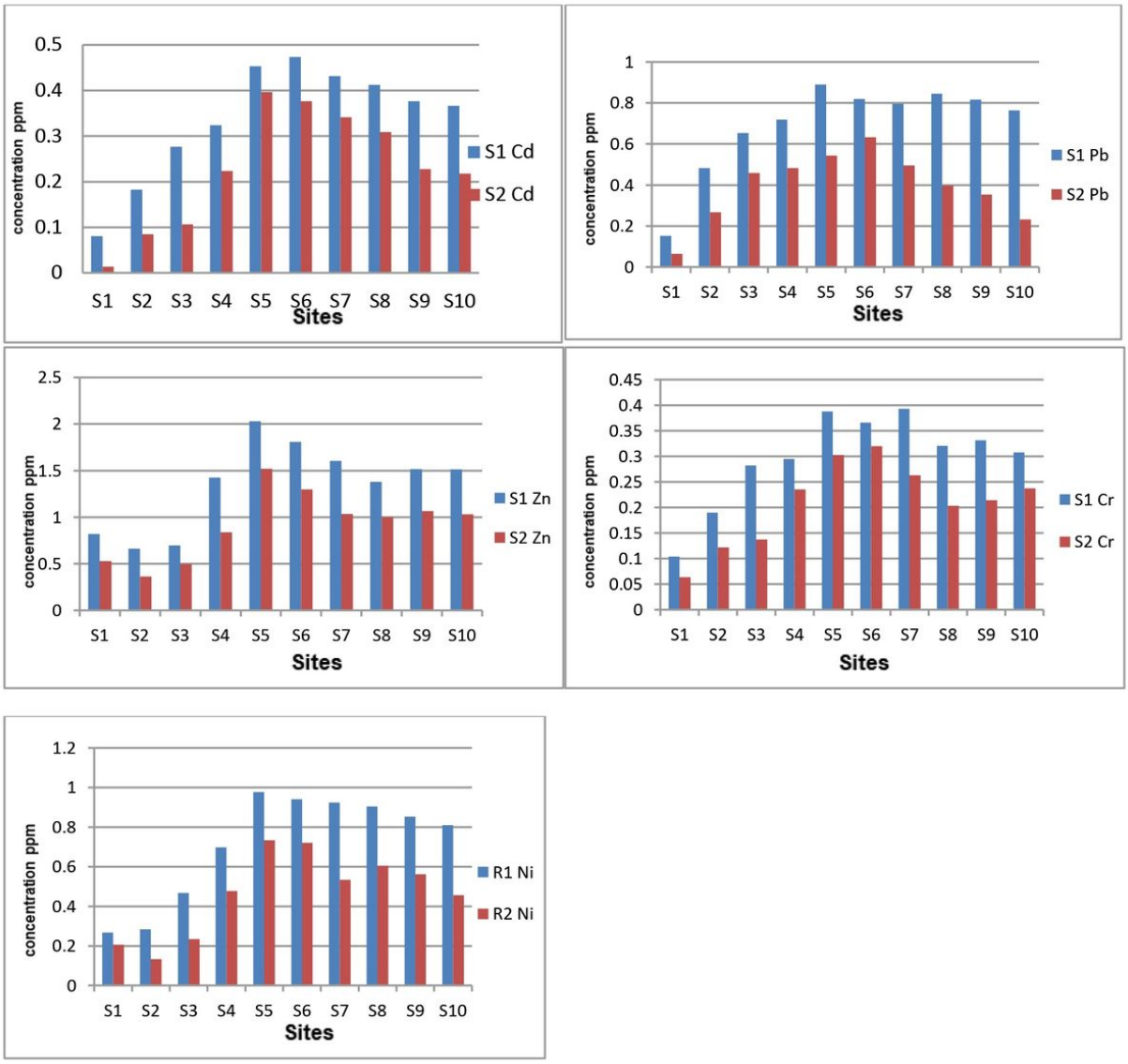

Figure 2

Average values of heavy metal concentrations (in ppm) in river water during first and second series at the water sites

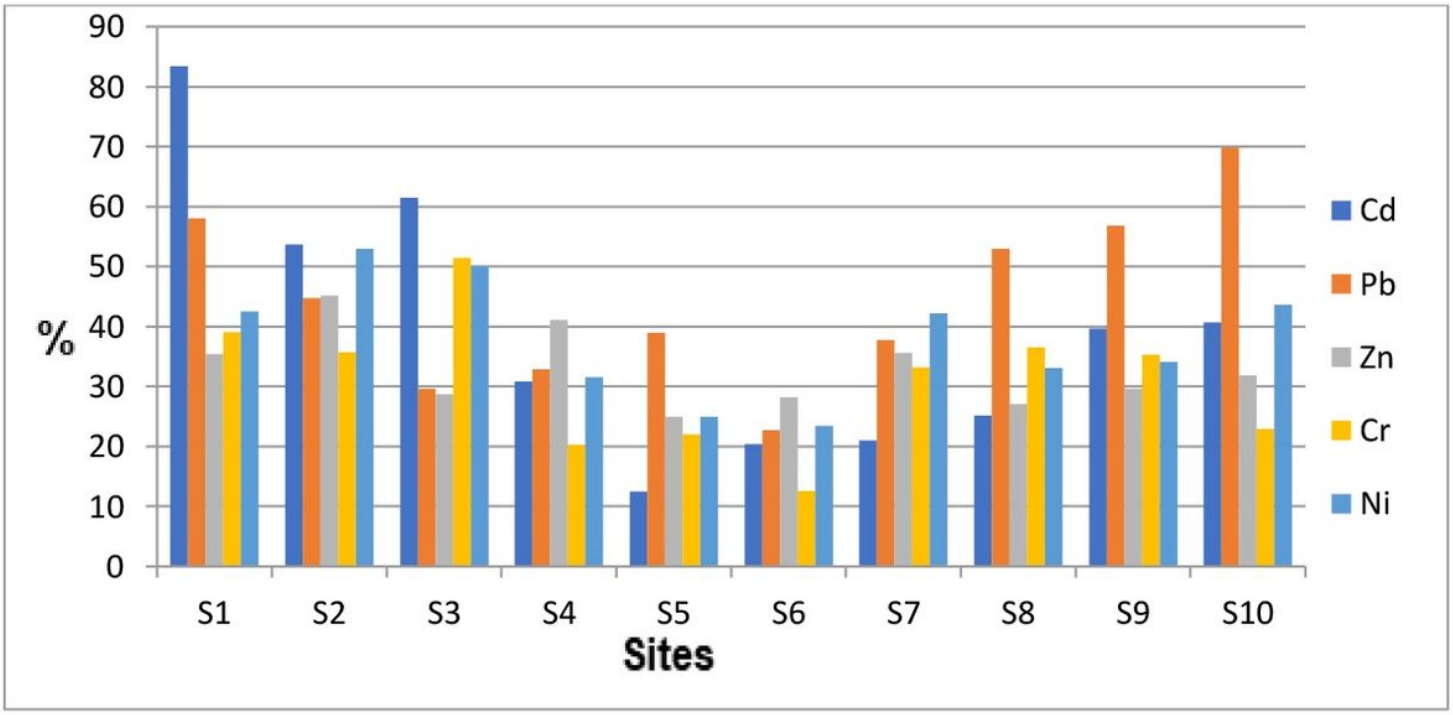

Figure 3

Percentage deviation of concentrations between first and second Series for heavy metals at all sites

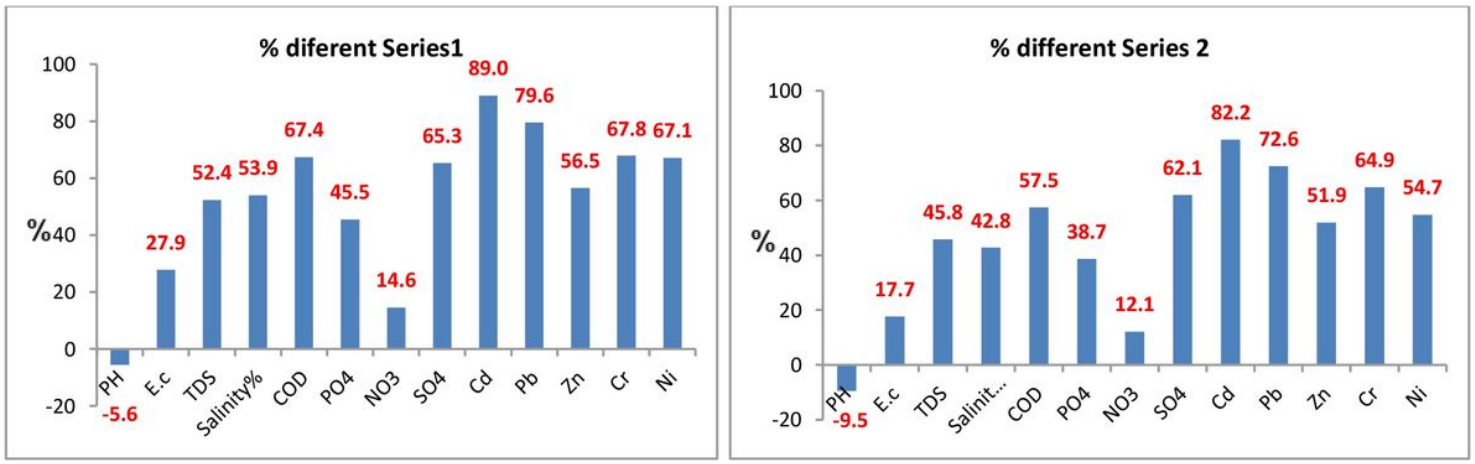

Figure 4

Percentage deviation of concentrations between upstream (S1) and downstream (S10) at both series

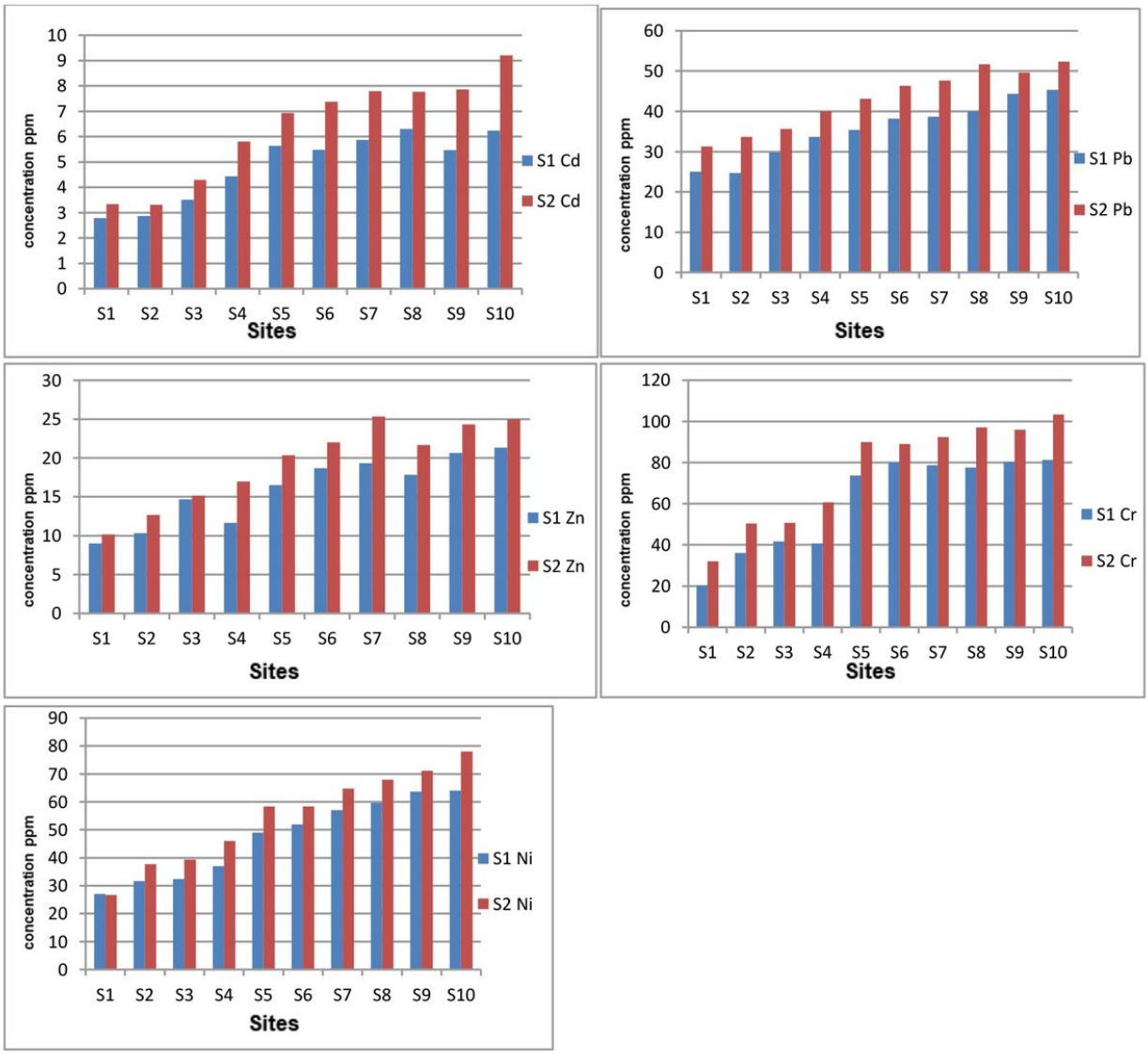

Figure 5

Average values of heavy metal concentrations (in ppm) in the first and second series of sediment samples of relevant sites.

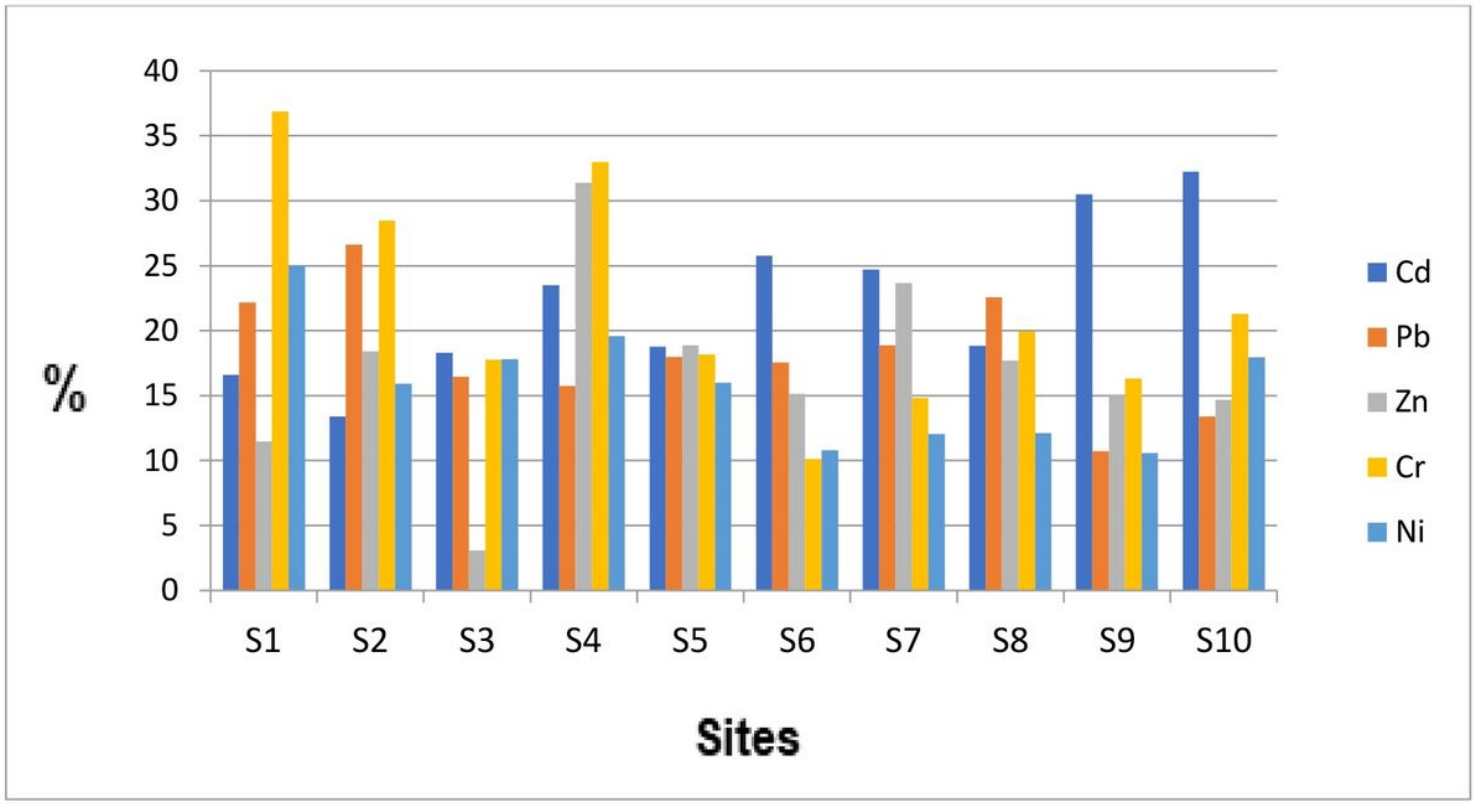

Figure 6

Figure 7: The percentage deviation of heavy metal concentrations between first and second series in sediment samples of all sites

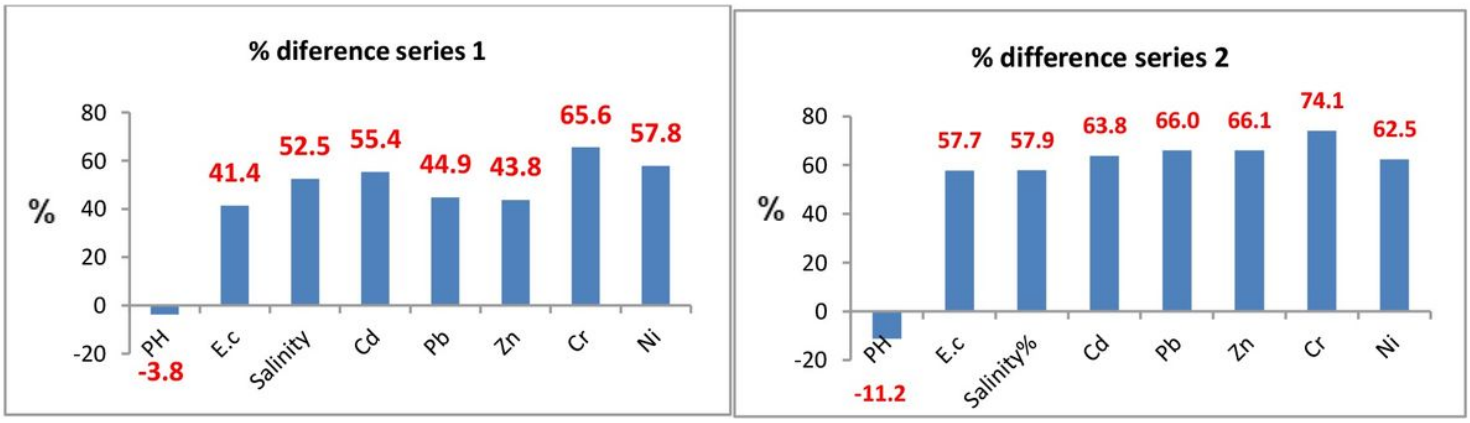

Figure 7

Figure 8: Percentage deviation of contaminant concentrations between upstream (S1) and downstream (S10) in sediment samples of both series