Micro-essential and toxic heavy metals in surface water of Harike wetland - India

Nafiaah Naqash[1], *Sarita* Devi[2], *Rahul* Singh[1,*]

[1]School of Bioengineering & Biosciences, Lovely Professional University, Phagwara, Punjab, India – 144411
[2]GD Goenka University, Sohna, Haryana, India

*Corresponding author: rahulsingh.mlkzoology@gmail.com

Abstract. Wetlands are consequential habitats with diverse flora and fauna. The overutilization of these habitats led to the implementation of conservational strategies which includes Ramsar convention. Harike wetland is the largest wetland in northern India, declared as a Ramsar site. However, the recent industrialization and anthropogenic activities may result in the ecological degradation of this wetland. The wetland is eminently polluted due to industrial discharges, therefore the present study was undertaken to investigate heavy metal status of Harike wetland. The analysis included bio-essential (chromium, manganese, iron, cobalt, nickel, copper, zinc) and non-essential (arsenic, mercury, cadmium, lead) heavy metals. The concentration of heavy metals in the surface water was determined using inductively Coupled Plasma-Mass Spectrometry and the recovered values were compared to the global and national standards. Among all the heavy metals analysed the mean concentrations of iron (437.983 μg/l), mercury (4.011 μg/l) and lead (156.719 μg/l) were recorded to be alarming. The higher concentration of heavy metals may be directly related to the inlet from river Sutlej, as the water is contaminated by various industrial, domestic and agricultural sources. Therefore, continuous monitoring and adequate measures need to be implemented to improve the water quality of internationally recognized wetland.

1 Introduction

Wetlands serve as ecologically beneficial habitats, establishing a crucial connection between land and water. The varied ecosystem within wetlands significantly contributes to both biotic and abiotic factors. These ecosystems play a vital role in providing essential ecological services, such as flood control, groundwater replenishment, biodiversity maintenance, water purification, water conservation, and other critical functions [1]. Wetlands are categorized into natural and manmade types, with manmade wetlands undergoing similar processes but within controlled environments. These exceptionally rich habitats are intended for preservation through various initiatives. The Ramsar Convention represents an international treaty initiated in 1971, signed in Ramsar, Iran. This convention aims to address and mitigate the global loss and degradation of wetlands [2]. India has signed the convention in 1982 with 75 Ramsar sites till date [3]. The wetland ecosystem in India spans a total area of 4,050,536 hectares, with 23,000 hectares designated to natural and manmade wetlands in Punjab [4]. Harike wetland is the largest one in Northern India covering an area of 4100 ha [5]. The wetland came into existence during barrage construction in 1952 [6]. Recognised as a biodiverse wetland, Harike plays a crucial role in supporting a vast array of plant and animal species. However, despite its significant environmental functions, the wetland is under threat due to industrial and sewage discharges. Situated at the confluence of the Beas and Sutlej rivers, the wetland faces degradation as polluted water from these rivers adversely affects its ecological health. Additionally, industrial effluents from the nearby city of Ludhiana further contribute to the pollution of the Sutlej River through Buddha Nullah [7]. The polluted water of river Sutlej finally reaches Harike wetland from where its impact spreads to southern Punjab and Rajasthan.

In recent decades, swift industrialization and urban development have led to significant pollution in aquatic as well as terrestrial systems [8]. Among the various pollutants, heavy metals emerge as a major concern due to their recognized potential for toxicity, bioaccumulation, and persistent presence over extended periods [9], [10], [11], [12]. Metal pollution is a prevalent issue in the environment, stemming from various sources like industrial discharges, mining activities, metal extraction processes, and electronic waste [13], [14], [15]. Elevated concentrations of metals and metalloids have been documented in various fish species and freshwater systems

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globally [16], [17], [18]. Moreover, heavy metals have the capability to adhere to aquatic debris, including plastic particles, leading to secondary exposure [19], [20]. Earlier investigations have examined crucial concentrations of heavy metals in the Harike wetland using an Atomic Absorption Spectrophotometer (AAS) [21], [22]. These reports proposed the continuous monitoring of the wetland so that specific measures could be implemented to conserve it. Heavy metals were categorized into two groups: micro-essential metals, necessary in specific quantities for human metabolism, and non-essential metals, which lacked a known biological role. Micro-essential metals could result in toxicity both at higher concentrations and deficiencies. [23]. Hence, the current study focuses on assessing the concentrations of both biologically essential and non-essential heavy metals in the surface water of Harike wetland. The analysis was conducted using Inductively Coupled Plasma-Mass Spectrometry (ICP-MS), a technique known for its precision [24]. In present work micro-essential elements chromium (Cr), manganese (Mn), iron (Fe), cobalt (Co), nickel (Ni), copper (Cu), zinc (Zn) and the toxic elements arsenic (As), mercury (Hg), cadmium (Cd) and lead (Pb) have been considered for heavy metal analysis. The focus of this study was to gather the baseline information regarding heavy metal concentration in internationally recognized wetland.

2 Methodology

2.1 Study area

Spanning an extensive area of 4100 hectares, the Harike wetland stretches across three districts in Punjab—Kapurthala, Tarn-Taran, and Ferozepur—located at approximately 31.17ºN latitude and 75.2º longitude. Originally designed for water storage and supply for drinking and irrigation purposes to regions in Punjab and Rajasthan, this wetland holds significant ecological importance. Acknowledged as a critical habitat for waterfowl, it gained recognition as a wildlife sanctuary in 1982, designated by the Ministry of Environment and Forests, Government of India. Renowned as both a habitat and breeding ground, Harike wetland hosts a diverse array of migratory and resident bird species, including the Indian Skimmer, Yellow-eyed Pigeon, Sykes's Knightjar, and Rufous-vented Prinia [7]. In 1992 the wetland was declared as bird sanctuary by the State government of Punjab [25]. The wetland is also recognized for the significant role in conserving fish biodiversity which includes about 61 fish species [26]. The selection of sampling sites was meticulously undertaken, identifying key locations adjacent to Gurudwara Nanaksar Sahib (Site 1), Harike Bird Sanctuary (Site 2), and Harike Wildlife Sanctuary (Site 3) as illustrated in Figure 1.

Fig.1 Map of Harike wetland marked with sampling sites

2.2 Water sampling

Surface water samples were systematically collected through grab sampling at these three designated sites within the Harike wetland. Utilizing 1000ml borosilicate glass bottles, water samples were collected from the surface up to a depth of 25cm. To ensure the integrity of the samples, a thorough filtration process was employed, employing Whatman filter paper No. 42, followed by acidification. A protective measure against the precipitation of heavy metals involved the addition of 1 ml of HNO_3 to each sample. Subsequently, the acidified samples were meticulously stored in a refrigerator at 4ºC to preserve their chemical composition until further analysis.

The comprehensive heavy metal analysis of all collected samples was conducted utilizing ICP-MS, a highly precise analytical technique known for its accuracy in detecting and quantifying trace elements. This rigorous sampling and analysis methodology ensures the reliability and accuracy of the data obtained from the study of heavy metal concentrations in the surface water of the Harike wetland.

3 Results and Discussion

The results obtained from ICP-MS recorded different concentrations of heavy metals at three different sampling sites of the wetland. The concentrations of heavy metals in surface water at each site have been presented in Table 1 along with the possible sources of these elements. The basic requirement is the good quality drinking water for the betterment of human physiology. Therefore the concentrations obtained were compared to the prescribed limits by the World Health Organization (WHO) and Bureau of Indian Standards (BIS) as mentioned in Table 2. The permissible limits of selected heavy metals in drinking water have been prescribed just to clarify the tolerance limit of these elements. The over dosage of these metals may result in severe health hazards which have been also mentioned in the Table 2.

Table. 1 Concentration of micro-essential and toxic heavy metals in different sampling sites analyzed through ICP-MS and their common sources

Heavy Metals		Site 1 (ppb)	Site 2 (ppb)	Site 3 (ppb)	Common sources
Micro-essential heavy metals	Cr	2.839	0.092	1.377	Discharge from steel and pulp mills.
	Mn	75.165	50.440	121.382	Metallurgic processes, fossil fuels and fertilizers.
	Fe	193.463	214.700	905.787	Iron and steel industry effluents, iron pipes
	Co	0.886	1.172	2.407	Chemical and electrical industries.
	Ni	10.174	1.695	8.437	Electroplating, steel industry, storage batteries.
	Cu	65.312	15.612	53.997	Mining, storage batteries and fertilizer production.
	Zn	950.283	146.734	253.101	Plastic, steel, printing ink and rubber production.
Toxic heavy metal	As	2.475	5.156	5.775	Runoff from glass and electronic production wastes.
	Cd	5.697	0.813	1.627	Erosion of galvanized pipes, runoff from paint and batteries waste.
	Hg	6.322	3.483	2.229	Discharge from refineries and factories.
	Pb	374.690	33.663	61.806	Paint industries, manufacturing of Pb coated goods.

*Note: the concentrations exceeding the permissible limits have been highlighted

The mean concentration of micro-essential elements followed the order as Cr<Co<Cu<Ni<Mn<Fe<Zn and in case of toxic heavy metals it was recorded as Cd<Hg<As<Pb. Furthermore when compared to the permissible limits it was observed that among micro-essential elements only Fe exceeded the concentration limits at Site 3. In case of toxic heavy metals both Pb and Hg were present in high concentrations at all three sites, whereas Cd concentration

exceeded the permissible limits only at Site 1. Though toxic metal Arsenic was recorded under permissible limits at all three sites of the wetland.

Table. 2 Prescribed limits for metal concentration (ppm) in water by World Health Organization (WHO) and Bureau of Indian Standards (BIS). Potential health effects on concentration above permissible limits.

Heavy Metals		Prescribed Limits WHO (ppm)	Prescribed Limits BIS (ppm)	Negative Health Impacts
Micro-essential heavy	Cr	0.05	-	Allergic dermatitis, carcinogenic, liver and kidney damage.
	Mn	0.4	0.1	Neurological effects
	Fe	0.3	0.3	Hepatic problems, gastric and esophageal ulceration
	Co	0.05	-	Dermatitis, lung and heart effects
	Ni	0.07	0.02	Skin allergies, nasal sinusitis, nasal mucosal injury
	Cu	2	1.5	Neurological complications, hypertension, liver damage
	Zn	3	15	Hematological disorders, human metabolism deterioration
Toxic heavy metals	As	0.01	0.05	Circulatory problem, risk of cancer, skin damage.
	Cd	0.003	0.003	Nausea, vomiting, respiratory difficulties
	Hg	0.001	0.001	Kidney damage, Neurological and gastrointestinal complications.
	Pb	0.01	0.10	Hypertension, tiredness, anemia, behavioral changes, mental retardation

The mean concentrations of each element in three different sites was calculated in order to compare it with the permissible concentrations by the WHO standards (Figure 2). Thereby depicting that the concentration of Cr, Mn, Co, Ni, Cu, Zn, As and Cd were within the permissible limits. Whereas Fe, Pb and Hg concentrations are exceeding the prescribed concentrations for drinking water as established by WHO.

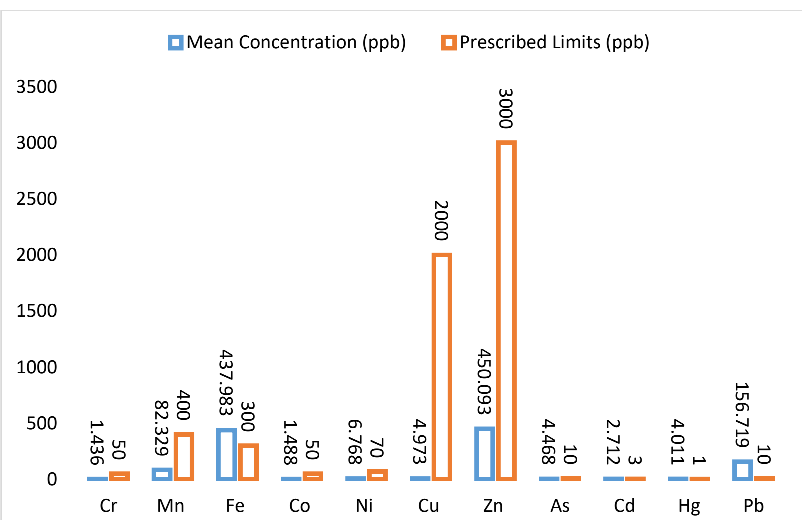

Fig. 2 Chart representing mean of heavy metal concentrations at different sites in comparison with the permissible limits by WHO

Among micro-essential heavy metals Zn was recorded at highest concentration but it was within the range prescribed by WHO. Zinc is regarded as a micro-nutrient in living organisms, therefore its deficiency results in equivalent negative impacts as its increased concentration [23]. Fe was found to be the second dominant micro-essential element in surface water and its concentration was beyond the permissible limits. The exposure of chronic Fe concentrations may result in esophageal and gastric ulceration in humans and animals [27]. Micro-essential heavy metals Cu, Mn, Ni, Co and Cr were found to be within the limits prescribed by WHO. The trace quantities of these metals are biologically essential for certain physiological and biochemical functions in humans and animals. These metals are required for oxidation reduction reactions as they are essential constituents of certain key enzymes [28]. Therefore deficiency of these micro-nutrients may result in adverse effects causing various diseases. Cu, a micronutrient serves as an important cofactor for various stress related enzymes which includes superoxide, peroxidase, catalase, dismutase and cytochrome. However, at higher concentration copper has reported to cause cellular damage in humans [29]. Similarly other essential heavy metals are also required for biological metabolism and their excess exposure leads to the adverse diseases. Toxic heavy metals As, Cd, Hg and Pb have no established biological role and are known to induce multiple organ damage even at lower concentrations. International agency for research on cancer have reported these metals as human and animal carcinogens [30]. In present study mean concentration of toxic metals Pb and Hg as analysed through ICP-MS is beyond the permissible limits. Whereas mean concentrations of Cd and As are below the prescribed limits by WHO. The concentration of Pb above permissible limits causes neurological damage and in case of children it results in mental retardation and blood disorders [31]. Further the poisonous metal, Pb has serious biochemical effects as it interferes with the haemo synthesis which causes haematological disorders [32]. Mercury is also a toxic heavy metal which induces hazardous effects both in animals and humans at concentrations above the tolerance limit. The toxic effects of mercury may lead to neurotoxicity, gastrointestinal toxicity and nephrotoxicity [33]. Overall the heavy metal pollution in Harike wetland may have negative effect on flora and fauna as it is surrounded by wildlife and bird sanctuaries. It has been reported that maximum pollution is brought by the river Sutlej which is contaminated with hazardous chemicals which may further affect the diversity of the wetland adversely [34]. The higher concentration of toxic metals is alarming even for the people living in the vicinity of the wetland. Therefore the present study focuses on the establishment of certain criteria for conserving Harike wetland to impede further deterioration.

4 Comparative assessment with previous metal analysis in Harike

Very few studies have been undertaken to assess the heavy metal status in surface waters of Harike wetland. First analysis included 6 micro-essential (Cr, Mn, Co, Ni, Cu & Zn) and 2 toxic heavy metals (Cd & Pb) where, the concentration of heavy metals exceeding the limits provided by WHO were Cr, Cd & Pb [21]. Another study included 4 micro-essential (Cr, Fe, Cu & Zn) and 2 toxic heavy metals (Hg & Pb), with Cr, Fe, Cu, Hg & Pb exceeding the WHO prescribed limits [22]. Present study determined the concentration of 7 micro-essential (Cr, Mn, Fe, Co, Ni, Cu & Zn) and 4 toxic heavy metals (As, Cd, Hg & Pb) in surface waters of wetland. The results obtained when compared to past studies revealed that the concentration of heavy metals has decreased (Table 3). Thus, reducing the risk posed by the higher concentrations of heavy metals to the numerous diversities present in and surrounding Harike wetland. Although, the decrease in concentrations has been observed in present study however, few metals including Fe, Hg & Pb concentrations are above permissible limits by WHO. Among these Hg & Pb are toxic heavy metals which may accumulate in the food chain. As such these metals may have toxicological effects on the flora and fauna of the wetland. The ecological benefits provided by the wetland may also be affected due to deterioration of wetland. The contaminated water of the wetland needs immediate attention, so that strict management is implemented for the maintenance of water quality. Therefore, continuous monitoring is essentially required to avoid environmental degradation of the wetland.

Table. 3 Present and previous heavy metal analysis in surface waters of Harike wetland

	2015 (ppm) [21]	**2017 (ppm)** [22]	**Present study (ppb converted to ppm)**	**WHO (ppm)**
Cr	0.12	0.121	0.001	0.05
Zn	0.69	2.589	0.450	3
As	-	-	0.004	0.01
Cd	0.01	-	0.002	0.003
Pb	0.53	0.704	0.156	0.01

*Note: the concentrations exceeding the permissible limits have been highlighted.

5 Conclusion

Heavy metal analysis of wetlands is mandatory to investigate the possibility of adverse effects of pollution on most productive habitats. Therefore the present study deals with analyzing surface water of Harike wetland to determine concentration of heavy metals. The increasing trend of micro-essential heavy metal concentrations was recorded as Cr<Co<Cu<Ni<Mn<Fe<Zn and in case of toxic heavy metals it was reported as Cd<Hg<As<Pb using ICP-MS. The exceeded concentrations of iron, mercury, cadmium and lead at one site or the other is clearly indicating heavy metal pollution in the wetland. The heavy metals at higher concentrations are deteriorating the water quality which may in turn cause threat to the biodiversity of the wetland. The study recommends intense monitoring and reduction of industrial and anthropogenic discharges into the rivers. Further the relevant authorities must design cost effective and time saving way for the continuous monitoring of heavy metals and other pollutants

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