

Potentially Toxic Trace Metals in Water and Lake-Bed Sediment of Panchpokhari, an Alpine Lake Series in the Central Himalayan Region of Nepal

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Abstract This study assessed the level of potentially toxic trace metals (PTMs), seasonal variations, and their possible sources from the surface water and lake-bed sediment of Panchpokhari lake series, an alpine and glacial lake at 4160 m a.s.l. in Central Nepal. The lake series have five lakes, with Lake-1 larger than others. So, Lake-1 was investigated thor-

oughly during pre-monsoon and post-monsoon seasons. Sediment core was collected from the deepest basin of the Lake-1 during pre-monsoon. Most of the PTM concentrations were higher in the pre-monsoon season; however, Sc, Cr, Cu, Zn, As, and Ag were higher in the post-monsoon. This is an indication that the lake has been impacted either by natural or long-range transported atmospheric pollutants. Ti, Sb, and Ag had extremely high enrichment factor (EF) in waters, whereas Cd, Zn, and As had high EF in sediments indicating that these metals originated from anthropogenic sources. Furthermore, PTM concentrations in the sediment were in the increasing order of $Hg < Cd < Ag < Mo < Sb < Sn < As < U < Sc < Co < Cs < Cu < Pb < Ni < Cr < V < Zn < Rb < Mn < Ti < Fe$ and showed that the upper layer (top 10 cm) of lake sediment has been receiving a higher load of PTMs in the recent period. he observed EF values also suggested that major sources of PTMs in the sediment were from crustal origin except for a few metals (Ti, V, Sb, and Ag) which were enriched anthropogenically due to long-range transport of atmospheric pollutants, deposited at the higher elevations. Nevertheless, the level of pollution in sediments was low as indicated both by EF and geo-accumulation index.

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1 Introduction

Potentially toxic trace metals (PTMs) are the major environmental pollutants (Nriagu and Pacyna 1988), and their contamination in water and sediment can pose biological hazards. Such toxic contamination has become a major environmental problem of modern times in both developing and developed countries throughout the world (Heikens et al. 2001; Zhang et al. 2007; Sun et al. 2010). In addition, PTMs have varying natural concentrations and different levels of toxicity, which affects the environment and the whole ecosystems. Some of them like Cr, Cu, Co, Mn, Fe, and Se are important constituents of living organisms in very small amounts and necessary for their health, growth, and development while their deficiency is often detrimental for living beings but their presence in high concentrations is very harmful and can be the cause of stunted growth or even mortality (Goldhaber 2003; WHO 2004). Trace amounts of Zn, Cu, Co, and Fe are essential and play important biochemical roles in the metabolism of aquatic organisms (Mason 2002; Kim et al. 2010), whereas Pb, Cd, and Hg metals are highly toxic. Therefore, they are serious pollutants in nature and can pose threats to the ecosystems at both local and global scales due to their toxicity, persistence, and solubility as well as bio-accumulating and bio-magnifying nature (Tam and Wang 2000). The presence of PTM loads in pristine water bodies arises from a number of factors like atmospheric pollution, lithology of terrestrial catchment, and climatic conditions (Tarvainen et al. 1997; Román-Ross et al. 2002; Jin et al. 2010). Sometimes, scavenged metals during weathering and other processes are emitted into the atmosphere (Smol 2002) and could pollute the environment as well. Often times, contamination could be due to direct anthropogenic activities such as improper disposal of sewage, industrial pollution, petroleum spills, and toxic waste dumping (Santos et al. 2005).

Lake-bed and marine sediments are the final pathways of both natural and anthropogenic components produced or derived from the environment (Saeed and Shaker 2008), finally deposited to the bottom with autochthonous constituents forming the total sediment of lakes, which act as a reservoir of trace metals. Moreover, PTMs tend to be more closely associated with sediment than water and hence accumulate there in. Thus, the concentration of substances in sediment can serve as a good indicator of pollution in the water column of a lake system. In fact, lake sediments are natural archives of

the climate and environment proxies for the response of a lake and its catchment to anthropogenic and climate change (Lami et al. 1998; Silva and Rezende 2002). According to Smol (2006), the study of sediment enables collecting the baseline information for trace metals deposited and assessing their contamination levels which provides a record of spatial and temporal history of a particular region.

In recent decades, long-range transported trace metals through the atmosphere have become a major global concern and most of the high-altitude water bodies in the remote locations have been impacted by the deposition of pollutants (Kyllönen et al. 2009; Zhou et al. 2012). Extensive research has been carried out to investigate this phenomenon along with PTM contamination in precipitation since the 1970s (Peirson et al. 1973). Global trace metal concentrations in the atmosphere increased in the nineteenth century due to human activities following the industrial revolution (Galloway et al. 1982). PTM concentrations in atmospheric wet deposition indicate that long-range transport of pollutants has a significant impact on the high-altitude remote area in the southern side of Central Himalayan regions of Nepal (Tripathee et al. 2016) as well as in the northern side of the Tibetan Plateau (Guo et al. 2014; Guo et al. 2016). Moreover, depositions of atmospheric PTMs in snow were detected on the southern part of the Tibetan Plateau (Huang et al. 2013). These studies have suggested lower PTM concentrations in the Tibetan Plateau and higher amounts along the southern slopes of Himalayas. The transport of anthropogenic pollutants and air borne dust from south and central Asia is largely a result of the monsoon climatic system (Huang et al. 2007; Ming et al. 2008; Tripathee et al. 2016) which influences the ecosystems of these regions.

High elevation and shallow water bodies as well as pristine background environment is suitable for understanding the impacts of long-range transport of atmospheric pollutant studies. Such environments are more susceptible to atmospheric inputs (Mosello et al. 1995; Galassi et al. 1997; Lami et al. 1998) and are also good indicators of regional and global pollution (Camarano and Manca 1997). Panchpokhari lake series is situated in a small valley surrounded by a mountainous ridge; being an alpine, remote, and oligotrophic lake (Raut et al. 2015), it was chosen as an environmentally significant site for the present research. Although the Panchpokhari area is frozen for almost half of the year, it is culturally significant and famous religious

pilgrimage site as well. Every year, more than thousands of Hindus and Buddhist pilgrims gather over there on the occasion of *Janai Purnima* (full moon day in August) to perform the rituals. Besides the scenic beauty, the lakes are the origin of water flowing to the Indrawati River and also provide electricity for the nearby villages through a micro-hydropower plant at Tipeni in the Sindhupalchowk district. Due to its natural pristine environment, this area is important for the conservation of the water catchment and biodiversity resources (Pradhan et al. 2009), as well as tourism promotion. It is very important to understand the status of these pristine lakes in the region. Therefore, the main aim of the present work was to assess contamination levels, sources, and implications of the major PTMs in surface water and lake-bed sediment of Panchpokhari lake series.

2 Materials and Methods

2.1 Study Area

The study area Panchpokhari (*five lakes*) is located in the Langtang National Park in the Central Himalayan region of Nepal between the longitude of 85° 42.96'–85° 43.25' E and latitude of 28° 2.41'–28° 2.54' N at an elevation of 4160 m a.s.l. (Fig. 1). Being glacial in origin (Bhujju et al. 2010; Krstić et al. 2012), it is one of 58 lakes ranging from 3000 to 4999 m a.s.l. in the Sindhupalchowk district of Nepal about 100 km north-east of Kathmandu, the capital city of Nepal. The mean annual temperature is 2 °C recorded from the nearest meteorological station, i.e., in Kyanjin Gompa (Barnard et al. 2006); the lake is generally frozen until the end of April.

2.2 Sampling Sites

The Panchpokhari lake system has five shallow lakes (Lake-1, Lake-2, Lake-3, Lake-4, and Lake-5) of less than 11 m depth that are well mixed. Among the five, Lake-1 is the largest compared to other lakes and shaped like a human footprint. Lake-5 is located adjacent to the right of the Lake-1, whereas Lake-2 is located near the rest house, and Lake-3 and-4 are located towards the middle of the basin. Thus, Lake-1 was studied systematically and water samples were collected from five different locations, i.e., the inlet, outlet, littoral zone, middle (center), and the deepest point to represent the complete water quality of the lake (Table 1), whereas

samples were collected from the inlet, outlet, littoral, and human influence area (near the rest house) for Lake-2 and only from the inlet and outlet sites for other centrally located lakes (Lake-3, Lake-4, and Lake-5).

2.3 Surface Water and Lake-Bed Sediment Samples

Altogether, a total of 21 PTMs were analyzed in the surface water samples during pre-monsoon ($n = 15$) and post-monsoon ($n = 15$) season including alkali metals (Rb and Cs), transition metals (Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cd, Cu, Zn, Mo, and Ag), metalloids (As and Sb), other metals (Al, Sn, and Pb), and rare-earth metal (U). Same number of PTMs (Hg instead of Al) were analyzed ($n = 13$) in the bed sediments of Lake-1 (0–26 cm) during pre-monsoon season.

2.4 Sample Collection, Preservation, and Storage

Water samples were taken from 10 to 20 cm below the lake surface free from aquatic vegetation and collected in ultra clean HDPE bottles for PTM analysis. All the collected samples were immediately preserved with concentrated HNO₃ to make pH < 2 and kept in an icebox during transport and stored in the refrigerator at 4 °C before lab analysis. Similarly, a UWITEC-CORER model was used for collecting the lake-bed sediment samples. The diameter of core tube was \varnothing (60 mm) and the length was 60 cm. Sediment core samples of 26 cm were retrieved from the deepest part of Lake-1 in May 2013, the pre-monsoon season. The core sediment was sectioned into 2-cm increments and each of segments was stored in a Ziploc polyethylene-bag separately and kept in the icebox for transportation to the Soil and Water Analysis Laboratory at Aquatic Ecology Centre (AEC) of Kathmandu University, Dhulikhel, Nepal, where it was stored under refrigeration in a frozen state until analysis.

2.5 Water and Sediment Sample Analysis

The water samples were measured directly by inductively coupled plasma-mass spectrometry (ICP-MS, X-7; Thermo-Elemental) at room temperature at the Institute of Tibetan Plateau Research, Chinese Academy of Sciences. A series of pre-treatment processes were followed during the sediment sample analysis. First, sediment samples were dried at 40 °C for 48 h and sieved through 2 mm mesh size. Then, the samples were digested by

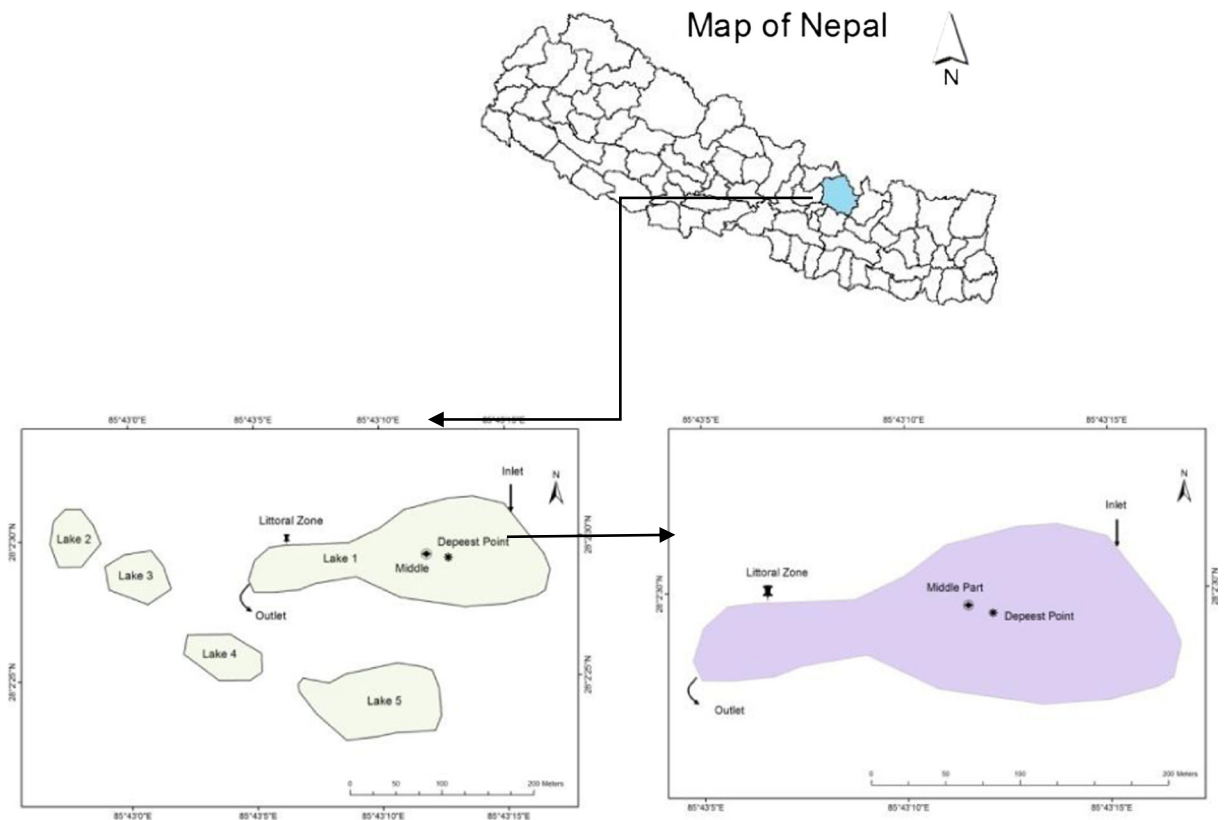


Fig. 1 Location map of the Panchpokhari lake series and detailed layout of the five lakes

pressurized acid digestion using a mixture of HNO_3 and HF in a sealed Teflon vessel. The solution was then transferred to a polypropylene bottle and the concentrations of trace metals were measured by ICP-MS. The complete analysis was carried out at the key laboratory of the Institute of Tibetan Plateau Research, Chinese Academy of Sciences, Beijing, China. However, for the Hg analysis, Leeman Hydra-IIC Direct Hg Analyzer (Leeman Lab Hydra, Teledyne Leeman Laboratories, and Hudson, NH) which involves thermal decomposition

and detection by atomic absorption spectroscopy, following the US EPA Method 7473, was used. The minimum detection limit (MDL) for lake sediment was <0.1 ppt which was calculated with standard deviation by the measurement of blank solution. Quality assurance and quality control of this analysis were conducted by using the samples (GSS-9) as standard reference material (Huang et al. 2016; Kang et al. 2016).

2.6 Quality Control and Assurance

Maximum precautions were taken to avoid the contamination during sampling in the field as well as laboratory analysis. All quartz glassware and PTFE wares were cleaned carefully before use. They were soaked in 50% HNO_3 (v/v) overnight. After boiling for 10 h, they were washed with deionized water. Finally, they were dried in a dustless oven and stored in a plastic bag before use. The HNO_3 (CMOS, JT Baker, USA) and HF (B VIII, Beijing Institute of Chemical Reagent Research, Republic of China) used were all ultrapure grade. The ultrapure deionized water (18.2 M Ω) was used. The

Table 1 Sampling sites (locations) with latitude and longitude of Lake-1

S. N.	Longitude (east)	Latitude (north)	Sampling sites
1.	85° 43. 141'	28° 02. 521'	Inlet
2.	85° 42. 958'	28° 02. 481'	Outlet
3.	85° 42. 971'	28° 02. 520'	Littoral zone
4.	85° 43. 056'	28° 02. 523'	Middle (central)
5.	85° 43. 057'	28° 02. 526'	Deepest

Table 2 Measurement of accuracy in some PTMs of certified reference materials (GSS-9 and AGV-2)

	Fe (ppm)		Mn (ppm)		Cu (ppm)		Zn (ppm)		Pb (ppm)	
	GSS-9	AGV-2	GSS-9	AGV-2	GSS-9	AGV-2	GSS-9	AGV-2	GSS-9	AGV-2
Certified value	33,600	46,830	520.0	770.0	25.00	53.00	61.00	86.00	25.00	13.00
Measured value	32,080	45,780	506.1	754.5	26.35	52.26	60.93	91.49	22.91	12.57
Recovery (%)	95.5	97.8	97.3	97.9	105.4	98.6	99.9	106.4	91.6	96.7

MDLs which are defined as three times the standard deviation of replicate blank measurements were Al (0.061 ppb), Sc (0.008 ppb), Ti (0.096 ppb), V (0.009 ppb), Cr (0.043 ppb), Mn (0.012 ppb), Fe (1.280 ppb), Co (0.005 ppb), Ni (0.070 ppb), Cu (0.070 ppb), Zn (0.040 ppb), As (0.155 ppb), Rb (0.009 ppb), Mo (13.76 ppt), Ag (3.135 ppt), Cd (2.018 ppt), Sn (7.262 ppt), Sb (6.597 ppt), Cs (2.040 ppt), Pb (2.758 ppt), and U (0.118 ppt).

For the sediment, GSS-9 and AGV-2 (from CNS and USGS, respectively) were analyzed as certified reference materials (CRM) for quality assurance of sediment analysis. The percentage recovery rates ranged from 91% for most of the PTMs except Mn which was calculated by actual value with obtained value (Table 2).

2.7 Data Analysis and Risk Assessments

The obtained data of various PTMs were used to calculate enrichment factor (EF) to find out the potential source of PTMs load in the Panchpokhari lake. EF is defined as the minimum factor by which the weight percentage of mineral in an ore body is greater than the average occurrence of that mineral in the Earth's crust. It describes about the origin of PTMs, whether crustal or anthropogenic. Thus, it is often used to evaluate the degree of anthropogenic influence (Kyllönen et al. 2009; Cong et al. 2010), comparing the concentrations of trace metals with a reference metals (Blaser et al. 2000; Reimann and de Caritat 2000). EF was calculated using the following equation:

$$EF_X = \frac{\left(C_X/C_R\right)_{sample}}{\left(C_X/C_R\right)_{reference}}$$

where X represents the metal of interest. EF_X represents the enrichment factor of X , C_X is the concentration of X , and C_R is the concentration of reference metal in the

Earth's crust. There are some common reference metals like Al, Fe, and Sc that are used (Schropp et al. 1990; Schiff and Weisberg 1999) for EF calculations. However, rubidium (Rb) was selected as a reference metal to calculate the EF value in the present study as it is not affected by anthropogenic sources (Sharma et al. 2015). The EF value was calculated based on the average upper continental crustal (UCC) compositions (e.g., Taylor and McLennan 1995) and many recent studies also followed the same due to lack of particular local references but it might be different from region to region. According to Kyllönen et al. (2009), metals with EF values close to unity indicate a strong natural influence and EF (>4) indicates some other sources but higher EF (>10) is regarded as the anthropogenic origin. Generally, the EF is used to assess soil contamination (enrichment) and its interpretation was given by Sutherland (2000) which has been followed by recent researchers as well (Table 3).

In addition, geo-accumulation index (I_{geo}) values were also calculated to identify the pollution status of sediment. This index was first proposed by Müller (1969), to identify and classify the pollution status of sediments and soils, which is a simple quantitative measurement of metal pollution. It has seven levels from unpolluted to extremely polluted quality of the sediment. I_{geo} was calculated using the following equation:

Table 3 Enrichment factor class in sediments

Value	Sediment quality
EF < 2	Depletion to minimal enrichment
EF 2 < 5	Moderate enrichment
EF 5 < 20	Significant enrichment
EF 20 < 40	Very high enrichment
EF > 40	Extremely high enrichment

Source: Sutherland 2000

Table 4 Description of geo-accumulation index (I_{geo}) to give sediment quality

Value	Class	Sediment quality
$I_{geo} < 0$	0	Practically uncontaminated
$0 < I_{geo} < 1$	1	Uncontaminated to moderately contaminated
$1 < I_{geo} < 2$	2	Moderately contaminated
$2 < I_{geo} < 3$	3	Moderately to heavily contaminated
$3 < I_{geo} < 4$	4	Heavily contaminated
$4 < I_{geo} < 5$	5	Heavily to extremely contaminated
$I_{geo} > 5$	6	Extremely contaminated

$$I_{geo} = \log_2 \left(\frac{C_n}{1.5B_n} \right)$$

where C_n is the measured concentration of the PTMs and B_n is the geochemical background value of given metal in the shale (Turekian and Wedepohl 1961) of element n ; 1.5 is the background matrix correction

factor that are due to lithogenic variations and very small anthropogenic influences. I_{geo} of trace elements in sediment and grading of pollution levels are shown in Table 4.

3 Results and Discussion

3.1 Water

The average concentrations of PTMs of surface water during pre-monsoon and post-monsoon season of Panchpokhari lake series are presented in Table 5. The obtained results were in the increasing order of $Cs < U < Cd < As < Sc < Mo < Ag < Co < V < Sn < Cr < Pb < Rb < Cu < Ni < Sb < Ti < Mn < Zn < Al < Fe$ and $Cd < Cs < U < Co < V < Sc < Mo < Ag < As < Sn < Pb < Rb < Ni < Sb < Cr < Cu < Ti < Mn < Zn < Al < Fe$ during the pre-monsoon and post-monsoon seasons, respectively. Most of the PTM concentrations were found within the WHO guideline value for drinking water quality and European

Table 5 Seasonal variation of PTMs level (ppb mean, STDEV, min, and max) in the surface waters

PTMs	Pre-monsoon (May 2013)				Post-monsoon (October 2013)				WHO GV	EU
	Average	STDEV (±)	Min	Max	Average	STDEV (±)	Min	Max	2011	1998
Al	159.4	231.6	29.4	913.9	139.6	117.7	42.09	495.6	200	200
Sc	0.175	0.070	0.080	0.382	0.248	0.097	0.072	0.448		
Ti	7.969	14.22	0.898	54.97	5.857	6.71	1.216	28.64		
V	0.319	0.467	0.019	1.663	0.242	0.221	0.074	0.986		
Cr	0.686	0.421	0.073	1.538	3.530	3.982	0.335	14.11	50	50
Mn	18.85	15.78	6.978	53.16	7.238	2.247	1.816	9.808	500	50
Fe	231.1	217.1	47.74	820	176.2	126.0	29.71	587.9	No guideline	200
Co	0.309	0.136	0.159	0.665	0.230	0.086	0.094	0.405		
Ni	1.963	1.837	0.212	5.607	1.584	1.234	0.305	4.502	20	20
Cu	1.831	1.735	0.234	5.886	3.598	4.292	0.332	14.04	2000	2000
Zn	12.74	14.56	3.785	60.76	31.55	27.77	2.830	92.92	3000	Not mentioned
As	0.173	0.007	0.167	0.181	0.748	0.668	0.155	2.268	10	10
Rb	1.635	0.812	0.855	3.85	1.054	0.44	0.438	2.001		
Mo	0.201	0.263	0.02	1.016	0.266	0.161	0.090	0.642	70	Not mentioned
Ag	0.224	0.091	0.039	0.410	0.476	0.081	0.377	0.611	No guideline	Not mentioned
Cd	0.068	0.089	0.014	0.305	0.023	0.007	0.011	0.036	3	5
Sn	0.558	0.586	0.11	1.895	0.771	0.751	0.170	3.372	No guideline	
Sb	4.184	0.586	3.313	5.331	2.892	0.888	1.539	4.726	5	5
Cs	0.047	0.076	0.008	0.297	0.026	0.03	0.007	0.118		
Pb	1.075	0.901	0.208	3.181	0.956	0.814	0.085	2.479	10	10
U	0.065	0.095	0.010	0.374	0.042	0.033	0.012	0.124	1400	Not mentioned

Union standards (Lenntech 1998; WHO 2011). The concentrations generally followed the same pattern in both seasons without any remarkable seasonal variations. However, some of the PTM values were lower in the post-monsoon as compared to those in the pre-monsoon season. This could likely be due to the diluting effect of monsoon precipitation being expressed just after the rainy season. In fact, the concentrations of Al, Ti, V, Mn, Fe, Co, Ni, Rb, Cd, Sb, Cs, Pb, and U were

higher in the pre-monsoon than those in the post-monsoon period, which was again probably due to a dilution effect during the latter because of heavy rainfall at the time of sampling in October 2014. On the other hand, concentrations of Sc, Cr, Cu, Zn, As, Mo, Ag, and Sn were higher in post-monsoon as compared to those in pre-monsoon. Mastoi et al. (2008) also reported the similar observation in a natural shallow Manchar Lake in Pakistan. The average values were similar in most of

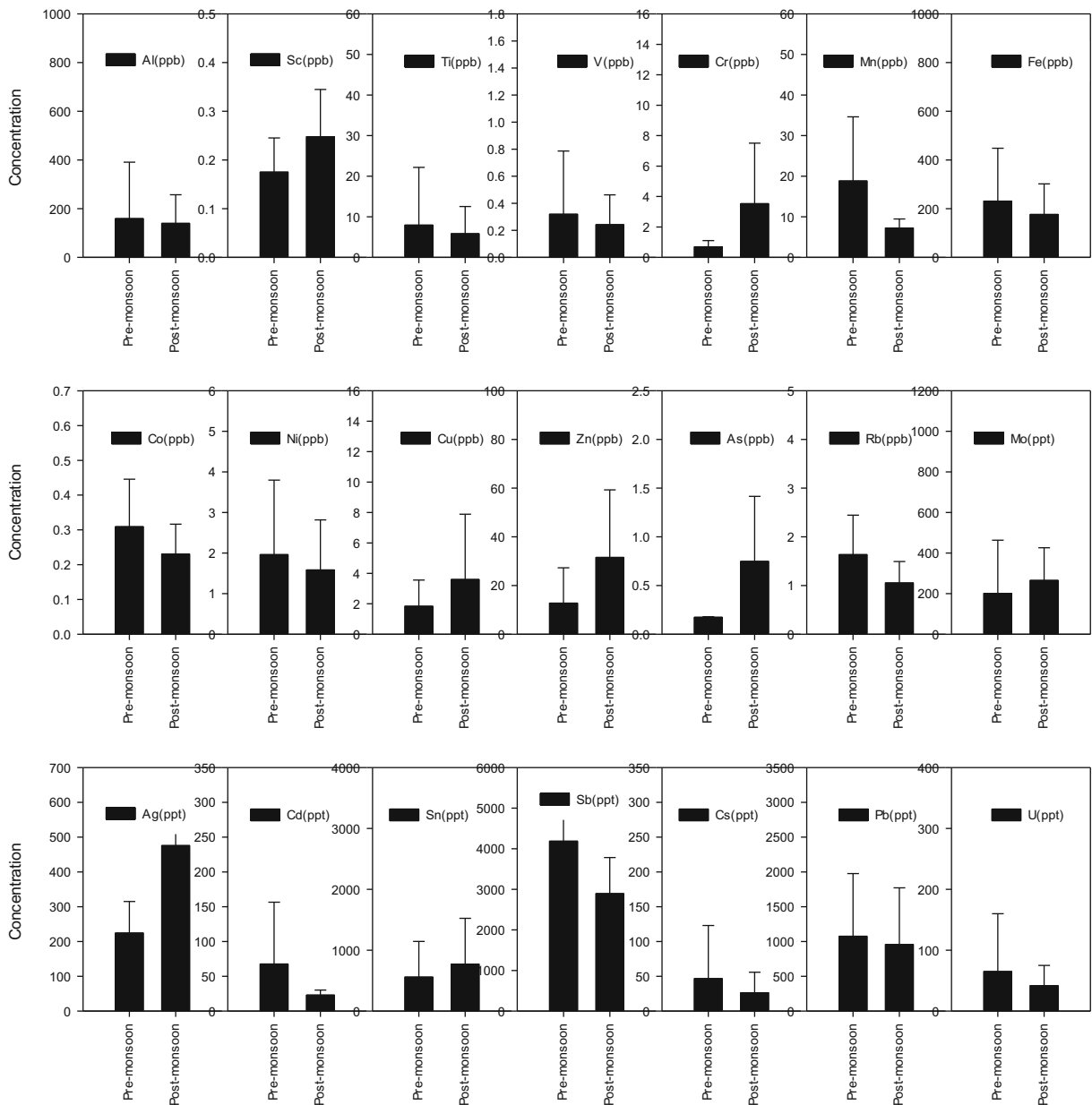


Fig. 2 Seasonal variations of average PTM concentrations of five lakes (ppb and ppt) with standard error in surface waters of Panchpokhari lake series

the PTMs but a maximum value of Al (913.9 ppb) in Lake-2 outlet and Al (495.6 ppb) in the littoral zone was noted during the pre-monsoon and post-monsoon seasons, respectively. With increasing acidification, aluminum (Al) concentration commonly increases in surface water (Lydersen et al. 2002), and the acidic nature of Panchpokhari lake waters has already been observed by Raut et al. (2015).

A high concentration of Al is toxic to aquatic biota (Baker and Schafeld 1982; Hutchinson et al. 1989) and can be expected to have an adverse effect on the biodiversity of the lake. Average values of Mn lies within the guideline value but exceeded the max limit (53.16 ppb) during the pre-monsoon. High concentrations of Al and Mn are harmful to aquatic organisms (Burton et al. 2013). Similarly, average values of Fe were maximum during both pre-monsoon (820 ppb) and post-monsoon (587.9 ppb), which even exceeded guideline values suitable for the protection of aquatic ecosystems (CBS 2013). Maximum accumulations of Fe, Mn, Al, Zn, As, Ba, Ni, and Cr were also recorded by Saygi and Yigit (2012). Metals such as Cr (0.686 ppb), Cu (1.831 ppb), Zn (12.74 ppb), Cr (3.53 ppb), Cu (3.598 ppb), and Zn (31.55 ppb) were recorded quite high both during pre-monsoon and post-monsoon seasons which was also recorded by Tripathee et al. (2014) in the nearby region. Therefore, this might be an indication of long-range transported atmospheric pollution because the main sources of As, Cr, Cu, Ni, Pb, and Zn are via the atmosphere (Nriagu 1996; Nriagu and Pacyna 1988; Burton et al. 2013; Tripathee et al. 2014).

Seasonal variations of PTMs are shown in Fig. 2. These elements could have a toxic effect on the aquatic

life of the lakes. Al, Fe, Hg, and Pb tend to react with humic acid but generally, the free metal ion concentrations are associated with toxicity to aquatic organisms (Gundersen and Steinnes 2003).

The concentrations of some of the PTMs were slightly higher than reported in previous studies of high-altitude lakes in Nepal as shown in Table 6, which indicate that depositions of pollutants seem to be gradually increasing either naturally or from anthropogenic sources through long-range transport.

The EF values of analyzed PTMs in the surface water of lake series varied from very low to extremely high, i.e., Al (0.261 ppb) to Ti (3457 ppb) during the pre-monsoon and Al (0.262 ppb) to Ti (2829 ppb) during the post-monsoon seasons, respectively. Among the analyzed PTMs, Ti, Sb, and Ag showed excessively high values, which could not be shown on the same graph; hence, with the exception of these metals, the EF values of all other PTMs are presented in Fig. 3. Similarly, Cd, Zn, and As also showed high EF values and belong to the class (EF > 40) having extremely high enrichment (Sutherland 2000) which is an indication that they are from anthropogenic sources and are of major concerns in the water bodies.

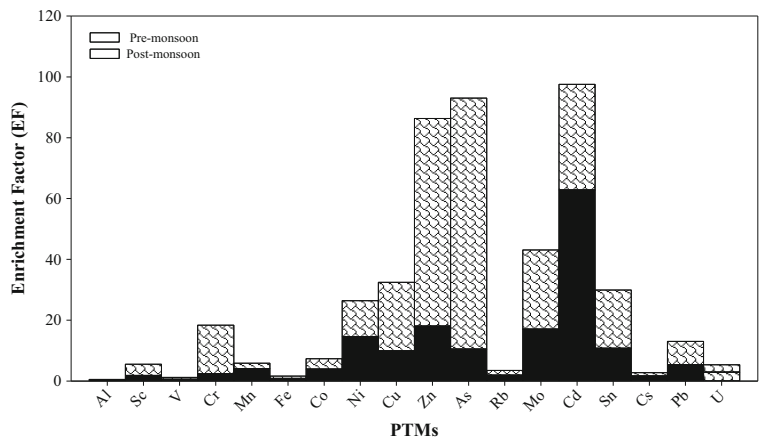
Similar results were also reported by Sharma et al. (2015) in Lake Gosainkunda and by Tripathee et al. (2014) on wet deposition in the Central Himalaya. PTMs such as Al, V, Fe, Cs, Sc, Rb, and Mn were found in the group having non-enrichment value (EF < 2) which clearly suggests that they originated from crustal sources. EF values between 2 and <5 were observed for Co and U showing moderate enrichment, but metals like Mn, Cr, and Sc exhibited seasonal fluctuation which may be from the crustal sources. Furthermore, Ni, Cu,

Table 6 Comparative study results of high-altitude lakes of Nepal during dry season

Lakes	Study year	PTMs (ppb)												References
		Al	V	Fe	Mn	Co	Ni	Cu	Zn	Cr	Cd	Pb	As	
Everest	1992	40.80	–	–	–	–	–	–	–	0.03	–	0.02	–	Reynolds et al. (1995)
Gokyo 2nd	2009	–	–	739	BDL	–	–	BDL	61.00	–	5.00	27.00	BDL	Sharma et al. (2012)
Gokyo 3rd	2009	–	–	846	57.00	–	–	BDL	63.00	–	5.00	17.00	BDL	Sharma et al. (2012)
Gokyo 4th	2009	–	–	1597	55.00	–	–	BDL	BDL	BDL	41.00	BDL	BDL	Sharma et al. (2012)
Gosainkunda	2010	91.93	0.26	129.5	4.3	0.11	0.76	1.76	13.79	0.21	0.02	0.65	–	Sharma et al. (2015)
Panchpokhari	2013	159	0.32	231.1	18.85	0.31	1.96	1.83	12.74	0.69	0.07	1.08	0.17	Present study

BDL below detection limit

Fig. 3 Seasonal variation of EF value in the surface waters of lake series (Panchpokhari)



Zn, As, Mo, and Pb showed significant enrichment of EF value, suggesting their anthropogenic origin.

3.2 Lake-Bed Sediment

The average concentrations of PTM profile from lake-bed sediment (core of Lake-1) of Panchpokhari are shown in Fig. 4. Although depth-wise distributions varied, general pattern of PTMs was in the increasing order of Hg<Cd<Ag<Mo<Sb<Sn<As<U<Sc<Co<Cs<Cu<Pb<Ni < Cr < V < Zn < Rb < Mn < Ti < Fe. With the

exception of Fe, Mn, As, Zn, Pb, Mo, and Hg, other analyzed PTMs followed a similar pattern of distribution in between (18–26 cm) depth. However, notable fluctuations seen between 10 and 18 cm depth might be due either to natural geological disturbances or possibly to the influence of atmospheric pollutants during that period (Fig. 4). In addition, ²¹⁰Pb dating of core sediment gave a history of 200 years with a sedimentation rate of 0.1 cm per annum (Fig. 4). Once again, PTM concentrations were distributed in nearly the same pattern in the upper-most layers between (0–10 cm) which could likely be due to recent atmospheric transport of pollutants over the past several decades.

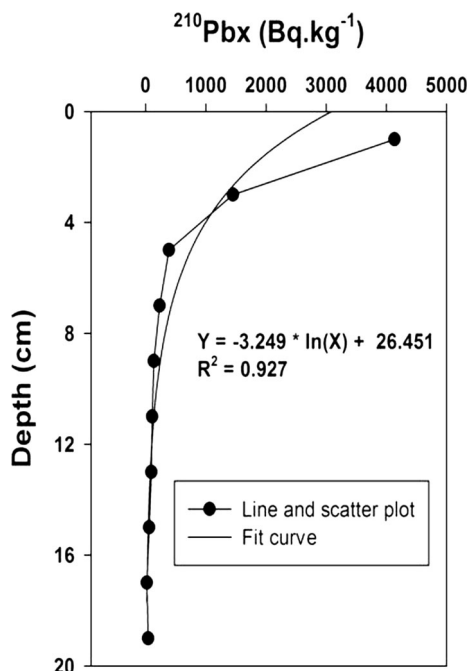


Fig. 4 ²¹⁰Pb dating of lake-bed sediment from Panchpokhari lake series

Expectantly, accumulations of PTM concentrations were much higher in the lake-bed sediment (Fig. 5) as compared to those in the water. Similar findings were reported by Nguyen et al. (2005), particularly result from the high adsorption of PTMs by particulate matter and its settling over time at the lake bottom. Variation of lake sediment reflected on the basis of PTM distribution could be due to more sand and low organic matter (Franca et al. 2005) that exhibit low metal enrichment. According to Tsai et al. (2003), concentrations of trace metals in sediment increase as the amount of organic matter increases. In the present study, higher accumulation of Fe, Ti, Mn, Rb, Zn, As, Pb, and Hg was found mostly in the top layer (0–10 cm) and such type of results was also reported by other researchers (e.g., Elghobashy et al. 2001; Ibrahim and El-Naggar 2006; Kang et al. 2016). Further, Swarnalatha et al. (2014) also reported the significant level of concentration of Cr, Ni, and Zn in the tropical shallow lake sediment of South India. The concentration of Fe was much higher in the sediment profile of Panchpokhari which can also be

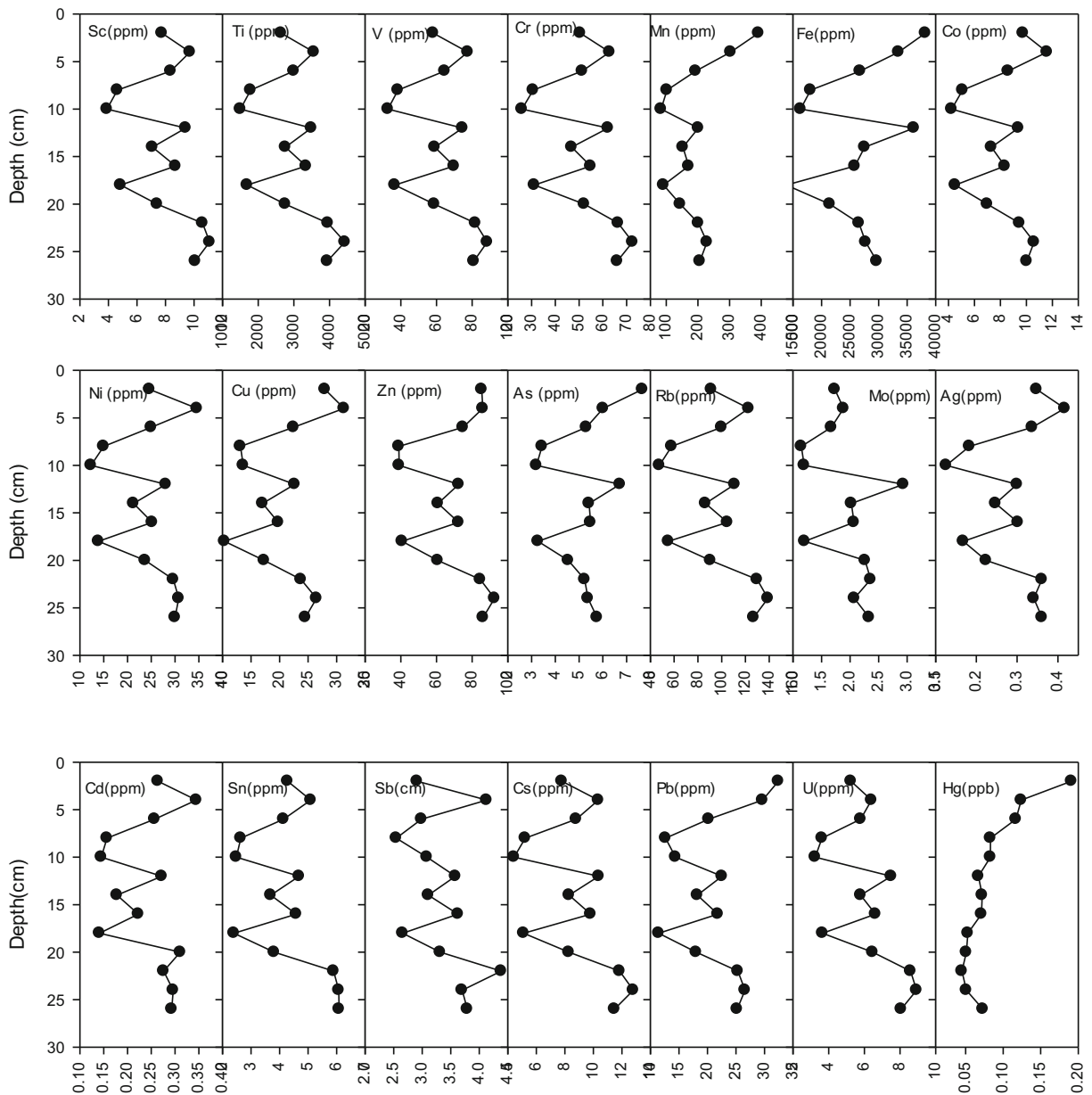


Fig. 5 Depth-wise distribution of PTMs (ppm/ppb) in the sediment of Lake-1 (Panchpokhari)

carcinogenic to aquatic fauna (Deugnier and Turlin 2001; Papanikolau and Pantopoulos 2005). Generally, Fe appears higher in the sediment and is an essential component of clay minerals which makes up a major proportion of the sediment in lakes (Carroll 1958). Similarly, As, Cd, and Pb are also toxic and serious pollutants not only by accumulating in the sediment but also by becoming sources of pollution within the water column (Papagiannis et al. 2004). Saygi and Yiğit (2012) suggested maximum

accumulation of Fe, Mn, Al, and Zn in sediment by natural processes during summer. Moreover, Hg was also found in increasing trend, which is potentially very toxic and one of the major anthropogenic contaminants. A recent study indicated higher accumulation of Hg in the sediment core of southern slopes of Himalaya (Kang et al. 2016) as compared to northern slopes in the Tibetan Plateau. Such a result was also reported by Sharma et al. (2015) in an alpine Himalayan lake, Gosainkunda of Nepal.

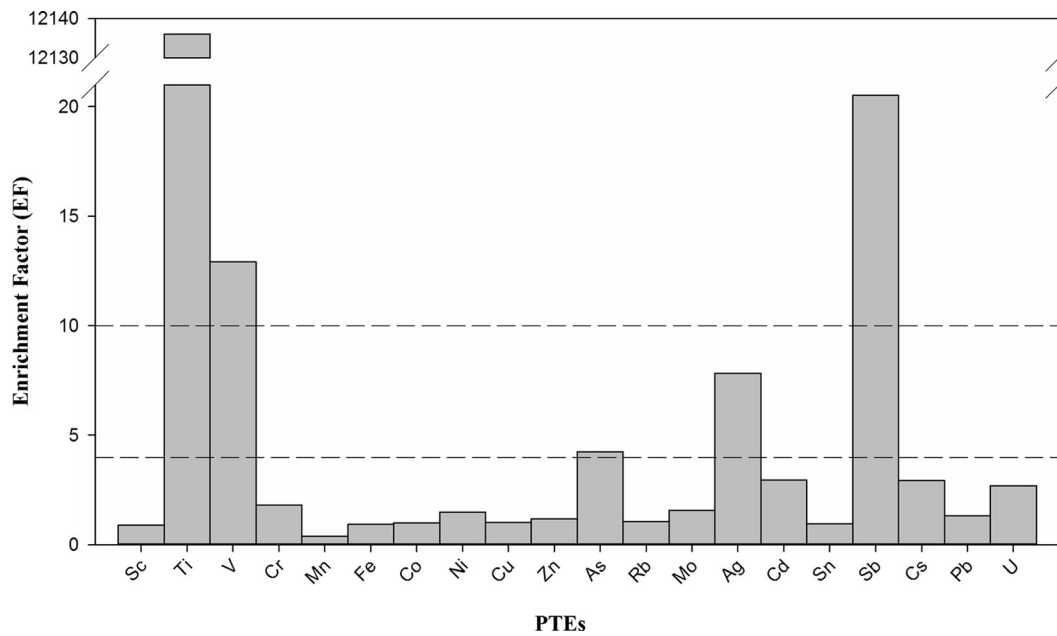


Fig. 6 Average EF values of PTMs in the lake-bed sediment of Lake-1 (Panchpokhari)

The average depth-wise EF values of analyzed PTMs are shown in Fig. 6. Hg was not plotted in the graph due to absence of reference value to calculate EF. Metals such as V, Ag, and Sb were highly enriched based on their EF values, whereas As, Cd, Cs, and U showed moderate enrichment (EF 2 < 5) value. The remaining metals were non-enriched based on the categories given by Sutherland (2000).

Majority of elements had negative I_{geo} values (Fig. 7) indicating minimal influence of anthropogenic

contaminants in the sediments. Such a result suggests an unpolluted sediment status in the alpine lake. Based on this estimate, we can conclude that the regional parent bedrock properties and natural accumulation process in wetlands are primary factors controlling the elemental concentrations in the sediments of wetland.

4 Conclusions

Some PTMs such as Fe, Mn, and Al were predominant in the lake waters which were mainly due to anthropogenic influences of pilgrims and visitors. On the other hand, concentration loads of Ni, Cu, Zn, As, Mo, and Pb were also high which could be the result of long-range transport of atmospheric pollutants through South-Asian Monsoon. Both EF and I_{geo} values of PTMs in sediments supported the possibilities of crustal origin from natural sources except for Ag, Sb, Cs, and U. The increasing trend of many PTMs in recent lake-bed sediment clearly indicates the continuous impacts of industrial revolution in the late 1800s and thereafter. These findings are clear indications that shallow to medium lakes in high-altitude mountains are important archives for studying the accumulation rate of various types of pollutants including potentially toxic metals, and require more research.

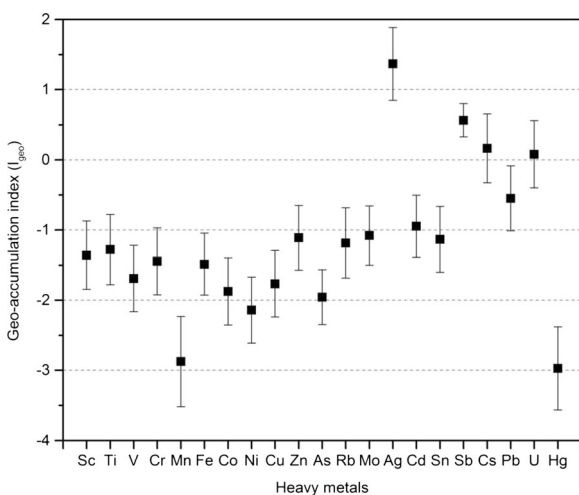


Fig. 7 Geo-accumulation index (I_{geo}) in the sediment of Lake-1 (Panchpokhari)

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