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Research Article

Spatial/Temporal Characterization and Risk Assessment of Trace Metals in Mangla Reservoir, Pakistan

Muhammad Saleem,[1] Javed Iqbal,[1] Gulraiz Akhter,[2] and Munir H. Shah[1]

[1] *Department of Chemistry, Quaid-i-Azam University, Islamabad 45320, Pakistan*
[2] *Department of Earth Sciences, Quaid-i-Azam University, Islamabad 45320, Pakistan*

Correspondence should be addressed to Munir H. Shah; mhshahg@qau.edu.pk

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Composite water samples were collected from different sites of Mangla reservoir, Pakistan, in premonsoon, monsoon, and postmonsoon seasons. The physicochemical parameters and trace/heavy metals were determined in all water samples. The results manifested significant seasonal variations among Co, Cr, Ni, and Pb and the metals exhibited highest contribution in premonsoon season except Mn. Principal component analysis (PCA) and cluster analysis (CA) revealed considerable anthropogenic intrusions in the reservoir. Probable risk associated with the metals levels on human health was also evaluated using hazard quotients (HQ) by ingestion and dermal routes for adults and children. It was noted that Cd, Co, Cr, Ni, and Pb ($HQ_{ing} > 1$) were the most important pollutants leading to noncarcinogenic concerns. The HQ_{derm} levels of all metals were below unity, suggesting that these metals posed no hazards via dermal absorption, while the oral intake was the major exposure pathway. The largest contributors to chronic risks were Cd, Co, Cr, Ni, and Pb in all the seasons. Therefore, immediate measures should be taken for sustaining the healthy aquatic ecosystem.

1. Introduction

Lakes are important and significant bodies in preserving freshwater, replenishing underground water, and adjusting local climate; consequently, they are considered one of the most versatile ecosystems in the world [1–3]. Recently, with the increasing urbanization and exhaustive development of industry, trace metals pollution in the lakes has become a worldwide problem because they are nondegradable and most of them have toxic effects on the organisms [4]. The accumulation of metals in aquatic ecosystems can lead to hazards on human and wildlife; therefore, researchers have been focusing on quantifying trace metals and their risk assessments on the aquatic environment [4–9]. Metals are derived from a variety of natural and anthropogenic sources, such as atmospheric deposition, geological weathering of rocks, untreated municipal wastes and industrial effluents, mining and mineral processing, electroplating, metal chelates from different industries, and agricultural activities mainly due to indiscriminate use of fertilizer [7, 10–18]. Multivariate statistical techniques are powerful tools for meaningful data reduction and the interpretation of geochemical data; hence, they are widely used in geochemical and ecochemical studies [6, 19, 20]. These methods have been frequently applied to identify pollution sources, to apportion natural versus anthropic contributions, and to describe the spatial distribution of pollutants [18, 21–25].

The Mangla Lake (Mirpur, Pakistan) was erected for hydroelectric power generation, irrigation, and flood control in 1967 across Jhelum River. Currently, the Lake water is also used for drinking and household purposes in adjoining areas. In this respect, the concentrations of trace metals in the Lake in different seasons were of great concern. The objectives of this study were (1) to quantify the dissolved concentrations of cadmium (Cd), cobalt (Co), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb), and zinc (Zn) in the Lake in premonsoon, monsoon, and postmonsoon seasons, (2) to determine the spatial and temporal

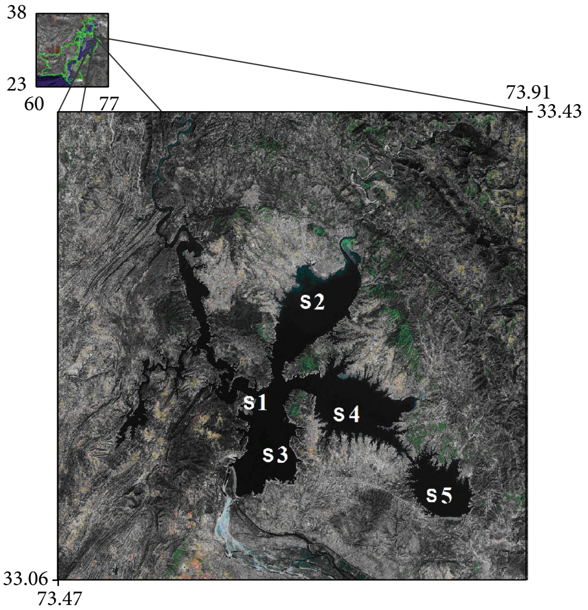

Figure 1: Map of the study area showing sampling sites.

variations of the metals in the Lake, (3) to evaluate water quality by comparing the measured levels with guidelines for drinking water and the protection of freshwater aquatic life, (4) to compare their concentrations with national and international reported levels, (5) to explore the natural and/or anthropogenic sources of these metals in the reservoir, and (6) to assess the human health risk associated with the metal levels. Ultimately, the study would help to develop water management and conservation strategies for the reservoir.

2. Materials and Methods

2.1. Study Area. Mangla reservoir is one of the largest freshwater resources in Pakistan (Figure 1). It was impounded in 1967 by damming Jhelum River near Mirpur city, Azad Jammu, and Kashmir, Pakistan. The reservoir is fed by two perennial rivers (Jhelum and Poonch) and two nonperennial rivers (Kanshi and Khad). It has five reservoir pockets: Jhelum, Poonch, Main Mangla, Khud, and Jari. The main dam is 3140 m long and 138 m high (above core trench) with a reservoir of 253 km^2. Since its construction, the water storage capacity of Mangla Dam has been reduced from 7,255 to 5,764 million cubic meters due to high sedimentation rates from the catchments areas [26]. The contribution of the Lake related to betterment of environment such as farming progression, employment, and enhanced living standard is noteworthy. Furthermore, provision of extra water and power production enlarges these encouraging influences. The Lake water is also being used for drinking purpose by the inhabitants. The untreated municipal and poultry wastes, industrial effluents, and agricultural runoffs from the villages, towns, and city around the reservoir are the major pollution sources for the Lake water. In addition, various streams also carry the pollutants into the reservoirs during the high flow period.

Table 1: Certified versus measured concentrations (μg/L) of trace metals in standard reference material (SRM 1643d).

Metal	Certified concentration	Measured concentration
Cd	6.47 ± 0.37	6.09 ± 0.28
Cr	18.53 ± 0.20	19.7 ± 0.24
Co	25.00 ± 0.59	22.3 ± 0.41
Cu	20.5 ± 3.8	19.2 ± 2.1
Fe	91.2 ± 3.9	92.6 ± 3.5
Mn	37.66 ± 0.83	36.1 ± 0.67
Ni	58.1 ± 2.7	61.2 ± 2.1
Pb	18.15 ± 0.64	19.7 ± 0.83
Zn	72.48 ± 0.65	69.6 ± 0.94

2.2. Sampling, Processing, and Analysis. Composite water samples were collected from five sites in the Lake (as shown in Figure 1) in premonsoon (May 2012), monsoon (August 2012), and postmonsoon (November 2012). Direct method was employed to collect the water samples (triplicate) in polyethylene bottles (1.5 L, volume capacity) following standard procedure [27]. Each water sample was composed of three equal volume subsamples from an area of 10–20 m^2. The samples were kept in airtight large plastic ice-cold containers and were transported to laboratory within 6 h of their collection for further processing. Suspensions in the water samples were removed through filtration. The initial portion of filtration was discarded and the next one was preserved in refrigerator after acidifying with HNO_3 ($pH < 2$) until chemical analysis [4, 28].

The water quality parameters including temperature (T), hydrogen ion concentration (pH), dissolved oxygen (DO), and total dissolved solids (TDS) were measured in the field/on site: pH was measured using a digital pH meter (model: Martini Mi 180); DO was estimated by digital DO meter (model: Martini Mi 190); TDS were measured by a digital TDS meter (model: Jenway 470). Concentrations of the metals (Cd, Cr, Co, Cu, Fe, Mn, Ni, Pd, and Zn) were determined using a flame atomic absorption spectrophotometer (model: Shimadzu AA-670, Japan). Calibration line method was used for the quantification of metals. A reagent blank was analyzed to determine the contamination during processing/preserving of the water samples. All the measurements were made in triplicate. The reliability of the analytical data was ensured by using standard reference material (SRM-1643d) and the results are shown in Table 1. Instrument settings were as recommended in the manufacturer's manual, with wavelengths (nm) of 228.8 (Cd), 240.7 (Co), 357.9 (Cr), 324.8 (Cu), 248.3 (Fe), 279.5 (Mn), 232.0 (Ni), 217.0 (Pb), and 213.9 (Zn).

Chemical reagents (AR grade, certified purity > 99.99%) used during chemical processing and analysis were purchased from E-Merck (Darmstadt, Germany). Doubly distilled water was used for the preparation of working standards from stock solution (1000 mg/L) and for the dilution of water samples whenever required [29]. Glassware was decontaminated by washing with tap water and detergent solution (5%, w/v), soaked in nitric acid (5%, v/v) overnight, and rinsed with doubly distilled water, respectively. If some adhering organic matter was suspected, a rinse with acetone followed by doubly distilled water was also given. Finally, the glassware was dried in an oven maintained at 85°C for more than six hours prior to use.

2.3. Statistical Analyses. Possible sources of the metals in water reservoir were identified by principal component analysis (PCA) and cluster analysis (CA). PCA was applied on the dataset after varimax normalized rotation. It yielded significant principal components (PCs) which showed the contribution of major sources to total pollution index [29–32]. Cluster analysis (CA), an unsupervised configuration recognition method, disposed a set of variables into two or more jointly unidentified clusters established on grouping of internal variables. Its purpose was to ascertain a scheme of unifying variables where each cluster shared communal properties. Ward's method was applied to obtain dendrogram of CA showing grouping of the metals and sampling sites to assess the spatial variability [4]. Statistical analyses were carried out using STATISTICA software [33].

2.4. Human Health Risk Assessment. Human beings are exposed to trace metals through three possible ways: direct ingestion, inhalation, and dermal contact. Oral intake and dermal absorption routes are most common for drinking water [34–37]. The average daily doses through these pathways can be determined using the following equations:

$$\mathrm{ADD}_{\mathrm{ing}} = \frac{C_{\mathrm{water}} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}},$$
$$\mathrm{ADD}_{\mathrm{derm}} = \frac{C_{\mathrm{water}} \times \mathrm{SA} \times K_p \times \mathrm{ET} \times \mathrm{EF} \times \mathrm{ED} \times \mathrm{CF}}{\mathrm{BW} \times \mathrm{AT}}, \quad (1)$$

where $\mathrm{ADD}_{\mathrm{ing}}$ is the average daily dose by ingestion (μg/kg-day), $\mathrm{ADD}_{\mathrm{derm}}$ is the average daily dose through dermal absorption (μg/kg-day), C_{water} is the mean metals concentration in surface water (μg/L), IR is the ingestion rate (L/day, 2.2 for adults and 1.8 for children), EF is the exposure frequency (days/year, 350), ED is the exposure duration (years, 70 for adults and 6 for children), BW is the average body weight (kg, 70 for adults and 15 for children), AT is the averaging time (days, 25550 for adults and 2190 for children), SA is the exposed skin area (cm^2, 18000 for adults and 6600 for children), ET is the exposure time (hours/day, 0.58 for adults and 1 for children), CF is the unit conversion factor (L/cm^3, 0.001), and K_p is the dermal permeability coefficient (cm/h), 0.001 for Cd, Cu, Fe, and Mn, 0.002 for Cr, 0.004 for Co, Pb, and Ni, and 0.0006 for Zn [12, 35–39].

Risk evaluation related to the noncarcinogenic risks was quantified by computing hazard quotient (HQ). The HQ is a ratio of average intake of contaminants from exposure ways (oral intake/dermal) to the related reference dose (RfD) which was calculated using the following equation:

$$\mathrm{HQ}_{\mathrm{ing/derm}} = \frac{\mathrm{ADD}_{\mathrm{ing/derm}}}{\mathrm{RfD}_{\mathrm{ing/derm}}}. \quad (2)$$

Significant noncarcinogenic risk is associated with HQ > 1. Hazard index (HI) was computed to determine the total possible noncarcinogenic risks posed by multipathways. The HI was computed by adding the HQs from all probable pathways as below:

$$\mathrm{HI}_{\mathrm{ing/derm}} = \sum_{i=1}^{n} \mathrm{HQ}_{\mathrm{ing/derm}}, \quad (3)$$

where $\mathrm{HI}_{\mathrm{ing/derm}}$ is the hazard index via ingestion or dermal contact (unitless). HI > 1 indicated a potential for an adverse effect on human health [35].

3. Results and Discussion

3.1. Physical Characteristics. The physical condition of a water body strongly influences the chemical and biological processes that occur in the water column and consequently its ecological and chemical status. The mineral constituents existing in water determine the aptness of water. Water quality parameters (T, pH, TDS, and DO) in three seasons along with international/national water quality guidelines are given in Table 2. The data showed that average measured temperature values were 31.6, 24.3, and 13.8°C in premonsoon, monsoon, and postmonsoon, respectively. The pH values of the surface water ranged from 7.8 to 8.3, from 7.2 to 7.8, and from 6.4 to 6.8 in premonsoon, monsoon, and postmonsoon seasons, respectively. Consequently, the water samples were slightly alkaline in nature having carbonate species as $\mathrm{HCO_3}^{1-}$ in premonsoon and monsoon [40], whereas they were slightly acidic in postmonsoon which could be due to human activities such as use of fertilizer and timber harvesting [41]. The lowest pH (6.4) was found in postmonsoon at site S5 near to Khaliqabad and Kakra towns, while the highest pH (8.3) was found in the samples near the industrial area of Mirpur city during premonsoon. The solubility of metals is generally lowered by elevating pH and increased by lowering pH, discharging free metal ions into the overlying water [42].

Total dissolved solid (TDS) is an important parameter as it can affect the taste of water. Concentrations of TDS are generally related to human activities such as urban water runoffs, municipal wastewater discharges, and agricultural activities in the catchments areas. The contents of TDS ranged from 86.3 to 229 mg/L, from 99.2 to 385 mg/L, and from 74.2 to 139 mg/L with mean values of 92.0, 139, and 79.4 mg/L in premonsoon, monsoon, and postmonsoon seasons, respectively. Water having TDS less than 1000 mg/L is generally considered fresh/acceptable [43]. Consequently, surface waters from the Lake were considered fresh with respect to TDS levels in all seasons. In premonsoon and monsoon the highest and lowest values of TDS were found at sites S2 and S5, whereas in postmonsoon they were found at sites S1 and S5, respectively.

The dissolved oxygen (DO) in surface water ranged from 4.2 to 4.9 mg/L (mean: 4.3 mg/L), from 6.0 to 6.5 mg/L (mean: 6.1 mg/L), and from 6.4 to 7.4 mg/L (mean: 6.5 mg/L) in premonsoon, monsoon, and postmonsoon seasons, respectively. DO concentration of greater than 5 mg/L is recommended to support the biota in aquatic ecosystem [28]. Flow regime,

Table 2: Descriptive statistics for trace metals (μg/L) and water quality parameters in comparison with national/international guidelines.

	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	T	pH	TDS	DO
Premonsoon													
Mean	36.3	235	75.1	20.3	128	13.1	313	339	31.2	31.6	7.9	92.0	4.3
Median	30.5	231	52.5	16.0	127	10.0	322	241	30.5	31.5	8.1	82.4	4.6
Max	103	501	312	56.0	381	59.0	682	1501	76.0	32.5	8.3	229	4.9
Monsoon													
Mean	31.1	157	67.2	14.4	88.5	8.16	126	226	7.84	24.3	7.4	139	6.1
Median	30.5	116	54.0	11.0	66.5	8.00	127	228	5.00	24.4	7.4	131	6.1
Max	53.0	547	194	55.0	278	24.0	415	465	72.0	28.1	7.8	384	6.5
Postmonsoon													
Mean	18.3	103	21.0	13.7	109	18.9	124	129	11.7	13.8	6.4	79.4	6.5
Median	17.0	99.0	17.0	12.5	78.5	18.0	116	124	10.5	13.8	6.4	77.0	6.5
Max	40.0	254	63.0	30.0	454	43.0	405	267	31.0	14.2	6.8	139	7.4
Water quality criteria for drinking water													
WHO (2008)	3	40	50	2000	300	100	70	10	3000	—	6.5–8.5	1200	—
USEPA (2009), MCL	5		100	1300	300	50	700	15	5000	—	6.5–8.5	500	—
EC (1998)	5		50	2000	200		20	10	100		—	—	—
Pak-EPA (2008)	10		50	2000		500	20	50	5000	—	6.5–8.5	1000	—
Freshwater quality criteria for protection of aquatic life													
USEPA (2006), CMC (acute)	2		16	13	1000		470	65	120	—	—	—	—
USEPA (2006), CCC (chronic)	2.5		11	9	1000		52	3		—	—	—	—

T: temperature (°C); TDS: total dissolved solids (mg/L); DO: dissolved oxygen (mg/L); CMC: criterion maximum concentration; CCC: criterion continuous concentration.

seasonal influences, and anthropic effects can be the causes of DO variations [44]. The release of unprocessed solid wastes from poultry farms and farmlands and municipal wastes from surrounding areas could be the possible reason for lower DO levels in premonsoon. Consequently, the aquatic ecosystem was under stress due to low DO levels. Although same types of inputs were also observed in monsoon and postmonsoon, their effects were insignificant due to dilution with plenty of water. Moreover, the lower DO levels could also be due to relatively higher temperature in premonsoon [41].

Measured values of pH in surface water were found to be within permissible limits of WHO [45], USEPA [46], and Pak-EPA [47] in premonsoon and monsoon seasons, whereas in postmonsoon they were found to be below permissible limits in 55% of the samples. However, average values of TDS in the water samples were found to be within WHO, USEPA, and Pak-EPA guidelines (Table 2). It indicated that the water from Mangla Lake was fresh having low salinity and minerals [48].

3.2. Spatial Variations of Trace Metals. During the study period, all trace metals did not show significant spatial variations ($p < 0.05$) in the water reservoir. Highest levels of the metals were observed at sites S3 and S4 in all seasons. The total concentrations of trace metals in premonsoon season showed an average level of 132 μg/L, with following order: S4 (181 μg/L) > S3 (147 μg/L) > S5 (125 μg/L) > S1 (106 μg/L) > S2 (103 μg/L); meanwhile, in monsoon season, the average level was 81.0 μg/L, and the relative order was S4 (105 μg/L) > S3 (89.0 μg/L) > S5 (80.0 μg/L) > S1 (68.0 μg/L) > S2 (61.0 μg/L). Likewise, the total concentrations of trace metals in postmonsoon exhibited an average value of 61.0 μg/L, followed in order of abundance by site S4 (80.0 μg/L) > S3 (65.0 μg/L) > S5 (56.0 μg/L) ≈ S1 (56.0 μg/L) > S2 (48.0 μg/L). Overall order of total metal concentrations remained the same in all seasons: S4 > S3 > S5 > S1 > S2 (Figure 2).

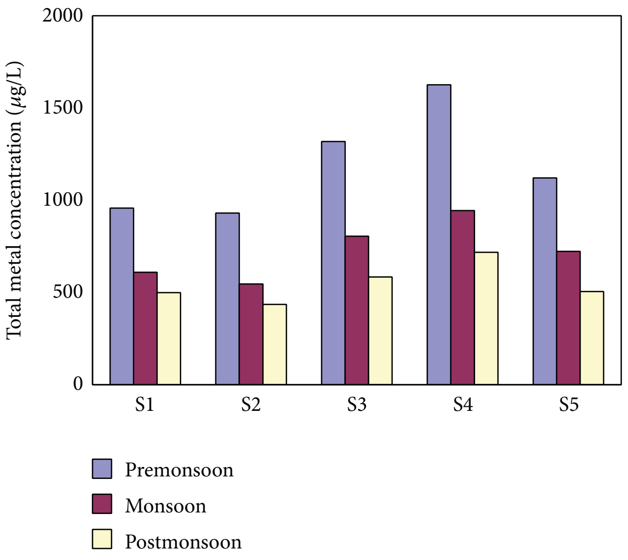

Figure 2: Total concentrations of trace metals at different sampling sites.

Spatial distribution of total concentrations of the metals showed a decreasing trend from the sites near highly urban areas to the sites near suburban areas. The higher total concentrations were measured at sites S4 and S3 which are located at highly urbanized area (Mirpur city), while the lower total concentrations were found at sampling sites S1 and S2 which are close to the less populated areas of the Lake in all seasons. Moreover, the water samples collected near the Lake outlet had the lowest concentrations. It could be due to dilution and transfer of metals from water column towards the sediments [4, 7, 49]. Among the metals, Fe, Co, Pb, and Ni were the most abundant elements in the reservoir, whereas Cu, Mn, and Zn were less abundant. Nickel and lead showed maximum concentrations at sites S3 and S4, while Fe indicated higher concentrations at sites S4 and S5 in all seasons. However, the maximum levels of Cd and Co were found at sites S3 and S4 in pre- and postmonsoon seasons and at sites S4 and S5 in monsoon. The mean order of metals level was Pb (339 μg/L) > Ni (331 μg/L) > Co (235 μg/L) > Fe (128 μg/L) > Cr (75.1 μg/L) > Cd (36.3 μg/L) > Zn (31.2 μg/L) > Cu (20.3 μg/L) > Mn (13.1 μg/L) in premonsoon, Pb (226 μg/L) > Co (157 μg/L) > Ni (126 μg/L) > Fe (88.5 μg/L) > Cr (67.2 μg/L) > Cd (31.1 μg/L) > Cu (14.4 μg/L) > Zn (7.84 μg/L) > Mn (8.16 μg/L) in monsoon, and Pb (129 μg/L) > Ni (124 μg/L) > Fe (109 μg/L) > Co (103 μg/L) > Cr (21.0 μg/L) > Mn (18.9 μg/L) > Cd (18.3 μg/L) > Cu (13.7 μg/L) > Zn (11.7 μg/L) in postmonsoon. For all the seasons, high concentrations of Co, Ni, and Pb were observed at sites S3 and S4 which are under the influence of urbanization and industrialization.

3.3. Seasonal Variations of Trace Metals. The average concentrations of trace metals in all seasons are shown in Table 2. Sum of concentrations of all the metals (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) in Lake water showed the average values of 1191 μg/L, 726 μg/L, and 549 μg/L in premonsoon, monsoon, and postmonsoon seasons, respectively. The results indicated that the metal concentrations were higher in premonsoon as compared to the other seasons irrespective of the sampling sites. It may be owing to the varying seasonal inputs to the Lake water due to hydrological regime and seasonal anthropogenic activities. This may be attributed to the high evaporation and intense anthropogenic activities (agriculture and high degree of domestic/industrial activities) in summer (premonsoon) to elevate the total concentrations of metals [50, 51]. High precipitation and large water inputs from the tributaries and rivers were the most plausible explanations for the lowest total concentrations in monsoon and postmonsoon seasons. High precipitation in monsoon mixes large volumes of uncontaminated runoff water with contaminated water which resulted in decline of total metal concentrations in Lake water [4].

Most of the trace metals showed significant temporal/seasonal variations ($p < 0.05$). Among the metals, Cd, Co, Cr, Cu, Ni, and Pb displayed the following order: premonsoon > monsoon > postmonsoon; meanwhile, for Zn and Fe, the order was premonsoon > postmonsoon > monsoon. However, Mn showed different order: postmonsoon > premonsoon > monsoon. About 80% of annual precipitation is associated with monsoon season resulting in dilution of the pollutants. Some of the metals (Fe, Mn, and Zn) showed higher contributions in varying hydrological seasonality, which may be attributed to mixed sources due to natural contributions and anthropogenic activities [12, 51]. Thus, precipitation and seasonal anthropogenic activities played important roles in distribution of metal levels in surface water of the Lake [12].

3.4. Comparison with National and International Studies. Average metal concentrations in water samples from Mangla Lake were also compared with the reported studies from different reservoirs in national and international studies (Table 3). Some of the metals (Cd, Co, Cr, Ni, and Pb) showed higher contents in the Mangla Lake compared with other studies in all seasons except Co in postmonsoon levels which were lower than those reported for the Legnica Lake (Southwest Poland), Dil Deresi Stream (Turkey), Anthropogenic Lake (West Poland), and Lake Gilow (Poland) [4, 7, 8, 52–56]. Cu and Zn exhibited lower concentrations than those reported for the Dil Deresi Stream (Turkey), Kanyaboli Lake (Kenya), Anthropogenic Lake (West Poland), Legnica Lake (Southwest Poland), and Lake Gilow (Poland) in all seasons, while Fe showed lower concentrations than those reported for the Dil Deresi stream (Turkey) and Anthropogenic Lake (West Poland) in all seasons. Manganese showed lower average levels than those reported for Kanyaboli Lake (Kenya), Anthropogenic Lake (West Poland), Legnica Lake (Southwest Poland), and Lake Gilow (Poland) in all seasons.

Average metal levels measured in the present study were compared with previously reported levels [57] from Mangla reservoir which showed almost comparable levels for Cd, Cu, Mn, Ni, Pb, and Zn with few exceptions; nevertheless, in the present study, average levels of Cr were significantly higher than the reported levels. In comparison with other national reported studies related to surface water (Table 3), the mean concentrations of Cd were relatively higher in premonsoon and monsoon than levels reported in other studies [58–61]. The average levels of Cr and Ni were found to be elevated in all seasons compared to others; Co levels (premonsoon and monsoon) were noted to be higher than levels in Rawal lake (summer) and Manchar lake; and Pb average levels were elevated compared to others except Mangla Lake in premonsoon and postmonsoon. However, Fe, Mn, and Zn contents were noted to be lower than others. Moreover, Cu levels were comparable to other reported levels.

3.5. Comparison with Guideline/Standard Values. Metal concentrations in different seasons were compared with water quality guidelines for drinking water [45–47, 62] and the protection of freshwater aquatic life [63] (Table 2). The maximum concentrations of Cu and Zn and the average values of Fe and Mn in the reservoir were lower than the maximum permitted concentrations established by EC, USEPA, WHO, and Pak-EPA guidelines. The maximum concentrations of Mn in premonsoon were higher than USEPA, while the maximum value of Fe in premonsoon and postmonsoon exceeded WHO, USEPA, EC, and Pak-EPA guidelines. The maximum and average values of Cd and Pb in the Lake

Table 3: Comparison of mean trace metals concentrations (μg/L) in surface water of Mangla Lake with national and international studies.

	Cd	Co	Cr	Cu	Fe	Mn	Ni	Pb	Zn	Reference
Mangla Lake Premonsoon	36.3	75.1	235	20.3	128	13.1	313	339	31.2	This study
Mangla Lake Monsoon	31.1	67.2	157	14.4	88.5	8.16	126	226	7.84	This study
Mangla Lake Postmonsoon	18.3	21.0	103	13.7	109	18.9	124	129	11.7	This study
Kralkızı Dam Reservoir, Turkey	0.036	—	22.06	2.83	58.63	—	15.75	2.56	5.02	[4]
Dicle Dam Reservoir, Turkey	0.030	—	18.58	2.12	62.07	—	15.86	1.84	4.12	[4]
Batman Dam Reservoir, Turkey	0.044	—	16.5	ND	57.66	—	15.96	1.56	4.09	[4]
Danjiangkou Reservoir, China	1.17	1.08	6.29	13.32	19.14	5.69	1.73	10.59	2.02	[7]
Dil Deresi stream, Turkey	8	21	42	37	4030	—	—	120	700	[8]
Taihu Lake, China	0.06	—	0.99	5.81	—	—	5.34	2.74	15.86	[52]
Kanyaboli Lake, Kenya	4.4	6.06	21.54	23.95	—	284.27	16.38	20.65	32.79	[53]
Kainji Dam, Nigeria	—	1.2	2.2	1.3	13	9	0.90	1.2	0.90	[54]
Anthropogenic Lake in West Poland	2.9	24	3.3	48	154	1230	4	111	207	[55]
Legnica Lake in Southwest Poland	1.72	65	1.1	29	6	670	68.2	0.21	204	[56]
Lake Gilow, Poland	0.58	27	0.9	48	6	390	76	0.5	167	[56]
Mangla Lake, Pakistan Summer	30	250	80	20	150	10	130	380	30	[57]
Mangla Lake, Pakistan Winter	30	160	70	20	130	20	110	340	30	[57]
Rawal Lake, Pakistan Summer	6	11	9	10	93	4	—	162	14	[58]
Rawal Lake, Pakistan Winter	25	204	97	17	76	13	—	223	22	[58]
Khanpur Lake, Pakistan	20	114	46	9	51	11	—	221	15	[59]
Manchar Lake, Pakistan Summer	6.14	41.0	8.23	20.4	3228	75.8	35.8	87.6	774	[60]
Manchar Lake, Pakistan Winter	4.22	34.7	6.84	18.2	2780	64.7	31.0	78.7	683	[60]
Manchar Lake, Pakistan	5.3	38.9	7.64	18.9	2960	72.6	35.0	82.4	730	[61]

ND: not detected.

were higher than the water quality guidelines set by WHO, USEPA, EC, and Pak-EPA in three seasons. The maximum and average concentrations of Co in three seasons were higher than WHO guidelines, while maximum value of Cr in all seasons and its average value in premonsoon and monsoon were higher than WHO, EC, and Pak-EPA guidelines, whereas maximum concentration of Cr exceeded USEPA permissible limits in premonsoon and monsoon seasons. The maximum and average concentrations of Ni in three seasons were higher than WHO, EC, and Pak-EPA guidelines, while they were below USEPA guidelines. When the measured metal concentrations were compared with USEPA guidelines, for aquatic life protection [63], it was found that the maximum concentrations of Cd, Cr, Cu, and Pb exceeded the criterion maximum concentrations (CMC) and criterion continuous concentration (CCC) values, while Ni was higher than the criterion continuous concentration (CCC) values in all seasons.

In this study, 100% of the water samples in monsoon and >90% samples in premonsoon and postmonsoon for Cd and Pb exceeded EC, WHO, USEPA, and Pak-EPA guidelines. About 58% of the samples for Cr in premonsoon and monsoon and >92% for Ni in all seasons were found to exceed WHO, EC, and Pak-EPA guidelines. Cobalt concentrations were higher than WHO guidelines in 96%, 86%, and 84% of samples in premonsoon, monsoon, and postmonsoon, respectively. However, the measured concentrations of Fe in more than 90% of samples were lower, whereas Cu, Mn, and Zn levels were found to be lower in 100% of the water samples than the national and international guidelines in all seasons. Thus Cd, Co, Ni, and Pb were potential pollutants in the reservoir and they might pose health risks for the local population. For example, Cd causes renal tubular dysfunction, bone fragility, kidney dysfunction, skeletal damage, and reproductive disorders; Co causes polycythemia, heart dysfunctions, thyroid alterations, testicular deterioration and

Table 4: Principal component loadings* for trace metals in surface water of Mangla Lake.

	PC1	PC2	PC3	PC4
Eigenvalue	1.85	1.47	1.23	1.22
Total variance (%)	38.6	20.4	15.7	10.6
Cumulative variance (%)	38.6	59.0	74.7	85.3
Cd	**0.83**	—	—	—
Co	**0.83**	—	—	—
Cr	—	**0.83**	—	—
Cu	—	0.35	**0.67**	0.25
Fe	0.29	—	—	**0.67**
Mn	—	—	—	**0.61**
Ni	0.26	**0.67**	—	—
Pb	—	**0.73**	—	—
Zn	—	0.33	**0.69**	—

*PC loadings <0.25 are omitted.

wither, lesser development, and persistence of progenies; Ni may cause skin allergies, lung fibrosis, dermatitis, and cancer of the respiratory tract; and Pb may affect central nervous system especially in young children, kidney, and cardiovascular system [64–66].

3.6. Multivariate Statistical Analyses. Principal component analysis (PCA) was used to find out the plausible contributing sources of selected metals in water reservoir. This method allows identifying different groups of metals that correlate and thus can be considered as having a similar behavior and common origin [67]. The numbers of significant PCs were selected on the basis of Kaiser criterion with eigenvalue greater than 1 [68]. Four PCs with eigenvalues >1 which explained about 85.3% of the total variance were obtained. The corresponding PCs, variable loadings, and the explained variance are shown in Table 4. First component (PC 1), which accounted for 38.6% of the total variance, showed strong positive loadings (>0.70) for Cd and Co. The second component (PC 2) contributed to Cr, Ni, and Pb elucidating 20.4% of total variance. These two components could be attributed to anthropogenic activities in the area, such as agricultural activities, discharge of untreated poultry, municipal/industrial wastes, and atmospheric deposition [69–72]. The studied area is under tremendous pressure of urbanization, industrialization, land cultivation, reservoir upgradation, and vehicular load. The third component (PC 3) which accounted for 15.7% of the total variance was associated with Cu and Zn, whereas PC 4 (10.6% of total variance) showed elevated loadings for Fe and Mn. The association of Cu, Fe, Mn, and Zn in the Lake reflected their natural background levels. Table 2 revealed that mean concentrations of these metals never exceeded the maximum permitted concentrations established by EC, WHO, Pak-EPA, and USEPA water quality guidelines. Therefore, the metals were predominantly derived from natural sources such as weathering of parent material and subsequent pedogenesis.

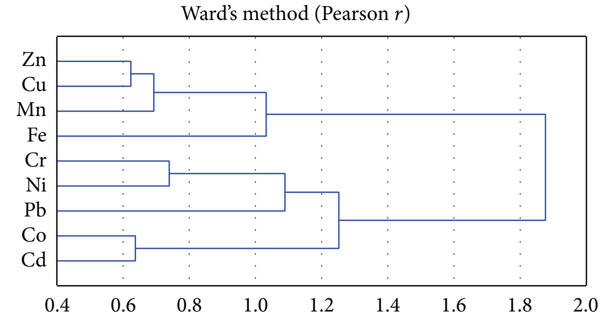

Figure 3: Dendrogram showing clustering of trace metals in Mangla Lake.

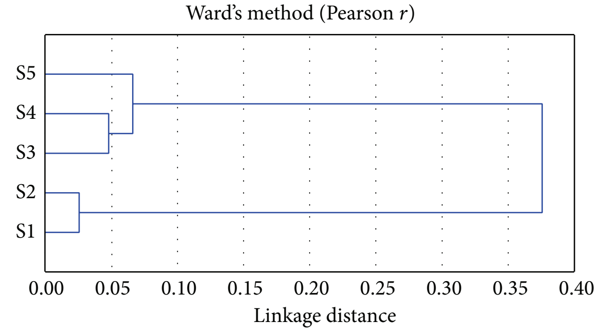

Figure 4: Dendrogram showing clustering of sampling sites in Mangla Lake.

Cluster analysis (CA) of the metal data was performed to explore the grouping of trace metals in surface water of Mangla Lake, Pakistan. It is shown as a dendrogram in Figure 3, where the metals were grouped into two main clusters. Cluster 1 consisted of Cu-Zn-Mn-Fe, while cluster 2 was composed of Cd-Co-Pb-Ni-Cr. The metals in each cluster had similar characteristic features and possibly similar inputs. Consequently, the metals grouped in cluster 1 were considered to be contributed by natural inputs, whereas the metals in cluster 2 were likely to be included by anthropic intrusions in surface water of Mangla Lake, Pakistan. In this study, exactly similar elemental grouping was shown by PCA and CA; Cd shared cluster and PC with Co, Cu with Zn, Mn with Fe, and Cr with Ni-Pb.

Cluster analysis (CA) was also applied to the dataset to group the similar sampling sites (spatial variability). Spatial CA rendered a dendrogram (Figure 4), where all five sampling sites were grouped into three statistically significant clusters. Cluster 1 consisted of site S5; cluster 2 consisted of two sites, S3 and S4; and cluster 3 consisted of two sites, S1 and S2. The sites in each cluster had similar characteristic features and possibly similar sources. The results manifested that sites S3 and S4 had the highest concentrations of Cd, Co, Cr, Ni, and Pb; site S5 exhibited moderate metal concentrations, while sites S1 and S2 had the lowest concentrations of the studied metals. Consequently, sites S3 and S4 were relatively heavily polluted by the metals; site S5 was moderately polluted, whereas sites S1 and S2 indicated low metal pollution in the Lake.

Table 5: Hazard quotient (HQ) and hazard index (HI) for each metal in surface water of Mangla Lake.

Metal	RfD_{ing} (μg/kg-day)	RfD_{derm} (μg/kg-day)	HQ_{ing} Child	HQ_{ing} Adult	HQ_{derm} Child	HQ_{derm} Adult	$\text{HI} = \sum \text{HQs}$ Child	$\text{HI} = \sum \text{HQs}$ Adult
Premonsoon								
Cd	0.5	0.025	8.3E + 00	2.2E + 00	6.1E − 01	2.1E − 01	9.0E + 00	2.4E + 00
Co	0.3	0.06	9.0E + 01	2.4E + 01	6.6E − 01	2.2E − 01	9.1E + 01	2.4E + 01
Cr	3	0.075	2.9E + 00	7.5E − 01	8.4E − 01	2.9E − 01	3.7E + 00	1.0E + 00
Cu	40	8	5.8E − 02	1.5E − 02	1.1E − 03	3.6E − 04	5.9E − 02	1.6E − 02
Fe	700	140	2.1E − 02	5.5E − 03	3.9E − 04	1.3E − 04	2.1E − 02	5.6E − 03
Mn	24	0.96	5.6E − 02	1.6E − 02	5.8E − 03	2.0E − 03	6.1E − 02	1.8E − 02
Ni	20	5.4	1.5E + 00	4.7E − 01	9.8E − 03	3.3E − 03	1.5E + 00	4.8E − 01
Pb	1.4	0.42	2.2E + 01	7.3E + 00	1.4E − 01	4.6E − 02	2.2E + 01	7.3E + 00
Zn	300	60	1.2E − 02	3.1E − 03	1.3E − 04	4.5E − 05	1.2E − 02	3.2E − 03
Monsoon								
Cd	0.5	0.025	7.2E + 00	1.9E + 00	5.3E − 01	1.8E − 01	7.7E + 00	2.1E + 00
Co	0.3	0.06	6.0E + 01	1.6E + 01	4.4E − 01	1.5E − 01	6.1E + 01	1.6E + 01
Cr	3	0.075	2.6E + 00	6.7E − 01	7.6E − 01	2.6E − 01	3.3E + 00	9.3E − 01
Cu	40	8	4.1E − 02	1.1E − 02	7.6E − 04	2.6E − 04	4.2E − 02	1.1E − 02
Fe	700	140	1.5E − 02	3.8E − 03	2.7E − 04	9.0E − 05	1.5E − 02	3.9E − 03
Mn	24	0.96	3.5E − 02	1.0E − 02	3.6E − 03	1.2E − 03	3.8E − 02	1.1E − 02
Ni	20	5.4	6.0E − 01	1.9E − 01	3.9E − 03	1.3E − 03	6.1E − 01	1.9E − 01
Pb	1.4	0.42	1.5E + 01	4.9E + 00	9.1E − 02	3.1E − 02	1.5E + 01	4.9E + 00
Zn	300	60	3.0E − 03	7.9E − 04	3.3E − 05	1.1E − 05	3.0E − 03	8.0E − 04
Postmonsoon								
Cd	0.5	0.025	4.2E + 00	1.1E + 00	3.1E − 01	1.0E − 01	4.5E + 00	1.2E + 00
Co	0.3	0.06	3.9E + 01	1.0E + 01	2.9E − 01	9.8E − 02	4.0E + 01	1.0E + 01
Cr	3	0.075	8.1E − 01	2.1E − 01	2.4E − 01	8.0E − 02	1.0E + 00	2.9E − 01
Cu	40	8	3.9E − 02	1.0E − 02	7.2E − 04	2.4E − 04	4.0E − 02	1.1E − 02
Fe	700	140	1.8E − 02	4.7E − 03	3.3E − 04	1.1E − 04	1.8E − 02	4.8E − 03
Mn	24	0.96	8.0E − 02	2.4E − 02	8.3E − 03	2.8E − 03	8.8E − 02	2.7E − 02
Ni	20	5.4	6.0E − 01	1.9E − 01	3.9E − 03	1.3E − 03	6.0E − 01	1.9E − 01
Pb	1.4	0.42	8.4E + 00	2.8E + 00	5.2E − 02	1.8E − 02	8.4E + 00	2.8E + 00
Zn	300	60	4.5E − 03	1.2E − 03	4.9E − 05	1.7E − 05	4.5E − 03	1.2E − 03

3.7. Human Health Risk Assessment. Table 5 shows the hazard quotient (HQ), hazard index (HI), and risk values for the oral and dermal exposures in premonsoon, monsoon, and postmonsoon seasons relating to children and adults. In premonsoon season, HQ$_{\text{ing}}$ (hazard quotient by ingestion) of Cd, Co, Cr, Ni, and Pb for children was more than 1 implying that it may cause adverse health effects and potential noncarcinogenic concern. In case of adults, HQ$_{\text{ing}}$ of Cd (2.2), Co (24), and Pb (7.3) was much higher than unity and Cr (0.75) was nearing unity indicating its serious health concerns. However, HQ$_{\text{ing}}$ of the remaining metals for children and adults was less than 1, suggesting that these metals posed little/no hazard through direct intake of the Lake water. The HQ$_{\text{derm}}$ (hazard quotient by dermal absorption) of all the metals for adults and children was below unity, indicating that these metals posed little/no hazards via dermal absorption. The largest values of HQ$_{\text{derm}}$ were 0.84, 0.66, and 0.61 which were for Cr, Co, and Cd for children, respectively, demonstrating that these metals could cause potential adverse health effects via dermal absorption. Overall, HI of Cd, Co, Cr, and Pb for adults and children exceeded 1, and HI of Ni for children was near 1. It can be concluded that the highest contributors to chronic risks were Cd, Co, Cr, Ni, and Pb, while the lowest were Fe, Mn, and Zn for both the adults and children.

In monsoon season, HQ$_{\text{ing}}$ for Cd, Co, Cr, and Pb for children was higher than unity, while in case of adults, HQ$_{\text{ing}}$ for Cd, Co, and Pb was higher than 1. However, HQ$_{\text{derm}}$ was lower than unity for children and adults demonstrating that these metals posed no significant adverse health effects via dermal absorption. Generally, HI of Cd, Co, and Pb for children and adults exceeded 1 and HI of Cr was higher than unity for children and was near unity (0.93) for adults. Cd, Co, Cr, and Pb emerged as major pollutants in this season. In postmonsoon season, HQ$_{\text{ing}}$ for children of Cd, Co, and Pb was higher than one and Cr and Ni were nearing one indicating potential noncarcinogenic adverse health risks via oral intake. For adults, HQ$_{\text{ing}}$ for Cd, Co, and Pb was higher than unity.

As a whole, it was revealed that Cd, Co, Cr, Ni, and Pb could pose severe health effects to the inhabitants through ingestion pathway in the studied seasons, whereas the remaining metals through oral pathway and all the studied metals via dermal pathway could cause little/no health concerns. Consequently, distinctive consideration should be paid to manage these toxic elements and to support healthy aquatic ecosystem. Nonetheless, uncertainties related to methodological features, such as water and dermal contact factor (K_p), different exposure conditions, and temporal and spatial disparities in pollutants levels were not quantified. Furthermore, employed parameters were taken from USEPA, WHO, and elsewhere; they could not be definite to local situations. Therefore, a more detailed risk characterization related to the risk levels by the metals in the Mangla Lake is suggested.

4. Conclusions

Concentrations of trace metals (Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) in the water of the Mangla Lake, Pakistan, demonstrated significant seasonal variations. Irrespective of the sampling sites, the metal concentrations were lower in postmonsoon season compared to other seasons due to marked dilution effect. The measured concentrations of Cd, Co, Cr, Ni, and Pb were recorded higher than water quality guidelines. PCA and CA indicated that both geogenic and anthropogenic activities were contributing factors to metal abundance in the Lake. Cluster analysis grouped all sampling sites into three major clusters. Higher concentrations of metals were found near urban areas revealing that their concentrations had been strongly affected by anthropogenic influences. Thus, it was logical to conclude that the elevated concentrations of metals in Lake water were considerably due to direct discharge of untreated municipal/industrial wastes into the Lake. Human health risk was assessed using exposure risk assessment model which indicated Cd, Co, Cr, Ni, and Pb were the most prevalent pollutants causing noncarcinogenic concerns in all seasons and the oral ingestion was the major exposure pathway. Therefore, special attention should be paid to manage Cd, Co, Cr, Ni, and Pb in the study area and measures needed to be taken for sustaining the healthy aquatic ecosystem.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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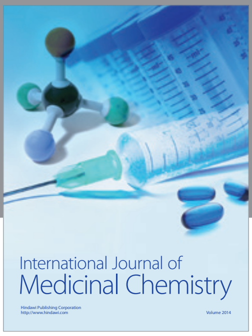
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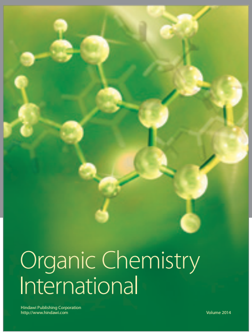
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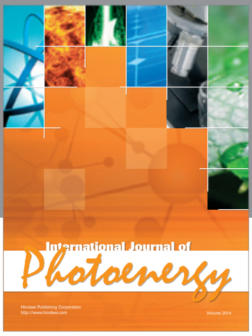
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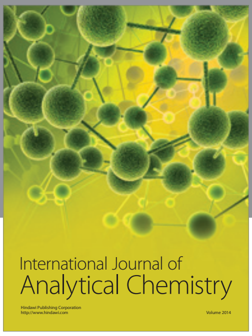
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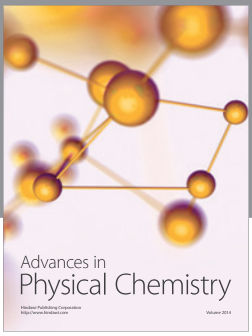
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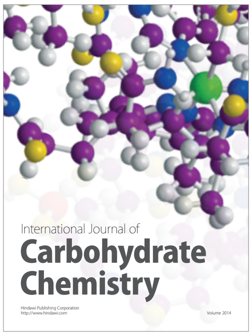
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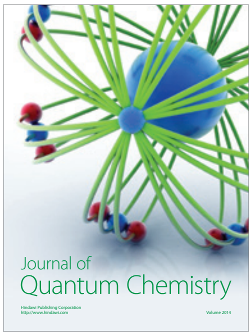
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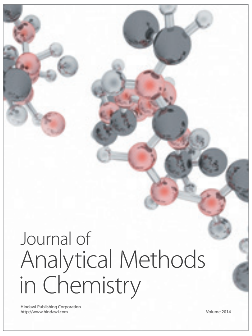
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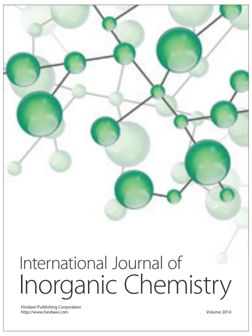
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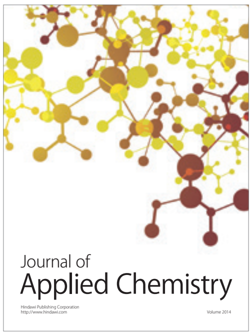
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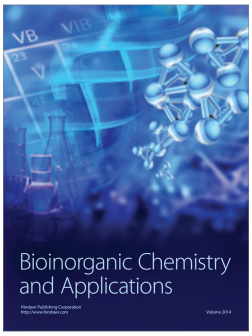
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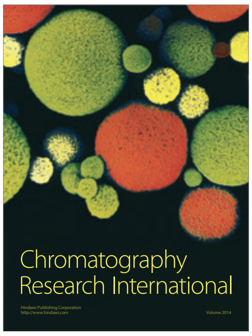
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