



## Article

# Multivariate Statistical Methods and GIS-Based Evaluation of Potable Water in Urban Children's Parks Due to Potentially Toxic Elements Contamination: A Children's Health Risk Assessment Study in a Developing Country

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**Abstract:** Contamination of potentially toxic elements (PTEs) has received widespread attention in urban children's parks (UCPs) worldwide in the past few decades. However, the risk assessment of PTEs in drinking water sources of UCPs is still unknown particularly in developing countries. Hence, the present study investigated the spatial distribution, sources for PTEs (Cd, Cr, Pb, Ni, and Cu), and health risk assessment in drinking water sources of UCPs in Khyber Pakhtunkhwa, Pakistan. Among PTEs, Cd, Cr, and Pb had low to high concentrations and exceeded the safe limits of WHO and PAK-EPA in most UCPs. PCA results showed high anthropogenic and low natural sources, contributing to the release of PTEs in all UCPs. Heavy-metal pollution index (PTE-PI) results showed low to high pollution levels for all UCPs, with the highest values of 113 and 116 for Sardaryab Park Charsadda (SPC) and Zoo Park Peshawar (ZPP), respectively. Heavy-metal evaluation index (PTE-EI) results also showed low to high pollution levels for all UCPs. UCPs samples (50%) showed low pollution levels in PTE-PI results. To the contrary, UCPs samples (50%) exhibited high pollution levels in PTE-EI results. The non-carcinogenic risk of HQ and HI values of all PTEs were below the permissible limit ( $<1$ ) for adults and children via ingestion and dermal contact. CR and TCR results showed that PTEs (Cr, Cd, Pb, and Ni) had the highest carcinogenic risk ( $>1.00 \times 10^{-4}$ ) for both adults and children in all UCPs, except Cd and Ni for adults via the ingestion route, while Cr values ( $>1.00 \times 10^{-4}$ ) were exceeded for children in some of the UCPs via the dermal route. Consequently, long-term exposure to toxic PTEs could pose a carcinogenic risk to the local population. Thus, the present study suggests that the government should implement enforcement with firm protocols and monitoring guidelines of environmental regulations to mitigate PTEs originating from anthropogenic sources in order to reduce health risks and improve public health safety in urban areas.

**Keywords:** toxic elements; pollution indices; source identification; water quality; risk assessment

## 1. Introduction

According to the world urbanization prospects of the United Nations (UN), 55% of the world's population resides in urban areas [1]. Urban parks play a critical role in maintaining the urban ecosystem with the rapid development of urbanization [2,3]. As a result, high amounts of pollutants are accumulated in the urban environment due to rapid urbanization, high energy consumption, and migrations of people into big cities [4,5]. Pollutants like potentially toxic elements (PTEs) are released into the urban park soils and the surrounding environment as a consequence of high urbanization and industrialization [6–8]. The primary sources of PTEs are heavy traffic and intensive human activities (such as the dumping of municipal and industrial wastes, home heating, industrial emissions, and energy generation), leading to contamination of parks and roadside lawns in urban environments [9–11]. PTE contamination in urban children's parks (UCPs) is associated with traffic emissions, fuel combustion, tire and brake wear, particles of weathered street dust, waste discharge from industrial activities, weathering particles from buildings and pavement surfaces, urban infrastructure development, wastewater disposal systems, and dumping of municipal solid waste [12–17]. Moreover, urban soils are exposed to PTE input from natural sources, wastewater irrigation, and atmospheric deposition [18–21], resulting in being discharged into water bodies by natural weathering, the erosion process, and anthropogenic activities [22]. Typically, urban runoff contains a range of pollutants that could contaminate water quality [23,24], and such pollutants like PTEs degrade the drinking water sources and leach into groundwater system.

Residents can be exposed to PTEs by ingestion, inhalation, and skin contact, especially children and adults who often visit urban parks [3]. PTEs can pose substantial risks to the public's health once they have bioaccumulated, especially to the most susceptible groups, such as children and senior citizens [13]. Furthermore, visitors and locals commonly engage in recreational activities in public parks, and exposure to elevated PTEs can hinder a child's development, cause abnormal behavior, or lead to other chronic disorders, while others have the potential to cause cancer when exposed to high concentrations [25]. In general, prolonged exposure to PTEs may cause a variety of harmful health effects, including cancer, immune system disruption, neurological problems, cardiovascular and liver diseases, kidney and bone malfunction, and immune system disruption [26]. Due to the fact that children and other park visitors are more susceptible to PTE toxicity, it is crucial to identify the risks of exposure to drinking water sources in urban parks [27]. So far, multivariate statistical analyses like principle component analysis multiple linear regression (PCA-MLR) have been widely used for source identification of PTEs [28,29]. Therefore, it is crucial to identify the potential sources of PTEs and high risk areas in urban parks in order to prevent and control PTE pollution [30].

Several studies have shown contamination by PTEs such as Cr, Ni, Cd, As, Cu, Pb, and Zn in urban soils in parks, sports playgrounds, and schools [31–34], with most of the recent studies focused on PTE concentration, potential sources, and health risk assessment in UCP soils. PTE pollution was also reported in various research in some of the urban lakes [35,36]. The PTEs in urban environments have been a major concern for researchers and environmentalists worldwide. Research on PTE contamination in drinking water sources of urban parks and the urban environment is very limited. However, to the best of our knowledge, this will be the first study to assess PTE contamination, its potential source, and health risk evaluation in UCP drinking water sources of the Khyber Pakhtunkhwa (KPK) province in Pakistan. Parks in this province serve as popular recreation areas and tourism destinations for the local citizens. PTEs in the UCP drinking water could pose a serious threat to human health. As the proportion of people living in urban areas is predicted to expand from 50% today to 66% by 2050 worldwide [29], identifying and remediating PTEs-contaminated soils and drinking water may become more critical. Evidently, research into the health risks posed by PTEs in drinking water sources of UCPs is still unknown in developing countries like Pakistan. To fill this research gap, a scientific approach is needed to evaluate the PTEs pollution in UCPs and its origins to minimize

the health risks via drinking water ingestion and dermal contact of exposure pathways for children and adults. This study could be important to provide scientific support for controlling and prevention of PTEs contamination in order to improve healthy urban environments worldwide, as proposed by the World Health Organization (WHO).

Therefore, we have selected a total of 20 UCPs in famous regions of the KPK province. The primary objectives of the present study were as follows: (1) to investigate the spatial distribution of PTEs (Cd, Cr, Pb, Ni, and Cu) in selected UCPs; (2) to assess the PTEs contamination and potential sources by PCA-MLR; and (3) to assess their possible risks (carcinogenic and non-carcinogenic) to human health, via ingestion and dermal contact, for the local population (adults and children). This research can be used as a reference basis in future studies for proper management of PTE contamination in urban parks. The findings of this study can help decision makers and can alert inhabitants around the world to manage and remediate PTEs contamination in soil and drinking water sources with the guidelines and remediation strategies in urban environments.

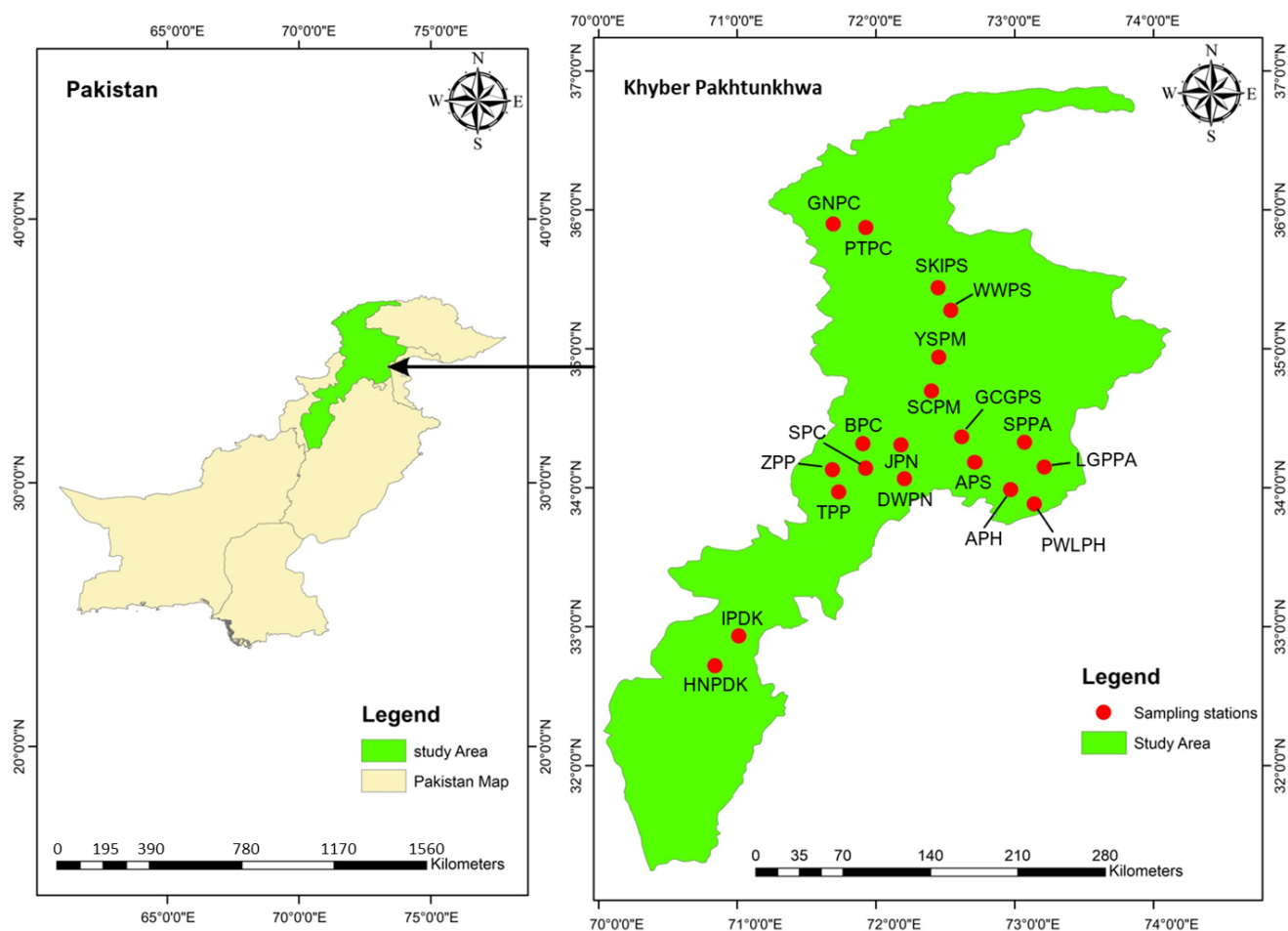
## 2. Materials and Methods

### 2.1. Study Area, Water Sampling, and Analysis

The water samples were collected from a total of 20 UCPs of 10 cities in the Khyber Pakhtunkhwa province of Pakistan in 2019, as shown in Figure 1. All the UCPs were selected based on high population and industrialized areas, and the parks were also used for different recreational activities by the local children and adults. The UCPs are categorized with specific acronyms, including Baghicha Park Charsadda (BPC), Sardaryab Park Charsadda (SPC), Zoo Park Peshawar (ZPP), Tatara Park Peshawar (TPP), Jannah Park Nowshera (JPN), Disney Water Park Nowshera (DWPN), Pak Wonder Land Park Haripur (PWLPH), Ayub Park Haripur (APH), Sher Khan International Park Swat (SKIPS), Wonder World Park Swat (WWPS), Albel Park Swabi (APS), Gohati Cricket Ground Park Swabi (GCGPS), Sports Complex Park Mardan (SCPM), Younus Stadium Park Mardan (YSPM), Gol National Park Chitral (GNPC), Pakistan Tour Park Chitral (PTPC), Haq Nawaz Park Dera Ismail Khan (HNPDK), Insaf Park Dera Ismail Khan (IPDK), Lady Garden Public Park Abbottabad (LGPPA), and Shimla Pahari Park Abbottabad (SPPA).

The research methodology (see Figure 2) was followed for drinking water sample collection with the proper guidelines, and all the samples were collected in the selected UCPs. The water samples were randomly collected from different drinking water sources including bore wells, hand pumps, and tap water in UCPs. The total geographical area of each park was varied, and water samples were collected from the abovementioned sources. Based on the total area of each UCP, 10 water samples were collected from drinking water sources. A total of ( $n = 200$ ) drinking water samples were collected from all the 20 UCPs in all selected cities. The drinking water resources in these UCPs are regularly used for drinking and other domestic purposes. Before sample collection, water was allowed to run for a few minutes during the sampling time from the specific target in each park. The polyethylene bottles were first cleaned with distilled water, then water samples were taken. A few drops of acid ( $\text{HNO}_3$ , 0.5%  $v/v$ ) were added to bottles prior to PTE analysis. All the water samples were filtered by Whatman filter paper (0.45  $\mu\text{m}$ ). All the samples were sealed, labeled, and well preserved using the method used by [37].

The water samples were directly used for the physicochemical parameters analysis (Figure 2). pH and total dissolved solids (TDS) were determined using a digital pH meter (Model C93, Turnhout, Belgium) and a TDS meter (Model S518877), respectively, while electrical conductivity (EC) was measured by a conductivity meter (Model HI98303). The water samples were analyzed for PTEs including cadmium (Cd), chromium (Cr), lead (Pb), nickel (Ni) and copper (Cu) by graphite furnace atomic absorption spectrophotometry (PerkinElmer, Waltham, MA, USA, ASS-PEA-700) using standard procedures adopted by [15].

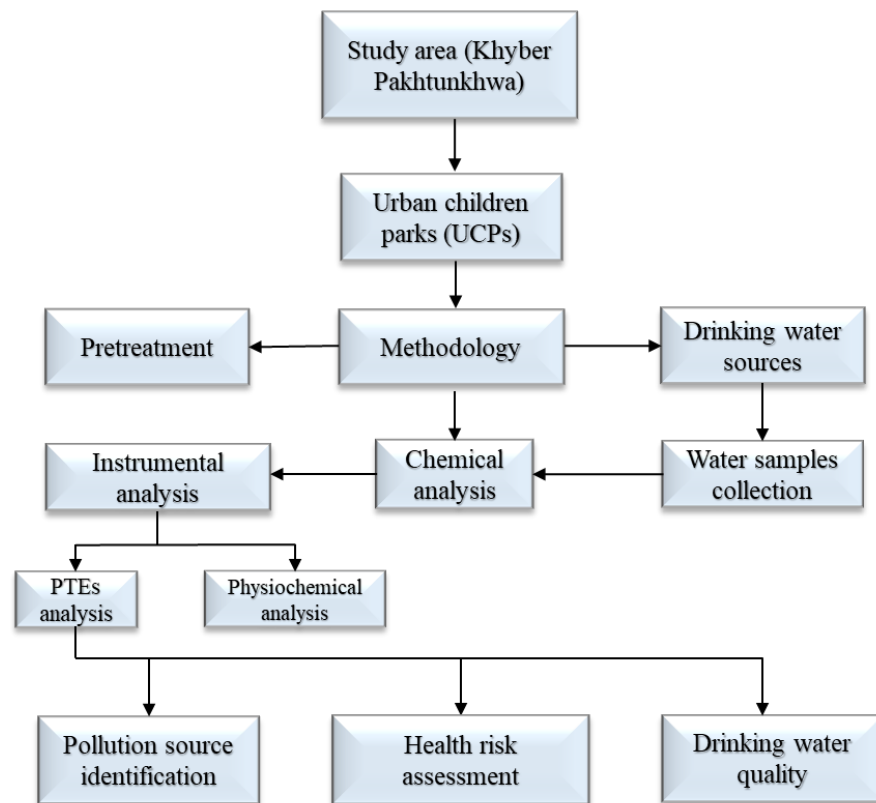


**Figure 1.** Study area map showing the sampling points of 20 urban children's parks in Khyber Pakhtunkhwa province, Pakistan. Baghicha Park Charsadda (BPC), Sardaryab Park Charsadda (SPC), Zoo Park Peshawar (ZPP), Tatara Park Peshawar (TPP), Jannah Park Nowshera (JPN), Disney Water Park Nowshera (DWPN), Pak Wonder Land Park Haripur (PWLPH), Ayub Park Haripur (APH), Sher Khan International Park Swat (SKIPS), Wonder World Park Swat (WWPS), Albela Park Swabi (APS), Gohati Cricket Ground Park Swabi (GCGPS), Sports Complex Park Mardan (SCPM), Younus Stadium Park Mardan (YSPM), Gol National Park Chitral (GNPC), Pakistan Tour Park Chitral (PTPC), Haq Nawaz Park Dera Ismail Khan (HNPDK), Insaf Park Dera Ismail Khan (IPDK), Lady Garden Public Park Abbottabad (LGPPA), and Shimla Pahari Park Abbottabad (SPPA).

## 2.2. PTEs Pollution Indices

### 2.2.1. Potentially Toxic Elements Pollution Index (PTE-PI)

The PTE-PI is a measurement that reflects the combined impact of several dissolved PTEs and indicates the overall quality of water with regard to PTE concentration [38]. PTE-PI is an effective technique used to assign a specific rating/weighting ( $W_i$ ) for each selected parameter. The rating is based on a value between 0 and 1, indicates the relative importance of each parameter, and can be interpreted as inversely related to the suggested standard ( $S_i$ ) for individual parameter [39,40].



**Figure 2.** Research flow chart of the PTEs study in drinking water sources of UCPs.

Different PTE-PI ranges have been specified and used to classify water quality as “poor”, “good”, “very good”, and “excellent”. The critical level of the PTE-PI value is 100, and it has been reported that PTE-PI levels above 100 have a stronger negative impact on health [41]. PTE-PI can be calculated by using Equations (1) and (2):

$$\text{PTE-PI} = \frac{\sum_{i=1}^n (Q_i \times W_i)}{\sum_{i=1}^n W_i} \quad (1)$$

$$Q_i = \frac{M_i}{S_i} \times 100 \quad (2)$$

where  $Q_i$  and  $W_i$  represent the sub-index and unit weightage of the  $i$ th parameter, respectively, and  $n$  is the total number of the parameters.  $M_i$  is the concentration of the measured  $i$ th parameter, and  $S_i$  is the concentration of standard maximum permissible values ( $\mu\text{g/L}$ ) of WHO [42], respectively, as shown in Table S1, described by the European Directive on water quality [43]. The weightage unit ( $W_i$ ) signifies an inverse proportional to the maximum concentration level.

$$W_i = \frac{1}{S_i} \quad (3)$$

### 2.2.2. Potentially Toxic Elements Evaluation Index (PTE-EI)

The PTE-EI is also used to evaluate the combined impact of measured PTEs on the overall drinking water quality [39]. In the present study, five PTEs (Cd, Cr, Pb, Ni, and Cu) were selected and assessed for PTE-PI and PTE-EI calculation. HEI was computed by the given Equation (4):

$$\text{PTE-EI} = \sum_{i=1}^n \frac{H_c}{H_{mac}} \quad (4)$$

where  $H_c$  is the observed value of the  $i$ th parameter ( $\text{mg/L}$ ), while  $H_{mac}$  is the maximum permissible concentration of the  $i$ th parameter [44].

### 2.3. Human Health Risk Model

#### 2.3.1. Non-Carcinogenic Risk Assessment

The health risk model is used to determine the extent of human exposure to toxic PTEs, developed by the United States Environmental Protection Agency (USEPA) [45], in the drinking water of UCPs. Risk assessment is described as the method of estimating the likelihood of any given likely amount of detrimental health impacts occurring over a specified time period [46]. Human health risk assessment was estimated in recent studies to demonstrate the potential health risks posed by PTEs in water, soil, and air [47,48]. The risk assessment is one of the most effective ways of providing crucial information, highlighting the most critical metals among heavy metals, which have a negative influence on human health and the environment due to their toxicity. The most useful aspect of health risk assessment is that it provides information and depicts an inhabitant's health status. Children and adults are usually exposed to toxic PTEs in UCPs drinking water by two main exposure pathways, i.e., ingestion and dermal contact. Furthermore, adults are regarded as the general population, while children are considered the sensitive group. The daily metal intake (*DMI*) was used to quantify the exposure to PTEs in UCPs water through ingestion and dermal contact. The daily metal intake via ingestion (*DMI<sub>ing</sub>*) and the daily metal intake via dermal contact (*DMI<sub>der</sub>*) of PTEs in UCP water were calculated according to Equations (5) and (6), respectively.

$$DMI_{ing} = \frac{C \times IR \times EF \times ED}{BW \times AT} \quad (5)$$

$$DMI_{der} = \frac{C \times SA \times Kp \times ET \times ED \times EF \times CF \times ABS}{BW \times AT} \quad (6)$$

The descriptions of the parameters for the health risk model are presented in Table S2. Non-carcinogenic toxic risk was estimated by calculating the hazard quotients (*HQ*) and hazard index (*HI*). The hazard quotient (*HQ*) was calculated as the proportion of the *DMI* and the reference dose (*RfD*) for a specific PTE. The total non-carcinogenic risks (*HI*) were identified by the sum of the *HQ* values of all selected PTEs in the UCPs water. The *HQ<sub>ing</sub>*, *HQ<sub>der</sub>*, and *HI* of all PTEs in UCPs water was calculated according to Equations (7)–(9).

$$HQ_{ing} = \frac{DMI_{ing}}{RfD_{ing}} \quad (7)$$

$$HQ_{der} = \frac{DMI_{der}}{RfD_{der}} \quad (8)$$

$$HI = \sum HQ = \sum \frac{DMI}{RfD} \quad (9)$$

where *DMI* is the average daily exposure dose through ingestion and dermal contact. *RfDs* are the reference doses of selected PTEs for ingestion and dermal contact. All *RfD* values of non-carcinogenic risk for both exposure pathways are presented by the United States Department of Energy (USDOE) [49], as shown in Table S2. The values of *HQ* and *HI* greater than 1 indicate non-carcinogenic risk to human health in the exposed population, while *HQ* and *HI* values less than 1 are considered safe.

#### 2.3.2. Carcinogenic Risk Assessment

The carcinogenic risk of toxic PTEs was calculated by using the carcinogenic risk (*CR*), and the total cancer risk (*TCR*) represents the sum of the potential carcinogenic risks. The estimated *CR* value shows the probability of developing cancer risk for an individual during a lifetime exposure to carcinogenic toxic chemicals. The *CR* was calculated for PTEs (Cr, Cd, Pb, and Ni) in the present study, based on their observed corresponding *DMI* and available cancer slope factor (*CSF*) values. The non-carcinogenic and total carcinogenic

risks can be identified by combining the overall risks for three exposure routes in the UCPs drinking water. The  $CR$  for two exposure pathways and  $TCR$  were calculated by using Equations (10)–(12) [50]:

$$CR_{ing} = DMI_{ing} \times CSF \quad (10)$$

$$CR_{der} = DMI_{der} \times CSF \quad (11)$$

$$TCR = \sum CR \quad (12)$$

where  $CSF$  is the cancer slope factors (per mg/kg-day) as shown in Table S2. In general, the carcinogenic risk is unacceptable if the  $CR$  values exceed ( $>10^{-4}$ ), while  $CR$  values ( $1 \times 10^{-6} < CR < 1 \times 10^{-4}$ ) are assumed to be acceptable with no carcinogenic risk [51], whereas the  $CR$  values ( $<10^{-6}$ ) imply that the carcinogenic risk can be negligible [45].

#### 2.4. Quality Assurance and Quality Control

The standard procedure was used to certify the PTEs quality in order to confirm the accuracy of the results. Double distilled water and certified standard solution (1000 mg/L) of Fluka Kamica (Buchs, Switzerland) were used for quality control of the standard solution and analysis of PTEs. The analytical chemical spectroscopic purity of 99.9% (Merck Darmstadt, Germany) was also used for the sample formation and PTEs analysis. The reagent blanks, duplicate samples, and standards were used with different concentrations. Analytical estimated error was less than or equal to 10%, and the reproducibility of the analytical results was within 5%.

#### 2.5. Statistical Analysis

Descriptive statistics were performed using Microsoft Excel (2016) and SPSS (version 21). PCA-MLR was used for PTEs contamination sources using XLSTAT (2022). Origin Lab (2018) and SigmaPlot (14.0) were used to make all the figures. Arc Geographic Information System (Arc-GIS 10) software was used for the study area map and distribution maps of PTEs.

### 3. Results

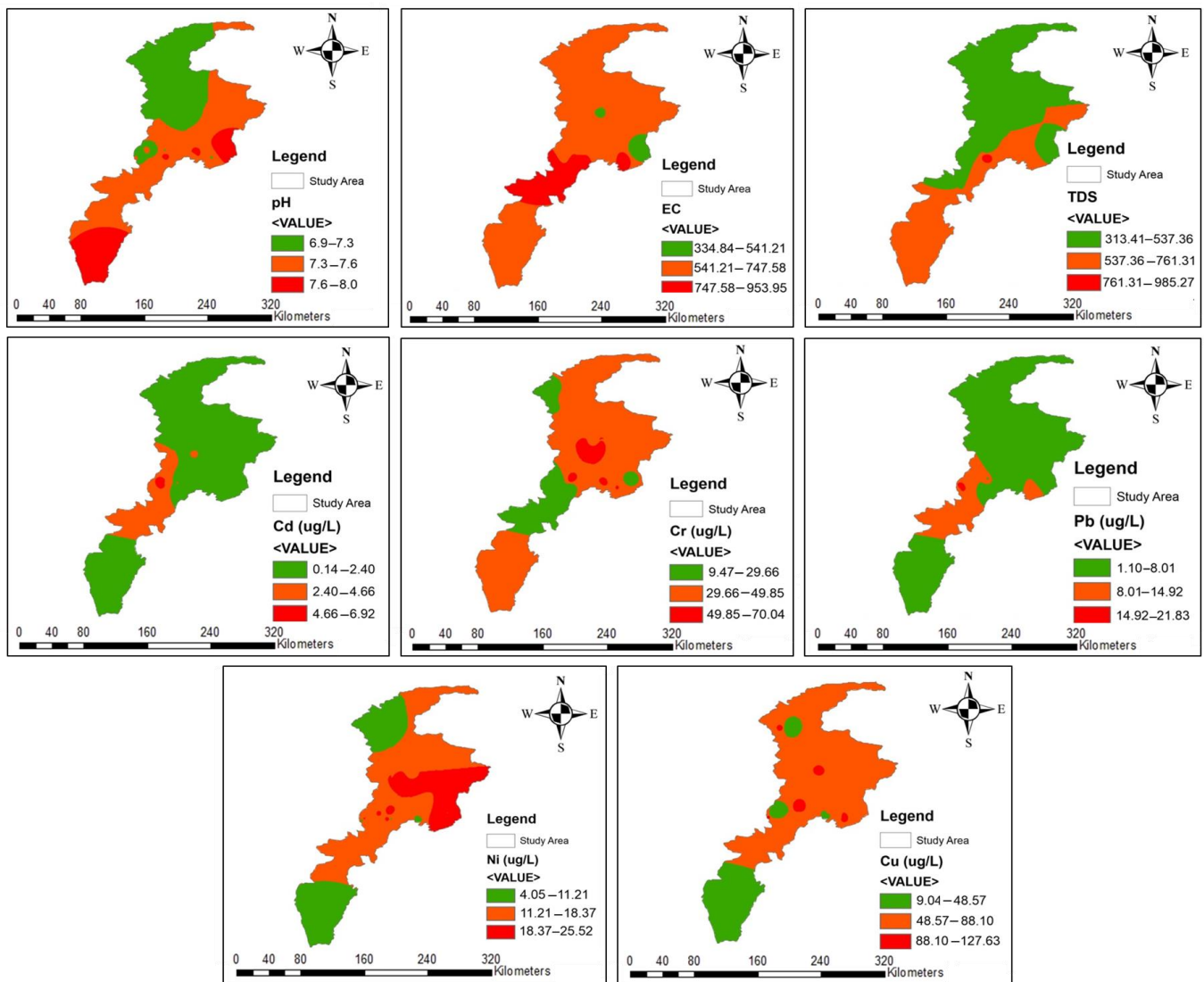
#### 3.1. Physiochemical Characteristics of UCPs Water

The physiochemical properties of all UCPs water samples are presented in Table S3 and Figure 3. There is a substantial variation in the concentration of physiochemical parameters in drinking water of all UCPs. The overall pH varied from 6.10 to 8.46 for drinking water of all the UCPs, suggesting that it is neutral to alkaline in nature. The lowest mean value of pH (6.95) was recorded for BPC and PTPC, while a high pH value (8.03) was observed for LGPPA. The EC values were varied for drinking water of all UCPs, ranged from 192 to 1394  $\mu\text{S}/\text{cm}$ . The lowest EC mean value was recorded for LGPPA (331  $\mu\text{S}/\text{cm}$ ), and the highest EC mean value was detected for APH (959  $\mu\text{S}/\text{cm}$ ). Furthermore, the concentration of total dissolved solids (TDS) of all UCPs samples was observed between 220–1245  $\mu\text{g}/\text{L}$ . The highest TDS value was recorded for DWPB, with a mean value of 986  $\mu\text{g}/\text{L}$ , while the lowest TDS was observed for BPC with a mean value of 313  $\mu\text{g}/\text{L}$ . These physiochemical parameters possibly influence and control the occurrence and bioavailability of PTEs in groundwater, which is further discussed in Section 4 below.

#### 3.2. Spatial Distribution of PTEs in UCPs Water

The basic descriptive statistics and spatial distribution of PTEs concentrations in UCPs drinking water are presented in Table S3 and Figure 3. The distribution levels of PTEs in the drinking water of all UCPs are greatly varied. Among PTEs, Cu had the highest concentration for all UCPs, followed by Cr, Ni, Pb, and Cd. Cu was observed with elevated concentration, ranging from 38.8–222  $\mu\text{g}/\text{L}$  with a mean value of 130  $\mu\text{g}/\text{L}$  for PWPB. The lowest concentration was recorded for Cd, ranged from 0.09–0.19  $\mu\text{g}/\text{L}$  with a mean

value of 0.14  $\mu\text{g/L}$  in the UCP water of PTPC. Cr concentrations exceeded the permissible limit (50  $\mu\text{g/L}$ ) set by WHO [52] in UCPs water of SPC, PWLPH, WWPS, SCPM, and YSPM, while its concentrations were found to be within acceptable limits of WHO [52] and PAK-EPA [53].



**Figure 3.** Spatial distribution of pH, EC, TDS, and PTEs (Cd, Cr, Pb, Ni, and Cu) in UCPs.

For BPC and SPC, Cu exhibited high mean concentrations (31.1 and 19.0  $\mu\text{g/L}$ ), followed by Ni (19.5 and 14.6  $\mu\text{g/L}$ ) and Cr (11.6 and 12.5  $\mu\text{g/L}$ ), respectively. A low mean concentration was recorded for Pb (8.49 and 9.37  $\mu\text{g/L}$ ) and followed by Cd (4.35 and 6.95  $\mu\text{g/L}$ ), respectively. For ZPP, Pb showed a high mean concentration of 21.9  $\mu\text{g/L}$ , followed by Ni (19.2  $\mu\text{g/L}$ ) and Cu (14.0  $\mu\text{g/L}$ ), respectively. The concentrations of all selected PTEs were found to be within allowable limits of WHO in all the UCPs water, indicating low contamination, except Cd. Noticeably, Cd concentration was above the permissible limit (3  $\mu\text{g/L}$ ) of WHO, with mean values of 4.35 and 4.05  $\mu\text{g/L}$  in UCPs water of BPC and ZPP, respectively. Similarly, Cu showed high abundance with mean values of 106 and 75.5  $\mu\text{g/L}$ , while Cd and Pb showed lowest concentrations with mean values (3.60 and 1.12  $\mu\text{g/L}$ ) and (9.54 and 2.45  $\mu\text{g/L}$ ) in TPP and JPN, respectively. Likewise, Cu exhibited high mean concentration (80.6  $\mu\text{g/L}$ ), while the lowest mean concentration (0.42  $\mu\text{g/L}$ ) was recorded for Cd in DWPN. Furthermore, the PTEs such as Cr and Ni concentrations were relatively comparable for PWLPH, APH, and SKIPS, with mean values



(51.4, 49.2, and 46.1  $\mu\text{g/L}$ ) and (25.8, 18.2 and 16.5  $\mu\text{g/L}$ ), respectively. To the contrary, the concentrations of Cu were comparatively higher than the other PTEs, with mean values of 130, 54.2, and 98.8  $\mu\text{g/L}$ , respectively, in the above-mentioned UCPs. Cr and Pb mean concentrations (51.4 and 15.2  $\mu\text{g/L}$ ) in PWLPH were found to be higher than the threshold values of WHO [52] and PAK-EPA [53], respectively. The mean concentrations of PTEs in drinking water showed high variation between different UCPs of KPK, indicating low to high contamination of Cd, Cr, and Pb, which exceeded the permissible limits of WHO [52] and PAK-EPA [53]. The PTEs concentrations were noticeably low to moderate level for WWPS and APS. Similarly, Cr was remarkably observed with high mean values of 68.3, 71.1, 61.5, and 61.3  $\mu\text{g/L}$  for WWPS, GCGPS, SCPM, and YSPM, respectively and exceeded the threshold limits of WHO [52] and PAK-EPA [53]. Cd exhibited low concentration levels (0.67 and 0.99  $\mu\text{g/L}$ ), followed by Ni (2.60 and 19.2  $\mu\text{g/L}$ ), for APS and YSPM, respectively. Among UCPs, the PTEs concentrations were comparatively lower, especially for Cd, Pb, and Ni in UCPs drinking water of GNPC, PTPC, HNPK, and IPDK. Cu showed high concentration for GNPC with mean value of 104  $\mu\text{g/L}$ , while to the contrary, PTPC, HNPK, and IPDK showed low Cu concentration with mean values of 8.5, 10.0, and 19.1  $\mu\text{g/L}$ , respectively, and Cr had moderate mean concentration in the aforementioned UCPs.

Similarly, LGPPA and SPPA showed relatively similar mean values of PTEs such as Cd (0.55 and 0.58  $\mu\text{g/L}$ ) and Cu (60.1 and 51.1  $\mu\text{g/L}$ ), respectively. The spatial distribution of high abundance of PTEs (especially Cu, Cr, Pb, and Cd) in all UCPs water confirms high contamination levels that could be subjected to the input of potential sources in the study areas. Overall, in the present study, the mean concentrations of PTEs occurred in descending order of  $\text{Cu} > \text{Cr} > \text{Ni} > \text{Pb} > \text{Cd}$  for all UCPs.

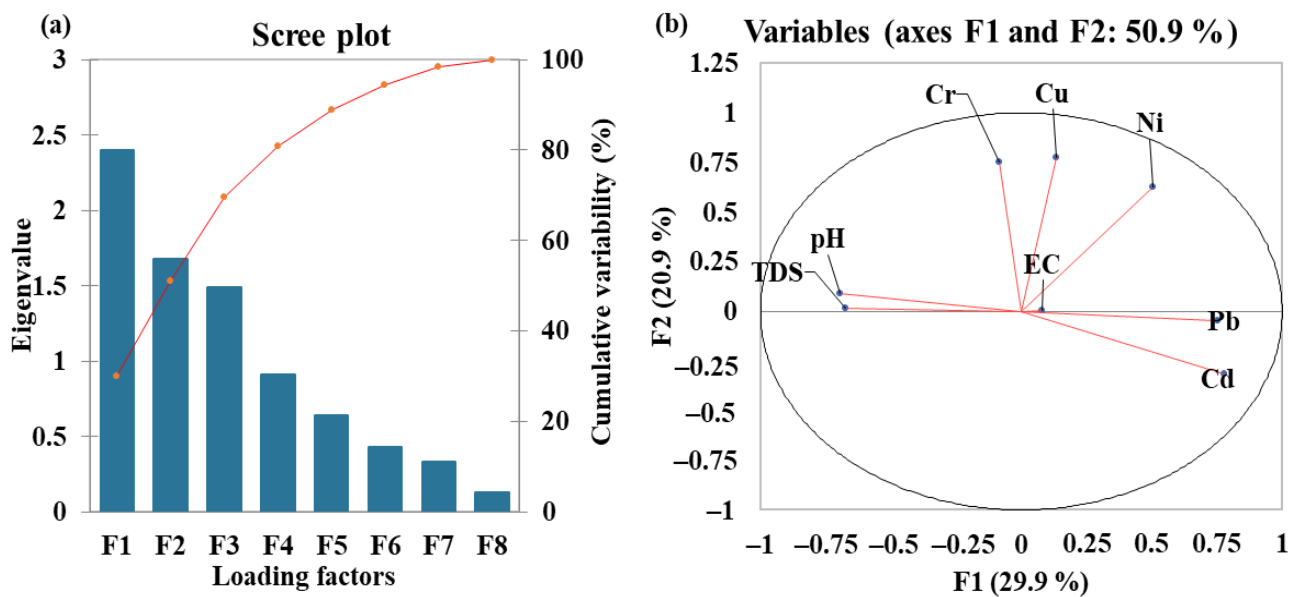
### 3.3. Source Apportionment of PTEs

Principle component analysis (PCA) is an effective technique used for source apportionment of PTEs [54]. In this study, we used PCA multilinear regression (PCA-MLR) to extract three major component factors in terms of eigenvalues (eigenvalue  $> 1$ ) and estimated total variance. The positive and negative loadings for different datasets of PTEs in UCPs are presented in Table 1. Overall, four significant factors (F1, F2, F3, and F4) were obtained, and the first two significant loading factors were observed for PTEs in UCPs, as presented in Figure 4. The positive loading factors imply that the presence of the water variables could influence the groundwater or surface water samples. Contrarily, the negative loading factors show that the groundwater and surface water quality are not affected by the water variables. The four factors of the UCPs water sources described 80.8% of the total variation with an eigenvalue of 6.56, as shown in Table 1.

**Table 1.** Principal components analysis of selected parameters of UCPs.

Parameters	F1	F2	F3	F4
pH	<b>-0.69</b>	0.09	0.14	<b>0.62</b>
EC	0.08	0.00	<b>0.90</b>	-0.33
TDS	<b>-0.68</b>	0.02	<b>0.66</b>	0.15
Cd mg/L	<b>0.78</b>	-0.31	0.15	0.26
Cr mg/L	-0.09	<b>0.75</b>	-0.22	-0.05
Pb mg/L	<b>0.75</b>	-0.05	0.34	0.21
Ni mg/L	<b>0.50</b>	<b>0.63</b>	0.10	0.45
Cu mg/L	0.13	<b>0.78</b>	0.18	-0.28
Eigenvalue	2.40	1.67	1.49	1.00
Variability (%)	29.9	20.9	18.6	11.3
Cumulative %	29.9	50.9	69.5	80.8

Note: Bold values are the main contributors to PCA.



**Figure 4.** (a) Significant loading factors and (b) overall loading factors of results of UCPs.

Factor 1 (F1) described 29.9% of the total variance with an eigenvalue of 2.40. F1 is predominantly loaded with PTEs such as Cd, Pb, and Ni, showing moderate positive loadings (0.78), (0.75), and (0.50), respectively, except pH and TDS have moderate negative loadings (−0.69) and (−0.68), respectively. F1 contributed to the moderate loadings of PTEs for the UCPs drinking groundwater, suggesting anthropogenic and natural sources. The negative loadings of pH and TDS could be associated with the features and mobility of PTEs [55]. Factor 2 (F2) described 20.9% of the total variance with an eigenvalue of 1.67. F2 had strong positive loadings for Cr (0.75), Ni (0.63), and Cu (0.78), respectively. The contribution of the strong loadings of PTEs in F2 results showed anthropogenic origin in drinking water sources of UCPs.

Thus, F1 and F2 results showed moderate to high loadings of physicochemical and PTEs contribution, suggesting anthropogenic and natural sources in the drinking water sources of UCPs. Factor 3 (F3) accounted for 18.6% of the total variance with an eigenvalue of 1.49 and was strongly characterized with EC (0.90) and moderate loading with TDS (0.66). The high loading of EC and moderate TDS reflect anthropogenic sources in drinking water sources of UCPs. Factor 4 (F4) explained 11.3% of the total variance with an eigenvalue of 1.00. F4 showed a low contribution in the PCA results, and only pH had a positive moderate loading of 0.62 in F4. Moderate loadings of the physicochemical parameters might be attributed to their moderate to high concentration in the study area.

### 3.4. Pollution Assessment of PTEs

Pollution indices (PTE-PI and PTE-EI) of PTEs were carried out for the drinking water quality of UCPs. PTE-PI and PTE-EI represent different classes with low to high risk levels. The PTE-PI values were calculated based on all PTEs for each UCP (Table 2). The mean concentration value of selected PTEs (Cd, Cr, Pb, Ni, and Cu) were considered while calculating the PTE-PI and PTE-EI of the UCPs water.

Results showed that PTE-PI values of PTEs were observed from low to high values for all the drinking water samples of UCPs, ranging from 11.4 to 116 with the mean PTE-PI value (48.2), which is below the critical threshold pollution index value of 100. The lowest PTE-PI value was recorded for GNPC, while the highest PTE-PI value was observed for ZPP. Overall, the PTE-PI values for all the UCPs water samples were found to be below the critical limit of PTE-PI (100), except SPC (113) and ZPP (116), which exceeded the threshold critical value. These UCPs (SPC and ZPP) showed high PTE-PI values, indicating contamination in drinking water in comparison with other UCPs. In most of the cases (16

UCPs), the mean PTE-PI was much lower than the allowable index value of 100 suggested for drinking water [56]. The scales were slightly altered utilizing multiples of the median as a criterion in order to apply these PTE-PI indices. To distinguish between different levels of contamination, the data are divided into three classes: low (<40), medium (40–80), and high (>80). According to the PTE-PI results, 50% of 10 UCPs samples showed low pollution levels (low risk) with “Excellent” water quality, 40% of eight UCPs had medium pollution levels (medium risk) with “Good” and “Very Good” water quality, and 10% of two UCPs indicated high pollution levels (high risk) with “Poor” water quality.

**Table 2.** Overall Potentially Toxic Elements Pollution Index (PTE-PI), Potentially Toxic Elements Evaluation Index (PTE-EI), and quality classification (as per PTE-PI scale) of all PTEs of UCPs water.

UCPs	PTE-PI	PTE-EI	Quality as per PTE-PI Scale
BPC	79.8	2.25	Good
SPC	113	2.80	Poor
ZPP	116	3.48	Poor
TPP	73.7	2.15	Good
JPN	23.9	1.16	Excellent
DWPN	22.9	1.19	Excellent
PWLPH	73.4	3.35	Good
APH	57.4	2.55	Very good
SKIPS	33.1	1.95	Excellent
WWPS	55.5	2.74	Very good
APS	24.1	1.11	Excellent
GCGPS	44.2	2.55	Very good
SCPM	74.3	3.24	Good
YSPM	43.2	2.55	Very good
GNPC	11.4	0.810	Excellent
PTPC	17.1	1.16	Excellent
HNPDK	28.7	1.59	Excellent
IPDK	20.9	1.43	Excellent
LGPPA	31.5	1.50	Excellent
SPPA	20.0	1.30	Excellent
Average	48.2	2.04	

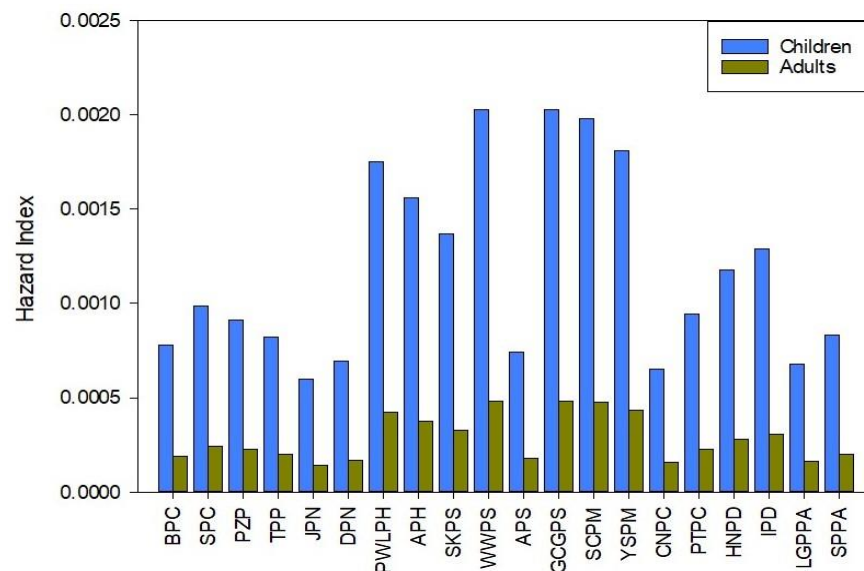
Furthermore, PTE-EI was also used for the brief interpretation of the pollution index [57] of PTEs in UCPs water. The PTE-EI values of PTEs ranged from 0.810 to 3.48 for all UCPs water, with a mean value of 2.04 (Table 2). The respective mean values of the samples were used to construct different PTE-EI values, and the various levels of contamination are characterized by the mean values. Moreover, the PTE-EI values were classified on the basis of pollution levels as low (PTE-EI < 1), medium (PTE-EI = 1–2), and high (>2) as described previously by [50]. Therefore, the results were observed according to the proposed PTE-EI criteria as follows: 5% of samples (1 UCP) showed low pollution level (low risk), 45% of (9 UCPs) samples had medium pollution levels (medium risk), and 50% of (10 UCPs) samples exhibit high pollution levels (high risk), as shown in Table 2. Similar observations and results of PTE-PI and PTE-EI were found in agreement with the study reported by [58].

### 3.5. Health Risk Assessment

The health risks of selected PTEs in UCPs drinking water sources were evaluated for the adults and children via two exposure pathways, i.e., ingestion and dermal contact. The statistical results of DMI for both adults and children via the two exposure pathways are shown in Tables S4 and S5. The results showed that DMI values were higher for children than adults in both exposure routes. Cd had the lowest DMI values for both exposure routes as compared to other PTEs. The non-carcinogenic risks of PTEs also showed low HQ values for adults in comparison with children. Similarly, the ingestion route is the dominant exposure pathway for non-carcinogenic risks of PTEs, followed by the dermal route. The highest HQ mean value ( $2.67 \times 10^{-3}$ ) was observed for Cr in GCGPS, while the

lowest HQ mean value ( $3.77 \times 10^{-6}$ ) was recorded for Ni in APS via the ingestion route. For the dermal route, Cr had the highest HQ mean value ( $6.18 \times 10^{-3}$ ) for GCGPS, while Ni showed the lowest HQ mean value ( $9.13 \times 10^{-8}$ ) for SKIPS. Based on non-carcinogenic risk results, the HQ values of all PTEs for both exposure routes were less than the standard permissible limit ( $HQ < 1$ ), as shown in Tables S6 and S7, suggesting no non-carcinogenic risk to the local population. The decreasing trend of the HQ mean values of PTEs was  $Cr > Pb > Cd > Ni > Cu$ , via ingestion for both children and adults, while for the dermal route, it was as follows:  $Cr > Cd > Pb > Cu > Ni$ , for both children and adults.

The HI values of all PTEs in UCPs were less than 1 for both the adults and children, as shown in Figure 5. HI values of PTEs ranged from  $2.03 \times 10^{-3}$  to  $5.98 \times 10^{-4}$  for children, and for adults they were  $4.83 \times 10^{-4}$  to  $1.44 \times 10^{-4}$ , via both exposure pathways, respectively. This indicates that exposure of PTEs in the UCPs could not pose non-carcinogenic risks to adults and children through the two exposure routes. Among UCPs, HQ and HI values were relatively lower in JPN, DWPN, APS, GNPC, and PTPC than the others. Generally, the HQ and HI values of children were relatively higher than those of adults in all UCPs, signifying that the non-carcinogenic risk of children is higher than that of adults via both exposure pathways (Tables S6 and S7 and Figure 5).



**Figure 5.** Hazard index (HI) of non-carcinogenic effects of all PTEs in drinking water of UCPs.

Furthermore, CR results showed that PTEs (Cr, Cd, Pb, and Ni) had the highest potential carcinogenic risk for both adults and children and surpassed the threshold value ( $1.00 \times 10^{-4}$ ) in all UCPs, except Cd and Ni for adults ( $<1.00 \times 10^{-4}$ ) via the ingestion route (Table 3). The highest value was recorded for Cr ( $3.28 \times 10^{-1}$ ) in GCGPS via the ingestion route, while the lowest CR value was observed for Ni ( $6.34 \times 10^{-5}$ ) in APS. Moreover, CR values of all PTEs were lower than the standard limit, except Cr had a high CR value ( $>1.00 \times 10^{-4}$ ) for children via the dermal route. High CR values of Cr ( $>1.00 \times 10^{-4}$ ) were recorded for all the UCPs, except BPC, SPC, ZPP, TPP, and JPN via the dermal route. The carcinogenic risk levels of PTEs in UCPs are found to be acceptable, except Cd and Ni for adults ( $<1.00 \times 10^{-4}$ ) via the ingestion route and Cr ( $>1.00 \times 10^{-4}$ ) for children via the dermal route are prone to cause potential carcinogenic risk. The TCR mean values were identified for selected PTEs (Cr, Cd, Pb, and Cu) and were found to be varied in the present study. The TCR values for Cr, Cd, Pb, and Cu exceeded the acceptable limit ( $1.00 \times 10^{-4}$ ) except Cd and Ni for adults (Table 3). High TCR values of Cr, Cd, Pb, and Cu indicate high total carcinogenic risk for children and adults exposed to drinking water in all UCPs, and were found to be in agreement with the results of high cancer risk observed for adults and children [59].

**Table 3.** Carcinogenic risk of selected PTEs (Cr, Cd, Pb, and Ni) via ingestion and dermal route in drinking water of UCPs.

UCPs	Children								Adults							
	CRing	CRder	CRing	CRder	CRing	CRder	CRing	CRder	CRing	CRder	CRing	CRder	CRing	CRder	CRing	CRder
	Cr		Cd		Pb		Ni		Cr		Cd		Pb		Ni	
BPC	$5.36 \times 10^{-2}$	$6.22 \times 10^{-4}$	$2.99 \times 10^{-3}$	$1.73 \times 10^{-5}$	$8.12 \times 10^{-3}$	$4.71 \times 10^{-5}$	$1.84 \times 10^{-3}$	$2.13 \times 10^{-6}$	$1.36 \times 10^{-2}$	$1.42 \times 10^{-4}$	$7.58 \times 10^{-4}$	$3.96 \times 10^{-6}$	$2.06 \times 10^{-3}$	$1.08 \times 10^{-5}$	$4.67 \times 10^{-4}$	$4.88 \times 10^{-6}$
SPC	$5.76 \times 10^{-2}$	$6.68 \times 10^{-4}$	$4.77 \times 10^{-3}$	$2.77 \times 10^{-5}$	$8.96 \times 10^{-3}$	$5.20 \times 10^{-5}$	$1.38 \times 10^{-3}$	$1.60 \times 10^{-6}$	$1.46 \times 10^{-2}$	$1.53 \times 10^{-4}$	$1.21 \times 10^{-3}$	$6.32 \times 10^{-6}$	$2.28 \times 10^{-3}$	$1.19 \times 10^{-5}$	$3.50 \times 10^{-4}$	$3.66 \times 10^{-6}$
ZPP	$4.34 \times 10^{-2}$	$5.03 \times 10^{-4}$	$2.78 \times 10^{-3}$	$1.61 \times 10^{-5}$	$2.10 \times 10^{-2}$	$1.22 \times 10^{-4}$	$1.81 \times 10^{-3}$	$2.10 \times 10^{-6}$	$1.12 \times 10^{-2}$	$1.15 \times 10^{-4}$	$7.06 \times 10^{-4}$	$3.68 \times 10^{-6}$	$5.34 \times 10^{-3}$	$2.79 \times 10^{-5}$	$4.61 \times 10^{-4}$	$4.81 \times 10^{-6}$
TPP	$6.25 \times 10^{-2}$	$7.24 \times 10^{-4}$	$2.47 \times 10^{-3}$	$1.43 \times 10^{-5}$	$9.12 \times 10^{-3}$	$5.29 \times 10^{-5}$	$9.15 \times 10^{-4}$	$1.06 \times 10^{-6}$	$1.59 \times 10^{-2}$	$1.66 \times 10^{-4}$	$6.27 \times 10^{-4}$	$3.28 \times 10^{-6}$	$2.32 \times 10^{-3}$	$1.21 \times 10^{-5}$	$2.32 \times 10^{-4}$	$2.43 \times 10^{-6}$
JPN	$7.60 \times 10^{-2}$	$8.82 \times 10^{-4}$	$7.69 \times 10^{-4}$	$4.46 \times 10^{-6}$	$2.34 \times 10^{-3}$	$1.36 \times 10^{-5}$	$2.03 \times 10^{-3}$	$2.35 \times 10^{-6}$	$1.93 \times 10^{-2}$	$2.02 \times 10^{-4}$	$1.95 \times 10^{-4}$	$1.02 \times 10^{-6}$	$5.95 \times 10^{-4}$	$3.11 \times 10^{-6}$	$5.15 \times 10^{-4}$	$5.37 \times 10^{-6}$
DWPN	$9.72 \times 10^{-2}$	$1.13 \times 10^{-3}$	$2.88 \times 10^{-4}$	$1.67 \times 10^{-6}$	$4.75 \times 10^{-3}$	$2.76 \times 10^{-5}$	$8.61 \times 10^{-4}$	$9.99 \times 10^{-7}$	$2.47 \times 10^{-2}$	$2.58 \times 10^{-4}$	$7.32 \times 10^{-5}$	$3.82 \times 10^{-7}$	$1.21 \times 10^{-3}$	$6.30 \times 10^{-6}$	$2.19 \times 10^{-4}$	$2.28 \times 10^{-6}$
PWLPH	$2.37 \times 10^{-1}$	$2.75 \times 10^{-3}$	$1.15 \times 10^{-3}$	$6.69 \times 10^{-6}$	$1.46 \times 10^{-2}$	$8.48 \times 10^{-5}$	$2.44 \times 10^{-3}$	$2.82 \times 10^{-6}$	$6.02 \times 10^{-2}$	$6.29 \times 10^{-4}$	$2.93 \times 10^{-4}$	$1.53 \times 10^{-6}$	$3.71 \times 10^{-3}$	$1.94 \times 10^{-5}$	$6.18 \times 10^{-4}$	$6.46 \times 10^{-6}$
APH	$2.27 \times 10^{-1}$	$2.63 \times 10^{-3}$	$1.45 \times 10^{-3}$	$8.40 \times 10^{-6}$	$8.08 \times 10^{-3}$	$4.69 \times 10^{-5}$	$1.72 \times 10^{-3}$	$1.99 \times 10^{-6}$	$5.76 \times 10^{-2}$	$6.01 \times 10^{-4}$	$3.68 \times 10^{-4}$	$1.92 \times 10^{-6}$	$2.05 \times 10^{-3}$	$1.07 \times 10^{-5}$	$4.36 \times 10^{-4}$	$4.55 \times 10^{-6}$
SKIPS	$2.12 \times 10^{-1}$	$2.46 \times 10^{-3}$	$5.56 \times 10^{-4}$	$3.22 \times 10^{-6}$	$5.44 \times 10^{-3}$	$3.16 \times 10^{-5}$	$1.56 \times 10^{-3}$	$1.81 \times 10^{-6}$	$5.40 \times 10^{-2}$	$5.63 \times 10^{-4}$	$1.41 \times 10^{-4}$	$7.37 \times 10^{-7}$	$1.38 \times 10^{-3}$	$7.21 \times 10^{-6}$	$3.96 \times 10^{-4}$	$4.14 \times 10^{-6}$
WWPS	$3.15 \times 10^{-1}$	$3.65 \times 10^{-3}$	$1.89 \times 10^{-3}$	$1.09 \times 10^{-5}$	$4.16 \times 10^{-3}$	$2.41 \times 10^{-5}$	$2.33 \times 10^{-3}$	$2.70 \times 10^{-6}$	$8.00 \times 10^{-2}$	$8.35 \times 10^{-4}$	$4.79 \times 10^{-4}$	$2.50 \times 10^{-6}$	$1.06 \times 10^{-3}$	$5.51 \times 10^{-6}$	$5.92 \times 10^{-4}$	$6.18 \times 10^{-6}$
APS	$1.12 \times 10^{-1}$	$1.30 \times 10^{-3}$	$4.60 \times 10^{-4}$	$2.67 \times 10^{-6}$	$4.20 \times 10^{-3}$	$2.43 \times 10^{-5}$	$2.49 \times 10^{-4}$	$2.89 \times 10^{-7}$	$2.85 \times 10^{-2}$	$2.98 \times 10^{-4}$	$1.17 \times 10^{-4}$	$6.10 \times 10^{-7}$	$1.07 \times 10^{-3}$	$5.57 \times 10^{-6}$	$6.34 \times 10^{-5}$	$6.61 \times 10^{-7}$
GCGPS	$3.28 \times 10^{-1}$	$3.80 \times 10^{-3}$	$1.20 \times 10^{-3}$	$6.97 \times 10^{-6}$	$4.36 \times 10^{-3}$	$2.53 \times 10^{-5}$	$1.85 \times 10^{-3}$	$2.14 \times 10^{-6}$	$8.33 \times 10^{-2}$	$8.69 \times 10^{-4}$	$3.05 \times 10^{-4}$	$1.59 \times 10^{-6}$	$1.11 \times 10^{-3}$	$5.78 \times 10^{-6}$	$4.69 \times 10^{-4}$	$4.90 \times 10^{-6}$
SCPM	$2.84 \times 10^{-1}$	$3.29 \times 10^{-3}$	$1.83 \times 10^{-3}$	$1.06 \times 10^{-5}$	$1.09 \times 10^{-2}$	$6.32 \times 10^{-5}$	$1.84 \times 10^{-3}$	$2.14 \times 10^{-6}$	$7.20 \times 10^{-2}$	$7.52 \times 10^{-4}$	$4.64 \times 10^{-4}$	$2.42 \times 10^{-6}$	$2.77 \times 10^{-3}$	$1.44 \times 10^{-5}$	$4.68 \times 10^{-4}$	$4.89 \times 10^{-6}$
YSPM	$2.83 \times 10^{-1}$	$3.28 \times 10^{-3}$	$6.79 \times 10^{-4}$	$3.94 \times 10^{-6}$	$7.35 \times 10^{-3}$	$4.27 \times 10^{-5}$	$1.82 \times 10^{-3}$	$2.11 \times 10^{-6}$	$7.18 \times 10^{-2}$	$7.49 \times 10^{-4}$	$1.73 \times 10^{-4}$	$9.01 \times 10^{-7}$	$1.87 \times 10^{-3}$	$9.75 \times 10^{-6}$	$4.62 \times 10^{-4}$	$4.82 \times 10^{-6}$
GNPC	$9.87 \times 10^{-2}$	$1.14 \times 10^{-3}$	$2.95 \times 10^{-4}$	$1.71 \times 10^{-6}$	$1.03 \times 10^{-3}$	$5.99 \times 10^{-6}$	$8.07 \times 10^{-4}$	$9.36 \times 10^{-7}$	$2.51 \times 10^{-2}$	$2.62 \times 10^{-4}$	$7.49 \times 10^{-5}$	$3.91 \times 10^{-7}$	$2.62 \times 10^{-4}$	$1.37 \times 10^{-6}$	$2.05 \times 10^{-4}$	$2.14 \times 10^{-6}$
PTPC	$1.60 \times 10^{-1}$	$1.86 \times 10^{-3}$	$9.61 \times 10^{-5}$	$5.57 \times 10^{-7}$	$3.53 \times 10^{-3}$	$2.05 \times 10^{-5}$	$4.42 \times 10^{-4}$	$5.13 \times 10^{-7}$	$4.06 \times 10^{-2}$	$4.24 \times 10^{-4}$	$2.44 \times 10^{-5}$	$1.27 \times 10^{-7}$	$8.96 \times 10^{-4}$	$4.68 \times 10^{-6}$	$1.12 \times 10^{-4}$	$1.17 \times 10^{-6}$
HNPDK	$1.92 \times 10^{-1}$	$2.23 \times 10^{-3}$	$3.98 \times 10^{-4}$	$2.31 \times 10^{-6}$	$5.27 \times 10^{-3}$	$3.06 \times 10^{-5}$	$5.26 \times 10^{-4}$	$6.11 \times 10^{-7}$	$4.88 \times 10^{-2}$	$5.09 \times 10^{-4}$	$1.01 \times 10^{-4}$	$5.28 \times 10^{-7}$	$1.34 \times 10^{-3}$	$6.99 \times 10^{-6}$	$1.34 \times 10^{-4}$	$1.40 \times 10^{-6}$
IPDK	$2.20 \times 10^{-1}$	$2.55 \times 10^{-3}$	$4.60 \times 10^{-4}$	$2.67 \times 10^{-6}$	$2.15 \times 10^{-3}$	$1.25 \times 10^{-5}$	$7.13 \times 10^{-4}$	$8.28 \times 10^{-7}$	$5.59 \times 10^{-2}$	$5.83 \times 10^{-4}$	$1.17 \times 10^{-4}$	$6.10 \times 10^{-7}$	$5.46 \times 10^{-4}$	$2.85 \times 10^{-6}$	$1.81 \times 10^{-4}$	$1.89 \times 10^{-6}$
LGPPA	$8.62 \times 10^{-2}$	$1.00 \times 10^{-3}$	$3.77 \times 10^{-4}$	$2.19 \times 10^{-6}$	$6.93 \times 10^{-3}$	$4.02 \times 10^{-5}$	$1.66 \times 10^{-3}$	$1.92 \times 10^{-6}$	$2.19 \times 10^{-2}$	$2.29 \times 10^{-4}$	$9.59 \times 10^{-5}$	$5.00 \times 10^{-7}$	$1.76 \times 10^{-3}$	$9.19 \times 10^{-6}$	$4.21 \times 10^{-4}$	$4.40 \times 10^{-6}$
SPPA	$1.28 \times 10^{-1}$	$1.48 \times 10^{-3}$	$3.98 \times 10^{-4}$	$2.31 \times 10^{-6}$	$2.73 \times 10^{-3}$	$1.58 \times 10^{-5}$	$2.05 \times 10^{-3}$	$2.38 \times 10^{-6}$	$3.24 \times 10^{-2}$	$3.39 \times 10^{-4}$	$1.01 \times 10^{-4}$	$5.28 \times 10^{-7}$	$6.92 \times 10^{-4}$	$3.61 \times 10^{-6}$	$5.21 \times 10^{-4}$	$5.43 \times 10^{-6}$
TCR	$1.66 \times 10^{-1}$		$1.27 \times 10^{-3}$		$6.79 \times 10^{-3}$		$1.44 \times 10^{-3}$		$4.20 \times 10^{-2}$		$3.23 \times 10^{-4}$		$1.72 \times 10^{-3}$		$3.67 \times 10^{-4}$	

CRing: Carcinogenic risk via ingestion, CRder: Carcinogenic risk via dermal contact.

#### 4. Discussion

The physiochemical properties play a critical role in influencing the bioavailability of PTEs in groundwater. All the drinking water samples had relatively low to high pH levels in the present study of UCPs. By comparison, the mean pH values in UCPs water were slightly varied, reflecting the existence of alkalizing products such as calcium-magnesium carbonate or calcium carbonate in flagstones, gravel, concrete, cement, and mortar as well as the atmospheric particulate deposition [60,61], resulting in release into the groundwater system. As a result, the drinking water sources could be affected by low to high pH and influence the PTEs concentration. TDS mean values were high for most of the UCPs water, and the presence of high TDS could be attributed to the leaching of ions into the groundwater system [62].

The comparison of PTEs concentrations in all samples with the permissible limits of WHO revealed varying degrees of contamination in drinking water sources of UCPs. Our results indicated that PTEs (Cu, Cr, and Cd) concentrations were generally higher in the UCPs drinking water than in the previous studies. For instance, the PTEs (Cu, Cd, Ni) concentrations (Figure 3) in the present study were relatively higher in most of the UCPs than in a previous study of lakes along urban gradient lakes in Wuhan, China [63]. Similarly, high Cu, Cr, Pb, Ni, and Cd were found to be higher than the previous study of the urban Houguan Lake in Wuhan, China [64], and also higher than the reported values of PTEs (Cu, Pb, and Cd) in urban groundwater of Nnewi, Southeast Nigeria [65]. In contrast, the mean values of PTEs concentrations were relatively lower than the results of the urban Mariout Lake, Egypt, reported by [66], and were lower than the mean values of Cu, Pb, Ni, and Cr (except Cd) in the groundwater of an industrial park situated at the southeast of Zhejiang Province, China [67]. In another study, PTEs such as Cr, Cu, and Cd were relatively higher, while Pb concentrations were lower than in the previous study of drinking water sources in the industrial city of Sialkot, Punjab Pakistan [26], and were higher than in our recent studies of [68–70]. The variation in PTEs concentration level demonstrates numerous factors depending upon various potential sources of anthropogenic activities and natural geological backgrounds, resulting in UCPs water contamination. This variation in PTEs concentration also reflects the impact of physiochemical properties of the soil, which might affect the mobilization and bioavailability of PTEs in the groundwater system. For instance, the bioavailability and mobility of PTEs can potentially be influenced by soil pH, organic matter, and cation-exchange mechanism. In addition, due to the precipitation mechanism between PTEs and anions, PTEs can be more readily accessible and mobilized in comparison to high pH [71]. Another reason is that urbanization in UCPs areas could also affect the soil texture, soil pH, cation-exchange capacity, bulk density, and thus PTEs discharge and deposit in the soils [72] and release into the groundwater system. Moreover, long-term disposal of solid waste and wastewater irrigation in parks result in UCPs soil contamination with high level of PTEs. Additional factors that contribute to the contamination of the soil with PTEs include inadequate sludge and septic tank treatment plans, sanitary system leakage, and raw sewage water from installations [73].

Some other factors like pH may have an impact on bioavailability of PTEs in the drinking water of UCPs. Evidently, Li et al. also reported that the physiochemical properties of the soil ecosystem (soil pH, cation exchange mechanism, and organic matter) could influence bioavailability of PTEs. The dissolution of PTEs occur in soil due to redox potential under acidic to alkaline conditions (pH 5.0 to 8.0) and reach into the groundwater through leaching [74]. In the present study, some of the UCPs (BPC, SKIPS, and PTPC) had low pH level with moderate to high PTEs concentration as compared to the other UCPs. PTEs are bioavailable and easier to mobilize at low pH levels compared to high pH levels because of soil precipitations between various anions and metals [75]. Some of the anthropogenic sources such as domestic waste disposal contribute to release toxic pollutants [76] in the UCPs. According to a previous study of PTEs in Peshawar urban parks in Pakistan, Cd originates from industrial facilities (power stations, coal combustion, and the metallurgical sector), whereas Ni is derived from agricultural practices including

wastewater irrigation and solid waste disposal [73], and Pb is primarily found in urban soil due to vehicle exhaust emissions [77]. Furthermore, ref. [5] reported that the soil's characteristics, pollutant emissions, and the geochemical background had a significant impact on the spatial distribution of PTEs.

In the present study, moderate and high loadings of selected PTEs in PCA results demonstrate numerous factors as shown in Figure 4, showing their anthropogenic and natural sources. The loading factors of pH in F1 and F4 factors reflect the presence of alkaline salts such as calcium-magnesium carbonate  $\text{CaMg}(\text{CO}_3)_2$  and calcium carbonate ( $\text{CaCO}_3$ ) in flagstones, concrete, gravel stones, and cement in soils [71], resulting in influencing the PTEs availability in UCPs. Despite the anthropogenic sources, previous research demonstrated that soil parent materials had an impact on the distribution of PTEs like Cd [3]. The PTEs such as Ni could originate from waste disposal and agricultural activity of wastewater irrigation [73], which could be released to the drinking water sources of UCPs. Other PTEs (Cu and Pb) in urban parks are mainly derived from vehicular exhaust emissions [77].

The results of health risk assessment showed that children are more exposed to average daily exposure doses due to smaller skin surface area and lower body weight. Children are also more susceptible to external environment during their developmental stages and growth. This could be a reason that children are more exposed to drinking water sources in UCPs via ingestion and dermal contact than adults. Previous studies also reported that children are more vulnerable to average daily exposure dose and high non-carcinogenic risks than adults [78]. We concluded that long-term exposure to these toxic PTEs (Cd, Ni, and Cr) through drinking water consumption of UCPs presents a carcinogenic risk to the local population. Our results of HQ, HI, and CR are consistent with the recent study conducted by [79] and [34]. TCR results also showed high values of Cr, Cd, Pb, and Cu, indicating high total carcinogenic risk for children and adults exposed to drinking water in all UCPs. In the present study, children present high non-carcinogenic risk and carcinogenic risk for both exposure routes. In general, children are typically more susceptible than adults to exposure to toxic contaminants due to their continuous outdoor activity [80] and their propensity to play on the ground, put objects in their mouth, and use their hands [81,82], confirming the risk of drinking water contaminated with PTEs in UCPs.

Additionally, heavy traffic and industrialized regions can influence and prominently increase these contaminants in the soil [83], resulting in contaminating the drinking water sources as well in local UCPs. The public urban parks are more likely accessed and visited by children and adults, and contaminated soil can be introduced in UCPs by their footsteps. The soil in these locations also has a significant impact on the population's health, posing a concern if pollutant concentrations are high [84]. As a result, PTEs can possibly reach the groundwater system through leaching due to contaminated soil and continuous wastewater irrigation. A high content of PTEs is released as a result of long-term wastewater discharge and solid waste disposal in parks. Additionally, improper treatment and leakage in sludge and septic tank schemes, sanitary system seepage from installations, and discharge of raw sewage water [73] also contribute to contaminating the drinking water sources with PTEs in UCPs. Therefore, with the primary attention on prevention, and gaining control of potential health concerns needs to be undertaken related to exposure to PTEs, particularly for children in UCPs of Khyber Pakhtunkhwa, Pakistan.

## 5. Conclusions

In the present study, the mean concentrations of PTEs (Cd, Cr, Pb, Ni, and Cu) showed high variation in drinking water sources of UCPs. PTEs such as Cd, Cr, and Pb had high contamination and exceeded the permissible limits of WHO and PAK-EPA. The PCA results of PTEs revealed high anthropogenic sources of traffic and industrial emissions, solid waste disposal, wastewater irrigation, and soil parent materials. The PTE-PI and PTE-EI results showed low to high pollution levels for all the UCPs, with high values for Sardaryab Park Charsadda (SPC) and Zoo Park Peshawar (ZPP), indicating unhealthy drinking water

quality in comparison with other UCPs. The HQ and HI values of all PTEs were less than the permissible limit ( $<1$ ), while CR values surpassed the threshold value ( $1.00 \times 10^{-4}$ ) of PTEs for both adults and children in all UCPs, except Cd and Ni for adults via the ingestion route. For the dermal route, CR values of Cr were higher than the standard limit among PTEs in most of the UCPs. The TCR values of PTEs were higher for children and adults via drinking water consumption in all UCPs. Our results revealed high carcinogenic risk of PTEs (Cd, Ni, and Cr) exposed to the local population in all UCPs. Therefore, the present study proposed that the government should control and mitigate the PTEs pollution in drinking water sources by enforcing environmental regulations as well as implementing the proper management strategies to remediate its pollution in urban children's parks and reduce potential health risk in urban areas. The assessment of PTEs contamination and the health risk evaluation used in this work should be used in future risk assessments, as it can aid in the implementation of more appropriate metal risk management in urban environments worldwide.

**Supplementary Materials:** The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/su151713177/s1>: Table S1. The mean  $W_i$  and  $Q_i$  calculation for groundwater samples of the study area. Table S2. Parameter values used for daily metal intake in health risk models. Table S3. Physiochemical and PTEs mean concentration ( $\mu\text{g/L}$ ) in drinking water sources of UCPs in KPK, Pakistan. Table S4. The DMI values of selected PTEs via ingestion for drinking water in UCPs. Table S5. The DMI values of selected PTEs via dermal contact for drinking water in UCPs. Table S6. Non-carcinogenic risks posed by each PTE via ingestion of UCPs water in KPK. Table S7. Non-carcinogenic risks posed by each PTE via dermal exposure pathway of UCPs water in KPK.

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