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Health Risk Associated with Some Trace and Some Heavy Metals Content of Harvested Rainwater in Yatta Area, Palestine

Issam A. Al-Khatib [1,*], Ghadeer A. Arafeh [2], Mutaz Al-Qutob [3], Shehdeh Jodeh [4,*], A. Rasem Hasan [5], Diana Jodeh [6] and Michael van der Valk [7]

[1] Institute of Environmental and Water Studies, Birzeit University, Birzeit 00970, Palestine
[2] Faculty of Graduate Studies, Birzeit University, Birzeit 00970, Palestine; Ghadir.Arafa@hotmail.com
[3] Department of Earth and Environmental Studies, Faculty of Science and Technology, Al-Quds University, East Jerusalem 00970, Palestine; qutob@planet.edu
[4] Department of Chemistry, An-Najah National University, P. O. Box 7, Nablus 00970, Palestine
[5] Department of Civil Engineering, An-Najah National University, P.O. Box 7, Nablus 00970, Palestine; mallah@najah.edu
[6] Division of Plastic and Reconstructive Surgery, Johns Hopkins All Children's Hospital, St. Petersburg, FL 33701, USA; djodeh1@jhmi.edu
[7] Hydrology.nl—International Hydrology and Water Resources, P.O. Box 61003, NL-1005 HA Amsterdam, The Netherlands; info@hydrology.nl
* Correspondence: ikhatib@birzeit.edu (I.A.A.-K.); sjodeh@hotmail.com (S.J.); Tel.: +970-22982120 (I.A.A.-K.); +970-599590498 (S.J.); Fax: +970-22982120 (I.A.A.-K.); +970-92345982 (S.J.)

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Abstract: Rainwater is considered a dependable source for domestic purposes within rural areas in Palestine. Harvested rainwater stored in cisterns is used to leverage deficits from municipal water supplies. Harvested rainwater in areas surrounded with industrial and agricultural activities is usually contaminated with heavy and trace metals. To study the effects of human exposure to heavy and trace metals, 74 harvested rainwater samples of rain-fed cisterns were collected from different localities in the Yatta area of Palestine in the months of January and February of 2016. The water samples were analysed for Ca, Mg, Al, Fe, K, Na, Ag, Li, Co, Ba, Bi, Sr, Ga, V, Rb, Mo, Beand Tl elements utilizing ICP-MS (inductively coupled plasma mass spectrometry). The selected trace metals were found within the concentration limits of the acceptable values, in accordance with WHO and Palestinian standards, except for K and Al, which were found above the allowed limits. The potential risks of the selected trace metals on the health of the local residents, as well as the possible sources of such heavy metals, were also studied. The Chronic daily intake (CDI) of each metal and health risk indexes (HRI) were calculated for both adults and children residents. The oral ingestion pathway was studied, including exposure via drinking water. The values for CDI were found in the descending order of: Ca > Mg > Na > K > Sr > Fe > Al > Ba > Li > V > Rb > Ag > Mo > Ga > Co > Bi > TI > Be. The values of HRI were below 1 for most of the selected heavy metals, expect for Li for children, indicating potential health risk. The study also predicted that the local residents have a higher chance of developing cancer in their lifetime, especially children, with respect to the carcinogenic risk (CR_{ing}) values for Na, Mg, Al, Ba, K, Ca, Fe and Sr, which were greater than standardized limits ($>10^{-6}$). The rest of the selected elements were within the acceptable limit in the five different studied locations. Furthermore, univariate, multivariate and statistical analysis depending on one-way ANOVA, inter-metal correlation, cluster analysis (CA) and principal component analysis (PCA) results revealed that geogenic and anthropogenic activities were major sources of drinking water contamination by heavy metals in the Yatta area.

Keywords: cisterns; heavy metals; rainwater harvesting; human health risk; Yatta area

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1. Introduction

The existence of heavy metals in drinking water results from two independent factors. The first one is naturally occurring through the weathering of rocks and soil erosion, which are often present at low levels in the environment [1–3]. The second factor results from continuously increasing human activities such as agricultural activities; agrochemicals and livestock manures uses, fertilizers, intensive animal practices, pesticides and other human activities, such as: mining and melting of minerals (which potentially have an impact on human health), [4–8].

Drinking water sources include groundwater wells, rivers, lakes, reservoirs, rainwater harvesting cisterns, ponds and so forth. These various sources of water pose the greatest risk to human health, due to their contamination by heavy metals [9]. In Palestine, despite the increase in water shortage and the water crisis, the rainwater harvesting cisterns are considered as a partial solution to minimizing and managing these crises. In this case, the advantage of using the rainwater harvesting cisterns is in providing an additional domestic water source; one which meets basic human needs [10].

The risk of exposure to heavy metals and the high toxicity level is determined depending on the type of the metal, its concentration and its biological role [9]. The chronical exposure and accumulation of heavy metals in the human body can lead to many health threats such as lung fibrosis, cardiovascular and kidney diseases, irregularity in blood composition and mutagenic/carcinogenic effects to the human body. Furthermore, they can cause physical, muscular and neurological degenerative processes that may develop into Alzheimer's disease, Parkinson's disease and multiple sclerosis (a nervous system disease that affects white matter of the brain and spinal cord). In some cases, heavy metals accumulating in human body may damage and destroy the mental and central nervous function, leading to a death famed as "fatal effects of heavy metals toxicity" [8,11–14].

Most heavy metals (As, Cd, Cr, Pb and Hg) are considered to be cancer-inducing agents [15]. Although several heavy metals serve as enzymes that are essential for intracellular process and have DNA-binding domains, almost all heavy metals induce various cancers and diseases [16,17]. It has been reported that heavy metals such as cobalt (Co), iron (Fe), magnesium (Mg) and molybdenum (Mo) are required for many biochemical and physical functions, as they are essential nutrients for these functions [15,18]. Inadequate supply of these micro-nutrients results in a variety of deficiencies and diseases or syndromes [15,18]. Other metals such as aluminium (Al), lithium (Li), barium (Ba), beryllium (Be), bismuth (Bi), gallium (Ga), strontium (Sr), thallium (Tl), silver (Ag) and vanadium (V) are considered to be non-essential metals since they have no identified biological functions [19].

Although required in very small amounts, trace elements such as iron, iodine, fluoride, copper, zinc, chromium, selenium, manganese and molybdenum are vital for maintaining health. Also referred to as micro minerals, these trace elements are part of enzymes, hormones and cells in the body. Insufficient intake of trace minerals can cause symptoms of nutritional deficiency.

In Palestine, limited studies have been conducted for detection of heavy metal levels in the polluted rainwater harvested in cisterns. In a study conducted by Malassa et al. [20], two remarkable findings were discovered during analysis of different trace heavy metals in 44 rainwater harvesting samples. These samples were collected in the western part of Hebron. First, the concentrations of some heavy metals were higher than the permissible limits for drinking water, according to WHO standards. Second, most of the rainwater harvested in cisterns was contaminated by heavy metals. One limitation of the study is that it did not mention the probable sources of the heavy metals.

In this paper, an effort is made to monitor and evaluate the levels of heavy metal concentrations in the harvested rainwater, in five different localities of the Yatta area (South Hebron): Al-Hadidya, Khallet El Mayya, Khallet Salih, Al-Hila and Yatta Centre. The harvested rainwater is used mainly for drinking and other domestic uses. Eighteen common and uncommon heavy metals were determined including Calcium (Ca), Magnesium (Mg), Aluminium (Al), Iron (Fe), Potassium (K), Sodium (Na), Silver (Ag), Lithium (Li), Cobalt (Co), Barium (Ba), Bismuth (Bi), Strontium (Sr), Gallium (Ga), Vanadium (V), Rubidium (Rb), Molybdenum (Mo), Beryllium (Be) and Thallium (Tl). The relationship between contaminated water with heavy metals and human health has been investigated. Different univariate and multivariate statistical analyses, such as one-way analysis of variance (ANOVA), inter-metals correlation and principal component analysis (PCA), were applied to demonstrate the relationship between the contaminated water and human health using the aforementioned descriptive statistics methods.

The main objectives of the current study are to evaluate the heavy metal levels in rainwater harvesting cisterns in the Yatta area, to assess the human health risk and to identify the potential sources of contamination.

2. Materials and Methods

2.1. Study Area

Yatta is a Palestinian area located in the Hebron Governorate, approximately 8 km south of Hebron city, which is located in the southern part of the West Bank, [21,22]. Geographically, the Yatta area is bordered by Zif and Khallet El Mayya to the East, ArRihiya, Al Fawwar Camp and Wadi as Sada to the North, Beit 'Amra to the West and As Samu' to the South [23]. The overall climate of the Yatta area is classified as arid to semi-arid, with an increase in aridity toward the south and the east (Figure 1) [24]. The Yatta area is located in the mountainous area with an elevation of around 793 m above mean sea level, while the mean annual rainfall is about 303 mm and the average annual temperature is 18 °C, with an average annual humidity of 61% [24]. According to the Palestinian Central Bureau of Statistics [25] in 2016 Yatta town had a population of 64,277 people, Khallet El Mayya1 had 1865 people, Khallet Salih 443 people and Al-Hila had a population of 1686 persons.

The Yatta area has had a water network since 1974. In referring to the results of the survey that was conducted by ARIJ in 2009, nearly 85% of the households are connected. The Israeli Water Company (Mekkorot) is the main provider for water coming from Toque wells. Water is supplied intermittently on average three days weekly, with the worst situation in summer occurring once a week. To leverage deficits of municipal water supply, rainwater is usually harvested from house rooftops during the winter and stored in cisterns to be used when needed throughout the year. As a summary, the main water-obstacles facing the Yatta area can be summarized as: general shortage in water supply, inability of the existing water network to meet public demand, water pollution and an increase in water losses [23].

Rainwater harvesting is the main practice for domestic and agriculture purposes in the Yatta area. There are two types of rainwater collection systems in Yatta. The first one uses a large scale to collect storm water runoff from the ground and then is stored in large reservoirs. Another system uses a small scale of reservoirs, known as cisterns to collect and store rainwater during the winter season from controlled surfaces or roofs of the houses. These cisterns are constructed underneath or next to houses, with an average capacity of 70 m^3 [24]. A field survey data indicated that more than 60% of the Yatta area inhabitants have and depend on rainwater harvesting cisterns in their houses. This is because 15% of the households are not connected to the water distribution networks. In case the house is connected, the suffering from frequent shortages of water encourages the local inhabitants to construct rainwater harvesting cisterns.

The geology of the study area is mainly covered by limestone with dolomite and marl of Late Cretaceous (Cenomanian-Turonian) age and marl limestone with sandstone. Conglomerates of the Pliocene age (Tertiary) cover the eastern part of the Yatta area. Furthermore, chalk-marl with conglomerates (Pleistocene to recent) covers a small area of the south-eastern part of the area. The major soil type associations found in the Yatta area are classified as Terra Rossa, Brown Rendzinas and Pale Rendzinas which dominate most of the study area. Brown lithosols are found in pockets among the rocks and loessial arid brown soils are found on flat hilltops, plateaus and foot slopes as well as

Dark Brown Soils which are distributed in a portion of the western and eastern part of the study area. Furthermore, Brown lithosols and loessial Serozems soils are originally formed from limestone, chalk, dolomite and flint. These soil associations suffer from extensive erosion due to runoff, especially in steep slopes and also suffer from salt accumulation due to limited salt leaching capabilities [21].

The major quarrying and stone processing industries are considered to be the leading industries in the Yatta town. Processing and manufacturing industries—mainly food and agro-processing, clothing, textile, leather products and furniture—are concentrated in the Hebron city and a short distance near Yatta town. Approximately 31% of industrial facilities are located in residential areas within Yatta town [24]. The generated wastewater from the industries facilities has been continuously increasing over the last few years, which is expected to increase the potential health risk in the coming years.

2.2. Cistern Material and Type of Roof Material

The cistern material and type of roof material were not surveyed during the field work but they were surveyed by another researcher [26,27] for the same study area. The roof material consisted of reinforcement concrete zinc sheets and asbestos sheets, with percentages of 97.2%, 1.8% and 1.0% respectively. Regarding the cistern material, the highest percentage was reinforced concrete (30%) or rock (23%), whereas 18.8% of the cistern material was rock and concrete and the lowest percentage (0.3%) was popular.

2.3. Sampling and Analysing Approach

Based on the geologic setting criteria, elevation, shortage of water supply, population density and coexistence of the local residents near industrial facilities with an increase of cancer cases, five locations have been chosen as study areas; including Al-Hadidya, Khallet El Mayya, Khallet Salih, Al-Hila and Yatta Centre which all are located in the southern part of Hebron City. 74 Water samples were randomly collected from rainwater harvesting cisterns in households from the five localities. The water was stored in the cistern for a period of one to two months before sampling. Figure 1 shows the localities where the samples were collected from. The water samples were collected during January and February 2016 in one-litre high-density polyethylene bottles (pre-cleaned with 10% nitric acid followed by repeated rinsing with bi-distilled water), stabilized with a few drops of ultrapure nitric acid (0.5% HNO_3) that were added to prevent additional microbial growth. All water samples were then preserved in a refrigerator at 4 °C and transported to the lab of Al-Quds University for the examination and laboratory analysis purposes.

ICP-MS (Agilent Technologies 7500 Series, Agilent, Santa Clara, CA, USA) was used for analysis of the selected heavy metals including: Ca, Mg, Al, Fe, K, Na, Ag, Li, Co, Ba, Bi, Sr, Ga, V, Rb, Mo, Be and Tl. The analytical technique and operating conditions of ICP-MS adapted the following principle: nebulizer gas (argon) flow rate, 0.9 L/min; auxiliary gas (argon) flow rate, 0.3 L/min; plasma (argon) gas flow rate, 15 L/min; reaction gas flow (helium) rate, 4 mL/min; lens voltage, 7.25 V; and ICP RF power, 1100 W.

In terms of quality assurance concepts, an internal standard (Er) and a multi standard calibration method (29 metal standards with a concentration of matrix 5% HNO_3) were used to obtain an accurate quantitative determination for each of selected heavy metals in water samples. Water samples were prepared by dilution of 1.0 mL of the water samples to 10.0 mL with 0.3% ultrapure nitric acid and they were analysed by ICP-MS (Agilent, Santa Clara, CA, USA). The descriptive analytical approach has been used to analyse data, as well as to describe the obtained results.

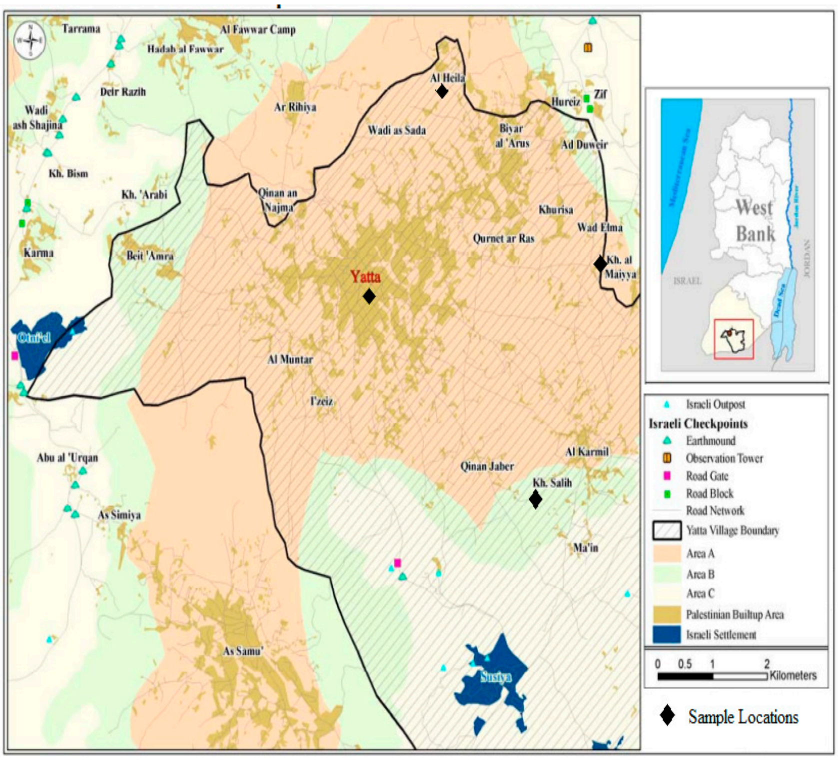

Figure 1. Study area including sampling localities in Yatta area [22].

2.3.1. Human Health Risk Assessment

Human Health Risk Assessment (HHRA) is defined as the processes of estimating the likelihood of occurrence of any given probable magnitude of adverse health effects, over a specified time period that may result from exposure to certain health hazards. Health risk assessment of heavy metals in rainwater harvesting cisterns used for drinking was examined via oral route exposure for adults and children recipients, based on the United States Environmental Protection Agency (US EPA) risk assessment methodology. Based on this premise, the human health risk assessment equations were calculated, as derived from the US EPA [28–31].

According to the samples which were analysed it is clear that most the collected samples of springs and wells are of fresh water with the Total Dissolved Solids (TDS) values of 300–900 mg/L.

According to the results, which were conducted during the study, showed that the Electrical Conductivity (EC) values ranges from 500–2000 µS/cm in the springs, while in the wells the values ranges from 500–1000 µS/cm. The range of pH for the whole studied samples was between 6.1–7.23.

Chronic Daily Intake (CDI)

Exposure to heavy metals can occur through an assortment of routes. Heavy metals may be inhaled as dust or fume or may also be ingested through food and drink. Once a heavy metal is absorbed, it distributes within tissues and organs. The most influential rout is via oral intake of drinking contaminated water. Oral ingestion was considered to be effectively contributing to residential exposure [32]. At all events, in comparison to other routes, all of them are considered negligible. Risk assessment via oral route was evaluated for all selected heavy metals.

The Chronic Daily Intake (CDI) was used to calculate the health risk analysis for drinking water from rainwater harvesting cisterns, associated with heavy metals exposure, using the following equation:

$$\mathrm{CDI} = \frac{\mathrm{C_m} \times \mathrm{I_w}}{\mathrm{W_b}} \tag{1}$$

where, CDI is Chronic Daily Intake (µg/kg/day), C_m is the concentration of heavy metal in drinking water (µg/L), I_w is the average daily intake of water (assumed to be 2 L/day for adult and 1 L/day for child) and W_b is the average body weights (assumed to be 72 kg for adult and 32.7 kg for child) [33].

Health Risk Indexes (HRI)

Health risk assessment classifies elements mainly as having a carcinogenic risk or non-carcinogenic risk. Non-carcinogenic chemicals are considered to have dose thresholds, below which no adverse health effects will occur.

In regard to non-carcinogenic health effects posed by heavy metals in drinking water, the Health Risk Index (HRI) was calculated using the following equation:

$$\mathrm{HRI} = \frac{\mathrm{CDI}}{\mathrm{RfD}} \tag{2}$$

The EPA has calculated a Reference Dose (RfD) as the acceptable safety level for chronic non-carcinogenic and developmental effects. According to US Environmental Protection Agency [31], the Oral Reference dose (RfD, µg/kg/day) values for Li, Be, Tl, V, Mo and Ag are 2×10, 5×10^{-3}, 8×10^{-4}, 7×10^{-3}, 5.0, and 5×10^{-3}, respectively.

When the HRI value is less than 1 ($\mathrm{HRI} < 1$) for a certain chemical, this indicates safe water regarding the consumption of the studied chemical but if HRI value exceeds 1 ($\mathrm{HRI} > 1$), this indicates a significant risk level [29,34].

For carcinogenic health effects, carcinogens are not considered to have an effective threshold. This consideration implies that there is a risk of cancer developing with exposures at low doses and, therefore, there is no safe threshold for exposure to these carcinogenic chemicals [35]. Furthermore, the risks are estimated as the incremental probability of an individual developing cancer over a lifetime as a result of exposure to the potential carcinogen [36].

Carcinogenic risks (CR) were calculated using the following equation and the detailed calculating process was followed by DE Miguel, Wu, Iqbal and Shah et al. [33,37,38].

$$\mathrm{CR_{ing}} = \frac{\mathrm{Exp_{ing}}}{\mathrm{SF_{ing}}} \tag{3}$$

$$\mathrm{Exp_{ing}} = \frac{\mathrm{CDI} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{AT}} \tag{4}$$

whereas $\mathrm{CR_{ing}}$ is carcinogenic risk via oral route (dimensionless); $\mathrm{Exp_{ing}}$ is the exposure dose through ingestion of water (µg/g/day); $\mathrm{SF_{ing}}$ is the carcinogenic slope factor, ingestion (µg/g/d) and EF is Exposure frequency (360 days/year); ED is Exposure duration (=70 year) and AT is Averaging time (10,950 days). In order to show the lifetime carcinogenic risk to the local residents, $\mathrm{CR_{ing}}$ values were calculated for all the selected metals. The slope factor converts the estimated daily intake of the heavy metal averaged over a lifetime of exposure directly to incremental risk of an individual developing cancer [28]. Based on our research study, the $\mathrm{SF_{ing}}$ for Adult is 0.4 and for Child is 2.0 (µg/g/day). The ranges of carcinogenic risks acceptable or tolerable by US EPA [39] was 10^{-6} to 10^{-4}.

2.3.2. Statistical Analysis

All human health risk equations and basic statistics such as min., max., mean and standard deviation were computed by using Microsoft Office-Excel, ver. 2010. Univariate and multivariate statistical analyses were processed by using the Statistical Package for the Social Sciences (SPSS) statistic software version 20. One-way ANOVA procedure, inter-metals correlation and principal component analyses (PCA) were used to identify the possible heavy metal sources. PCA was carried out by the method of Varimax normalized rotation on the dataset. Cluster analysis (CA) was applied by using hierarchical methods to produce a dendrogram that provides the clustering groups, which describes the beginning with the most similar pair of objects and forming higher clusters stepwise.

3. Results and Discussion

3.1. Drinking Water Characteristics

The concentrations of different heavy metals, which have been detected in the selected rainwater harvesting cisterns samples in the five locations, are shown in Table S1 in the Supplementary Materials. The concentrations were found in the order of Ca > Mg > Al > Na > K > Fe respectively. Most of the studied samples had Aluminium levels much above the permissible limit of 200 µg/L, in accordance with WHO standards [40]. In Khallet Salih's cisterns, Aluminium was recorded at 9.37 as a minimum, 181.83 µg/L as a mean and 1619.71 µg/L as a maximum. In other cisterns, Al-Hila, Aluminium was measured at concentrations of 4.90 as a minimum, 156.84 as a mean and 1166.18 as a maximum, all in µg/L. In rainwater harvesting cisterns surveys at the Yatta Centre, Aluminium was reported as 7.14 µg/L for a minimum, 52.21 µg/L for a mean and 264.55 µg/L as a maximum. It is clear that AL in Al-Hila and Khallet Salih's cisterns samples were above the WHO and Palestinian standards but that Al was within the Palestinian limit in the Yatta centre which may have to do with some contaminated wastewater coming from the industry located close bye. For Li, the value for the permissible limit has been estimated to be 0.05 µg/L. Lithium was detected in almost all the samples from rainwater harvested cisterns and the highest concentration (6.59 ± 6.22) µg/L was detected in Khallet Salih but the values were within the maximum concentration standard of 5000 µg/L, as suggested by WHO [40].

Calcium and magnesium levels in the rainwater harvesting cisterns samples were found to be safe when compared with WHO permissible limit values and Palestinian standards. Ca and Mg are needed by the body in larger quantities and deficiencies in the human system will lead to adverse health effects. Levels of Na and K in all the water samples were found to be below the WHO Permissible limit values with the exception of K levels in sampling localities within Khallet El Mayya and Al-Hadidya, in the Yatta area.

The results of the study indicated that the selected heavy metal concentrations in rainwater harvesting cisterns samples of Al-Hadidya, Khallet El Mayya, Khallet Salih, Al-Hila and Yatta Centre are within the acceptable limits, when compared with WHO and Palestinian Standards, except for K and Al. K and Al were above the permissible limits which lead to potential health risks for the local residents.

3.2. Comparison of Heavy Metals Concentrations with Other Studies

A few studies have been conducted worldwide related to the detection of heavy metal concentrations in harvested rainwater, particularly including the selected heavy metals in this study [15,20,41].

In order to obtain an obvious judgment about the level of contamination by heavy metals in harvested rainwater consumed by local people in the Yatta area, the obtained data should be compared with other literature reported data, obtained from different locations around the world. Because of the lack of literature about most of the selected heavy metals, concerning with rainwater harvesting cisterns used for drinking water, we have picked some of the selected heavy metals out in this study and compared the results with other studies in the world.

Table S2 in the Supplementary Materials shows a comparison between the concentrations of heavy metals in the present study (Yatta Centre's case) with concentrations in another study conducted in Kosovo. There were no differences between the metal concentrations of Al and Fe in the present study and Kosovo study. Concentrations of Ba, Co, Ca, K, Mg and Li in Kosovo were found to be higher than in the current study levels. Generally, the variations between the two different studies were not significant and both of them gave a reasonable level of results. Moreover, the obtained results in the current study compared with Kosovo study were within the acceptable limits when compared with WHO and Palestinian standards, except for K and Al, which were above the permissible limits, posing potential health risks on the local habitants.

3.3. Human Health Risk Assessments

3.3.1. Chronic Daily Intakes

Table S3 in the Supplementary Materials summarizes the calculated CDI values of selected heavy metals for consumption of drinking water from monitored rainwater harvesting cisterns for both adults and children. In the selected five locations, the range of the mean calculated chronic daily intake (CDI) values of selected heavy metals for adults who consumed drinking water from monitored rainwater harvesting cisterns for both adults and children are presented in Table S3. CDI values for Li, Be, Ba (except Khallet Salih and Khallet El Mayya samples), Al (for Khallet El Mayya samples), Tl, Bi, V, Co, Ga, Rb, Mo and Ag for both adult and children were found to be less than 1. The remaining calculated CDI values of heavy metals parameters and samples for both adult and children were observed to be more than one. Therefore, CDI indices for heavy metals were found in the order of: Ca > Mg > Na > K > Sr > Fe > Al > Ba > Li > V > Rb > Ag > Mo > Ga > Co > Bi >Ti > Be, through all selected areas within monitored rainwater harvesting cisterns, respectively. Furthermore, the CDI values for Li, Be, Tl, V, Mo and Ag were within or less than their respective RfD limits, in accordance to the United States Environmental Protection Agency [42].

3.3.2. Health Risk Indexes (HRI's)

The Health risk indexes values associated with drinking water from monitored rainwater harvesting cisterns are summarized in Table 1 for the five localities. The study showed that the inhabitants who consumed water from the monitored rainwater harvesting cisterns in all selected areas containing Li, Be, Ba, Tl, V, Sr, Mo and Ag had HRI values, for both adults and children, were found to be less than 1.This finding distinctly indicates that the oral exposure route of these selected heavy metals by drinking from rainwater harvested cisterns has no potential adverse effects; suggesting no health risk on local residence in the region. Only the HRI values of Li for children in Khallet Salih samples were more than 1. This indicates that the risk of the adverse effects of Li contamination in rainwater harvesting cisterns, in its respective area, was not acceptable.

Table 1. Health risk indexes (HRIs) of heavy metals through drinking water (n = 57) consumption.

Parameter	Receptor	RfD µg/(kg/day)	Location				
			Al-Hila	**Yatta Centre**	**Khallet Salih**	**Khallet El Mayya**	**Al-Hadidya**
Li	Adults	0.2	0.8	0.65	0.9	0.75	0.6
	Children		0.9	0.75	1.0	0.8	0.65
Be	Adults	5.0	6×10^{-5}	2×10^{-5}	4×10^{-5}	0.0	6×10^{-5}
	Children		8×10^{-5}	2×10^{-5}	4×10^{-5}	0.0	6×10^{-5}
Ba	Adults	70.0	120	1.2×10^{-2}	1.4×10^{-2}	1.4×10^{-2}	1.1×10^{-2}
	Children		130	1.4×10^{-2}	1.5×10^{-2}	1.6×10^{-2}	1.2×10^{-2}
Tl	Adults	0.08	100	2.5×10^{-3}	0.13	1.3×10^{-2}	1.3×10^{-2}
	Children		130	2.5×10^{-3}	0.13	1.3×10^{-2}	1.3×10^{-2}
V	Adults	7.0	200	1.4×10^{-2}	1.9×10^{-2}	1.3×10^{-2}	1.4×10^{-2}
	Children		210	1.6×10^{-2}	2.1×10^{-2}	1.3×10^{-2}	1.6×10^{-2}
Fe	Adults	700	8.89×10^{-3}	5.47×10^{-3}	8.96×10^{-3}	6×10^{-3}	6.54×10^{-3}
	Children		9.75×10^{-3}	6.01×10^{-3}	14.16×10^{-3}	6.61×10^{-3}	7.21×10^{-3}
Sr	Adults	600	13.5×10^{2}	18.2×10^{-2}	14.4×10^{-2}	19.4×10^{-2}	14.2×10^{-2}
	Children		14.9×10^{2}	16.7×10^{-2}	15.8×10^{-2}	21.3×10^{-2}	15.6×10^{-2}
Mo	Adults	5.0	6×10^{-3}	6×10^{-3}	8×10^{-3}	8×10^{-3}	6×10^{-3}
	Children		6×10^{-3}	6×10^{-3}	0.01	8×10^{-3}	8×10^{-3}
Ag	Adults	5.0	120	4×10^{-4}	4.7×10^{-3}	4×10^{-3}	4×10^{-3}
	Children		1.2×10^{-2}	4×10^{-4}	5×10^{-3}	4×10^{-3}	4×10^{-3}

RfD: Values for the rest of the metals were not available.

As shown in Table 2, the CR_{ing} value was calculated for all selected heavy metals depending on the estimating value of cancer slope factor as provided by US EPA IRIS, database. The CR_{ing} value for Na, Mg, Al, Ba K, Ca, Fe and Sr for both adults and children was found to exceed the safe limit of cancer risk. Only the CR_{ing} values for the rest of the selected heavy metals were within the acceptable limit in all different five localities in the Yatta area. Heavy metal-induced toxicity and carcinogenicity involves many mechanistic aspects, some of which are not clearly elucidated or understood. However, each metal is known to have unique features and physic-chemical properties that confer to its specific toxicological mechanisms of action [15]. Accordingly, the CR_{ing} values for selected heavy metals, which are greater than the acceptable limit, are considered to be significant by the US EPA.

Table 2. CR_{ing} projection of heavy metals through rainwater harvesting cisterns (n = 74).

Parameter	**Receptor**	**Location**				
		Al-Hila	**Yatta Centre**	**Khallet Salih**	**Khallet El Mayya**	**Al-Hadidya**
Li	Adults	1.84×10^{-4}	7×10^{-4}	1×10^{-3}	9×10^{-4}	7×10^{-4}
	Children	2.1×10^{-4}	1×10^{-4}	2×10^{-4}	2×10^{-4}	2×10^{-4}
Be	Adults	1.73×10^{-6}	5.75×10^{-7}	1.15×10^{-6}	0.00	1.73×10^{-6}
	Children	4.6×10^{-7}	1.15×10^{-7}	2.3×10^{-7}	0.00	3.45×10^{-7}
Na	Adults	1.82	1.99	1.81	2.44	2.3
	Children	0.402	0.44	0.4	0.54	0.5
Mg	Adults	1.83	2.1	2.1	3.22	2.3
	Children	0.4	0.5	0.5	0.71	0.5
Al	Adults	0.03	8×10^{-3}	0.03	5×10^{-3}	7×10^{-3}
	Children	5.5×10^{-3}	2×10^{-3}	6×10^{-3}	1×10^{-3}	1.5×10^{-3}
Ba	Adults	4.7×10^{-3}	5×10^{-3}	6×10^{-3}	1.2×10^{-3}	5×10^{-3}
	Children	1.02×10^{-3}	1×10^{-3}	1×10^{-3}	1×10^{-3}	1×10^{-3}
Tl	Adults	4.6×10^{-6}	1.15×10^{-6}	5.75×10^{-5}	5.75×10^{-6}	5.75×10^{-6}
	Children	1.2×10^{-6}	2.3×10^{-7}	2.3×10^{-5}	1.15×10^{-6}	1.15×10^{-6}
Bi	Adults	5.8×10^{-6}	1.73×10^{-6}	2.3×10^{-5}	5.75×10^{-6}	5.75×10^{-6}
	Children	2.3×10^{-6}	1.15×10^{-7}	2.3×10^{-5}	1.15×10^{-6}	1.15×10^{-6}
K	Adults	0.55	0.5	0.5	0.4	0.4
	Children	0.12	0.12	0.1	9.4×10^{-2}	0.1
Ca	Adults	5.74	6.2	5.96	8.6	7.2
	Children	1.14	1.4	1.3	1.9	1.6
V	Adults	8×10^{-4}	6×10^{-4}	7×10^{-4}	5.2×10^{-4}	6×10^{-4}
	Children	1×10^{-4}	1×10^{-4}	1×10^{-4}	1×10^{-4}	1.3×10^{-4}
Fe	Adults	4×10^{-2}	0.02	4×10^{-4}	2.4×10^{-2}	0.03
	Children	7×10^{-3}	5×10^{-3}	8×10^{-3}	5.3×10^{-3}	6×10^{-3}
Co	Adults	5.75×10^{-6}	1.15×10^{-5}	5.75×10^{-5}	2.3×10^{-5}	1.15×10^{-5}
	Children	1.15×10^{-6}	2×10^{-4}	1.15×10^{-5}	6×10^{-4}	2.3×10^{-6}
Ga	Adults	3×10^{-4}	3×10^{-4}	4×10^{-4}	3.4×10^{-4}	3×10^{-4}
	Children	6.9×10^{-5}	6.9×10^{-5}	9.21×10^{-5}	8.05×10^{-5}	5.75×10^{-5}
Rb	Adults	6×10^{-4}	45×10^{-4}	8×10^{-4}	5.2×10^{-4}	3×10^{-4}
	Children	1×10^{-4}	1×10^{-4}	1×10^{-4}	1.3×10^{-4}	6.9×10^{-5}
Sr	Adults	0.05	0.05	0.05	0.07	0.05
	Children	0.02	0.01	0.01	1.5×10^{-2}	1.1×10^{-2}
Mo	Adults	1×10^{-4}	1×10^{-4}	2×10^{-4}	2.3×10^{-4}	1.7×10^{-4}
	Children	3.45×10^{-5}	3.45×10^{-5}	5.75×10^{-5}	4.6×10^{-5}	4.6×10^{-5}
Ag	Adults	1×10^{-4}	1.15×10^{-5}	2×10^{-3}	1.2×10^{-4}	1.2×10^{-4}
	Children	6.9×10^{-5}	1.15×10^{-6}	3.4×10^{-4}	2.3×10^{-5}	2.3×10^{-5}

3.4. Statistical Analysis

3.4.1. One-Way ANOVA Comparison

A one-way ANOVA was conducted to statistically compare the effect of selected heavy metal contamination in various sampling locations. The Analysis of variance for each parameter separately showed that the effect of Li, Na, Mg, Al, Ba, Bi, K, V, Fe, Co, Ga, Rb, Sr and Ag contamination in the five different selected sampling locations, with a p-value < 0.05 level set as not significant. Another contradictory finding from analytical one way ANOVA showed that there was a highly significant effect for each parameter of Be, Tl, Ca and Mo contamination at the $p < 0.05$ level for the different five selected sampling locations: ($F(4, 69) = 3.088$, $p = 0.021$), ($F(4, 69) = 2.527$, $p = 0.048$), ($F(4, 69) = 3.009$, $p = 0.024$), ($F(4, 69) = 2.746$, $p = 0.035$) and ($F(4, 69) = 1.897$, $p = 0.121$) respectively as shown in Table 3.

Table 3. One-Way ANOVA comparison of selected heavy metals contamination for different localities in the target area.

Heavy Metals		Sum of Squares	df *	Mean Square	F **	Significance ***
Ca	Between Groups	2,895,177,129.700	4	723,794,282.425	3.009	0.024
	Within Groups	16,597,141,602.778	69	240,538,284.098		
	Total	19,492,318,732.478	73			
Be	Between Groups	0.001	4	0.000	3.088	0.021
	Within Groups	0.008	69	0.000		
	Total	0.009	73			
Tl	Between Groups	2.864	4	0.716	2.527	0.048
	Within Groups	19.548	69	0.283		
	Total	22.412	73			
Mo	Between Groups	4.858	4	1.214	2.746	0.035
	Within Groups	30.513	69	0.442		
	Total	35.371	73			

The mean difference is significant at a level of 0.05. *: Degree of freedom. **: Factor. ***: Bold values are significant.

A Tukey Post-hoc test was run to determine if there were a different significant variation in the selected heavy metal against the different sampling locations. As determined by a post-hoc comparison using the Tukey HSD test, the concentration of Be had a statistically significant difference between Al-Hila area, compared to the Khallet El Mayya area (p = 0.033) at the 0.05 level of significance. However, the Be concentrations in the other sampling locations was not significantly different from Al-Hila and Khallet El Mayya (Supplementary Material Figure S1b). Correspondingly, the concentrations of Tl were significantly higher (p = 0.046) in the rainwater harvesting cisterns samples that were collected from Khallet El Mayya, as compared to the Yatta centre (Supplementary Material Figure S1g). Likewise, a post-hoc test revealed that the concentrations of Ca were statistically significantly higher (p = 0.041) in differences between Khallet El Mayya samples compared to Al-Hila samples, which were collected from rainwater harvesting cisterns. All other comparisons between different locations were not significant. However, there were no significant variations found for Li, Na, Mg, Al, Ba, Bi, K, V, Fe, Co, Ga, Rb, Sr, Mo and Ag in rainwater harvesting cisterns samples collected from different sampling locations, as shown in the Supplementary Material Figure S1a–h.

3.4.2. Inter-Metals Correlation Coefficient

The inter-metal correlation analysis provides certain information about the degree of interrelation and association between the heavy metal concentrations sources and their respective pathways [43,44]. A correlation of +1 indicates a perfect positive relationship between two variables. A correlation of −1 indicates that one variable changes inversely in relation to the other. A correlation of zero indicates that there is no relationship between the two variables [45]. The Inter metal correlation co-efficient of selected heavy metals in rainwater harvesting cisterns samples are summarized in Table 4. A strong positive correlation analysis was observed between Li-Rb, Mg-Sr, Tl-Bi, Tl-Co, Tl-Ag, Bi-Co, Bi-Ag and Co-Ag with r values of 0.929, 0.940, 0.999, 0.935, 0.968, 0.931, 0.972 and 0.908, respectively. There were also positive correlations in some heavy metal pairs such as Na-Mg (r = 0.739), Mg-Ba (r = 0.841) and Co-Ga (r = 0.696). This correlation suggests contamination input is mainly due to anthropogenic input and agriculture activity. The average of p-values was 0.0032 and it was statistically significant.

On the contrary, a negative correlation was noted for some heavy metal pairs such as Li-Mg, Ba-V, Al-Ca and K-Sr. Through the obtained results, it was demonstrated that the inter-metal correlations, having a strong positive correlation, were likely to be distributed by the same source. Also, those inter-metal correlations, having a negative correlations, were found to have counteractive distributions. This finding interprets the increasing of trends based on heavy metal concentrations, which will play an important role in the future projection in rainwater harvesting cisterns in the Yatta area.

Table 4. Correlation matrixes of selected heavy metals in rainwater harvesting cisterns (n = 74).

Rainwater Harvesting Cisterns	Li	Be	Na	Mg	Al	Ba	Tl	Bi	K	Ca	V	Fe	Co	Ga	Rb	Sr	Mo	Ag
Li	1																	
Be	0.072	1																
Na	−0.087	−0.203	1															
Mg	−0.320 **	−0.281 *	0.739 **	1														
AL	0.087	0.889 **	−0.283 *	−0.325 **	1													
Ba	−0.266 *	0.058	0.658 **	0.841 **	0.008	1												
Tl	0.107	0.640 **	−0.194	−0.173	0.728 **	0.185	1											
Bi	0.111	0.646 **	−0.196	−0.178	0.727 **	0.182	0.999 **	1										
K	0.721 **	0.107	0.125	−0.324 **	0.065	−0.189	−0.061	−0.058	1									
Ca	−0.402 **	−0.191	0.819 **	0.894 **	−0.227	0.825 **	−0.132	−0.137	−0.244 *	1								
V	0.613 **	0.513 **	−0.014	−0.474 **	0.435 **	−0.234 *	0.250 *	0.256 *	0.735 **	−0.315 **	1							
Fe	−0.011	0.775 **	0.018	−0.062	0.862 **	0.205	0.671 **	0.668 **	0.010	0.059	0.357 **	1						
Co	0.124	0.663 **	−0.109	−0.173	0.746 **	0.203	0.935 **	0.931 **	0.023	−0.037	0.345 **	0.705 **	1					
Ga	−0.109	0.459 **	0.333 **	0.470 **	0.463 **	0.808 **	0.722 **	0.721 **	−0.148	0.482 **	0.014	0.562 **	0.696 **	1				
Rb	0.929 **	−0.018	−0.021	−0.270 *	−0.003	−0.289 *	−0.034	−0.029	0.772 **	−0.363 **	0.608 **	−0.062	−0.008	−0.210	1			
Sr	−0.252 *	−0.288 *	0.620 **	0.940 **	−0.331 **	0.832 **	−0.185	−0.187	−0.292 *	0.791 **	−0.522 **	−0.145	−0.219	0.464 **	−0.242 *	1		
Mo	0.399 **	0.392 **	0.236 *	0.119	0.387 **	0.379 **	0.626 **	0.632 **	0.331 **	0.161	0.378 **	0.349 **	0.625 **	0.632 **	0.295 *	0.171	1	
Ag	0.130	0.716 **	−0.179	−0.165	0.760 **	0.186	0.968 **	0.972 **	−0.029	−0.132	0.275 *	0.697 **	0.908 **	0.711 **	0.001	−0.171	0.648 **	1

**: Correlation is significant at the 0.01 level (2-tailed); *: Correlation is significant at the 0.05 level (2-tailed).

3.4.3. Principle Component Analysis (PCA)

In order to qualitatively evaluate clustering behaviour and to enable interpretation, the distribution manner of heavy metals correlations in Yatta area and PCA with Varimax Kaiser Normalization were determined for rainwater harvesting cisterns samples. Table 4 summarizes the components and rotational component matrixes for rainwater harvesting cistern samples. Varimax factor loading coefficient with a correlation of: $r > 0.75$ are explained as strong significant factor loading; r values within 0.75–0.50 are considered as moderate factor loading; and r values within 0.50–0.30 are considered as weak factor loading.

The PCA of the obtained data indicates to the grouping of inter metals correlates with three factors in rainwater harvesting cisterns. The factor loading 1 based on component matrix accounting for 39.104% of the total variance, has high loadings of ($r > 0.76$) for Be, Al, Tl, Bi, Fe, Co and Ag and moderate loadings for V and Ga. The strong negative loadings on Na, Mg, Ca and Sr indicated an inverse relation for Be, Al, Tl, Bi, Fe, Co and Ag. From this perspective, based on PCA output on rainwater harvesting cisterns, it is interpreted that the sources of contamination by Fe and Co are largely anthropogenic inputs. Effluents from stone cutting activity are directly discharged onto surrounding land and near households of residents. The factor loading 2 based on component matrix, which accounted for 29.259% of the total variance, demonstrated high loadings of Mg, Ba, Ca and Sr and moderate loadings of Na and Ga. Mg and Ca contaminations could have resulted from the erosion of rocks. Mg and Ca arise principally from the weathering of rocks containing Ferro magnesium minerals and Marl Limestone with sandstone rock covers most of the Yatta area. In addition, Mg and Ca levels could be influenced more by agricultural activities. The results suggest that factor loading 2 may have both geologic and anthropogenic contributions, the factor loading 3 based on component matrix, which accounted for 15.812% of the total variance, displayed a high loading for K and Rb and moderate loading of Na and V. Accordingly, the source of contamination could be produced from both natural and anthropogenic input. Several common pesticides and fertilizers were used fairly extensively in agriculture contained substantial concentrations of metals such as Ca, K, Mg, Mo, Co and Fe, which contaminated the surrounding soils in the Yatta area.

On the other hand, in the case of rotated component matrix, PCA revealed that the total cumulative variance for factor loading 1 was 38.245% with a high loading on Be, Al, Tl, Bi, Fe, Co, Ga and Ag. The elements of Be, Tl, Bi and Ga have been known in the chemical industry for their toxicity. Thus, the source of contamination could have been produced from discharging effluents from industrial facilities input, located in the Yatta area surroundings.

Table 5 shows the factor loading for the selected heavy metals in rainwater harvesting cisterns. Factor loading 2 based on rotated component matrix contributed 26.410% to the total variance with a high loading on Na, Mg, Ba, Ca and Sr. Mg, Ca and Na levels could be affected by the erosion of milestone rocks, while Ba and Sr levels could be influenced by local industrial activities. The results suggest that factor loading 2 may also represent both geologic and anthropogenic sources. Factor loading 3 contributed 19.519% to the total variance with a high loading on Li, K, V and Rb. K levels could have been influenced by local agriculture activates in the study areas and weathering/leaching of sandstone rocks. Li, V and Rb levels could have been influenced by the industrial activities input. It means that factor loading 3 also has both anthropogenic and geogenic sources.

3.4.4. Cluster Analysis

Figure 2 shows the heavy metals diagram using median linkage of cluster analysis within rainwater harvesting cisterns in five different localities. Cluster analysis for heavy metals could be grouped into three clusters or groups. Tl, Bi, Ag and Co have good similarity and are clustered in one group, while Co exhibits individual forms and joined together with the first cluster. Be and Al with joining with Fe are clustered into another group. Li, K and V are clustered in one group and joined together with the group of Ga, Mo and Rb. Similarly, Mg, SR, Na, Ca and Ba are clustered with one group and joined together with the other three clusters of heavy metals. Strong similarities between the second and third group showed that relationships of these metals come from same origin. However, if selected heavy metals in the first and second group are taken as a pollution indicator, this would also indicate a strong anthropogenic input. The relation of selected heavy metals in the first group with other groups possibly came from natural and anthropogenic inputs such as industrial and agriculture activities.

Table 5. Factor loading for selected heavy metals in rainwater harvesting cisterns (n = 74).

Parameter	Component Matrix			Rotated Component Matrix		
	Factor 1	**Factor 2**	**Factor 3**	**Factor 1**	**Factor 2**	**Factor 3**
Rainwater Harvesting Cisterns						
Li	0.259	−0.516	0.707	0.048	−0.149	0.900
Be	0.825	−0.031	−0.121	0.811	−0.177	0.088
Na	−0.197	0.640	0.561	−0.141	0.846	0.164
Mg	−0.295	0.890	0.252	−0.142	0.931	−0.237
AL	0.875	−0.050	−0.188	0.865	−0.231	0.048
Ba	0.103	0.920	0.260	0.248	0.916	−0.155
Tl	0.929	0.158	−0.143	0.953	−0.034	0.008
Bi	0.931	0.153	−0.140	0.953	−0.037	0.013
K	0.169	−0.476	0.763	−0.039	−0.077	0.911
Ca	−0.211	0.882	0.242	−0.061	0.909	−0.223
V	0.514	−0.481	0.506	0.330	−0.241	0.765
Fe	0.791	0.183	−0.099	0.818	0.024	0.005
Co	0.926	0.149	−0.070	0.938	−0.007	0.075
Ga	0.640	0.723	0.104	0.751	0.609	−0.089
Rb	0.135	−0.535	0.753	−0.083	−0.130	0.921
Sr	−0.304	0.845	0.258	−0.162	0.895	−0.214
Mo	0.657	0.249	0.492	0.618	0.375	0.462
Ag	0.942	0.150	−0.111	0.959	−0.027	0.043
Total	7.039	5.267	2.846	6.884	4.754	3.513
Variance (%)	39.104	29.259	15.812	38.245	26.410	19.519
Cumulative (%)	39.104	68.362	84.174	38.245	64.655	84.174

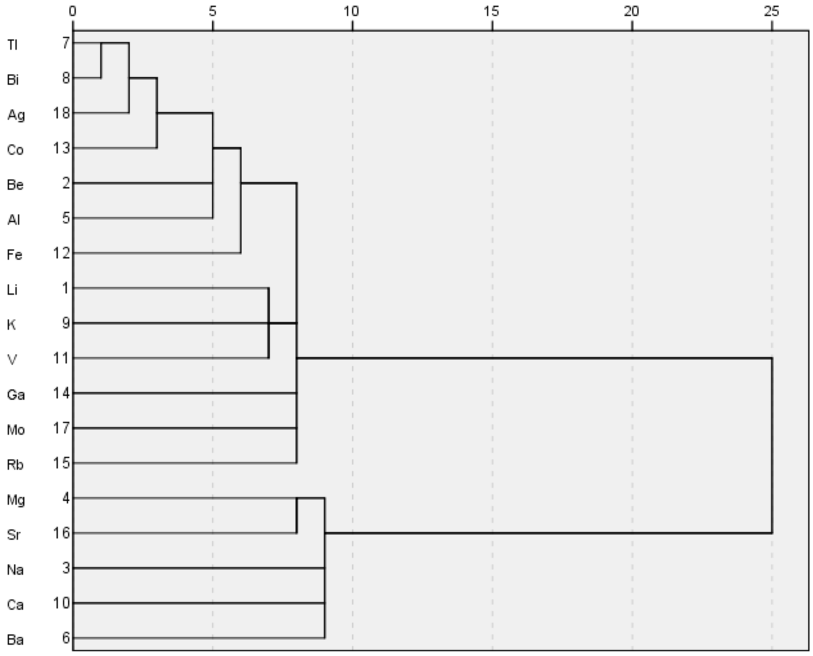

Figure 2. Heavy metals diagram using median linkage of cluster analysis within rainwater harvesting cisterns.

4. Conclusions

This study has indicated that selected heavy metal concentrations in rainwater harvesting cisterns samples of Al-Hadidya, Khallet El Mayya, Khallet Salih, Al-Hila and Yatta Centre are within the acceptable limits, in accordance with WHO and Palestinian standards. The exceptions are K and Al. The health risk index (HRI) of respondents in these areas were considered as low for all selected heavy metals, except the HRI value for Li for children in the Khallet Salih area, which was more than 1; indicating potential health risk and adverse effects on local residents.

This study revealed the following elements: Na, Mg, Al, Ba, K, Ca, Fe and Sr were elevated for both adults and children ($>10^{-6}$), through the ingestion of rainwater harvesting cisterns used for drinking. However, this does not imply that they pose a carcinogenic risk due to their concentrations alone. Al concentrations reported in this study do indicate a potential increased risk for dementia/Parkinson's disease, if it is obtained solely from the aforementioned source of water. Furthermore, Li concentrations reported in this study also indicate a potential increased risk of toxicity (causing diarrhoea, vomiting, stomach pains, muscle weakness and other symptoms), again if this is the only source of water available to the population of concern. The rest of the selected heavy metals were within the acceptable limit in the five different localities in the study area. These findings make a substantial contribution to perception of the linkage of the contamination of rainwater harvesting cisterns used for drinking and their direct collaboration with public health in the context of major anthropogenic activities in the Yatta area. Through this conclusion, we could predict that the levels of contaminants in the drinking water increases the risk for local residents in developing cancer, especially children.

The study has found that most of the heavy metals formed from anthropogenic input showed an almost regular pattern of depth concentrations. Higher heavy metal concentrations were observed at the Yatta area that may be attributed to the impact of anthropogenic activities in the study areas. However, geogenic sources such as natural weathering and erosion may also have contributed to the accumulation of selected heavy metals. The presence of geogenic and anthropogenic sources of heavy metals input was confirmed by the Principle component analysis and cluster analysis of the metals.

Based on the conclusion of this study, efforts should therefore be made by local authorities to eliminate the risks by applying technical and non-technical control measurements. The anthropogenic sources for heavy metals include mining, industrial productions, untreated sewage sludge and combustion by-products produced by coal burning power plants. Atmospheric emissions are probably the most preoccupant to human health and to the environment due to either the great quantity involved or their widespread dispersion.

In addition, there is a need for further case studies that will deepen the results of this research study, including human health risks in different exposure groups within different localities in the West Bank.

Supplementary Materials: Supplementary Materials: The following are available online at http://www.mdpi.com/2073-4441/11/2/238/s1, Figure S1: One-way ANOVA box plots comparison for Ca, Mg, Al, Fe, K, Na, Ag, Li, Co, Ba, Bi, Sr, Ga, V, Rb, Mo, Be and Tl, Table S1: The Concentrations (μg/L) of heavy metals in the selected rainwater harvesting cisterns (n total = 74), Table S2: Comparison of heavy metals concentrations (μg/L) with another study conducted in Kosovo, Table S3: Chronic daily intakes (CDIs, μg/ (kg·day)) of heavy metals in rainwater harvested in Yatta area cisterns.

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