

## Article

# Are Fresh Water and Reclaimed Water Safe for Vegetable Irrigation? Empirical Evidence from Lebanon

Marie Therese Abi Saab <sup>1,2,\*</sup> , Ihab Jomaa <sup>3</sup>, Rima El Hage <sup>1</sup>, Sleiman Skaf <sup>3</sup>, Salim Fahed <sup>1</sup>, Ziad Rizk <sup>1</sup>, Randa Massaad <sup>3</sup>, Dany Romanos <sup>1</sup> , Yara Khairallah <sup>1</sup> , Valerie Azzi <sup>1</sup>, Rhend Sleiman <sup>1</sup>, Roula Abi Saad <sup>1</sup>, Celine Hajjar <sup>1</sup>, Mohamed Houssemeddine Sellami <sup>4</sup> , Rodolph Aziz <sup>2</sup>, Rita Sfeir <sup>2</sup>, Marie Helene Nassif <sup>5</sup> and Javier Mateo-Sagasta <sup>6</sup>

<sup>1</sup> Lebanese Agricultural Research Institute, Fanar P.O. Box 90-1965, Lebanon; mrelhage@lari.gov.lb (R.E.H.); salimfahed@gmail.com (S.F.); zrizzk@lari.gov.lb (Z.R.); danny\_romanos@hotmail.com (D.R.); yara.khairallah@hotmail.com (Y.K.); valerie.azzi@hotmail.com (V.A.); rhend.t.sleiman@gmail.com (R.S.); roula\_abi\_saad@hotmail.com (R.A.S.); celine-hajjar@hotmail.com (C.H.)

<sup>2</sup> Department of Agriculture, The School of Engineering, The Holy Spirit University of Kaslik, Kaslik P.O. Box 446, Lebanon; rodolphaziz@live.com (R.A.); ritasfeir1@outlook.com (R.S.)

<sup>3</sup> Lebanese Agricultural Research Institute, Tal Amara P.O. Box 287, Lebanon; ijomaa@lari.gov.lb (I.J.); sleimskaif@hotmail.com (S.S.); rmasaad@lari.gov.lb (R.M.)

<sup>4</sup> Interdepartmental Research Centre on the “Earth Critical Zone”, University of Naples Federico II, 80055 Portici, Italy; hsellemi@yahoo.fr

<sup>5</sup> International Water Management Institute—MENA Office 7 Abdel Hadi Saleh St., Off Nile St., Giza 12611, Egypt; mariehelene.nassif@gmail.com

<sup>6</sup> International Water Management Institute, 127, Sunil Mawatha, Battaramulla, Colombo 10120, Sri Lanka; j.mateo-sagasta@cgiar.org

\* Correspondence: mtabisaa@lari.gov.lb; Tel.: +961-390-2994



**Citation:** Abi Saab, M.T.; Jomaa, I.; El Hage, R.; Skaf, S.; Fahed, S.; Rizk, Z.; Massaad, R.; Romanos, D.; Khairallah, Y.; Azzi, V.; et al. Are Fresh Water and Reclaimed Water Safe for Vegetable Irrigation? Empirical Evidence from Lebanon. *Water* **2022**, *14*, 1437. <https://doi.org/10.3390/w14091437>

Academic Editor: Edward Laws

Received: 11 March 2022

Accepted: 26 April 2022

Published: 30 April 2022

**Publisher’s Note:** MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



**Copyright:** © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

**Abstract:** The use of polluted water to irrigate is an increasing problem in the developing world. Lebanon is a case in point, with heavily polluted irrigation waters, particularly in the Litani River Basin. This study evaluated the potential health risks of irrigating vegetables (radishes, parsley, onions, and lettuce) using three water sources (groundwater, river water, and treated wastewater) and three irrigation methods (drip, sprinkler, and surface) over two growing seasons in 2019 and 2020. Water, crop, and soil samples were analyzed for physicochemical parameters, pathogens, and metals (Cu, Cd, Ni, Cr, and Zn). In addition, the bioaccumulation factor, estimated dietary intakes, health risk index, and target hazard quotients were calculated to assess the health risk associated with metal contamination. The study showed that, for water with less than 2 log *E. coli* CFU/100 mL, no pathogens (*Escherichia coli*, *salmonella*, parasite eggs) were detected in irrigated vegetables, irrespective of the irrigation method. With over 2 log *E. coli* CFU/100 mL in the water, 8.33% of the sprinkler- and surface-irrigated vegetables, and 2.78% of the drip-irrigated root crops (radishes and onions), showed some degree of parasitic contamination. *E. coli* appeared only on root crops when irrigated with water having over 3 log CFU/100 mL. The concentrations of most metals were significantly lower than the safe limits of the FAO/WHO of the Food Standards Programme Codex, except for zinc and chromium. The trends in the bioaccumulation factor and the estimated dietary intakes of metals were in the order of Cu < Cd < Ni < Cr < Zn. The target hazard quotient values for all metals were lower than 1.0. Under trial conditions, the adoption of drip irrigation with water with less than 3 log *E. coli* CFU/100 mL proved to be safe, even for vegetables consumed raw, except for root crops such as onions and radishes that should not be irrigated with water having over 2 log *E. coli* CFU/100 mL. Treated wastewater had no adverse effect on vegetable quality compared to vegetables irrigated with other water sources. These results support efforts to update the Lebanese standards for water reuse in agriculture; standards proposed in 2011 by the FAO, and currently being reviewed by the Lebanese Institution of Standards. This research will inform a sustainable water management policy aimed at protecting the Litani River watershed by monitoring water quality.

**Keywords:** wastewater reuse; health risks; water pollution; water management; vegetable irrigation

## 1. Introduction

The safe use of reclaimed water is increasingly recognized by governments and international organizations as part of the solution to water scarcity and pollution [1]. Using reclaimed water becomes attractive when alternative freshwater sources are unavailable or contaminated [2], which is often the case in the Middle East and North Africa. The region is home to 6% of the global population with just 1% of the Earth's renewable freshwater sources [3]. Over 17% and 14% of ground and surface water bodies in the region are heavily contaminated by pathogens and organic pollutants [4]. Emerging water pollutants are a growing concern [5].

The Litani River Basin in Lebanon is a dramatic regional example of both pollution and scarcity. The Litani region is in the worst condition on record, with major water quality and quantity issues affecting agricultural productivity, food security, food safety, and public health. It was recently described as a “dead” river [6,7]. Most domestic and industrial water is untreated. Some of the main causative factors affecting basin health are solid waste bulk disposal sites on riverbanks, direct discharge of non-treated sewage and industrial waters, and irrigation with contaminated waters. Both surface and groundwater sources suffer from physiochemical and biological contamination, and the pollution levels exceed the international norms most of the time [8–12]. Research suggests that existing management approaches in the river basin are not enough to mitigate the environmental problems or achieve Sustainable Development Goal 6 [13].

There are still few studies on how deterioration of water quality affects crops grown in the Litani Basin that are consumed by local inhabitants and a wider range of consumers. There are also few studies in Lebanon on the agricultural use of treated water and its potential health, agronomic, and environmental risks, particularly in the Bekaa Valley, where conventional treatment plants are now operational. For example, some studies deal with irrigating table grapes with treated effluent [14,15], as well as eggplants [16], and spinach [9]. Investigating threats to public health, environment, food security, and safety is therefore a necessity.

An experimental trial was conducted in the Bekaa Valley to grow important crops that are consumed raw in the Mediterranean diet using different irrigation resources and methods. The objective was to assess the impact on yield, mineral content, and health risks. The findings would constitute local evidence regarding the risks associated with irrigation water. This is important because the reuse of treated water is expected to increase in the coming years. In addition, the study would inform revisions to the Lebanese guidelines for water reuse in agriculture proposed by the Food and Agriculture Organization of the United Nations (FAO) in 2011, which are under review by the Lebanese Institution of Standards (LIBNOR). Finally, research studies would support efforts to develop and implement long-term policies for managing and monitoring water quality in the basin, ultimately leading to its conservation and sustainable use [17].

## 2. Materials and Methods

### 2.1. Characteristics of the Study Area

A trial was conducted at the Lebanese Agricultural Research Institute (LARI) in Bekaa Valley of Lebanon (33°51'44" N latitude, 35°59'32" E longitude and 905 m above sea level), specifically in the upper Litani watershed, during two summer seasons in 2019 and 2020 (Figure 1). The deteriorating water quality and water scarcity problems in this watershed result from climate variability, increasing demographic pressure, unsustainable farming practices, and the unregulated discharge of wastewater and solid wastes. The location of the field was near the wastewater treatment plant in Ablah Village. The plant discharges a secondary-level treated effluent and is adjacent to the Litani River, the main irrigation

water source in the Bekaa Valley (Figure 1). The soil at the experimental trial site has a sandy loam texture, a pH with a mean value of  $7.23 \pm 0.15$ , electrical conductivity of  $0.016 \pm 0.02 \text{ dS} \cdot \text{m}^{-1}$ , and organic matter content of  $1.55\% \pm 0.30\%$ .



**Figure 1.** Location of the experimental field showing irrigation resources: Ablah Waste Water Treatment Plant and the Litani River.

Similar to other Mediterranean locations, the climate of the research area is characterized by a hot dry season from April to October. The major weather data were obtained from an agro-meteorological station at the LARI experimental site. Figure S1 shows the weather regime in terms of reference evapotranspiration ( $E_{To}$ ), precipitation ( $P$ ), maximum temperature ( $T_{max}$ ), minimum temperature ( $T_{min}$ ), and mean relative humidity ( $RH_{mean}$ ) for the seasons 2019 and 2020. For the 2019 season, overall average maximum and minimum air temperatures from August to October were  $30.97$  and  $11.97$  °C, respectively. The total precipitation was 8 mm. In the 2020 season, overall average maximum and minimum air temperatures for the same period were  $33.23$  and  $13.60$  °C. Rainfall was negligible, with a value of 0.4 mm.

## 2.2. Treatments and Agronomic Management

The field experiment was conducted on a  $2700 \text{ m}^2$  field area divided into nine plots of  $234 \text{ m}^2$  each. Each plot was further divided into 12 sub-plots of  $9 \text{ m}^2$  ( $3 \times 3 \text{ m}^2$ ). Four commonly used kitchen vegetables (parsley, lettuce, radishes, and green onions) grown in the Bekaa Valley were selected for this trial. The four vegetables were grown under three water source conditions (GW: groundwater extracted from an existing well at the LARI station; TW: secondary-level treated municipal water from Ablah municipality; RW: surface water from the Litani River with a perennial water regime and considered a major resource for irrigation in the region). Three irrigation methods were used (Dr: drip, Sp: sprinkler, and Sr: surface irrigation). Thirty-six treatments were replicated three times within 108 sub-plots. The experimental design was a strip-split-plot system in a randomized complete block with three replicates with (i) water resource as the horizontal strip, (ii) irrigation system as the vertical strip, and (iii) crops as the sub-plot factor. The experimental layout is shown in Figure S1 in Supplementary Materials.

The experiment featured separate reservoirs and head units for fresh water treated effluent, and Litani River water irrigation treatments. Each head unit consisted of a pump, disk filter, and pressure gauges. Filters were manually cleaned. Surface, sprinkler, and drip

methods drew water from three sources, each from a different reservoir. All the plots had a separate irrigation system. The drip-irrigated sub-plots consisted of low-polyethylene surface laterals with 16 mm external diameters. All laterals were provided with in-line drippers with emitters placed 0.40 m apart (theoretical discharge rate of  $4 \text{ L h}^{-1}$  at a pressure of 100 kPa). The distance between laterals was 1 m. The sprinkler-irrigated sub-plots had quarter-circle sprayers delivering water at a low angle, covering a radius of 3 m and providing a flow of  $150 \text{ L} \cdot \text{m}^{-1}$  each and were mounted on risers 30 cm high. These were installed on the corners of sub-plots for better water distribution. Ridges were constructed on all sub-plots to avoid flooding in nearby sub-plots. Irrigation management consisted of checking the meteorological conditions. The soil–water balance in the active root zone was measured. The Penman–Monteith equation was used to determine the daily reference evapotranspiration using meteorological data. No fertilizers were used. The planting dates were 19 August 2019 and 10 August 2020.

### 2.3. Data Collection

#### 2.3.1. Water Sampling and Analysis

Water samples were taken from the three water sources and analyzed for their physicochemical characteristics, pathogen content, and presence of metals. Samples were taken every two weeks during irrigation season in both years of study. The parameters analyzed included biochemical oxygen demand ( $\text{BOD}_5$ ), chemical oxygen demand (COD), total suspended solids (TSS), pH, electrical conductivity (EC), selected metals (Cd, Cr, Cu, Ni, Zn), fecal coliforms, *E. coli*, *salmonella*, and helminth eggs. The  $\text{BOD}_5$  was analyzed in accordance with standard methods (APHA, 1998) [18], and chemical oxygen demand (COD) and the total suspended solids (TSS) were determined by APHA (2005) [19]. The pH and EC measurements were obtained in accordance with APHA (2005) [19]. The bacterial analysis and test for the presence of metals were performed in accordance with standard methods [20]. The presence of helminth eggs in the three water sources was also assessed. The samples were decanted for 24 h in the laboratory, and the sediment was retrieved (100 to 300 mL) then centrifuged for 15 min at 1200 rpm (revolutions  $\text{min}^{-1}$ ). After concentration, helminth eggs were recognized at magnifications of 100 in a McMaster counting cell [21], with Sheater's solution as a flotation liquid as in [22].

#### 2.3.2. Soil Sampling and Analysis

Before and after each growing season (August and end of October in 2019 and 2020), soil samples from each sub-plot were collected from the upper horizon (0–30 cm) and analyzed for physicochemical properties and the presence of metals using standard procedures. The soil samples were dried and sieved at 2 mm before analysis. The parameters studied were as follows: soil pH analyzed following the method described by ISO 10390 (2005) [23] using a Thermo Scientific Orion Star pH meter, and electrical conductivity (EC) analyzed according to ISO 11265 (1994) [24] using a Thermo Scientific Orion Star conductivity meter. Nitrogen (N) was determined using the Kjeldahl distillation process (ISO 11261, 1995) [25]. Phosphorus was examined in accordance with ISO 11263 (1994) [26], while potassium was determined using a flame photometer. Organic matter was analyzed in accordance with [27]. Chromium (Cr), nickel (Ni), zinc (Zn), copper (Cu), and cadmium (Cd) were extracted with diethylenetriaminepentaacetic acid (DTPA) and identified using atomic absorption spectrophotometer [28,29].

The field soil had a sandy loam texture with 7% clay, 51% sand, and 42% silt, and organic matter content of  $1.5\% \pm 0.62\%$ . The soil pH mean value was  $7.23 \pm 0.15$  and the electrical conductivity was  $0.016 \pm 0.02 \text{ dS} \cdot \text{m}^{-1}$ .

#### 2.3.3. Plant Sampling and Analysis

##### Plant Yield and Mineral Content

From each sub-plot, plant samples were taken to determine plant-related parameters. Five randomly selected lettuce heads were harvested from each sub-plot while the radish,



parsley, and onion samples were taken from a 1 m<sup>2</sup> frame. The plant fresh yield (kg·m<sup>-2</sup>) and the percentage of dry matter were determined in each sub-plot. P, K, Ca, and Mg in vegetables contribute to the required daily human dietary intake by 11%, 35%, 7%, and 24%, respectively [30,31]. The harvested vegetables were analyzed for their mineral content following the procedure described in [32]. All samples were dried in an oven at 70 °C for 48 hours before being ground using a laboratory blender. Samples were microwave digested by adding a strong acid to dried powdered samples. Filtrates were taken from each sample to assess their mineral composition. Nitrogen was measured using the Kjeldahl method. Phosphorus was analyzed by spectrophotometry while potassium, calcium, and magnesium were analyzed by atomic absorption spectrometry [32].

#### Pathogen Contamination of Edible Plant Parts

Plants were randomly selected from the middle row of each sub-plot at maturity to determine pathogenic loading in edible parts (*E. coli*, *salmonella*, helminth eggs, etc.). In the laboratory, *E. coli* were analyzed in accordance with ISO 16649-2 (2001) [33]. The *salmonella* detection protocol was performed according to ISO 6579 (2017) [34]. In the 2020 season, the plant samples were also analyzed for the detection of *Listeria monocytogenes* according to ISO 11290-1 (2017) protocol [35], and *E. coli* 0157 (ISO/TS 13136, 2012) [36]. Crop samples were transferred to the laboratory in sterile plastic bags for parasitological analysis. A portion of 200 g from each sample was carefully washed with tap water, and the washing water was filtered and left to settle for 24 h. The top layer was removed, and the leftover washing water was centrifuged for 15 min at 1200 rpm. The supernatant was discarded, and the residue was carefully collected and examined by following the technique of [21]. Microscopic examination was carried out in a McMaster counting cell at a magnification of 100 times. Because radish tubercles form in the ground, they were weighed (200 g), then manually brushed and washed with tap water and treated using the procedure described in [22].

#### Metal Contamination of Plants

Randomly selected plants from each sub-plot were collected to determine heavy metal accumulation in edible parts of the four vegetables. The samples were ground and oven-dried at 70 °C until they reached a constant weight. For the microwave digestion, a half gram of each sample was measured and mixed with 5 mL of concentrated HNO<sub>3</sub> and 1 mL of HCl at 100 °C in microwave vessels. The obtained solution was filtered and brought to a 50 mL solution by adding deionized distilled water. The metal concentrations were determined by atomic absorption spectrometer [37]. The maximum permissible heavy metal limits for vegetable crops used in this study are reported in Table S1.

### 2.4. Data Analysis

#### 2.4.1. Statistical Analysis

All dependent variables were assessed for normal distribution and variance homogeneity using the Kolmogorov–Smirnov and Bartlett tests. If the normality assumption was broken, we used the Box–Cox transformation to treat the data. The field experiment plot was arranged in a strip-split-plot system in a randomized complete block with three replications [38], with water source as the horizontal strip, irrigation system as the vertical strip, and crops as the sub-plot factor. To examine the significance of the difference between means, the least significant difference (LSD) was determined. SAS University Edition was used for all statistical analyses (Cary, NC, USA).

#### 2.4.2. Bioaccumulation Factor (BAF)

The bioaccumulation factor (BAF) was calculated as described in [39,40]. It is defined as the ratio of the metal concentrations in the plant to those in the soil and was calculated as

$$\text{BAF} = C_{\text{plant}}/C_{\text{soil}} \quad (1)$$

where  $C_{\text{plant}}$  and  $C_{\text{soil}}$  are metal concentrations in the edible parts of vegetables and soils.

#### 2.4.3. Estimated Daily Intake (EDI) of Metals

The estimated daily intake (EDI) of metals was calculated based on the metal concentrations in crops and the consumption of the respective food crop, as described in [39,41].

The following equation was used:

$$\text{EDI} = C_{\text{metal}} \times W_{\text{food}} / B_w \quad (2)$$

where  $C_{\text{metal}}$  is the concentration of heavy metals in crops ( $\text{mg} \cdot \text{kg}^{-1}$ ),  $W_{\text{food}}$  is the daily average consumption of vegetables, and  $B_w$  is body weight. The average adult and child body weights were calculated to be 73 and 32.7 kg, respectively [39]. The average daily vegetable intakes for adults and children were taken as 0.345 and 0.232 kg, respectively [42]. Index values less than 1.0 indicate safe levels, whereas values over 1.0 are associated with negative health impacts.

#### 2.4.4. Health Risk Assessment

The health risk index (HRI) and target hazard quotients (THQ) were calculated to assess the health risk associated with metal contamination. Data were analyzed using the methodology reported in [39].

Usually, HRI depends on the daily intake of metals compared to a reference oral dose [43]. It is calculated according to the formula:

$$\text{HRI} = \sum_n (C_n \times D_n / \text{RfD} \times B_w) \quad (3)$$

where  $C_n$  denotes the mean metal concentrations in vegetables on a fresh weight basis ( $\text{mg} \cdot \text{kg}^{-1}$ ),  $D_n$  denotes the average daily consumption rate of a certain vegetable in a year,  $\text{RfD}$  denotes the safe limit of oral exposure for a lifetime, and  $B_w$  denotes the average body weight. The dietary reference intakes (DRI) of the elements were taken as  $\text{RfD}$  [44,45].

The THQ helps to assess the non-carcinogenic health risks for humans who consume metal-contaminated vegetables. It is a ratio of the determined dose of a pollutant to a reference dose level [39,46]. It is calculated as

$$\text{THQ} = C \times I \cdot 10^{-3} \times \text{EF}_r \times \text{ED}_{\text{tot}} / \text{RfD} \times \text{BW}_a \times \text{AT}_n \quad (4)$$

where  $C$  is the mean metal level in vegetables ( $\text{mg} \cdot \text{kg}^{-1}$ , fresh weight);  $I$  is the per capita ingestion rate ( $255 \text{ g} \cdot \text{d}^{-1}$ );  $\text{EF}_r$  is the exposure frequency ( $350 \text{ d} \cdot \text{yr}^{-1}$ );  $\text{ED}_{\text{tot}}$  is the total exposure duration (70 years);  $\text{BW}_a$  is the average body weight of an adult; and  $\text{AT}_n$  is the averaging time, non-carcinogens ( $\text{ED}_{\text{tot}} \times 365 \text{ d} \cdot \text{yr}^{-1}$ ). Table S2 shows the parameters used in the calculation of EDI, HRI, and THQ.

### 3. Results

#### 3.1. Water Quality

Table 1 shows the seasonal averages of the physicochemical, microbiological, and metal parameters of the fresh, river, and treated water evaluated during the experiment for the two growing seasons. According to the guidelines for the interpretation of water quality for irrigation [47], all water sources showed a slight to moderate salinity (values between 0.7 and 3  $\text{dS} \cdot \text{m}^{-1}$ ) but are still suitable for irrigation. The salinity of GW was the lowest ( $0.78 \pm 0.16 \text{ dS} \cdot \text{m}^{-1}$  in 2019 and  $0.92 \pm 0.09 \text{ dS} \cdot \text{m}^{-1}$  in 2020), while that of RW was the highest ( $2.29 \pm 0.81 \text{ dS} \cdot \text{m}^{-1}$  in 2019 and  $2.73 \pm 0.74 \text{ dS} \cdot \text{m}^{-1}$  in 2020). The salinity in TW presented mean values of  $1.14 \pm 0.15 \text{ dS} \cdot \text{m}^{-1}$  in 2019 and  $1.01 \pm 0.11 \text{ dS} \cdot \text{m}^{-1}$  in 2020. The RW contained higher levels of sodium compared to GW and TW ( $115.59 \pm 5.20 \text{ mg} \cdot \text{L}^{-1}$  in 2019 and  $90.93 \pm 26.55 \text{ mg} \cdot \text{L}^{-1}$  in 2020). The COD values were within the admissible limits of 125  $\text{mg} \cdot \text{L}^{-1}$  for the three water sources in 2019. RW was an exception in season 2020, with a COD mean value of  $273.12 \pm 219.21 \text{ mg} \cdot \text{L}^{-1}$ . The  $\text{BOD}_5$  in GW (mean

values of  $16.83 \pm 2.64 \text{ mg}\cdot\text{L}^{-1}$  and  $6.33 \pm 9.81 \text{ mg}\cdot\text{L}^{-1}$  for seasons 2019 and 2020) was within the admissible limits of Water Category I as proposed by the Lebanese guidelines (Table S3). The values in RW ( $47.33 \pm 15.63 \text{ mg}\cdot\text{L}^{-1}$  in 2019 and  $79.67 \pm 61.83$  in 2020) were higher than the limits of wastewater reuse Category I [48]. The BOD<sub>5</sub> in TW (mean value of  $27 \pm 8.65 \text{ mg}\cdot\text{L}^{-1}$  in season 2019) showed an improvement in season 2020 (mean value of  $20.01 \pm 14.79 \text{ mg}\cdot\text{L}^{-1}$ ) and fall within the FAO admissible limits of Water Category I. Phosphorus and potassium levels were highest in RW, followed by TW, then GW. The lowest nitrate content was observed in RW ( $9.66 \pm 4.73 \text{ mg}\cdot\text{L}^{-1}$  in 2019 and  $9.12 \pm 11.40 \text{ mg}\cdot\text{L}^{-1}$  in 2020) but was higher in GW and TW with mean values of  $26.93 \pm 6.90$  and  $14.96 \pm 4.79 \text{ mg}\cdot\text{L}^{-1}$  in 2019 and  $20.42 \pm 8.11$  and  $16.50 \pm 13.83 \text{ mg}\cdot\text{L}^{-1}$  in 2020. These values were always within the admissible value of  $30 \text{ mg}\cdot\text{L}^{-1}$  of Water Category I. The higher content of nitrates in GW is mainly due to excessive nitrate applications in farm fields in the region. The unfortunate result is that nitrates end up in groundwater via the leaching process. This was confirmed by other local studies that analyzed groundwater in several wells in the Litani Basin and showed that nitrate concentrations sometimes exceeded  $300 \text{ mg}\cdot\text{L}^{-1}$  [12]. It has also been shown that the groundwater in the Litani Basin is highly vulnerable to all sources of contaminants, especially in a large area of the plain near Zahle City, where the water table is less than 2 m below ground level [13].

Fecal coliforms, mainly *E. Coli*, were present in all three water sources. The concentration of *E. coli* in GW was below the limits for Water Category I [48] in both seasons. However, they were highly present in RW ( $1.78 \times 10^5 \pm 1.78 \times 10^5 \text{ CFU}/100 \text{ mL}$  in 2019 and  $4.84 \times 10^5 \pm 6.00 \times 10^5 \text{ CFU}/100 \text{ mL}$  in 2020) and most often exceeded the limit of 1000 CFU/100 mL proposed by the WHO as sufficient for irrigation of all crops. The mean value of *E. coli* in TW in season 2019 was above the FAO limit of Water Category I but within the limit in season 2020. In season 2020, the bacterial quality of TW was better than GW with *E. coli* mean values of  $9.67 \pm 1.15 \text{ CFU}/100 \text{ mL}$  and  $1.84 \times 10^2 \pm 1.02 \times 10^2 \text{ CFU}/100 \text{ mL}$ , respectively, for both water sources. *Salmonella* was not detected in GW in either year but was present in RW. In season 2019, parasites were present in all water sources and the concentration of some species in TW and RW exceeded the limit value of  $1 \text{ egg}\cdot\text{L}^{-1}$  [48]. In season 2020, some parasites found in GW and RW were above the FAO limits.

Most metals were present in all water sources but did not exceed the limits in the Lebanese guidelines [48]. During both years, metal concentrations were highly variable and fluctuated. This finding agrees with other studies which have reported that, contrary to soil and plants subjected to metal enrichment and accumulation, the quantity of metal pollutants varies in water depending on the presence of pollutants such as batteries, various types of waste, and dust [49].

When comparing the quality of the three water sources in both seasons, it was found that GW is of Category I while RW is of Category III, as proposed by the Lebanese guidelines. This suggests that GW can be used to irrigate vegetables that are only eaten when cooked and that RW is not suitable for irrigating any vegetables [49]. TW was classified as Category III in season 2019 but quality substantially improved in season 2020 and the effluent was upgraded to Category I [49]. The microbial quality of TW in season 2020 became better than GW. This confirms the importance of the treatment efficiency of effluent and that treatment plants must be well maintained and upgraded to provide an effluent that is consistently suitable for irrigation.

### 3.2. Assessment of Yield and Mineral Content of Crops

The contribution of vegetables to the human dietary intake of total P, K, Ca, and Mg is normally 11%, 35%, 7%, and 24%, respectively [30,31]. In the context of the current study, the effects of irrigating with different water sources and methods were assessed on four crops for two growing seasons (2019 and 2020) in relation to their fresh yield ( $\text{g}\cdot\text{m}^{-2}$ ) and mineral content. The results are presented in Table 2.

**Table 1.** Average quality of groundwater, treated effluent, and river water for growing seasons 2019 and 2020.

	GW				TW				RW				Allowable Limits *		
	2019		2020		2019		2020		2019		2020		Category I	Category II	Category III
	Value	STDEV	Value	STDEV	Value	STDEV	Value	STDEV	Value	STDEV	Value	STDEV			
Physicochemical Parameters (mg·L <sup>-1</sup> )															
pH	7.21	0.14	7.80	0.21	7.72	0.30	7.68	0.19	7.31	0.18	7.62	0.12			
Electrical Conductivity (dS·m <sup>-1</sup> )	0.78	0.16	0.92	0.09	1.14	0.15	1.01	0.11	2.29	0.81	2.73	0.74			
COD	36.33	11.74	20.24	18.21	66.95	19.51	20.13	18.82	105.25	34.02	273.12	219.21	125	250	250
BOD <sub>5</sub>	16.83	2.64	6.33	9.81	27.00	8.65	20.01	14.79	47.33	15.63	79.67	61.83	25	100	100
Total suspended Solids	4.11	1.47	3.40	2.96	27.51	6.34	3.93	2.13	33.55	7.90	13.83	4.36	60	200	200
Nitrates	26.93	6.90	20.42	8.11	14.96	4.79	16.50	13.83	9.66	4.73	9.12	11.40	30	30	30
Phosphates	0.24	0.56	0.01	0.01	0.77	0.30	2.09	2.64	4.88	3.83	3.01	1.63			
Potassium	2.33	0.55	1.67	0.66	28.58	12.47	14.68	14.64	78.56	13.87	38.63	7.64			
Sodium	15.96	2.55	14.78	1.10	26.00	3.15	29.02	17.04	115.59	5.20	90.93	26.55			
Pathogens in Water															
E. coli (CFU/100 mL)	6.58 × 10	3.53 × 10	1.84 × 10 <sup>2</sup>	1.06 × 10 <sup>2</sup>	1.57 × 10 <sup>3</sup>	5.75 × 10 <sup>2</sup>	9.67	1.15	1.78 × 10 <sup>5</sup>	1.65 × 10 <sup>5</sup>	4.84 × 10 <sup>5</sup>	6.00 × 10 <sup>5</sup>	<200	<1000	
Salmonella	Absence		Absence		Sometimes present		Absent		Sometimes present		Sometimes present		Absent	Absent	Absent
Parasites (ova/ /L)															
Ascaris	0.00	0.00	0.00	0.00	2.00	2.76	0.00	0.00	2.67	3.67	1.00	1.00			
Chilomastix Mesnili	0.50	0.55	1.00	1.00	0.83	0.98	0.00	0.00	1.50	1.97	1.00	2.00			
Blastocystis	0.67	0.52	2.00	1.00	1.00	1.55	0.55	0.30	1.67	2.73	1.00	2.00	<1	<1	<1
Endolimax Nana	0.00	0.00	1.00	2.00	0.67	1.03	0.00	0.00	1.00	1.67	2.00	2.00			
Trace metals (mg·L <sup>-1</sup> )															
Cr	1.15	0.00	0.25	0.36	1.16	0.01	0.10	0.13	1.15	0.01	0.13	0.17		2	
Ni	0.20	0.00	0.03	0.08	0.21	0.00	0.13	0.22	0.22	0.01	0.04	0.08		0.5	
Zn	0.36	0.13	0.02	0.02	0.16	0.03	0.00	0.00	0.15	0.04	0.22	0.30		5	
Cu	0.47	0.20	0.07	0.16	0.40	0.28	0.05	0.12	0.38	0.04	0.10	0.24		0.5	
Cd	0.00	0.00	0.02	0.04	0.00	0.00	0.05	0.12	0.01	0.00	0.08	0.12		0.2	

Note: \* Effluent specifications for wastewater reuse in irrigation based on proposed Lebanese guidelines (FAO, 2011). Further description is provided in Supplementary Table S3. GW: groundwater; RW: river water; TW: treated water; COD: chemical oxygen demand; BOD<sub>5</sub>: Biological oxygen demand. Values in bold exceed the limits of FAO effluent Category I.



**Table 2.** Effects of water source, irrigation method, and crop on yield and nutritional value.

Treatment	Fresh Weight (Kg·m <sup>-2</sup> )		N (mg 100 g <sup>-1</sup> FW *)		P (mg 100 g <sup>-1</sup> FW)		K (mg 100 g <sup>-1</sup> FW)		Ca (mg 100 g <sup>-1</sup> FW)		Mg (mg 100 g <sup>-1</sup> FW)	
	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
<b>Water Sources</b>												
Groundwater	2.59 ± 2.29 b	2.76 ± 2.43 b	146.52 ± 26.14 a	78.12 ± 15.96 a	22.97 ± 7.92 b	11.76 ± 7.56 b	164.74 ± 27.72 b	259.56 ± 64.66 b	60.19 ± 21.38 b	106.68 ± 56.28 b	21.38 ± 7.92 b	26.88 ± 25.2 b
River—Litani	2.73 ± 2.82 a	3.49 ± 2.59 a	63.018 ± 28.00 c	70.05 ± 21.1 c	28.79 ± 7.78 a	14.35 ± 13.50 a	210.06 ± 42.79 a	285.27 ± 227.88 a	83.25 ± 14.78 a	124.91 ± 91.99 a	29.56 ± 5.45 a	37.98 ± 49.78 a
Treated Water—Ablah	2.65 ± 2.70 b	3.45 ± 2.63 a	126.55 ± 30.68 b	74.802 ± 32.88 b	31.45 ± 10.74 a	11.50 ± 10.68 b	213.226 ± 88.97 a	279.19 ± 234.27 a	80.54 ± 16.88 a	121.66 ± 69.05 a	29.91 ± 6.14 a	32.13 ± 23.84 ab
<b>Irrigation Methods</b>												
Drip	4.34 ± 3.27 a	3.70 ± 2.70 a	119.92 ± 37.95 a	76.12 ± 19.03 a	30.36 ± 8.35 a	12.71 ± 9.08 ab	214.80 ± 78.18 a	289.66 ± 207.93 ab	81.21 ± 14.42 a	99.91 ± 66.61 b	30.36 ± 6.07 a	40.44 ± 17.46 a
Sprinkler	2.34 ± 1.77 b	3.34 ± 2.61 a	100.50 ± 38.43 b	66.97 ± 27.15 b	25.12 ± 8.13 b	8.72 ± 6.35 b	187.94 ± 56.21 c	237.90 ± 168.90 b	66.51 ± 14.78 b	115.31 ± 56.30 b	23.65 ± 6.65 b	23.53 ± 12.67 c
Surface	1.30 ± 1.35 c	2.64 ± 2.27 b	114.94 ± 57.90 ab	88.98 ± 27.24 a	29.36 ± 12.59 b	18.10 ± 15.38 a	184.75 ± 45.08 b	329.42 ± 237.11 a	75.51 ± 29.37 b	152.95 ± 97.74 a	26.85 ± 7.39 b	29.06 ± 20.08 b
<b>Crops</b>												
Parsley	1.32 ± 0.95 c	1.90 ± 1.16 c	145.95 ± 36.00 a	104.70 ± 41.88 a	31.14 ± 7.78 a	18.85 ± 11.51 a	275.36 ± 104.11 a	501.51 ± 107.85 a	95.35 ± 30.16 b	217.78 ± 67.00 a	35.02 ± 8.76 a	47.95 ± 63.87 a
Lettuce	4.32 ± 0.83 a	3.77 ± 0.89 b	135.45 ± 74.45 b	85.58 ± 23.96 b	29.84 ± 7.02 b	14.98 ± 3.57 b	149.18 ± 25.74 c	472.00 ± 127.62 b	55.58 ± 17.55 c	92.42 ± 47.92 c	21.06 ± 7.02 c	23.53 ± 9.27 b
Onion	0.65 ± 0.36 d	0.63 ± 0.34 d	91.85 ± 32.18 c	69.16 ± 22.10 c	29.99 ± 7.24 b	13.42 ± 2.28 b	234.72 ± 85.83 b	233.08 ± 27.38 c	101.33 ± 21.71 a	136.18 ± 72.01 b	33.09 ± 8.27 b	23.96 ± 4.56 b
Radish	4.34 ± 0.38 b	6.60 ± 1.85 a	71.40 ± 32.02 d	48.21 ± 6.88 d	16.80 ± 4.20 c	10.33 ± 1.72 c	128.62 ± 40.42 d	180.93 ± 99.30 d	48.30 ± 12.60 d	47.64 ± 21.23 d	17.85 ± 5.77 d	14.59 ± 1.15 c
<b>Significance</b>												
Water Sources (W)	*	***	****	**	**	*	**	*	****	****	****	****
Irrigation Methods (I)	****	***	*	***	***	*	**	**	**	**	***	**
W × I	**	**	****	**	**	****	****	****	****	****	****	**
Crops (C)	****	****	****	****	****	****	****	****	****	****	****	****
C × W	****	****	****	****	****	****	****	****	****	****	****	****
C × I	****	****	****	****	****	****	****	****	****	****	****	****
C × W × I	****	****	****	****	****	****	****	****	****	****	****	****

Note: ns, \*, \*\*, \*\*\*, \*\*\*\* Non significant or significant at  $p \leq 0.05$ , 0.01, 0.001 and 0.0001. respectively. Means followed by a different letter in each column are significantly different according to the LSD test ( $p = 0.05$ ) FW: fresh weight basis.

Considering the water source as the source of variance, the crops irrigated with RW presented the highest mean yield ( $2.73 \pm 2.82 \text{ g} \cdot \text{m}^{-2}$  in 2019 and  $3.49 \pm 2.59 \text{ g} \cdot \text{m}^{-2}$  in 2020) and mineral content of phosphorus, potassium, calcium, and magnesium. There was no significant difference in terms of yield among the treatments irrigated with GW and TW in season 2019, while GW showed the lowest yield in 2020 ( $2.76 \pm 2.43 \text{ g} \cdot \text{m}^{-2}$ ). These results could be attributed to TW and RW being richer in phosphorus and potassium than GW. Overall, the study indicates that vegetables irrigated with TW have as good a mineral content as those irrigated from other water sources. Crops irrigated with GW showed the highest nitrogen content and the results were statistically different from the RW and TW treatments.

Considering the irrigation method as the source of variance, the results showed that the drip method gave the highest yields (Table 2) and differed significantly from sprinkler and surface irrigation treatments. The lowest yield was recorded for crops irrigated with the surface method. In both seasons, the mineral content of crops was the highest under drip and surface irrigation. K was the predominant macronutrient in all crops.

Our results on the mineral content of crops are in agreement with those reported by the USDA National Nutrient Database for Standard References, which found that parsley is the richest in nutrients [50,51]. However, some values differed from those reported by the USDA database and also varied from season to season. Such differences could be attributed to farming practices, environmental conditions and cultivars. Disparities in the mineral content of crops from one season to another may also be influenced by the light intensity during sample harvesting in each season [31].

### 3.3. Health Risk Assessment

#### 3.3.1. Pathogen Loading in Food Crops

Vegetables are thought to be the primary source of bacterial, protozoan, and helminth parasite infection in humans [52]. The average concentrations of the detected parasites and bacteria, as affected by the source of water and irrigation method, are presented in Figure 2 for the four crops grown in the two seasons.

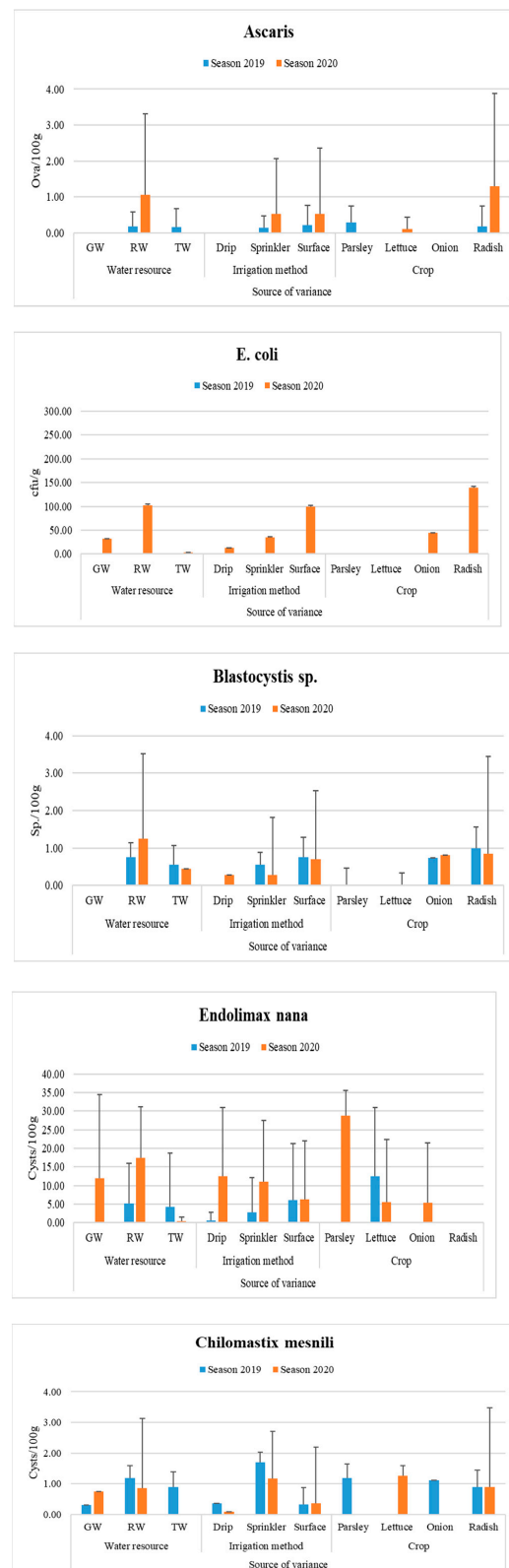
Concerning the parasitic contaminants, in 2019, *Chilomastix* was the most common parasitic contaminant in the study area (7.41% of total samples), followed by *Endolimax nana* (6.48%), *Ascaris* (3.70%), and *Blastocystis* spp. (1.85%). In the 2020 season, *Endolimax nana* was the most abundant parasitic contaminant (10.19% of total samples) followed by *Chilomastix* (3.70%), *Blastocystis* spp. (3.70%), and *Ascaris* (2.78%). *Chilomastix* and *Endolimax nana* are not pathogenic parasites but are considered commensal parasites and their presence is an indicator of fecal contamination and inadequate sanitation. However, there is ongoing debate about the commensal or pathogenic nature of *Blastocystis* spp. Recent studies have indicated that not all strains are pathogenic [53]. In this study, the occurrence of pathogenic parasites (*Ascaris* and *Blastocystis*) is considered low on irrigated crops.

All the examined vegetable samples irrigated with GW in both seasons were free from pathogenic parasites (*Ascaris* and *Blastocystis*). For TW-irrigated vegetables, 8.33% of the samples collected in season 2019 were contaminated, while 2.78% of total samples in season 2020 were infected only by *Blastocystis* sp. No *Ascaris* was found on crops. Vegetables irrigated with RW showed the highest percentage of contamination among the samples with 8.32% in 2019 and 16.67% in 2020.

Vegetables irrigated either with sprinkler or surface methods presented the highest percentage of contamination, with 8.33% of the total samples in each season. For vegetables grown under drip irrigation, there was no contamination with pathogenic parasites in season 2019, while only 2.78% of the vegetables grown in season 2020 were infected. The contaminated drip-irrigated crops were root crops (radish and onion).

The highest rate of parasitic contamination was detected in radishes (7.40% in 2019 and 11.10% in 2020). This was followed by parsley which showed 11.11% in 2019 and 2.70%

in 2020. Lettuce showed only 3.7% contamination in 2020. Onions presented percentages of contamination of 2.78% and 2.70% for seasons 2019 and 2020.



**Figure 2.** Mean concentrations of detected pathogens in relation to water source, irrigation method, and crop in seasons 2019 and 2020 (data elaboration based on 180 vegetables samples collected for each water source used for irrigation).

The mean concentrations of parasitic eggs in vegetables were (in descending order) *Endolimax nana* > *Chilomastix* > *Blastocystis* spp. > *Ascaris* (Figure 2), with values not exceeding 12.5, 1.7, 1, and 0.3 eggs/100 g, respectively. The obtained values were much lower than those reported in Morocco [22].

The results for bacterial contaminants showed that in season 2019, there was no bacterial loading on harvested crops in terms of *E. coli*. However, *salmonella* was detected on 3.70% of the collected vegetables, particularly on parsley with surface-irrigated river water. In season 2020, contamination of vegetables with *E. coli* was present. The highest and lowest rates of *E. coli* contamination were observed in RW (19.44% of total collected samples) and TW (5.56%), while 16.67% of the vegetables irrigated with GW were contaminated. The crops contaminated with *E. coli* in season 2020 were root crops (radish: 34.04% of total collected samples; onion: 18.52%). Contamination was detected on surface, sprinkler, and drip-irrigated crops. The variability in the occurrence of *E. coli* from season to season could be explained by the fact that in 2019, the harvest was in November, while in 2020, it was in October. It could be that the harvesting period affects pathogen loading on food crops since most pathogens are easily killed by heat, cold, sunlight, and lack of water. The vegetable samples were analyzed for the presence of *E. coli* 0157 and *Listeria monocytogenes*. The results showed that these bacteria were not present.

### 3.3.2. Metal Contamination in Food Crops

Heavy metal concentrations in the edible portions of food crops are shown in Table 3. The results show that the concentration values of most metals, except Cr, were higher in season 2019 than in 2020. This could be related to the three irrigation sources also showing a substantial improvement with respect to their content in metals during the second year of the trial.

Considering water as the source of variance, the highest concentrations of most metals in season 2019 were detected in TW followed by RW. The lowest concentrations were detected in GW, except for Zn and Cu. However, in season 2020, the crops irrigated with RW showed the highest content of metals, especially Cr and Cd, while those irrigated with GW continued to have the highest content of Zn.

Considering irrigation method as the source of variance, in season 2019, the highest concentrations of metals were reported under drip irrigation, while there was no significant difference among plots irrigated with surface or sprinkler methods for most tested metals. In season 2020, surface-irrigated crops were not significantly different from those that were drip-irrigated, particularly for Cr, Ni, and Cd.

Parsley showed the highest contamination for most metals, and radish the lowest levels for Cr, Zn, Cu, and Cd. This could be attributed to the shorter growing season for radishes than for other crops. In season 2020, only parsley contained Cd and Cu. Except for Zn and Cr, where some of the obtained values surpassed the limit values of 60 and 0.5 ppm, respectively, the findings from this study show that concentrations of most metals in vegetables are substantially lower than the safe limits for heavy metals given by FAO/WHO [54].

### 3.3.3. Bioaccumulation Factor

Table 4 shows the heavy metal transfer from soils to food crops through bioaccumulation (BAF).

Considering the water resource as the source of variance, in season 2019 there was a significant difference among the treatments with the highest BAF values obtained for TW, except for Zn and Cu that presented the highest BAF in GW. In season 2020, the highest BAF for Cr, Cu and Cd were detected in RW while BAF values for Ni was not significant among the three considered water resources. For irrigation methods, the highest BAF values were obtained under drip-irrigation and also surface-irrigated treatments, especially in season 2020.

**Table 3.** Effects of water source, irrigation method, and crop on metal contamination.

Treatment	Cr ppm		Ni ppm		Zn ppm		Cu ppm		Cd ppm	
	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
<b>Water Sources</b>										
Groundwater	1.131 ± 0.380 c	1.999 ± 1.430 b	4.032 ± 1.421 c	1.903 ± 4.654	85.561 ± 37.477 a	39.960 ± 21.919 a	1.408 ± 0.605 a	0.267 ± 0.695	0.033 ± 0.017 c	0.002 ± 0.006 c
River—Litani	1.392 ± 0.182 b	2.412 ± 1.169 a	9.520 ± 1.260 b	1.838 ± 4.206	47.150 ± 6.239 c	35.345 ± 17.456 b	0.980 ± 0.150 b	0.193 ± 0.438	0.084 ± 0.012 b	0.016 ± 0.037 a
Treated Water—Ablah	1.456 ± 0.213 a	1.516 ± 1.014 c	9.651 ± 2.176 a	1.481 ± 3.627	52.140 ± 7.539 b	35.311 ± 17.616 b	0.991 ± 0.111 b	0.180 ± 0.414	0.100 ± 0.024 a	0.005 ± 0.017 b
<b>Irrigation Methods</b>										
Drip	1.463 ± 0.296 a	1.939 ± 1.054 a	8.583 ± 2.533 a	3.780 ± 3.986 a	75.583 ± 42.818 a	35.275 ± 17.617 b	1.350 ± 0.634 a	0.110 ± 0.244 b	0.078 ± 0.023 a	0.010 ± 0.021 a
Sprinkler	1.236 ± 0.245 b	1.768 ± 1.034 b	7.379 ± 3.046 b	1.063 ± 2.844 b	52.987 ± 12.439 c	29.937 ± 15.201 b	0.983 ± 0.160 c	0.093 ± 0.313 b	0.068 ± 0.035 b	0.007 ± 0.023 b
Surface	1.281 ± 0.327 b	2.220 ± 1.602 a	7.241 ± 3.561 b	4.159 ± 5.909 a	56.281 ± 10.153 a	45.405 ± 21.018 a	1.046 ± 0.129 b	0.547 ± 0.760 a	0.066 ± 0.038 b	0.016 ± 0.034 a
<b>Crops</b>										
Parsley	1.246 ± 0.253 a	1.651 ± 0.714 b	7.291 ± 3.204 b	3.604 ± 4.784 a	66.466 ± 31.012 a	42.921 ± 18.167 b	1.186 ± 0.466 a	0.507 ± 0.580 a	0.094 ± 0.030 a	0.030 ± 0.042 a
Lettuce	1.304 ± 0.364 b	1.644 ± 0.509 b	8.019 ± 3.495 a	0.325 ± 1.042 b	56.554 ± 10.327 b	51.836 ± 16.495 a	1.078 ± 0.130 b	0 ± 0 b	0.072 ± 0.036 b	0 ± 0 b
Onion	1.414 ± 0.361 a	3.448 ± 1.541 a	8.067 ± 2.617 a	0.301 ± 0.789 b	66.402 ± 41.107 c	22.907 ± 9.524 d	1.187 ± 0.618 a	0 ± 0 b	0.075 ± 0.034 b	0 ± 0 b
Radish	1.341 ± 0.198 b	1.158 ± 0.472 c	7.560 ± 3.154 b	3.034 ± 6.023 a	57.046 ± 19.313 c	29.824 ± 16.643 c	1.054 ± 0.272 b	0 ± 0 b	0.072 ± 0.032 b	0 ± 0 b
<b>Significance</b>										
<b>Water Sources (W)</b>	****	***	****	ns	****	**	**	ns	****	****
<b>Irrigation Methods (I)</b>	**	ns	*	****	**	***	***	***	*	****
<b>W × I</b>	****	**	***	**	***	**	**	ns	***	****
<b>Crops (C)</b>	****	****	**	****	****	****	****	****	****	****
<b>C × W</b>	****	****	****	****	****	****	****	**	***	****
<b>C × I</b>	****	****	****	****	****	****	****	****	****	****
<b>C × W × I</b>	****	****	****	****	****	****	****	ns	****	****

Note: Ns, \*, \*\*, \*\*\*, \*\*\*\*. Non significant, significant at  $p \leq 0.05$ , 0.01, 0.001, and 0.0001, respectively. Means followed by a different letter in each column are significantly different according to the LSD test ( $p = 0.05$ ).

**Table 4.** Effects of water source, irrigation method, and crop on bioaccumulation factors.

Treatment	Cr ppm		Ni ppm		Zn ppm		Cu ppm		Cd ppm	
	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
<b>Water Sources</b>										
Groundwater	0.1070 ± 0.033 c	0.2133 ± 0.156 b	0.0783 ± 0.025 c	0.0716 ± 0.174	0.4699 ± 0.174 a	1.1835 ± 1.57 a	0.0807 ± 0.032 a	0.0246 ± 0.058 ab	0.2346 ± 0.121 c	0.0625 ± 0.225 b
River—Litani	0.1288 ± 0.020 b	0.2529 ± 0.13 a	0.1671 ± 0.024 b	0.0595 ± 0.136	0.2355 ± 0.036 c	0.5548 ± 0.404 c	0.05 ± 0.009 c	0.0351 ± 0.092 a	0.4496 ± 0.067 b	0.2099 ± 0.477 a
Treated Water—Ablah	0.1437 ± 0.025 a	0.1640 ± 0.114 c	0.1913 ± 0.050 a	0.0524 ± 0.127	0.2880 ± 0.061 b	0.6849 ± 0.398 b	0.0558 ± 0.010 b	0.0198 ± 0.046 b	0.4957 ± 0.144 a	0.541 ± 0.19 b
<b>Irrigation Methods</b>										
Drip	0.1340 ± 0.032 a	0.2203 ± 0.121 a	0.147 ± 0.044 a	0.0345 ± 0.088 b	0.3720 ± 0.217 a	0.8656 ± 1.492 b	0.0702 ± 0.037 a	0.0633 ± 0.090 a	0.431 ± 0.106 a	0.2344 ± 0.398 a
Sprinkler	0.1146 ± 0.018 b	0.1737 ± 0.105 b	0.139 ± 0.053 b	0.0360 ± 0.093 b	0.2960 ± 0.096 c	0.6502 ± 0.760 b	0.0551 ± 0.011 c	0.0155 ± 0.052 b	0.3713 ± 0.151 b	0.1097 ± 0.369 b
Surface	0.1309 ± 0.035 a	0.2362 ± 0.174 a	0.1501 ± 0.078 a	0.1475 ± 0.210 a	0.3253 ± 0.082 b	0.9072 ± 0.413 a	0.0613 ± 0.011 b	0.064 ± 0.095 a	0.3776 ± 0.208 b	0.2168 ± 0.412 a
<b>Crops</b>										
Parsley	0.1308 ± 0.020 a	0.3643 ± 0.175 a	0.1298 ± 0.043 c	0.1217 ± 0.158 a	0.3587 ± 0.164 a	1.5875 ± 1.696 a	0.0649 ± 0.029 a	0.0636 ± 0.075 a	0.3599 ± 0.142 b	0.4354 ± 0.545 a
Lettuce	0.1248 ± 0.035 bc	0.1772 ± 0.077 b	0.1593 ± 0.071 a	0.0128 ± 0.041 b	0.3098 ± 0.059 b	0.7455 ± 0.398 b	0.062 ± 0.009 ab	0 ± 0 b	0.4086 ± 0.184 a	0 ± 0 b
Onion	0.1299 ± 0.037 ab	0.1767 ± 0.061 b	0.1474 ± 0.049 b	0.1101 ± 0.220 a	0.3594 ± 0.215 a	0.5396 ± 0.294 c	0.0630 ± 0.033 ab	0 ± 0 b	0.4179 ± 0.139 a	0 ± 0 b
Radish	0.1206 ± 0.027 c	0.1221 ± 0.056 c	0.1458 ± 0.070 b	0.0134 ± 0.050 b	0.2965 ± 0.097 b	0.3582 ± 0.128 d	0.0587 ± 0.016 b	0 ± 0 b	0.3867 ± 0.179 b	0 ± 0 b



Table 4. Cont.

Treatment	Cr ppm		Ni ppm		Zn ppm		Cu ppm		Cd ppm	
	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
Significance										
Water Sources (W)	****	***	****	ns	****	***	***	*	***	**
Irrigation Methods (I)	***	**	*	****	***	**	***	**	**	**
W × I	****	**	****	**	****	***	****	***	**	****
Crops (C)	**	****	****	****	****	****	*	****	***	****
C × W	****	****	****	****	****	****	****	***	****	****
C × I	****	****	****	****	***	****	****	****	***	****
C × W × I	****	****	****	****	****	****	****	****	****	****

Note: Ns, \*, \*\*, \*\*\*, \*\*\*\*: Non significant, significant at  $p \leq 0.05$ , 0.01, 0.001, and 0.0001, respectively. Means followed by a different letter in each column are significantly different according to the LSD test ( $p = 0.05$ ). Means followed by a different letter in each column are significantly different according to the LSD test ( $p = 0.05$ ).

The trends in the BAF for heavy metals in different vegetables were in the increasing order of  $\text{Cu} < \text{Cd} < \text{Ni} < \text{Cr} < \text{Zn}$ . Similar trends were found for okra irrigated with sewage water in Saudi Arabia. Parsley showed the highest BAF for most metals while radish presented the lowest values [55]. The variation in BAF levels across vegetables may be explained by how plant type and related physiological features affect heavy metal uptake [39].

#### 3.3.4. Estimated Dietary Intake (EDI) of Metals, Hazard Risk Index (HRI), and Target Hazard Quotient (THQ)

The estimated EDIs of heavy metals for adults and children through the consumption of irrigated vegetables are presented in Tables 5 and 6. The trends in EDIs for both adults and children were in the order of  $\text{Zn} > \text{Cr} > \text{Ni} > \text{Cu} > \text{Cd}$ , with intakes from onion and parsley taking the lead. Table 7 shows that children are at higher risk than adults. In season 2019, the highest EDIs of most metals were found in TW treatments. In season 2020, vegetables irrigated with RW showed the highest EDIs, except for Zn and Cu. The risk of metal intake by both adults and children was higher under drip and surface irrigation.

The overall health risk associated with eating vegetables cultivated with water of various qualities was also evaluated using THQ and HRI. Both measures have a health protection level of 1.0 for lifetime hazards [56]. The THQ values for most metals were much lower than 1.0 (Tables S4 and S5), indicating that local residents who consume these vegetables are not yet exposed to a potential health risk. However, if pollution levels in the Litani Basin continue to increase unabated, the risk exposure in relation to metal uptake through vegetable consumption can be expected to increase as well.

#### 3.3.5. The Impact of Water Resource and Irrigation Method on Soil Properties

The soil parameters under GW, TW, and RW are given in Table 7. At the end of the two seasons of trials, in soil irrigated with RW, the pH, EC, phosphorus, and potassium levels were significantly higher than in soil under TW and GW. Drip-irrigated soil showed the highest physicochemical values. This result has been discussed by other researchers who reported that applying wastewater via drip irrigation caused an increase in the EC, organic matter, and mineral content of the soil [55,57,58]. Higher electrical conductivity was found at the end of the season, especially for soil irrigated with TW and RW, but the values were still within crop tolerance ranges. For nitrogen, there is no high accumulation that seems to be established between the beginning and end of the season. Plant uptake of nitrogen from the soil may be the reason for no significant accumulation [59]. For potassium, soil analysis showed that element concentration increased at the end of the season with the highest values in RW treatments. These results agree with other findings showing that wastewater irrigation significantly affects soil chemical properties and increases soil salinity, organic matter, exchangeable minerals, plant available P, and microelements [55].

At the beginning of season 2019, all analyzed metals were present in the soil and showed concentrations within the permissible levels, as set by the World Health Organization [60]. There were no significant differences among the metal levels according to the different water sources and irrigation methods. After two years of trials, all tested metals in the soil were below permissible levels. There was, however, a significant difference among the three water sources, with the highest levels of most metals in soil irrigated with RW (except for Zn). Drip irrigation resulted in the highest levels of Cr, Zn, and Cd. The differences in the levels of heavy metals in the soil profile before and after trials varied, which suggests that the risk of soil pollution depends not only on the quality of irrigation water but also on other factors such as rain, river sedimentation, air pollution, and the geology of the area.

**Table 5.** Effects of water source, irrigation method, and crop on estimated daily intake of heavy metals for adults.

Treatment	Cr ppm		Ni ppm		Zn ppm		Cu ppm		Cd ppm	
	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
<b>Water Sources</b>										
Groundwater	0.0066 ± 0.004	0.0089 ± 0.005 b	0.0020 ± 0.001 c	0.0007 ± 0.001 b	0.0407 ± 0.029 a	0.0237 ± 0.012 a	0.0006 ± 0.00047 a	0.0001 ± 0.0003	0.00001 ± 0.00001 c	0.000001 ± 0.000 b
River—Litani	0.0067 ± 0.002	0.0134 ± 0.007 a	0.0041 ± 0.001 b	0.0011 ± 0.002 a	0.0234 ± 0.006 b	0.0178 ± 0.009 c	0.0005 ± 0.0001 ab	0.0001 ± 0.0003	0.000038 ± 0.00002 b	0.00001 ± 0.000 a
Treated Water—Ablah	0.0068 ± 0.002	0.0082 ± 0.004 c	0.0045 ± 0.001 a	0.0007 ± 0.001 b	0.0250 ± 0.009 b	0.02 ± 0.009 b	0.0004 ± 0.0001 b	0.00009 10 <sup>-4</sup> ± 0.0002	0.000047 ± 0.00002 a	0.000002 ± 0.000 b
<b>Irrigation Methods</b>										
Drip	0.0072 ± 0.004 a	0.0103 ± 0.007 a	0.0036 ± 0.001 a	0.0017 ± 0.003 a	0.0357 ± 0.003 a	0.0155 ± 0.007 c	0.0006 ± 0.0004 a	0.0002 ± 0.0004 a	0.000035 ± 0.00002 a	0.00009 10 <sup>-4</sup> ± 0.000 a
Sprinkler	0.0059 ± 0.002 b	0.0084 ± 0.003 b	0.0033 ± 0.001 b	0.0006 ± 0.001 b	0.0247 ± 0.008 b	0.0213 ± 0.012 b	0.0004 ± 0.0001 b	0.00007 ± 0.0002 b	0.00003 ± 0.00002 b	0.000004 ± 0.000 b
Surface	0.007 ± 0.002 ab	0.0118 ± 0.006 a	0.0037 ± 0.002 a	0.0019 ± 0.002 a	0.0288 ± 0.011 a	0.0246 ± 0.010 a	0.0005 ± 0.0002 a	0.0002 ± 0.0003 a	0.00003 ± 0.00003 a	0.000001 ± 0.000 a
<b>Crops</b>										
Parsley	0.0077 ± 0.002 b	0.0121 ± 0.006 a	0.004 ± 0.001 b	0.0022 ± 0.003 a	0.0395 ± 0.022 a	0.0275 ± 0.011 a	0.0007 ± 0.0003 a	0.0003 ± 0.0004 a	0.00003 ± 0.000002 b	0.00002 ± 0.000 a
Lettuce	0.0050 ± 0.001 c	0.0115 ± 0.008 b	0.0031 ± 0.001 c	0.0001 ± 0.0003 c	0.0215 ± 0.004 b	0.0155 ± 0.008 c	0.0004 ± 0.0001 b	0 ± 0 b	0.00002 ± 0.000001 c	0 ± 0 b
Onion	0.0098 ± 0.003 a	0.0117 ± 0.003 b	0.0052 ± 0.002 a	0.0011 ± 0.002 b	0.0408 ± 0.024 a	0.0163 ± 0.006 c	0.0007 ± 0.0003 a	0 ± 0 b	0.00005 ± 0.000002 a	0 ± 0 b
Radish	0.0043 ± 0.001 d	0.0054 ± 0.002 c	0.0019 ± 0.0009 d	0.00009 ± 0.0004 c	0.0172 ± 0.005 c	0.0226 ± 0.011 b	0.0003 ± 0.00009 c	0 ± 0 b	0.00001 ± 0.000001 d	0 ± 0 b
<b>Significance</b>										
Water Sources (W)	****	****	****	**	***	**	*	ns	****	***
Irrigation Methods (I)	**	**	*	**	*	**	*	*	*	*
W × I	***	***	****	ns	****	**	****	*	****	ns
Crops (C)	****	****	****	****	****	****	****	****	****	****
C × W	****	****	****	****	****	****	****	****	****	****
C × I	****	****	****	****	****	****	****	****	****	****
C × W × I	****	****	****	****	****	****	****	****	****	****

Note: \* Significant at  $p \leq 0.05$ ; \*\* significant at 0.01; \*\*\* significant at 0.001; \*\*\*\* significant at 0.0001. Means followed by a different letter in each column are significantly different according to the LSD test ( $p = 0.05$ ).

**Table 6.** Effects of water sources, irrigation methods, and crop on estimated daily intake of heavy metals for children.

Treatment	Cr ppm		Ni ppm		Zn ppm		Cu ppm		Cd ppm	
	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
<b>Water sources</b>										
Groundwater	0.006 ± 0.004 b	0.009 ± 0.005 c	0.0023 ± 0.001 c	0.0008 ± 0.002 b	0.0468 ± 0.033 a	0.0275 ± 0.014 a	0.0007 ± 0.0005 a	0.0001 ± 0.0003	0.00001 ± 0.0001 c	0.000003 ± 0 b
River—Litani	0.007 ± 0.002 a	0.0106 ± 0.006 b	0.0047 ± 0.002 b	0.0013 ± 0.003 a	0.0269 ± 0.007 b	0.0203 ± 0.01 b	0.0005 ± 0.0002 ab	0.0001 ± 0.0003	0.00004 ± 0.0002 b	0.00001 ± 0 a
Treated water—Ablah	0.007 ± 0.003 a	0.0145 ± 0.008 a	0.0052 ± 0.002 a	0.0008 ± 0.002 b	0.0288 ± 0.011 b	0.0223 ± 0.012 b	0.0005 ± 0.0002 b	0.0001 ± 0.0003	0.00005 ± 0.0003 a	0.000004 ± 0 b
<b>Irrigation methods</b>										
Drip	0.0081 ± 0.004 a	0.0118 ± 0.008 b	0.0042 ± 0.002 a	0.0020 ± 0.004 a	0.0410 ± 0.034 a	0.0276 ± 0.008 a	0.0007 ± 0.00054 a	0.0002 ± 0.0007 a	0.0000410 10 <sup>-4</sup> ± 0.0002 a	0.00001 ± 0.00002 a
Sprinkler	0.0064 ± 0.002 b	0.01 ± 0.004 c	0.0038 ± 0.002 b	0.0007 ± 0.002 b	0.0284 ± 0.010 b	0.0250 ± 0.014 b	0.0005 ± 0.00017 b	0.00008 ± 0.0003 b	0.00003 ± 0.00003 b	0.000005 ± 0.000002 b
Surface	0.0077 ± 0.003 a	0.0127 ± 0.007 a	0.0043 ± 0.002 a	0.0022 ± 0.003 a	0.0331 ± 0.013 a	0.0275 ± 0.013 a	0.0006 ± 0.00026 a	0.0003 ± 0.0004 a	0.00004 ± 0.00003 b	0.00001 ± 0.00002 a
<b>Crops</b>										
Parsley	0.0088 ± 0.002 b	0.0136 ± 0.009 a	0.0046 ± 0.001 b	0.0025 ± 0.003 a	0.0454 ± 0.025 a	0.0317 ± 0.013 a	0.0008 ± 0.00039 a	0.0003 ± 0.0004 a	0.00004 ± 0.00002 b	0.00002 ± 0 a
Lettuce	0.0057 ± 0.002 c	0.006 ± 0.002 c	0.0035 ± 0.001 c	0.0001 ± 0.0004 c	0.0247 ± 0.005 b	0.0194 ± 0.008 c	0.0004 ± 0.00013 b	0 ± 0 b	0.00003 ± 0.00002 c	0 ± 0 b
Onion	0.0108 ± 0.004 a	0.0135 ± 0.004 a	0.006 ± 0.002 a	0.0023 ± 0.002 a	0.0469 ± 0.027 a	0.0266 ± 0.014 b	0.0008 ± 0.00042 a	0 ± 0 b	0.00005 ± 0.00003 a	0 ± 0 b
Radish	0.0043 ± 0.001 d	0.0129 ± 0.007 b	0.0022 ± 0.001 d	0.0012 ± 0.002 b	0.0197 ± 0.006 c	0.0158 ± 0.008 d	0.0003 ± 0.00010 c	0 ± 0 b	0.00002 ± 0.00001 d	0 ± 0 b

Table 6. Cont.

Treatment	Cr ppm		Ni ppm		Zn ppm		Cu ppm		Cd ppm	
	2019	2020	2019	2020	2019	2020	2019	2020	2019	2020
Significance										
Water sources (W)	**	***	****	**	***	**	*	ns	****	***
Irrigation methods (I)	**	**	*	**	*	***	*	*	**	*
W × I	***	***	****	ns	***	**	****	*	****	ns
Crops (C)	****	****	****	****	****	****	****	****	****	****
C × W	****	****	****	****	****	****	****	****	****	****
C × I	****	****	****	****	****	****	****	****	****	****
C × W × I	****	****	****	****	****	****	****	***	****	***

Note: ns, \*, \*\*, \*\*\*, \*\*\*\*: Non significant, significant at  $p \leq 0.05$ , 0.01, 0.001, and 0.0001, respectively. Means followed by a different letter in each column are significantly different according to the LSD test ( $p = 0.05$ ). Means followed by a different letter in each column are significantly different according to the LSD test ( $p = 0.05$ ).

Table 7. Effects of water source and irrigation method on soil properties at the beginning and end of seasons.

Treatment	pH	ECe (dS/m)	N%	P2O5 ppm	K2O ppm	Cr ppm	Ni ppm	Zn ppm	Cu ppm	Cd ppm
At the Beginning of the Trials										
Water Sources										
Groundwater	7.25 ± 0.16	0.21 ± 0.03	0.13 ± 0.03	35.14 ± 4.84	204.03 ± 18.15	76.76 ± 14.26	40.77 ± 8.27	215.72 ± 51.54	12.26 ± 2.49	0.2 ± 0.04
River—Litani	7.31 ± 0.19	0.22 ± 0.02	0.11 ± 0.03	34.40 ± 5.94	219.5 ± 35.51	81.74 ± 17.7	44.23 ± 5.77	241.61 ± 31.5	13.3 ± 1.73	0.25 ± 0.04
Treated Water—Ablah	7.38 ± 0.35	0.23 ± 0.03	0.12 ± 0.04	32.28 ± 6.58	218.89 ± 15.32	77.67 ± 8.99	41.33 ± 5.77	225.74 ± 31.49	12.43 ± 1.73	0.23 ± 0.05
Irrigation Methods										
Drip	7.31 ± 0.3	0.24 ± 0.03	0.14 ± 0.04	33.65 ± 6.38	220.34 ± 17.79	84.42 ± 8.51	42.44 ± 3.94	246.68 ± 39.59	12.97 ± 1.18	0.23 ± 0.03
Sprinkler	7.31 ± 0.29	0.24 ± 0.02	0.12 ± 0.03	34.5 ± 6.23	219.5 ± 21.93	79.73 ± 15.07	41.28 ± 8.52	225.48 ± 46.55	12.41 ± 2.56	0.23 ± 0.07
Surface	7.15 ± 0.14	0.23 ± 0.04	0.10 ± 0.02	34.66 ± 5.71	218.58 ± 31.22	72.02 ± 15.02	38.61 ± 4.34	210.9 ± 23.72	11.61 ± 1.31	0.21 ± 0.02
Significance										
Water Sources (W)	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns
Irrigation Methods (I)	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns
W × I	ns	ns	ns	ns	ns	ns	ns	ns	ns	ns
At the End of the Trials										
Water Sources										
Groundwater	7.04 ± 0.06 b	0.32 ± 0.12 b	0.157 ± 0.02 a	30.97 ± 7.42 c	232.95 ± 11.31 b	92.071 ± 1.86 c	27.730 ± 1.29 b	73.237 ± 18.10 a	8.063 ± 1.95 a	0.041 ± 0.02 c
River—Litani	7.13 ± 0.11 a	0.43 ± 0.11 a	0.135 ± 0.02 c	36.85 ± 8.76 a	248.63 ± 11.01 a	98.581 ± 5.86 a	31.358 ± 1.78 a	53.887 ± 2.75 c	8.354 ± 1.15 a	0.101 ± 0.03 a
Treated Water—Ablah	7.07 ± 0.07 b	0.33 ± 0.05 b	0.144 ± 0.03 b	31.26 ± 9.22 b	244.92 ± 13.48 a	93.520 ± 7.74 b	21.818 ± 1.60 c	59.309 ± 6.11 b	7.596 ± 2.26 b	0.081 ± 0.02 b
Irrigation Methods										
Drip	7.11 ± 0.10 a	0.35 ± 0.08 ab	0.154 ± 0.01 a	43.29 ± 4.82 a	246.30 ± 16.29 a	97.690 ± 4.04 a	29.152 ± 1.07 b	65.300 ± 8.33 a	8.030 ± 2.05 b	0.078 ± 0.05 a
Sprinkler	7.09 ± 0.09 a	0.41 ± 0.12 a	0.108 ± 0.02 b	24.55 ± 2.93 b	241.59 ± 7.88 b	87.870 ± 3.15 c	30.269 ± 2.32 a	69.712 ± 17.10 a	6.763 ± 0.96 c	0.073 ± 0.03 b
Surface	7.04 ± 0.06 b	0.32 ± 0.10 b	0.105 ± 0.01 b	27.24 ± 4.95 b	228.60 ± 2.05 c	94.614 ± 4.79 b	28.485 ± 1.78 c	51.420 ± 5.33 b	9.219 ± 1.52 a	0.072 ± 0.03 b
Significance										
Water Sources (W)	*	***	**	****	****	**	**	**	*	****
Irrigation Methods (I)	*	*	****	****	****	****	**	****	**	***
W × I	**	****	***	****	****	****	**	**	****	****

Note: ns, \*, \*\*, \*\*\*, \*\*\*\*: Non significant, significant at  $p \leq 0.05$ , 0.01, 0.001, and 0.0001, respectively. Means followed by a different letter in each column are significantly different according to the LSD test ( $p = 0.05$ ). Means followed by a different letter in each column are significantly different according to the LSD test ( $p = 0.05$ ).

#### 4. Discussion

The results of this study show that the quality of irrigation water, crop selection, choice of irrigation method, and local environmental conditions affect produce safety. After two years of monitoring the water quality of three irrigation water resources in the Litani Basin, the quality of secondary-level treated municipal water from Ablah municipality (TW) improved substantially, while groundwater extracted from an existing well at the LARI station (GW) became increasingly contaminated with pathogens. The concentration of *E. coli* in TW decreased from over 3 to less than 1 log CFU/100 mL in season 2020, while the concentration in GW fluctuated from less than 1 to over 2 log CFU/100 mL. In surface water from the Litani River (RW), it remained mostly above 5 log CFU/100 mL. Parasitic eggs appeared in all three water sources at a concentration exceeding the limit of 1 egg/L proposed by WHO and adopted in the draft Lebanese standards proposed by FAO in 2011 [48,61] but only when the fecal load in any water resource was above 2 log *E. coli* CFU/100 mL. Metals were found in all three water sources although their concentrations were mostly within FAO safety limits [48]. The water quality results indicate increasing pollution in the Litani Basin, and this needs to be urgently addressed through an integrated water management approach [14]. The draft standards on water reuse for irrigation proposed by FAO for Lebanon in 2011 prohibit the use of treated water for the production of vegetables intended to be eaten raw. The standards do allow irrigating vegetables to be consumed after cooking if the fecal load in the water is less than 2 log CFU/100 mL [48]. Guidelines and standards adopted in other countries (Jordan, Iran, Morocco, Oman) or proposed by international organizations such as WHO [61] are more flexible [62]. For example, the WHO guidelines allow the irrigation of vegetables intended to be eaten raw with water having fecal loads of less than or equal to 3 log CFU/100 mL, while setting some barriers regarding the selection of crops and irrigation methods [61]. Our study shows that for water with less than 2 log *E. coli* CFU/100 mL, no pathogens (*E. coli*, *salmonella*, parasite eggs) were detected in irrigated vegetables, irrespective of the irrigation method. With water between 2 to 5 log *E. coli* CFU/100 mL and containing helminth eggs at a concentration of 1 to 2.67 eggs/L, 8.33% of the sprinkler- and surface-irrigated vegetables, in addition to 2.78% of the drip-watered root crops (radish and onion), showed some degree of contamination, but only with helminth eggs. With irrigation water having over 3 log CFU/100 mL, *E. coli* appeared, but only on root crops (radish, 34.04% of total collected samples, and onion, 18.52%). The results agree with similar studies that reviewed the risks associated with pathogens in water used to irrigate crops. For example, [63–67] found that water with less than or equal to 2 log *E. coli* CFU/100 mL was safe for vegetables irrespective of the irrigation method. However, other studies have highlighted the importance of crop selection and irrigation method for food safety when water has over 2 log *E. coli* CFU/100 mL. For example, if drip irrigation was used with water containing more than 4 log *E. coli* CFU/100 mL, *E. coli* was not detected on artichokes [63], broccoli [64], cucumbers [65], eggplants [16,64], fennel [67], grapes [14,68], lettuce [69], or tomatoes [64]. The literature and our results underline the importance of the irrigation method as a health risk reduction practice and have important implications for local guidelines.

Our study showed that the highest concentration of pathogens on contaminated crops was found on those grown with sprinkler and surface irrigation. These results are similar to other studies [70,71] that found *E. coli* on sprinkler-irrigated lettuce, and [72,73] reported detecting *E. coli* on furrow-irrigated onions with water having more than 4 log *E. coli* CFU/100 mL. Drip was not a good barrier for root crops irrigated with water containing more than 2 log *E. coli* CFU/100 mL. The results are also in line with findings that reported *E. coli* on drip-irrigated root crops and above-ground crops which are in contact with soil and water irrigated with effluent containing 2 log *E. coli* CFU/100 mL (for example, lettuce [67], radishes [39,55], and tomatoes [66,67]).



The results also showed the significance of local environmental conditions by indicating that a decrease in air temperature in the fall season or a delay in the harvest time would likely kill *E. coli* on harvested crops, especially those growing above the ground. The prevailing climatic conditions before harvest influence pathogens even if they appear on the leaves from soil or contaminated water [74–76].

The results also showed that parsley and onions make the biggest contribution in terms of bioaccumulation factors, estimated daily intakes, hazard risk index, and target hazard quotient. By choosing suitable crops, the danger of human exposure to metal pollution can be considerably decreased. Climate, air depositions, heavy metal concentrations in soil, and the soil type on which the vegetables are produced, plus the maturity of the plants at the time of harvest, all impact heavy metal bioaccumulation in vegetables [77–80]. Heavy metals are generally more mobile in soils with a pH less than 7 and can be easily accumulated by crops [55]. Luckily, the pH of the soils in the Litani Basin site are above 7 and consequently are not hazardous for agricultural purposes.

These results may inform the revision process of the Lebanese guidelines currently being undertaken by the Lebanese Institute for Standards. The results suggest that the use of drip irrigation with water concentrating less than 3 log CFU/100 mL on vegetables grown above ground and consumed raw would be safe under Lebanese conditions. Our results do support the prohibition on irrigating root crops with water having more than 2 log *E. coli* CFU/100 mL. The limit of 1 egg/L for parasitic eggs, as proposed by WHO, should remain in the Lebanese guidelines for all quality classes. If these findings are given consideration, the updated draft standards on water reuse for irrigation proposed for Lebanon would be less stringent and more realistic, especially for farmers, investors, and the whole water sector.

Finally, treated water should be seen as a source of irrigation water and would produce vegetables of similar quality to those grown using other water sources. The possibility of safely using reclaimed water for irrigating vegetable crops would increase the potential for conserving freshwater in areas where its availability for irrigation is limited [81].

## 5. Conclusions

In Lebanon, the food chain remains exposed to contaminants due to widespread pollution of soils and water sources. Irrigation with polluted water is a common practice that remains uncontrolled and increases risks to human health. The development of a long-term policy for monitoring and managing water quality, leading to its conservation and sustainable use, is critical. The creation of irrigation water quality standards, or, more broadly, water reuse criteria, that enhance health protection and food safety at reasonable prices is important.

Our research has shown that the adoption of drip irrigation with water of less than 3 log *E. coli* CFU/100 mL is safe, even for vegetables consumed raw, except for root crops such as onions or radishes that should not be irrigated with water having more than 2 log *E. coli* CFU/100 mL. Treated water does not harm vegetable quality compared to vegetables irrigated with other water sources. These results deserve to be considered in the formulation of the first Lebanese standards for water reuse aiming to protect environmental health while making more productive and efficient use of water sources.

**Supplementary Materials:** The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/w14091437/s1>, Figure S1. Rainfall (mm), maximum and minimum temperatures (oC) and reference evapotranspiration (mm) for the growing seasons 2019 and 2020, Figure S2. The experimental layout, Table S1. Heavy metal maximum permissible limits in vegetable crops, Table S2. Parameters used in the calculation of EDI, HRI, and THQ, Table S3. Treated water categories as proposed by the Lebanese guidelines, Table S4. Effects of water sources, irrigation methods, and crops on health risk index, Table S5. Effects of water sources, irrigation methods, and crops on target health quotient. References [39,46,54,82–87] are cited in the supplementary materials.

**Author Contributions:** Conceptualization, M.T.A.S. and J.M.-S.; methodology, M.T.A.S. and J.M.-S.; software, M.H.S. and R.S. (Rhend Sleiman); field work, S.F., S.S., R.M., R.S. (Rita Sfeir) and I.J.; laboratory analysis, R.E.H., Z.R., D.R., Y.K., V.A., R.A.S., C.H. and R.A.; writing: original draft preparation, M.T.A.S.; writing: review and editing, M.T.A.S. and J.M.-S.; supervision, M.H.N. and J.M.-S. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research received no external funding.

**Institutional Review Board Statement:** Not applicable.

**Informed Consent Statement:** Not applicable.

**Data Availability Statement:** Upon reasonable request.

**Acknowledgments:** The study was conducted within the SIDA-funded ReWater-MENA project coordinated by the International Water Management Institute (IWMI) and implemented locally by the Lebanese Agricultural Research Institute (LARI). The authors are grateful to Nisreen Lahham and Amgad El Mahdi from IWMI. In addition, the authors would like to thank the Municipality of Ablah Village, particularly Engineer Mohamed Bou Daya for the technical support and facilities he provided to get us connected to the Ablah treatment plant.

**Conflicts of Interest:** The authors declare no conflict of interest.

## References

1. United Nations World Water Assessment Programme. *Wastewater: The Untapped Resource: The United Nations World Water Development Report 2017*; UNESCO: Paris, France, 2017; ISBN 9789231002014.
2. Mateo-Sagasta, J.; Zadeh, S.M.; Turrall, H. (Eds.) *More People, More Food, Worse Water? A Global Review of Water Pollution from Agriculture*; Food and Agriculture Organization of the United Nations: Rome, Italy; International Water Management Institute on Behalf of the Water Land and Ecosystems Research Program of the CGIAR: Colombo, Sri Lanka, 2018; ISBN 9789251307298.
3. Wael, M. Water Demand Management Is a Must in MENA Countries ... But Is It Enough? *J. Geol. Resour. Eng.* **2018**, *6*, 59–64. [\[CrossRef\]](#)
4. UNEP. A Snapshot of the World's Water Quality: Towards a Global Assessment. *United Nations Environment Programme*. 2016. Available online: [https://uneplive.unep.org/media/docs/assessments/unep\\_wwqa\\_report\\_web.pdf](https://uneplive.unep.org/media/docs/assessments/unep_wwqa_report_web.pdf) (accessed on 1 June 2021).
5. Haddaoui, I.; Mateo-Sagasta, J. A Review on Occurrence of Emerging Pollutants in Waters of the MENA Region. *Environ. Sci. Pollut. Res.* **2021**, *28*, 68090–68110. [\[CrossRef\]](#) [\[PubMed\]](#)
6. Shaban, A.; Hamzé, M. (Eds.) *The Litani River, Lebanon: An Assessment and Current Challenges*; Springer Nature: Cham, Switzerland, 2018; Volume 85.
7. Shaban, A. *Water Resources of Lebanon, (World Water Resources Volume 7)*, 1st ed.; Springer: Cham, Switzerland, 2020; ISBN-13: 978-3030487195, ISBN-10: 3030487199.
8. Abi Saab, M.T.; Jammoul, D.; Makhoul, H.; Fahed, S.; Lebbous, N.; Hajjar, C.; Abi Saad, R.; Younes, M.; Hajj, M.; Todorovic, M. Assessing the performance of constructed wetland for water quality management of a Southern Mediterranean river. *Water Environ. J.* **2018**, *32*, 508–518. [\[CrossRef\]](#)
9. Mcheik, M.; Toufaily, J.; Akel, M.; Hamieh, T.; Abi Saab, M.T.; Haj Hassan, B.; Roupheal, Y. Reuse of Treated Municipal Wastewater under Different Growing Seasons for the Spinach Production. *Am. J. Anal. Chem.* **2018**, *9*, 482–499. [\[CrossRef\]](#)
10. Nehme, N.; Haidar, C. The physical, and chemical and microbial characteristics of Litani River water. In *The Litani River, Lebanon: An Assessment and Current Challenges*; Springer: Cham, Switzerland, 2018; pp. 57–70, ISBN 9783319763002.
11. Fadel, A.; Faour, G.; Slim, K. Assessment of the trophic state and Chlorophyll-a concentrations using Landsat OLI in Karaoun reservoir. *Lebanon. Leban. Sci.* **2016**, *17*, 130–145. [\[CrossRef\]](#)
12. Darwish, T.; Atallah, T.; Francis, R.; Saab, C.; Jomaa, I.; Shaban, A.; Sakka, H.; Zdruli, P. Observations on soil and groundwater contamination with nitrate: A case study from Lebanon-East Mediterranean. *Agric. Water Manag.* **2011**, *99*, 74–84. [\[CrossRef\]](#)
13. Darwish, T.; Shaban, A.; Masih, I.; Jaafar, H.; Jomaa, I.; Simaika, J.P. Sustaining the Ecological Functions of the Litani River Basin, Lebanon. *Int. J. River Basin Manag.* **2021**, 1–15. [\[CrossRef\]](#)
14. Abi Saab, M.T.; Makhoul, H.; Zaghrini, J.; Fahed, S.; Romanos, D.; Khairallah, Y.; Hajjar, C.; Abi Saad, R.; Sellami, M.H.; Todorovic, M. Table grapes irrigation with treated municipal wastewater in a Mediterranean environment. *Water Environ. J.* **2020**, *35*, 617–627. [\[CrossRef\]](#)
15. Mcheik, M.; Toufaily, J.; Hassan, B.H.; Hamieh, T.; Abi Saab, M.T.; Roupheal, Y.; Ferracin, E.; da Shio, B.; Bashabshah, I.; Al-Hadidi, L. Reuse of treated municipal wastewater in irrigation: A case study from Lebanon and Jordan. *Water Environ. J.* **2017**, *31*, 552–558. [\[CrossRef\]](#)
16. Abi Saab, M.T.; Bashour, I.; Daou, C.; Romanos, D.; Khairallah, Y.; Salaman, M.; Sellami, M.H.; Roukoz, S. Eggplant irrigation with treated municipal wastewater. *Aust. J. Crop Sci.* **2021**, *104*, 163–170. [\[CrossRef\]](#)

17. Darwich, T.; Shaban, A.; Hamzé, M. The national plan for Litani River Remediation. In *The Litani River; Lebanon: An Assessment and Current Challenges*; Shaban, A., Hamzé, M., Eds.; Springer: Cham, Switzerland, 2018; Volume 85. [\[CrossRef\]](#)
18. APHA. *Standard Methods for the Examination of Water and Wastewater*, 20th ed.; American Public Health Association: Washington, DC, USA, 1998.
19. APHA. *Standard Methods for the Examination of Water and Wastewater*, 21st ed.; American Public Health Association: Washington, DC, USA, 2005.
20. Rice, E.W.; Baird, R.B.; Eaton, A.D. *Standard Methods for the Examination of Water and Wastewater*, 23rd ed.; American Public Health Association, American Water Works Association, Water Environment Federation: Mumbai, India, 2017; ISBN 9780875532875.
21. Arther, R.G.; Fitzgerald, P.R.; Fox, G.C. Parasite ova in anaerobically digested sludge. *J. Water Pollut. Control Fed.* **1981**, *53*, 1333–1338.
22. Hajjami, K.; Ennaji, M.M.; Fouad, S.; Oubrim, N.; Cohen, N. Wastewater Reuse for Irrigation in Morocco: Helminth Eggs Contamination's Level of Irrigated Crops and Sanitary Risk (A Case Study of Settat and Soualem Regions). *J. Bacteriol. Parasitol.* **2013**, *4*, 163. [\[CrossRef\]](#)
23. ISO 10390; Soil Quality Determination of pH. ISO: Geneva, Switzerland, 2021. Available online: <https://www.iso.org/standard/40879.html> (accessed on 1 June 2021).
24. ISO 11265; Soil Quality. Chemical Methods. Determination of the Specific Electrical Conductivity. ISO: Geneva, Switzerland, 1994. Available online: <https://bsol.bsigroup.com/en/Bsol-Item-Detail-Page/?pid=000000000001120580> (accessed on 1 June 2021).
25. ISO 11261; Soil Quality. Chemical Methods. Determination of Total Nitrogen. Modified Kjeldahl Method. ISO: Geneva, Switzerland, 1995. Available online: <https://bsol.bsigroup.com/en/Bsol-Item-Detail-Page/?pid=000000000000577890> (accessed on 1 June 2021).
26. ISO 11263; Soil Quality. Determination of Phosphorus—Spectrometric Determination of Phosphorus Soluble in Sodium Hydrogen Carbonate Solution. ISO: Geneva, Switzerland, 1994. Available online: <https://www.iso.org/standard/19241.html?browse=tc> (accessed on 1 June 2021).
27. Walkley, A.; Black, I.A. An examination of Degtjareff method for determining soil organic matter and a proposed modification of the chromic acid titration method. *Soil Sci.* **1934**, *37*, 29–37. [\[CrossRef\]](#)
28. Lindsay, W.L.; Norvell, W.A. Development of a DTPA soil test for zinc, iron, manganese, and copper. *Soil Sci. Soc. Am. J.* **1978**, *42*, 421–428. [\[CrossRef\]](#)
29. ISO 14870; Soil Quality. Extraction of Trace Elements by Buffered DTPA Solution. ISO: Geneva, Switzerland, 2001. Available online: <https://www.iso.org/obp/ui/#iso:std:iso:14870:ed-1:v1:en> (accessed on 1 June 2021).
30. Levander, O.A. Fruit and vegetable contributions to dietary mineral intake in human health and disease. *HortScience* **1990**, *25*, 1486–1488. [\[CrossRef\]](#)
31. Colonna, E.; Roupael, Y.; Barbieri, G.; De Pascale, S. Nutritional quality of ten leafy vegetables harvested at two light intensities. *Food Chem.* **2016**, *199*, 702–710. [\[CrossRef\]](#)
32. Motsara, M.R.; Roy, R.N. *Guide to Laboratory Establishment for Plant Nutrient Analysis*. FAO Fertilizer and Plant Nutrition Bulletin 19; Food and Agricultural Organization: Rome, Italy, 2008.
33. ISO 16649-2; Food Microbiology. Microbiology of Food and Animal Feeding Stuffs. Horizontal Method for the Enumeration of Beta-Glucuronidase-Positive Escherichia Coli Part 2: Colony-Count Technique at 44 °C using 5-bromo-4-chloro-3-indolyl beta-D-glucuronide. ISO: Geneva, Switzerland, 2001. Available online: <https://www.iso.org/standard/29824.html> (accessed on 1 June 2021).
34. ISO 6579; Food Microbiology. Microbiology of the Food Chain. Horizontal Method for the Detection, Enumeration and Serotyping of Salmonella Part 1: Detection of Salmonella spp. ISO: Geneva, Switzerland, 2017. Available online: <https://www.iso.org/standard/56712.html> (accessed on 1 June 2021).
35. ISO 11290-1; Food Microbiology. Microbiology of the Food Chain. Horizontal Method for the Detection and Enumeration of Listeria Monocytogenes and of Listeria spp. Part 1: Detection Method. ISO: Geneva, Switzerland, 2017. Available online: <https://www.iso.org/standard/60313.html> (accessed on 1 June 2021).
36. ISO/TS 13136; Food Microbiology. Microbiology of Food and Animal Feed. Real-Time Polymerase Chain Reaction (PCR)-Based Method for the Detection of Food-Borne Pathogens. Horizontal Method for the Detection of Shiga Toxin-Producing Escherichia coli (STEC) and the Determination of O157, O111, O26, O103 and O145 Serogroups. ISO: Geneva, Switzerland, 2019. Available online: <https://www.iso.org/standard/53328.html> (accessed on 1 June 2021).
37. Estefan, G.; Sommer, R.; Ryan, J. *Methods of Soil, Plant, and Water Analysis: A Manual for the West Asia and North Africa Region: Third Edition*; International Center for Agricultural Research in the Dry Areas (ICARDA): Beirut, Lebanon, 2013.
38. Gomez, K.A.; Gomez, A.A. *Statistical Procedures for Agricultural Research*, 2nd ed.; John Wiley and Sons: New York, NY, USA, 1984; ISBN 9780471870920.
39. Qureshi, A.S.; Hussain, M.I.; Ismail, S.; Khan, Q.M. Evaluating heavy metal accumulation and potential health risks in vegetables irrigated with treated wastewater. *Chemosphere* **2016**, *163*, 54–61. [\[CrossRef\]](#)
40. Adesodun, J.K.; Atayese, M.O.; Agbaje, T.A.; Osadiaye, B.A.; Mafe, O.F.; Soretire, A.A. Phytoremediation potentials of sunflower (*Tithonia diversifolia* and *Helianthus annuus*) for metals in soils contaminated with zinc and lead nitrates. *Water Air Soil Pollut.* **2010**, *207*, 195–201. [\[CrossRef\]](#)

41. Santos, E.E.; Lauria, D.C.; Silveira, P.C.L. Assessment of daily intake of trace elements due to consumption of foodstuffs by adult inhabitants of Rio de Janeiro City. *Sci. Total Environ.* **2004**, *327*, 69–79. [CrossRef]
42. Arora, M.; Kiran, B.; Rani, S.; Rania, A.; Kaura, B.; Mittala, N. Heavy metal accumulation in vegetables irrigated with water from different sources. *Food Chem.* **2008**, *111*, 811–815. [CrossRef]
43. Pandey, R.; Shubhashish, K.; Pandey, J. Dietary intake of pollutant aerosols via vegetables influenced by atmospheric deposition and wastewater irrigation. *Ecotoxicol. Environ.* **2012**, *76*, 200–208. [CrossRef] [PubMed]
44. F.N.B. Dietary Reference Intakes (DRIs): Recommended Intakes for Individuals. Food and Nutrition Board, Institute of Medicine, National Academies. Available online: <http://www.iom.edu/Global/NewsAnnouncements/~{}media/Files/ActivityFiles/Nutrition/DRIs/DRISummaryListing2.ashx>. (accessed on 15 July 2011).
45. European Commission. Regulation Setting Maximum Levels for Certain Contaminants in Foodstuffs. Available online: [http://www.eurlex.europa.eu/LexUriServ/site/en/oj/2006/l\\_364/l\\_36420061220en00050024.pdf](http://www.eurlex.europa.eu/LexUriServ/site/en/oj/2006/l_364/l_36420061220en00050024.pdf) (accessed on 17 July 2011).
46. Yang, Q.W.; Xu, Y.; Liu, S.J.; He, J.F.; Long, F.Y. Concentration and potential health risk of heavy metals in market vegetables in Chongqing, China. *Ecotoxicol. Environ.* **2011**, *74*, 1664–1669. [CrossRef] [PubMed]
47. Ayers, R.S.; Westcot, D.W. *Water Quality for Agriculture*; FAO Irrigation and Drainage Paper 29 Rev. 1.; Food and Agriculture Organization of the United Nations: Rome, Italy, 1994; ISBN 9251022631.
48. FAO/UTF/LEB/019/LEB. *Proposed Guidelines for Wastewater Reuse in Agricultural Irrigation*; Food and Agricultural Organization of the United Nations: Beirut, Lebanon, 2011.
49. Iqbal, M.; Ahmed, S.; Rehman, W.; Menaa, F.; Ullah, M.A. Heavy Metal Levels in Vegetables Cultivated in Pakistan Soil Irrigated with Untreated Wastewater. *Preliminary Results. Sustainability* **2020**, *12*, 8891. [CrossRef]
50. Haytowitz, D.B.; Ahuja, J.K.C.; Wu, X.; Somanchi, M.; Nickle, M.; Nguyen, Q.A.; Roseland, J.M.; Williams, J.R.; Patterson, K.Y.; Li, Y.; et al. USDA National Nutrient Database for Standard Reference, Legacy Release. Nutrient Data Laboratory, Beltsville Human Nutrition Research Center, ARS, USDA. Available online: <https://data.nal.usda.gov/dataset/usda-national-nutrient-database-standard-reference-legacy-release> (accessed on 1 June 2021).
51. U.S. Department of Agriculture, Agricultural Research Service. USDA Food and Nutrient Database for Dietary Studies 2017–2018. Food Surveys Research Group Home Page. Available online: <http://www.ars.usda.gov/nea/bhnrc/fsrg> (accessed on 1 June 2021).
52. Zewdneh, T.; Dawit, K. Parasitological Contamination of Wastewater Irrigated and Raw Manure Fertilized Vegetables in Mekelle City and Its Suburb, Tigray, Ethiopia. *MEJS* **2012**, *4*, 77–89.
53. Zanetti, A.D.; Malheiros, A.F.; de Matos, T.A.; Longhi, F.G.; Moreira, L.M.; Silva, S.L.; Castrillon, S.K.I.; Ferreira, S.M.B.; Ignotti, E.; Espinosa, O.A.; et al. Prevalence of Blastocystis sp. infection in several hosts in Brazil: A systematic review and meta-analysis. *Parasites Vectors* **2020**, *13*, 1–15. [CrossRef] [PubMed]
54. FAO; WHO. *Food Standards Programme Codex Committee on Contaminants in Foods. Fifth Session*; WHO: Geneva, Switzerland, 2011; pp. 64–89.
55. Balkhair, K.S.; Asharaf, M.A. Field accumulation risks of heavy metals in soil and vegetable crop irrigated with sewage water in western region of Saudi Arabia. *Saudi J. Biol. Sci.* **2016**, *23*, S32–S44. [CrossRef]
56. USEPA. *Region III Risk-Based Concentration Table: Technical Background Information*; Unites States Environmental Protection Agency: Washington, DC, USA, 2006.
57. Najafi, P.; Nasr, S. Comparison effects of wastewater on soil chemical properties in three irrigation methods. *Res. Crops* **2009**, *10*, 277–280.
58. Mojiri, A.; Jalalian, A. Relationship between growth of Nitraria schoberi and some soil properties. *J. Anim. Plant Sci.* **2011**, *21*, 246–250.
59. Heidarpour, M.; Mostafazadeh-Fard, B.; Koupai, J.A.; Malekian, R. The effects of treated wastewater on soil chemical properties using subsurface and surface irrigation methods. *Agric. Water Manag.* **2007**, *90*, 87–94. [CrossRef]
60. WHO. *Standard Maxima for Metals in Agricultural Soils*; World Health Organization: Geneva, Switzerland, 1993.
61. WHO. *Guidelines for the Safe Use of Wastewater, Excreta and Grey Water*; World Health Organization: Geneva, Switzerland, 2006.
62. Shoushtarian, F.; Negahban-Azar, M. Worldwide Regulations and Guidelines for Agricultural Water Reuse: A Critical Review. *Water* **2020**, *12*, 971. [CrossRef]
63. Gatta, G.; Libutti, A.; Beneduce, L.; Gagliardi, A.; Disciglio, G.; Lonigro, A.; Tarantino, E. Reuse of treated municipal wastewater for globe artichoke irrigation: Assessment of effects on morpho-quantitative parameters and microbial safety of yield. *Sci. Hortic.* **2016**, *213*, 55–65. [CrossRef]
64. Libutti, A.; Gatta, G.; Gagliardi, A.; Vergine, P.; Pollice, A.; Beneduce, L.; Disciglio, G.; Tarantino, E. Agro-industrial wastewater reuse for irrigation of a vegetable crop succession under Mediterranean conditions. *Agric. Water Manag.* **2018**, *196*, 1–14. [CrossRef]
65. Obayomi, O.; Ghazaryan, L.; Ben-Hur, M.; Edelstein, M.; Vonshak, A.; Safi, J.; Berstein, N.; Gillor, O. The fate of pathogens in treated wastewater-soil-crops continuum and the effect of physical barriers. *Sci. Total Environ.* **2019**, *681*, 339–349. [CrossRef]
66. Cirelli, G.L.; Consoli, S.; Licciardello, F.; Aiello, R.; Giuffrida, F.; Leonardi, C. Treated municipal wastewater reuse in vegetable production. *Agric. Water Manag.* **2012**, *104*, 163–170. [CrossRef]
67. Lonigro, A.; Rubino, P.; Lacasella, V.; Montemurro, N. Faecal pollution on vegetables and soil drip irrigated with treated municipal wastewaters. *Agric. Water Manag.* **2016**, *174*, 66–73. [CrossRef]



68. Petousi, I.; Daskalakis, G.; Fountoulakis, M.S.; Lydakis, D.; Fletcher, L.; Stentiford, E.I.; Manios, T. Effects of treated wastewater irrigation on the establishment of young grapevines. *Sci. Total Environ.* **2019**, *658*, 485–492. [\[CrossRef\]](#)
69. Urbano, V.R.; Mendonca, T.G.; Bastos, R.G.; Souza, C.F. Effects of treated wastewater irrigation on soil properties and lettuce yield. *Agric. Water Manag.* **2017**, *181*, 108–115. [\[CrossRef\]](#)
70. Decol, L.T.; López-Gálvez, F.; Truchado, P.; Tondo, E.C.; Gil, M.I.; Allende, A. Suitability of chlorine dioxide as a tertiary treatment for municipal wastewater and use of reclaimed water for overhead irrigation of baby lettuce. *Food Control* **2019**, *96*, 186–193. [\[CrossRef\]](#)
71. Kaetzl, K.; Lübken, M.; Uzun, G.; Gehring, T.; Nettmann, E.; Stenchly, K.; Wichern, M. On-farm wastewater treatment using biochar from local agro residues reduces pathogens from irrigation water for safer food production in developing countries. *Sci. Total Environ.* **2019**, *682*, 601–610. [\[CrossRef\]](#)
72. Farhadkhani, M.; Nikaeen, M.; Yadegarfar, G.; Hatamzadeh, M.; Pourmohammadbagher, H.; Sahbaei, Z.; Rahmani, H.R. Effects of irrigation with secondary treated wastewater on physicochemical and microbial properties of soil and produce safety in a semi-arid area. *Water Res.* **2018**, *144*, 356–364. [\[CrossRef\]](#)
73. Araújo, S.; Silva, I.A.T.; Tacão, M.; Patinha, C.; Alves, A.; Henriques, I. Characterization of antibiotic resistant and pathogenic *Escherichia coli* in irrigation water and vegetables in household farms. *Int. J. Food Microbiol.* **2017**, *257*, 192–200. [\[CrossRef\]](#) [\[PubMed\]](#)
74. Keraita, B.; Konradsen, F.; Drechsel, P.; Abaidoo, R.C. Reducing microbial contamination on wastewater-irrigated lettuce by cessation of irrigation before harvesting. *Trop. Med. Int. Health* **2007**, *12*, 8–14. [\[CrossRef\]](#)
75. Sallach, J.B.; Zhang, Y.; Hodges, L.; Snow, D.; Li, X.; Bartelt-Hunt, S. Concomitant uptake of antimicrobials and *Salmonella* in soil and into lettuce following wastewater irrigation. *Environ. Pollut.* **2015**, *197*, 269–277. [\[CrossRef\]](#) [\[PubMed\]](#)
76. WHO; FAO. *Safety and Quality of Water Used in Food Production and Processing—Meeting Report. Microbiological Risk Assessment 2019 Series No. 33*; World Health Organization: Geneva, Switzerland; Food and Agriculture Organization of the United Nations: Rome, Italy, 2019; ISBN 9789241516402.
77. Ahmad, J.U.; Goni, M.A. Heavy metal contamination in water, soil, and vegetables of the industrial areas in Dhaka. *Bangladesh. Environ. Monit. Assess.* **2010**, *166*, 347–357. [\[CrossRef\]](#)
78. Bhargava, A.; Carmona, F.F.; Bhargava, M.; Srivastava, S. Approaches for enhanced phytoextraction of heavy metals. *J. Environ. Manag.* **2012**, *105*, 103–120. [\[CrossRef\]](#) [\[PubMed\]](#)
79. Ali, H.; Khan, E.; Sajad, M.A. Phytoremediation of heavy metals concepts and applications. *Chemosphere* **2013**, *91*, 869–881. [\[CrossRef\]](#) [\[PubMed\]](#)
80. Hu, W.; Chen, Y.; Huang, B.; Niedermann, S. Health risk assessment of heavy metals in soils and vegetables from a typical greenhouse vegetable production system in China. *Hum. Ecol. Risk Assess.* **2014**, *20*, 1264–1280. [\[CrossRef\]](#)
81. Psarras, G.; Chartzoulakis, K.; Kasapakis, I.; Kloppmann, W. Effect of different irrigation techniques and water qualities on yield, fruit quality and health risks of tomato plants. *Acta Hort.* **2014**, *1038*, 601–608. [\[CrossRef\]](#)
82. Abbasi, A.M.; Iqbal, J.; Khan, M.A.; Shah, M.H. Health risk assessment and multivariate apportionment of trace metals in wild leafy vegetables from Lesser Himalayas, Pakistan. *Ecotoxicol. Environ. Saf.* **2013**, *92*, 237–244. [\[CrossRef\]](#)
83. Amin, N.; Hussain, A.; Alamzeb, S.; Begum, S. Accumulation of heavy metals in edible parts of vegetables irrigated with waste water and their daily intake to adults and children, District Mardan, Pakistan. *Food Chem.* **2013**, *136*, 1515–1523. [\[CrossRef\]](#)
84. FAO/WHO. *Guidelines for the Safe Use of Wastewater and Food Stuff, Technical Report of the Joint FAO/WHO Volume 2 No. 1*; Food and Agriculture Organization (FAO) and Organization (WHO): Geneva, Switzerland, 2013.
85. Khan, S.; Cao, Q.; Zheng, Y.M.; Huang, Y.Z.; Zhu, Y.G. Health risks of heavy metals in contaminated soils and food crops irrigated with wastewater in Beijing, China. *Environ. Pollut.* **2008**, *152*, 686–692. [\[CrossRef\]](#) [\[PubMed\]](#)
86. Rehman, K.; Bukhari, S.M.; Andleeb, S.; Mahmood, A.; Erinle, K.O.; Naeem, M.M.; Imran, Q. Ecological risk assessment of heavy metals in vegetables irrigated with groundwater and wastewater: The particular case of Sahiwal district in Pakistan. *Agric. Water Manag.* **2019**, *226*. [\[CrossRef\]](#)
87. USEPA. *Reference Dose (RfD): Description and Use in Health Risk Assessments, Background Document 1A, Integrated Risk Information System (IRIS)*; United States Environmental Protection Agency: Washington, DC, USA, 2013.