

Article

Heavy Metal Concentrations in Maltese Potable Water

Roberta Bugeja * and Claire Shoemake

Department of Pharmacy, Faculty of Medicine and Surgery, University of Malta, Msida, MSD 2080, Malta; E-Mail: czer1@um.edu.mt

* Author to whom correspondence should be addressed; E-Mail: roberta.bugeja.01@um.edu.mt.

Academic Editor: Yu-Pin Lin

Received: 28 March 2015 / Accepted: 6 May 2015 / Published: 18 May 2015

Abstract: This study evaluates the levels of aluminum (Al), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), lead (Pb), nickel (Ni) and zinc (Zn) in tap water samples of forty localities from around the Maltese Islands together with their corresponding service supply reservoirs. The heavy metal concentrations obtained indicated that concentrations of the elements were generally below the maximum allowed concentration established by the Maltese legislation. In terms of the Maltese and EU water quality regulations, 17.5% of the localities sampled yielded water that failed the acceptance criteria for a single metal in drinking water. Higher concentrations of some metals were observed in samples obtained at the end of the distribution network, when compared to the concentrations at the source. The observed changes in metal concentrations between the localities' samples and the corresponding supply reservoirs were significant. The higher metal concentrations obtained in the samples from the localities can be attributed to leaching in the distribution network.

Keywords: heavy metals; potable water; Maltese Islands

1. Introduction

The pollution of heavy metals in water has become a question of considerable public and scientific concern in view of the facts proving their acute toxicity to human health and biological systems [1]. Heavy metal toxicity represents a rare, yet clinically-significant medical condition, which if overlooked or inadequately treated, results in significant morbidity and mortality [2]. Drinking water can be a possible source of human exposure to heavy metals. In general, heavy metals are systemic toxins with specific nephrotoxic, neurotoxic, teratogenic and fetotoxic effects [3]. Heavy metals can

directly influence behavior by influencing neurotransmitter production and utilization, impairing mental and neurological function, and altering numerous metabolic body processes [4].

The Maltese Islands are located in the central part of the Mediterranean Sea, and they represent one of the most densely-populated countries in the world. Natural water resources in the Maltese Islands are scarce, and this is mainly due to the low rainfall, high evapotranspiration rate, the long dry season accompanied by a relatively short period of rainfall and the Islands' small surface area [5,6]. The Food and Agriculture Organization of the United Nations (FAO) has classified Malta with the ten poorest countries in terms of water resources per inhabitant [7]. Historically, Malta has always had a lack of natural water resources in relation to its needs, since it does not have any significant natural surface water resources. The two main sources of urban water supply are groundwater and desalinated seawater [8].

The objective of this study is to provide an overview of heavy metal concentrations in potable water that is being consumed by the Maltese population who has been repeatedly shown to have higher blood lead levels than the reference values proposed by the EU [9,10]. Drinking water could be a possible source of exposure to lead, as demonstrated by Fertmann *et al.* [11] in an epidemiological study in Hamburg, where participants who consumed tap water with lead higher than the detection limit showed significantly higher blood lead levels. This study also attempts to determine variations in heavy metal concentrations between the service supply reservoirs and the consumers' taps by correlating these changes with the distribution network.

2. Methodology

2.1. Sampling Plan

The sampling program was designed in a way that the Maltese Islands would be well represented in this study (Figure 1). Heavy metal levels of 40 localities from around the Maltese Islands together with their respective service supply reservoirs were evaluated. The localities were defined according to the number of local councils established in the Maltese Islands. Samples were collected from each of the localities chosen in order to represent the whole population of each locality.

The reservoirs were then chosen according to the selection of localities. Table 1 shows that the number of reservoirs sampled also provides a clear representation of the whole population.

Table 1. Percentage of reservoirs sampled from the whole population.

	No. of Reservoirs Sampled	Total No. of Reservoirs	% of Reservoirs Sampled
Malta	15	19	78.9
Gozo	4	5	80.0
Total	19	24	79.2

The localities corresponding to the specific reservoir sources are represented in Table 2. A total of 59 samples were collected; 40 samples from the chosen localities and 19 samples from the reservoirs supplying them.

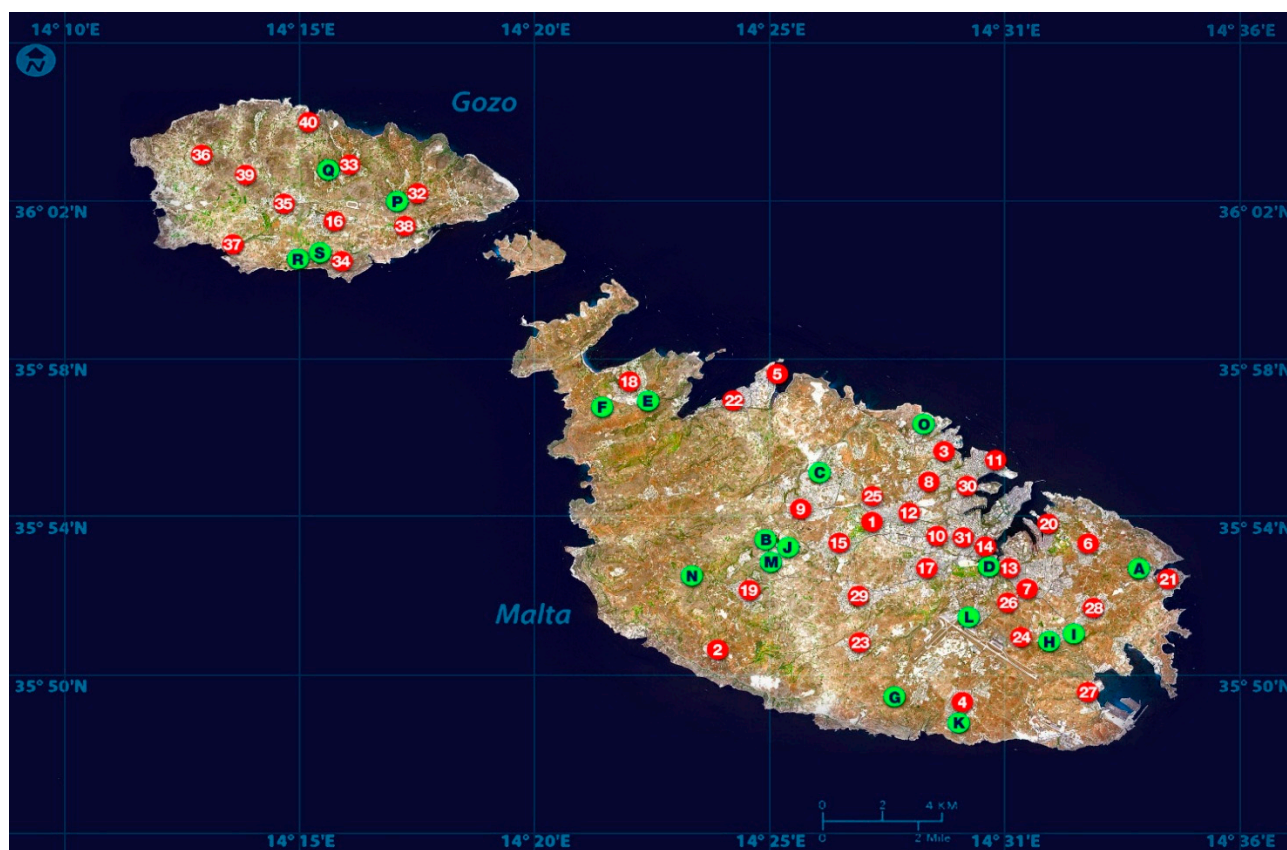


Figure 1. Map of the Maltese Islands showing the location of the samples taken from localities (marked in red) and supply reservoirs (marked in green). The Maltese Archipelago consists of two main inhabited islands: Malta and Gozo.

Table 2. List of localities and their corresponding reservoir sources.

Reservoir	A	B, J	C	D	E, F	G	H, I	K
Localities	21	1, 10, 12, 23, 29, 31	5, 9, 22, 25	14	18	6, 7, 13, 20, 24, 26, 28	27	4
Reservoir	L	M	N	O	P	Q	R, S	
Localities	17	15	2, 19	3, 8, 11, 30	32	33, 38	16, 34, 35, 36, 37, 39, 40	

Water samples from all localities were collected at the end point of distribution, *i.e.*, from the consumers’ taps. This was done in order to evaluate the quality of the water that is actually being consumed by the local population, thus reviewing the possibilities of heavy metal toxicity through potable water consumption.

Samples were collected in acid-washed containers and were acidified in 2% concentrated nitric acid on-site. Samples were transported in a cooler and stored in a refrigerator.

2.2. ICP-OES Analysis

Heavy metal levels were analyzed by ICP-OES using a method based on ISO 11885 [12]. The heavy metals analyzed were aluminum, cadmium, chromium, copper, iron, lead, nickel and zinc. Each

element was read off at the two best wavelengths similarly chosen as those instructed by the ISO 11885 method.

ICP-OES analysis was conducted by means of the Smart Analyzer (Ciros CCD) software. Each sample was introduced in the ICP-OES in its raw state and analyzed twice; each run generated a set of 3 readings together with the mean, standard deviation and the relative standard deviation. The instrument parameters are shown in Table 3.

Table 3. Instrumental parameters of the ICP-OES.

Parameter	Value
Radio Frequency (RF) generator power (W)	1400
Plasma gas flow rate (L/min)	12
Auxiliary gas flow rate (L/min)	0.8
Nebulizer flow rate (L/min)	1
Torch mode	Axial

Prior to use, the instrument was calibrated utilizing a fresh set of standard solutions, which were derived from a previously prepared mixed standard stock solution containing all of the elements analyzed (CPI International Peak Performance™ Certified Reference Materials were used to prepare calibrating solutions). The mixed standard stock solution was designed in accordance with the legal limits proposed in the Legal Notice 17 of 2009, as amended by Legal Notice 242 of 2009 of the Maltese Legislation; Water Intended for Human Consumption Regulations. The mixed standard stock solution was prepared in such a way that when dispensing different volumes to prepare the calibration standards, all elements involved would be set along the whole calibration range in a linear manner.

QC checks were performed before, during and after analysis using a set of QC standard solutions consisting of a blank solution containing tap water matrix, a QC low solution representing the lowest point on the calibration curve above zero and a QC high solution representing the highest point on the calibration curve. The QC low and QC high solutions ensured the consistency of the calibration curves, since the lowest and the highest points of the calibration curve tend to be the most variable. All standard solutions, both calibrating and QC check solutions, were prepared in a matrix similar to that of the water available locally in order to create a similar environment between the prepared solutions and the obtained samples. This was done to ensure that none of the elements would be masked during analysis due to the matrix effect.

In order to ensure the efficacy of the ICP-OES and the absence of interferences due to the matrix effect, two spiked sample solutions were also analyzed during the analysis. The spiked sample solutions were prepared by adding a known volume of the mixed standard stock solution, chosen between the volumes of the QC low and the QC high on the calibration curve, to a known volume of sample. Further confirmation of the method used and the data obtained was provided by the analysis of the European Reference Material ERM®-CA010a, which was routinely analyzed during each session of analysis. Typical percentage recoveries obtained before, during and after analysis are presented in Table 4.

The best wavelength for each element analyzed was chosen on the basis of the best and consistent percentage recoveries obtained in all QC checks performed during analysis. Thereby, the results presented are of the best wavelength analyzed for each element.

Table 4. Percentage recoveries of the QC standard solutions and ERM[®]-CA010a obtained before, during and after analysis.

Element	Wavelength	% Recovery							
		Before Analysis			During Analysis		After Analysis		
		ERM [®] CA010a	QC Low	QC High	Spiked Sample 32	Spiked Sample 40	ERM [®] CA010a	QC Low	QC High
Aluminum	394.401	96.7	99.0	95.5	101.4	105.4	97.9	100.2	97.0
Chromium	205.552	95.4	102.5	97.6	82.6	86.6	92.5	89.5	89.2
Iron	259.940	101.6	99.4	101.1	87.2	90.4	96.7	93.2	91.9
Nickel	231.604	96.9	99.7	99.7	86.9	91.4	91.0	90.5	91.2
Copper	327.396	-	102.1	98.1	93.2	98.9	-	97.2	95.2
Zinc	213.856	101.8	98.5	101.9	94.1	92.6	102.6	108.5	102.8
Cadmium	228.802	-	102.0	98.0	93.6	94.9	-	98.5	93.8
Lead	220.351	89.5	111.0	102.4	96.9	90.5	96.3	84.0	87.8

All of the raw data obtained were analyzed by means of the Biomedical Data Package (BMDP) Statistical Software. The sample results were compared to the legal concentration as proposed by the Maltese legislation by means of the Wilcoxon test, which is the non-parametric alternative of the one-sample *t*-test. One-way analysis of variance was used to compare the metal concentrations obtained in the localities with those obtained from the service reservoir samples feeding the respective localities sampled. This was done to show whether the concentrations in the reservoirs and the localities were significantly different or not.

Since the F ratio was significant in most cases, the mean results of the reservoirs and their corresponding localities were subsequently compared pairwise using the Bonferroni test.

3. Results and Discussion

3.1. Mean Concentrations of Localities and Their Corresponding Reservoirs

Out of forty localities, seven (17.5%) were not in the permitted range for a single metal.

Four of the localities had high concentrations of iron: Locality 3 had a concentration of 0.8 mg/L; Locality 2 and Locality 30 both had a concentration of 0.4 mg/L; and Locality 29 had a concentration of 1.2 mg/L.

Two of the localities, Locality 35 and Locality 36, both in Gozo, slightly exceeded the proposed legal limit for lead with a concentration of 0.01 mg/L in both villages, while the concentration of aluminum in Locality 21 was 0.36 mg/L.

Out of eighteen supply reservoirs, three (16.7%) were not in the permitted range for a single metal.

The legal limit of cadmium was exceeded by Reservoir H, with a concentration of 0.0136 mg/L. Reservoir E exceeded the legal limit of iron by 0.0754 mg/L, with an obtained mean concentration of 0.2754 mg/L, while Reservoir O had a concentration of 0.0622 mg/L of nickel.

Comparisons between all of the sample concentrations obtained and the Maltese legal limit were all statistically significant, since all of the *p*-values obtained from the operated Wilcoxon test were equal to zero and thus perfectly correlated.

A summary of the results obtained from samples of all localities and supply reservoirs is presented in Tables 5 and 6.

Table 5. Summary of the results in tap water sampled from localities, including the mean concentration, maximum mean value, limit of detection (LOD), limit of quantitation (LOQ), legal limit, standard deviation (SD), n (number of samples above LOD) and the best analytical wavelength.

Element	Mean	Max. Value	LOD	LOQ	Legal Limit	Unit	SD	n	Analytical Lines (nm)
Aluminum	0.026	0.356	0.006	0.018	0.2	mg/L	0.058	28	394.401
Cadmium	0.002	0.002	0.002	0.006	0.005	mg/L	0.0006	1	228.802
Chromium	<0.002	0.002	0.002	0.005	0.05	mg/L	0.0005	2	205.552
Copper	<0.02	0.028	0.02	0.05	2.0	mg/L	0.008	4	327.396
Iron	0.132	1.225	0.003	0.01	0.2	mg/L	0.227	40	259.94
Lead	<0.004	0.012	0.004	0.014	0.01	mg/L	0.003	14	220.351
Nickel	<0.003	0.006	0.003	0.008	0.02	mg/L	0.0009	1	231.604
Zinc	0.115	0.504	0.003	0.009	-	mg/L	0.136	39	213.856

Table 6. Summary of the results in water sampled from supply reservoirs, including the mean concentration, maximum mean value, limit of detection (LOD), limit of quantitation (LOQ), legal limit, standard deviation (SD), n (number of samples above LOD) and the best analytical wavelength.

Element	Mean	Max. Value	LOD	LOQ	Legal Limit	Unit	SD	n	Analytical Lines (nm)
Aluminum	0.01	0.043	0.006	0.018	0.2	mg/L	0.012	10	394.401
Cadmium	0.003	0.014	0.002	0.006	0.005	mg/L	0.003	10	228.802
Chromium	0.002	0.03	0.002	0.005	0.05	mg/L	0.007	3	205.552
Copper	<0.02	<0.02b	0.02	0.05	2.0	mg/L	0	-	327.396
Iron	0.06	0.275	0.003	0.01	0.2	mg/L	0.065	17	259.94
Lead	<0.004	<0.004	0.004	0.014	0.01	mg/L	0	-	220.351
Nickel	0.003	0.062	0.003	0.008	0.02	mg/L	0.014	1	231.604
Zinc	0.011	0.062	0.003	0.009	-	mg/L	0.023	4	213.856

3.2. Variations between Localities and Their Corresponding Supply Reservoirs

The two elements that exhibited the most significant variations between reservoirs and localities were iron and zinc, with 98% of the obtained concentrations being significantly different.

Aluminum and cadmium also exhibited a majority of significant variations between reservoirs and their corresponding localities, thus showing that these metal concentrations are somehow increasing following the passage of water in the distribution network. Significantly different concentrations between reservoirs and localities were also obtained for chromium and copper, followed by nickel and then lead. The data analysis results are presented in the Appendix.

3.3. General Trends of Heavy Metal Concentrations in the Maltese Islands

Aluminum was shown to exhibit higher concentrations in the south of the island, even though the concentrations obtained were minimal. In fact, the highest concentration was in Locality 21 (0.36 mg/L) situated in the south of Malta. Localities 20 and 24, also situated in the south of the island, showed higher concentrations of Al than the remaining localities analyzed. This was confirmed further by the Al concentration obtained in their corresponding Reservoir G, which was shown to have a higher concentration than the rest of the reservoirs.

No general trend for cadmium could be established, as concentrations were minimal and similar almost everywhere, with the exception of localities in Gozo, where nil concentrations above the detection limit were detected.

Chromium and copper did not show any general topographic trends, with concentrations occurring in minimal amounts throughout both islands.

Elevated levels of iron were mostly found in the central part of the island, even though iron was present in a random manner in almost every locality and reservoir.

Higher levels of lead were shown to be present in the northern part of Malta and in Gozo, with both Localities 35 and 36 slightly exceeding the proposed legal limit.

Nickel was absent in Gozo, while it was present in minimal amounts in Malta.

Zinc concentrations were very variable throughout both islands; however, reservoirs in Gozo contained trace amounts of Zn, while reservoirs in Malta contained nil.

The maximum values for the elements analyzed in Maltese tap water were comparable to the maximum results obtained in a study of tap water in Italy by Dinelli *et al.* [13], with the exception of the maximum value for iron, which was higher in the Maltese sample.

The maximum values obtained for cadmium and lead were also similar to the maximum values in drinking water samples from western Greece in a study conducted by Sotirios Karavoltos *et al.* [14]. However, maximum values for copper, chromium and nickel were lower in the Maltese samples.

The mean concentrations of cadmium, chromium, iron and lead in the Maltese samples were found to be lower than the mean concentrations in tap water samples from different districts of Shebin El-Kom city, Menoufiya in Egypt in a study by Badr *et al.* [15].

The general trend established in this study shows that metal concentrations tend to increase following distribution, since water collected from localities generally exhibited higher metal concentrations than their corresponding reservoirs, and this confirms other studies that indicate trace metal migration in drinking water from distribution networks [16]. Generally, metal concentrations are low when the water leaves the water supply, but during transportation and storage in the distribution network, the metal concentration may increase considerably [17]. Cadmium was an exception to this rule, with concentrations in reservoirs and at localities being quite similar. Iron was another exception, with the result that some reservoirs had a slightly higher concentration than their fed localities, although this was not always the rule.

4. Conclusions

This study has shown that the quality of potable water as regards heavy metals in the Maltese Islands is generally satisfactory, and most concentrations obtained were within the legal concentration range, with the exception of a few samples, which were found to have elevated levels of iron, aluminum and lead.

This study has also shown that metal concentrations in drinking water tend to increase from the supply reservoirs to the end-users, and this could be mainly related to corrosion in the distribution network. Corrosion occurs due to the instability of metals in the presence of water and due to the tendency to transform into a more stable and soluble form. Corrosion is characterized by partial solubilization of the materials constituting the treatment and supply systems, tanks, pipes, valves and pumps [18]. The main cause of metal pollution, such as lead, cadmium, copper, iron and zinc, in tap water is due to corrosion [19]. A study on tap water from Italy by Dinelli *et al.* also emphasized the possible effects of corrosion from the distributing tap water pipeline systems [13]. Parameters that significantly influence the corrosion and release of elements from distribution systems are pH and alkalinity. The solubility of corrosion products generally decreases with increasing pH [20]. Due to the fact that local water is hard, thus having a high pH value, the extent of corrosion in the local distribution network might be lower, and thus, this may contribute to keeping metal concentrations in trace amounts.

Acknowledgments

The research work disclosed in this publication is funded by the MASTER it! Scholarship Scheme (Malta). The scholarship is part-financed by the European Union – European Social Fund (ESF) under Operational Programme II – Cohesion Policy 2007-2013, “Empowering People for More Jobs and a Better Quality of Life”.

Author Contributions

Roberta Bugeja carried out the study and drafted the manuscript. Claire Shoemake was the supervisor of the project. Both authors read and approved the final manuscript.

Appendix

Variations between Localities and Their Corresponding Supply Reservoirs

The data analysis results are represented in this section. The *p*-values obtained from the Bonferroni test following one-way analysis of variance are represented in the tables of this section. Thus, any significant differences between the concentrations obtained in reservoirs and those in localities are shown here.

Table A1. Statistical variation between Reservoir A and its feeding Locality 21.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	A	0.0173	0.0019	N.D.	N.D.	0.1507	N.D.	N.D.	N.D.
Corresponding Locality (mg/L)	21	0.3557	0.0005	0.0011	N.D.	0.0255	N.D.	N.D.	0.0116
	<i>p</i>	<0.0001 *	<0.0001 *	0.0001 *	1.0000	<0.0001 *	0.8608	0.3632	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A2. Statistical variation between Reservoir C and its feeding Localities 5, 9, 22, 25.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	C	0.0031	0.0022	N.D.	N.D.	0.0385	N.D.	N.D.	N.D.
Corresponding Locality (mg/L)	5	0.0093	0.0012	N.D.	0.0003	0.1005	0.0007	0.0003	0.0925
	<i>p</i>	0.0160	<0.0001 *	0.3632	0.0902	<0.0001 *	0.1522	0.0256	<0.0001 *
Corresponding Locality (mg/L)	9	0.0027	0.0012	N.D.	N.D.	0.0984	0.0002	0.0002	0.0198
	<i>p</i>	0.7929	0.0001 *	1.0000	1.0000	<0.0001 *	0.3632	0.3632	<0.0001 *
Corresponding Locality (mg/L)	22	0.0028	0.0010	0.0007	N.D.	0.1175	0.0046	N.D.	0.3902
	<i>p</i>	0.2300	<0.0001 *	0.0003 *	1.0000	<0.0001 *	0.0021	0.3632	<0.0001 *
Corresponding Locality (mg/L)	25	0.0104	0.0008	0.0012	0.0022	0.0822	0.0027	N.D.	0.0322
	<i>p</i>	−8.820	<0.0001 *	0.0004 *	0.0015	<0.0001 *	0.1076	1.0000	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A3. Statistical variation between Reservoir D and its feeding Locality 14.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	D	0.0036	0.0022	N.D.	N.D.	0.0444	N.D.	N.D.	N.D.
Corresponding Locality (mg/L)	14	0.0131	0.0014	N.D.	0.0046	0.0392	0.0055	0.0008	0.0843
	<i>p</i>	<0.0001 *	0.0051	1.0000	<0.0001 *	0.0005 *	0.0088	0.1507	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A4. Statistical variation between Reservoirs B and J and their feeding Localities 1, 10, 12, 23, 29, 31.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	B	0.0014	0.0022	N.D.	N.D.	0.0249	N.D.	N.D.	N.D.
	J	0.0029	0.0015	N.D.	N.D.	0.0319	N.D.	N.D.	N.D.
Corresponding Locality (mg/L)	1	0.0108	0.0017	N.D.	0.0021	0.1164	0.0013	0.0003	0.1963
	<i>p</i>	(B) <0.0001 *	0.0052	0.6612	0.0009 *	<0.0001 *	0.0410	0.0560	<0.0001 *
	<i>p</i>	(J) <0.0001 *	0.2154	0.1852	0.0009 *	<0.0001 *	0.0525	0.0560	<0.0001 *
Corresponding Locality (mg/L)	10	0.0121	0.0016	0.0003	0.0283	0.0697	0.0022	0.0017	0.0710
	<i>p</i>	(B) 0.0003 *	−0.0019	0.2336	<0.0001 *	<0.0001 *	0.0339	0.0007 *	<0.0001 *
	<i>p</i>	(J) 0.0006 *	0.4973	0.1666	<0.0001 *	<0.0001 *	0.0459	0.0007 *	<0.0001 *
Corresponding Locality (mg/L)	12	0.0548	0.0014	N.D.	N.D.	0.0351	0.0014	N.D.	0.0431
	<i>p</i>	(B) <0.0001 *	0.0001 *	0.3632	1.0000	<0.0001 *	0.2670	1.0000	<0.0001 *
	<i>p</i>	(J) <0.0001 *	0.4162	1.0000	1.0000	<0.0001 *	0.3398	1.0000	<0.0001 *

Table A4. Cont.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Corresponding Locality (mg/L)	23	0.0215	0.0009	0.0008	0.0069	0.1316	0.0016	0.0001	0.0235
<i>p</i>	(B)	<0.0001 *	0.0001 *	0.0190	0.0014 *	<0.0001 *	0.1411	0.3632	<0.0001 *
<i>p</i>	(J)	<0.0001 *	0.0098	0.0155	0.0014 *	<0.0001 *	0.1917	0.3632	<0.0001 *
Corresponding Locality (mg/L)	29	0.0189	0.0006	0.0010	0.0029	1.2247	0.0060	0.0010	0.4179
<i>p</i>	(B)	<0.0001 *	<0.0001 *	<0.0001 *	0.0001 *	<0.0001 *	0.0071	0.0327	<0.0001 *
<i>p</i>	(J)	<0.0001 *	0.0004 *	0.0001 *	0.0001 *	<0.0001 *	0.0080	0.0327	<0.0001 *
Corresponding Locality (mg/L)	31	0.0138	0.0011	0.0010	N.D.	0.0721	0.0060	0.0007	0.1818
<i>p</i>	(B)	<0.0001 *	0.0004 *	-0.0030	1.0000	<0.0001 *	0.0075	0.0085	<0.0001 *
<i>p</i>	(J)	<0.0001 *	0.0600	-0.0028	1.0000	<0.0001 *	0.0084	0.0085	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A5. Statistical variation between Reservoir E and its feeding Locality 18.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	E	0.0434	0.0013	N.D.	N.D.	0.2754	N.D.	N.D.	N.D.
Corresponding Locality (mg/L)	18	0.0078	0.0006	0.0002	0.0017	0.0419	0.0087	N.D.	0.0952
<i>p</i>		<0.0001 *	0.0001 *	0.3041	0.2176	<0.0001 *	-0.0009	0.3632	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A6. Statistical variation between Reservoirs H and I and their feeding Locality 27.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Locality (mg/L)	27	0.0361	0.0004	0.0019	0.0100	0.1017	0.0031	0.0005	0.0213
Corresponding Reservoir (mg/L)	H	0.0020	0.0136	N.D.	N.D.	N.D.	N.D.	N.D.	N.D.
<i>p</i>		<0.0001 *	<0.0001 *	0.0005 *	<0.0001 *	<0.0001 *	0.0522	0.0476	<0.0001 *
Corresponding Reservoir (mg/L)	I	0.0119	0.0016	N.D.	N.D.	0.1040	N.D.	N.D.	N.D.
<i>p</i>		<0.0001 *	0.0001 *	0.0001 *	<0.0001 *	0.0494	0.0522	0.0476	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A7. Statistical variation between Reservoir K and its feeding Locality 4.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	K	0.0217	0.0013	N.D.	N.D.	0.0283	N.D.	N.D.	N.D.
Corresponding Locality (mg/L)	4	0.0389	0.0008	N.D.	N.D.	0.0222	N.D.	N.D.	0.0517
<i>p</i>		<0.0001 *	0.0475	1.0000	1.0000	<0.0001 *	0.5051	1.0000	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A8. Statistical variation between Reservoir G and its feeding Localities 6, 7, 13, 20, 24, 26, 28.

		Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	G	0.0331	0.0016	N.D.	N.D.	0.0280	N.D.	N.D.
Corresponding Locality (mg/L)	6	0.0154	0.0011	N.D.	0.0017	0.0230	N.D.	0.0198
<i>p</i>		<0.0001 *	0.0730	0.0323	0.2120	<0.0001 *	0.2031	0.1877

Table 8. *Cont.*

		Cd	Cr	Cu	Fe	Pb	Ni	Zn	
Corresponding Locality (mg/L)	7	0.0219	0.0008	N.D.	N.D.	0.0432	N.D.	0.0013	0.0600
	<i>p</i>	<0.0001 *	0.0038	1.0000	1.0000	<0.0001 *	0.2304	<0.0001 *	<0.0001 *
Corresponding Locality (mg/L)	13	0.0137	0.0011	N.D.	0.0011	0.0560	0.0005	0.0008	0.1358
	<i>p</i>	<0.0001 *	0.0307	1.0000	0.2136	<0.0001 *	0.2071	0.0676	<0.0001 *
Corresponding Locality (mg/L)	20	0.1148	0.0005	0.0022	0.0217	0.0374	N.D.	0.0014	0.0125
	<i>p</i>	<0.0001 *	0.0003 *	<0.0001 *	<0.0001 *	<0.0001 *	1.0000	0.0172	<0.0001 *
Corresponding Locality (mg/L)	24	0.0787	0.0004	0.0004	N.D.	0.0258	0.0008	N.D.	0.0154
	<i>p</i>	<0.0001 *	0.0002 *	0.0415	1.0000	<0.0001 *	0.3632	1.0000	<0.0001 *
Corresponding Locality (mg/L)	26	0.0322	0.0004	0.0009	N.D.	0.0193	0.0026	N.D.	0.0260
	<i>p</i>	0.3317	0.0002 *	0.0018	1.0000	<0.0001 *	0.0314	0.3632	<0.0001 *
Corresponding Locality (mg/L)	28	0.0164	0.0005	0.0004	N.D.	0.0212	0.0015	N.D.	0.0384
	<i>p</i>	<0.0001 *	0.0002 *	0.0207	1.0000	<0.0001 *	0.1688	1.0000	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A9. Statistical variation between Reservoir L and its feeding Locality 17.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	L	0.0022	0.0030	N.D.	N.D.	0.0644	N.D.	N.D.	N.D.
Corresponding Locality (mg/L)	17	0.0039	0.0013	0.0008	0.0020	0.0422	0.0009	0.0055	0.4059
	<i>p</i>	0.0534	<0.0001 *	0.0002 *	-0.0011	<0.0001 *	0.1763	<0.0001 *	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A10. Statistical variation between Reservoir M and its feeding Locality 15.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	M	0.0076	0.0021	N.D.	N.D.	0.0850	N.D.	N.D.	N.D.
Corresponding Locality (mg/L)	15	0.0160	0.0013	N.D.	0.0087	0.1110	0.0001	0.0017	0.0043
	<i>p</i>	0.0001 *	0.0003 *	0.2586	<0.0001 *	<0.0001 *	0.3632	0.0004 *	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A11. Statistical variation between Reservoir N and its feeding Localities 2 and 19.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	N	0.0074	0.0020	N.D.	N.D.	0.0421	N.D.	N.D.	N.D.
Corresponding Locality (mg/L)	2	0.0001	0.0015	N.D.	0.0012	0.4060	0.0003	N.D.	0.5041
	<i>p</i>	<0.0001 *	0.0217	1.0000	0.0018	<0.0001 *	0.1307	0.2257	<0.0001 *
Corresponding Locality (mg/L)	19	0.0047	0.0011	0.0002	0.0008	0.1483	0.0068	0.0001	0.0986
	<i>p</i>	0.0338	-0.0012	0.1357	0.0087	<0.0001 *	0.0003 *	0.3632	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A12. Statistical variation between Reservoir O and its feeding Localities 3, 8, 11, 30.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	O	0.0015	0.0013	0.0298	N.D.	0.0002	0.0019	0.0622	N.D.
Corresponding Locality (mg/L)	3	0.0038	0.0004	N.D.	N.D.	0.7928	N.D.	0.0004	0.2567
	<i>p</i>	0.0767	0.0791	<0.0001 *	0.3632	<0.0001 *	0.1290	<0.0001 *	<0.0001 *
Corresponding Locality (mg/L)	8	0.0151	0.0004	N.D.	N.D.	0.1611	N.D.	0.0002	0.0656
	<i>p</i>	<0.0001 *	0.0773	<0.0001 *	0.3632	<0.0001 *	0.0003 *	<0.0001 *	<0.0001 *
Corresponding Locality (mg/L)	11	0.0182	N.D.	N.D.	N.D.	0.0107	N.D.	0.0004	N.D.
	<i>p</i>	<0.0001 *	0.0362	<0.0001 *	0.3632	<0.0001 *	0.0003 *	<0.0001 *	1.0000
Corresponding Locality (mg/L)	30	0.0079	0.0001	0.0001	N.D.	0.4258	0.0030	N.D.	0.0682
	<i>p</i>	0.0003 *	0.0461	<0.0001 *	0.3632	<0.0001 *	0.4307	<0.0001 *	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A13. Statistical variation between Reservoirs R and S and their feeding Localities 16, 35, 35, 36, 37, 39, 40.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	R	0.0015	0.0026	0.0021	0.0002	0.0298	N.D.	N.D.	0.0622
	S	0.0013	0.0025	0.0019	0.0009	0.0272	N.D.	N.D.	0.0487
Corresponding Locality (mg/L)	16	N.D.	0.0020	N.D.	0.0025	0.1169	N.D.	N.D.	0.1138
	(R)	0.1214	0.0188	0.0004 *	<0.0001 *	<0.0001 *	0.2204	0.3632	<0.0001 *
	(S)	0.0362	-0.0022	0.0003 *	0.0002 *	<0.0001 *	1.0000	0.3632	<0.0001 *
Corresponding Locality (mg/L)	34	0.0048	N.D.	0.0004	0.0013	0.0689	0.0035	N.D.	0.1004
	(R)	0.0256	<0.0001 *	0.0005 *	0.0013 *	<0.0001 *	0.0545	0.3632	<0.0001 *
	(S)	0.0172	<0.0001 *	0.0003 *	0.0356	<0.0001 *	0.0139	0.3632	<0.0001 *
Corresponding Locality (mg/L)	35	0.0009	N.D.	N.D.	0.0027	0.0615	0.0103	N.D.	0.0301
	(R)	0.4454	<0.0001 *	0.0004 *	<0.0001 *	<0.0001 *	0.0041	0.3632	<0.0001 *
	(S)	0.2471	<0.0001 *	0.0003 *	0.0005 *	<0.0001 *	0.0035	0.3632	<0.0001 *
Corresponding Locality (mg/L)	36	0.0097	N.D.	0.0003	0.0150	0.0468	0.0121	N.D.	0.0397
	(R)	0.0001 *	<0.0001 *	0.0003 *	<0.0001 *	<0.0001 *	0.0001 *	0.3632	<0.0001 *
	(S)	0.0001 *	<0.0001 *	0.0002 *	<0.0001 *	<0.0001 *	0.0003 *	0.3632	<0.0001 *
Corresponding Locality (mg/L)	37	0.0093	N.D.	0.0002	0.0283	0.0368	0.0066	N.D.	0.02678
	(R)	0.0001 *	<0.0001 *	0.0003 *	0.0206	<0.0001 *	0.0357	0.3632	<0.0001 *
	(S)	<0.0001 *	<0.0001 *	0.0001 *	0.0222	<0.0001 *	0.0214	0.3632	<0.0001 *
Corresponding Locality (mg/L)	39	0.0090	N.D.	0.0008	0.0279	0.0171	0.0093	N.D.	0.1683
	(R)	0.0001 *	<0.0001 *	0.0028	<0.0001 *	<0.0001 *	<0.0001 *	0.6676	<0.0001 *
	(S)	<0.0001 *	<0.0001 *	-0.0023	<0.0001 *	<0.0001 *	<0.0001 *	0.6676	<0.0001 *
Corresponding Locality (mg/L)	40	0.0186	N.D.	0.0010	0.0027	0.0084	0.0043	N.D.	0.0305
	(R)	<0.0001 *	<0.0001 *	0.0071	<0.0001 *	<0.0001 *	0.0065	0.3632	<0.0001 *
	(S)	<0.0001 *	<0.0001 *	0.0064	0.0003 *	<0.0001 *	-0.0019	0.3632	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Table A14. Statistical variation between Reservoirs P, Q and their feeding Localities 32 and 33, 38.

		Al	Cd	Cr	Cu	Fe	Pb	Ni	Zn
Reservoir (mg/L)	P	0.0065	0.0032	0.0013	N.D.	0.0339	0.0022	N.D.	0.0467
Corresponding Locality (mg/L)	32	N.D.	N.D.	0.0004	0.0012	0.0668	0.0047	N.D.	0.0944
	<i>p</i>	0.0002 *	<0.0001 *	0.0258	0.0068	<0.0001 *	0.2093	0.1910	<0.0001 *
Reservoir (mg/L)	Q	0.0078	0.0031	0.0017	N.D.	0.0310	N.D.	0.0003	0.0569
Corresponding Locality (mg/L)	33	0.0042	N.D.	0.0002	0.0008	0.0962	0.0048	N.D.	0.0721
	<i>p</i>	0.0992	<0.0001 *	0.0001 *	0.0417	<0.0001 *	0.0322	0.0756	<0.0001 *
Corresponding Locality (mg/L)	38	0.0010	N.D.	N.D.	0.0006	0.1446	0.0077	N.D.	0.4672
	<i>p</i>	0.0002 *	<0.0001 *	0.0002 *	0.0532	<0.0001 *	0.0148	0.0756	<0.0001 *

All *p*-values (*p*) marked with an asterisk (*) denote statistically-significant differences in concentrations between the reservoir and its feeding locality.

Conflicts of Interest

The authors declare no conflict of interest.

References

1. Anazawa, K.; Kaida, Y.; Shinomura, Y.; Tomiyasu, T.; Sakamoto, H. Heavy-metal distribution in river waters and sediments around a “Firefly Village”, Shikoku, Japan: Application of multivariate Analysis. *Anal. Sci.* **2004**, *20*, 79–84.
2. Jan, A.T.; Ali, A.; Haq, Q.M.R. Glutathione as an antioxidant in inorganic mercury induced nephrotoxicity. *J. Postgrad. Med.* **2011**, *57*, 72–77.
3. Saxena, G.; Dube, S.N.; Flora, S.J.S. Heavy metals induced central cholinergic system disorders and their possible mechanisms. In *Recent Trends in the Acetylcholinesterase System*; Parveen, M., Kumar, S., Eds.; IOS Press: Amsterdam, The Netherlands, 2005; Chapter 3, p. 28.
4. Nava-Ruíz, C.; Méndez-Armenta, M. Cadmium, lead, thallium: Occurrence, neurotoxicity and histopathological changes of the nervous system. In *Pollutant Diseases, Remediation and Recycling*; Lichtfouse, E., Schwarzbauer, J., Robert, D., Eds.; Springer International Publishing: Cham, Switzerland, 2013; Chapter 6, p. 323.
5. Attard, G. Natural Resources in Malta. Available online: http://www.ciheam.org/old_website/images/CIHEAM/PDFs/Observatoire/NAN/nan22.pdf (accessed on 21 March 2015).
6. Malta: Use of Leakage Control in Water Management Strategy Case # 22. Available online: <http://ec.europa.eu/ourcoast/download.cfm?fileID=817> (accessed on 21 March 2015).
7. FAO. Review of world water resources by country. In *Water Reports*; FAO: Rome, Italy, 2003; p. 19.
8. FAO. *Malta Water Resources Review*; FAO: Rome, Italy, 2006; Chapter 6, p. 35.
9. Sammut, M.; Savona-Ventura, C. Petrol lead in a small island environment. *Int. J. Risk Saf. Med.* **1996**, *9*, 33–40.
10. Savona-Ventura, C.; Sammut, M.; Licari, L.; Vassallo, A. Blood lead levels in pregnant women and the neonate. *Malta Med. J.* **1997**, *9*, 42–45.

11. Fertmann, R.; Hentschel, S.; Dengler, D.; Janssen, U.; Lommel, A. Lead exposure by drinking water: An epidemiological study in Hamburg, Germany. *Int. J. Hyg. Environ. Health* **2004**, *207*, 235–244.
12. EN ISO 11885:1997(E). *Water Quality-Determination of 33 Elements by Inductively Coupled Plasma Atomic Emission Spectroscopy*; International Organization for Standardization (ISO): Geneva, Switzerland, 1997.
13. Dinelli, E.; Lima, A.; Albanese, S.; Birke, M.; Cicchella, D.; Giaccio, L.; Valera, P.; de Vivo, B. Major and trace elements in tap water from Italy. *J. Geochem. Explor.* **2012**, *112*, 54–75.
14. Karavoltzos, S.; Sakellari, A.; Mihopoulos, N.; Dassenakis, M.; Scoullou, M.J. Evaluation of the quality of drinking water in regions of Greece. *Desalination* **2008**, *224*, 317–329.
15. Badr, E.; Agrama, A.; Badr, S. Heavy metals in drinking water and human health, Egypt. *Nutr. Food Sci.* **2011**, *41*, 210–217.
16. Veschetti, E.; Achene, L.; Ferretti, E.; Lucentini, L.; Citti, G.; Ottaviani, M. Migration of trace metals in Italian drinking waters from distribution networks. *Toxicol. Environ. Chem.* **2010**, *92*, 521–535.
17. Fontenay, F.; Andersen, A.; Werner, W.; Erning, W. Nickel and Lead Release to Drinking Water—Danish and European Results. 8351. Available online: http://www.forcetechnology.com/NR/rdonlyres/68BF09BF-5C28-416E-9F89-E8C17208B753/2635/EUROCORR_2009_Nice_711September2009_web.pdf (accessed on 1 March 2015).
18. WHO. Chemical aspects. In *Guidelines for Drinking-Water Quality*, 4th ed.; WHO: Geneva, Switzerland, 2011; Chapter 8, p. 174.
19. Sorlini, S.; Gialdini, F.; Collivignarelli, C. Metal leaching in drinking water domestic distribution system: An Italian case study. *Int. J. Environ. Health Res.* **2014**, *24*, 497–514.
20. Kim, E.J.; Herrera, J.E.; Huggins, D.; Braam, J.; Koshowski, S. Effect of pH on the concentrations of lead and trace contaminants in drinking water: A combined batch, pipe loop and sentinel home study. *Water Res.* **2011**, *45*, 2763–2774.

© 2015 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 license (<http://creativecommons.org/licenses/by/4.0/>).