

Article

Household Water Filtration Technology to Ensure Safe Drinking Water Supply in the Langat River Basin, Malaysia

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Abstract: Populations in the Langat River Basin, Malaysia, frequently experience water supply disruption due to the shutdown of water treatment plants (WTPs) mainly from the chemical pollution as well as point and non-point sources of pollution. Therefore, this study investigated the aluminium (Al), arsenic (As), cadmium (Cd), chromium (Cr), and lead (Pb) concentrations in the drinking water supply chain at the basin because of its prolonged persistence and toxic characteristics in the aquatic environment. Three replicates of water samples were collected from the river, outlets of WTPs, household tap and filtered water, respectively, in 2015, for analysis by Inductively Coupled Plasma Mass Spectrometry. Higher concentration of these metals was found in household tap water than in the treated water at the WTPs; however, the concentration of these metals at the four stages of the drinking water supply chain conformed to the drinking water quality standard set by the World Health Organization. The Mann-Whitney and Kruskal-Wallis tests also found that metal concentration removal significantly varied among the eight WTPs as well as the five types of household water filtration systems. With regards to the investigated household filtered water, the distilled filtration system was found to be more effective in removing metal concentration because of better management. Therefore, a two-layer water filtration system could be introduced in the Langat River Basin to obtain safe drinking water supply at the household level.

Keywords: chemical pollution; water treatment plant; coagulation water treatment method; reverse osmosis; water quality



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1. Introduction

The Langat River is one of the 189 major river basins in Malaysia and has unique characteristics [1,2]. The Langat basin shares different jurisdictions—Selangor State (78.14%), Negeri Sembilan State (19.64%), and the Federal Territories of Putrajaya (1.9%) and Kuala Lumpur (0.33%) in Malaysia [1,3]. Therefore, the pollution of this transboundary river, both from point and non-point sources, is a serious concern because of pollution management by different states; state governments are the owners of river and land in Malaysia [1,4–11]. Inadequate collaboration and partnership in respect to pollution management of Langat River has also been reported among the agencies [12,13]. Langat River is one of the prime sources of drinking water in the basin and it provides drinking water to almost one-third of the population in Selangor State [7,9]. However, the nine water treatment plants (WTPs) based in the Langat River Basin have experienced several shutdown incidents because of pollution of the river, both from point and non-point sources, including chemical pollution [1,7,14,15]. A few studies have also reported high concentrations of arsenic (As), lead (Pb), cadmium (Cd), chromium (Cr), and aluminum (Al) in Langat River [4,9,16–18], compared to other rivers around the world, such as Huaihe, China [19]; Nile, Egypt [20]; Karnaphuli, Bangladesh [21], and Transylvania, Romania [22], among others. For instance, Al 46.28 ± 32.71 – $380 \mu\text{g/L}$ [4,23,24]; As 4 – $201.11 \mu\text{g/L}$ [4,23,25–27]; Cd 0.11 ± 0.12 – $35.56 \mu\text{g/L}$ [16,23,26,28]; Cr 0.67 ± 0.90 – $70 \mu\text{g/L}$ [4,16,23–25], and Pb

0.16 ± 0.23 – $57.78 \mu\text{g/L}$ [4,16,23,25,26] concentrations have been reported to be high in the Langat River since 1985. The highest concentration of Al, As, Cd, Cr, and Pb in the Langat River also crossed the Malaysian standard of drinking water quality— $200 \mu\text{g/L}$, $10 \mu\text{g/L}$, $3 \mu\text{g/L}$, $10 \mu\text{g/L}$, and $10 \mu\text{g/L}$, respectively [29]. Lim et al. [24] also reported a high level of Al $290.07 \pm 817.50 \mu\text{g/L}$ in the Langat River, possibly due to the natural weathering of aluminium and ferralsols clay minerals at the Langat basin, which exceeded the Malaysian standard for drinking water quality— $200 \mu\text{g/L}$.

Arsenic (As), Cadmium (Cd), Chromium (Cr), and Lead (Pb) are highly toxic contaminants, and have been reported by the United States Environmental Protection Agency [30,31] to have carcinogenic and non-carcinogenic health risks, if ingested for a long time even at trace level. Similarly, two epidemiological studies have globally confirmed the significant association between Al ingestion via drinking water and Alzheimer's disease [32,33]. Human health is highly susceptible to the exposure of heavy metals, even at trace levels, because of its prolonged persistence in environmental media and its acute toxicity. Researchers have already reported the abundance of toxic As, Cd, Cr, and Pb in the environment and its exposure on human health [34,35]. The exposure of these metals is widespread, especially in developing countries, mainly because of human activities as well as natural weathering of mineral rocks [4,28]. Hydrous aluminium in clay mineral [36] and erosion of ferralsols (i.e., oxisols and ultisols) enriched with Al [5,24] in the Langat River Basin is attributed to the higher dissolved concentration of Al in the Langat River. Moreover, the excess use of $\text{Al}_2(\text{SO}_4)_3$ (aluminium sulphate) in the conventional flocculation and coagulation method of tropical river water disinfection might also have contributed to the higher level of Al concentration in the water supply at the household level in the Langat River Basin in Malaysia [9,37]. Therefore, this study investigated Al, As, Cd, Cr, and Pb status in the drinking water supply chain at the Langat River Basin in order to suggest better management of drinking water quality.

2. Methods

2.1. Water Sample Collection and Analytical Method

Three replicates of each water sample were collected in polyethene containers from the eight stations of the river upstream to downstream, from precisely the same places where the water treatment plants (WTPs) in the basin collect water for drinking water treatment purposes. Three replicates of each water sample were also collected from the outlets of the eight WTPs along with from the kitchen tap of 15 households at the basin, based on the common use of five types of household filtration systems. Three replicates of each household filtration water were also collected from the same 15 households. The samples were collected once in 2015 to analyze Al, As, Cd, Cr, and Pb concentrations in the drinking water supply chain in the Langat River Basin, Malaysia.

The samples were acidified with concentrated HNO_3 to maintain a $\text{pH} < 2$ as soon as possible after collection, to avoid contamination as well as precipitation of trace elements. Similarly, all the glassware used for the analysis were acid-washed to avoid possible contamination. The raw samples were also filtered using $0.45 \mu\text{m}$ glass fibre filter paper (Whatman) to analyse the dissolved part. Chelex[®] 100 resin (i.e., 50–100 mesh, Bio-Rad, Hercules, CA, USA) column ion-exchange method was applied to analyze the three replicates of these 500 mL water samples. Ion-exchange columns were prepared by soaking 20 g of Chelex[®] 100 resin in 2.5 M HNO_3 for two hours and then decanting and soaking in clean 2.5 M HNO_3 for another two hours. This slurry mixture was then poured into a fritted glass column, allowed to drain, washed with 30 mL of 2.5 M HNO_3 twice, rinsed with 30 mL of distilled deionized water (DI- H_2O) twice, and then converted to the ammonium form by eluting with 10 mL of 1 M NH_4OH . Excess NH_4OH was removed by rinsing with 30 mL of DI- H_2O . The prepared columns were placed in a rack.

The weighed samples ($\text{pH} < 2$) were buffered with a mixture of the same volume of 10 M $\text{NH}_4\text{CH}_3\text{CO}_2$ and 10 M NH_4OH to adjust the pH to ~ 5.4 (approximately). Then, three replicates of the 500 mL water samples were drained in the prepared column and the

flow rate of the sample water was adjusted to 20 s per drop. When no solution remained above the column, the column was rinsed with 30 mL of 10 M $\text{NH}_4\text{CH}_3\text{CO}_2$ to remove excess salts and eluted with 25 mL of 2 M HNO_3 into a 30-mL (LPE) bottle. The eluent was analysed for the dissolved Al, As, Cd, Cr, and Pb concentrations (i.e., $\mu\text{g/L}$) by Inductively Coupled Plasma-Mass Spectrometry (ICP-MS, ELAN 9000 ICP-MS, PerkinElmer, Shelton, CT, USA) [9,38,39].

The locations of water sampling points were recorded by a Global Positioning System (GPS, GARMIN, GPSMAP 76CSx, Kansas, MO, USA) to prepare the water sample location map (Figure 1). Accordingly, in-situ physicochemical water quality parameters, such as dissolved oxygen (DO), conductivity, total dissolved solids (TDS), pH, temperature, salinity, etc. were recorded with a calibrated Professional Plus Water Quality Instrument (6050000, YSI Incorporated, Yellow Springs, OH, USA) from each sampling point.

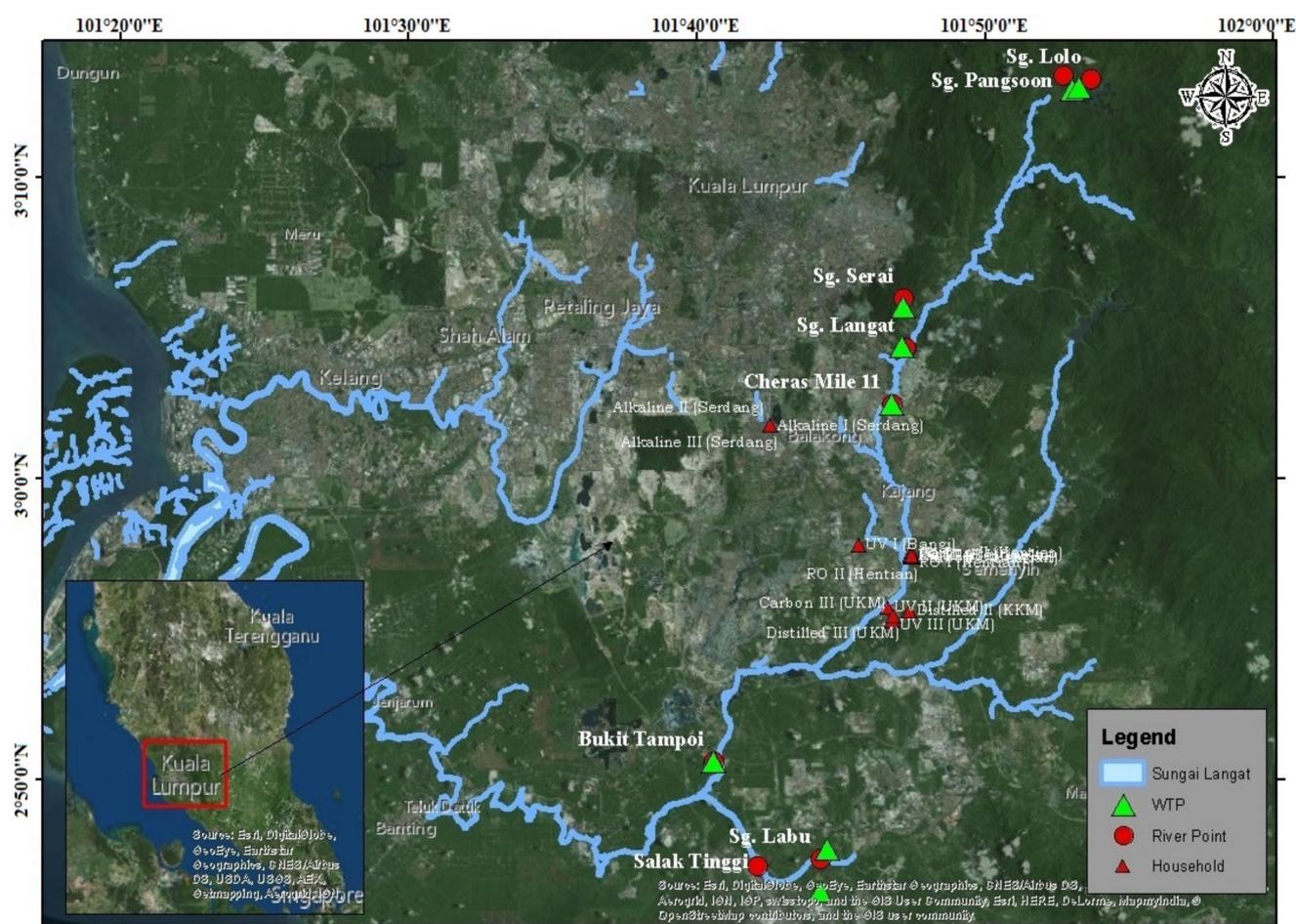


Figure 1. Location of Water Sample Collections in the Langkat River Basin, Malaysia.

2.2. Quality Assurance and Control

The quality of analytical data was controlled by the calibration of the ICP-MS, and analysis of replicates and blanks. Therefore, the standard of several concentrations was prepared with the Multi-Element Calibration Standard III (PerkinElmer, Lot # CL7-173YPY1, PE # N9300233, Waltham, MA, USA) with the same acid mixture used for sample dissolution to calibrate the metals analysis by ICP-MS. The accuracy of ICP-MS was analyzed with the standard curve ($r^2 = 0.999$) for Al, As, Cd, Cr, and Pb, respectively, and the precision of the analytical procedure was determined by relative standard deviation (RSD) i.e., Al 0.002%, As 5.800%, Cd 0.295%, Cr 0.005%, and Pb 0.003%. Accordingly, blanks were analyzed for background correction of these metals' concentrations. The mean SRM recoveries

were Al 99.972 ± 0.002 , As 75.992 ± 4.408 , Cd 94.966 ± 0.280 , Cr 99.803 ± 0.005 , and Pb $98.762 \pm 0.003\%$. The low As SRM recovery might be due to its vaporized characteristic. However, the National Institute of Public Health and the Environment of the Netherlands accepted the SRM recovery of As in the water sample between 75% and 125% as well as RSD 10% [40]. Since mean arsenic SRM was determined to be $75.992 \pm 4.408\%$, the remaining 24.008% was calculated for each sample to report the corrected As concentration in this study.

2.3. Statistical Analysis

One-way analysis of variance (ANOVA) were conducted with SPSS (Version 21.0, IBM Corp., Armonk, NY, USA) to indicate the significant mean differences of metals' concentration in the drinking water supply chain in the Langat River Basin, Malaysia. Pearson correlation analysis was conducted to measure the linear correlation between determined metal concentrations and physicochemical parameters in the Langat River. Non-parametric Mann-Whitney U test and Kruskal-Wallis H test were used to analyze the metal concentrations because these analyses do not require the assumption of normal distributions of data. Contaminants are rarely distributed normally because of both point and non-point sources of pollutions. Mann-Whitney U tests were used to measure the significant differences in the mean concentrations of metals at the two stages of the drinking water supply chain. Similarly, Kruskal-Wallis H tests were used to measure the significant differences in the mean concentrations of metals in the four stages of the drinking water supply chain.

3. Results and Discussions

3.1. Metal Concentrations in the Langat River

The higher dissolved concentration of Al (529.96 ± 70.45 $\mu\text{g/L}$) upstream of the Langat River (Figure 2) than downstream (77.65 ± 41.42 $\mu\text{g/L}$) is mostly from the natural weathering of iron and silica bedrock in the Titiwangsa Granite Hill Range. The high concentration of Al in the Langat River has crossed the highest limit of drinking water quality standard 200 $\mu\text{g/L}$ set by the Ministry of Health Malaysia [29] and the World Health Organization [41]. The hydrous aluminium in clay minerals (i.e., magnesium-aluminosilicate) of the basin [36,42] and erosion of ferralsols (i.e., oxisols and ultisols) enriched with Al [24] is attributed to the higher dissolved concentration of Al in the Langat River, apart from man-made activities such as discharges of Al-enriched wastewater into the river by water treatment plants [43].

Similarly, the primary source of As in the river is the natural weathering of arsenopyrite mineral [44] in the granite belt, along with anthropogenic activities in the basin. The anthropogenic inputs might be the main contributor to increase in concentration of As (1.81 ± 0.95 $\mu\text{g/L}$) in the midstream of the Langat River. Effluent discharge from the urban area (especially effluent from the sewerage treatment plants), industrial area, as well as runoff from the agricultural area might have increased the concentration of As in the Langat River. The mean concentration of As was 1.65 ± 0.93 $\mu\text{g/L}$, which was within the drinking water quality standard set by the Ministry of Health Malaysia as well as the World Health Organization (Table 1). The highest concentration of Cd 2.54 ± 0.02 $\mu\text{g/L}$ at Serai point, which is a hilly forest area, might be due to the natural weathering of Cd from zinc ores such as sphalerite (ZnS) as well as the cadmium mineral such as Greenockite (CdS) [45] in the Titiwangsa Granite Hill Range of the Langat Basin. Moreover, the point sources of pollution, such as the effluent from the sewerage treatment plants (STPs) near the Langat and Cheras point of the river, also attribute to the higher concentration of Cd, i.e., 1.25 ± 0.09 $\mu\text{g/L}$ and 1.23 ± 0.73 $\mu\text{g/L}$, respectively (Figure 2). The higher dissolved concentration of Cr was investigated at the upstream hilly forest area such as Pangsoon point 0.60 ± 0.56 $\mu\text{g/L}$, Lolo point 0.66 ± 0.36 $\mu\text{g/L}$, and Serai point 0.60 ± 0.04 $\mu\text{g/L}$ than the downstream mostly because of weathering of serpentinite rock-derived oxisols along the central belt of peninsular Malaysia [46]. However, the much lower Pb concentrations in the

downstream ($5.03 \pm 0.27 \mu\text{g/L}$) was probably due to lower atmospheric Pb inputs [47] in the downstream than the upstream ($20.71 \pm 3.67 \mu\text{g/L}$). Apart from the natural weathering process, the use of fertilizers such as arsenal herbicides (i.e., lead arsenate) in agricultural activities [4], mainly in palm oil plantation [5] and tin mining [26,48], are the essential sources of Pb concentrations in the Langat River.

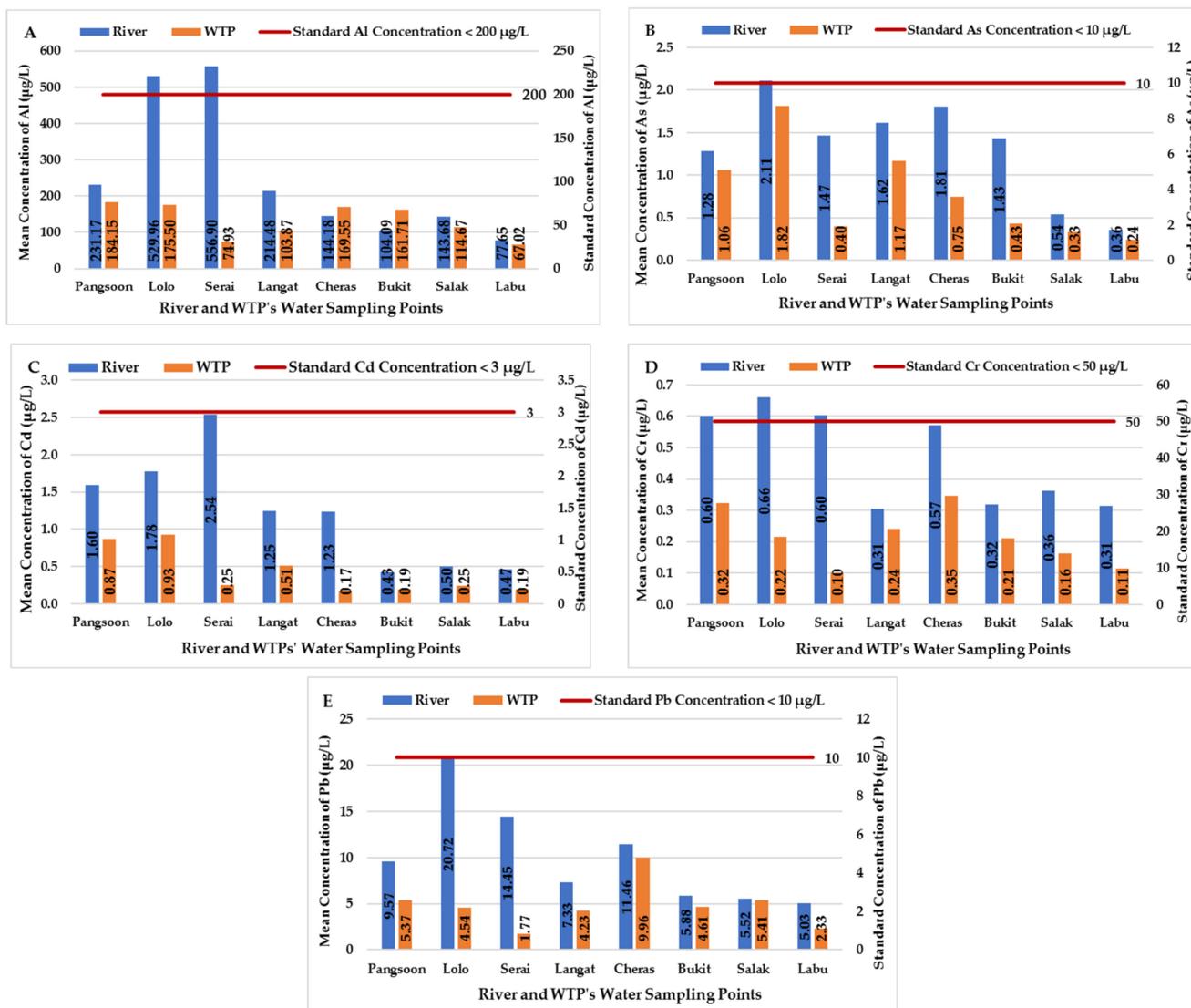


Figure 2. Al (A), As (B), Cd (C), Cr (D), and Pb (E) Concentrations ($\mu\text{g/L}$) from upstream to downstream in the Langat River and water treatment plants (WTPs).

Table 1. Mean Al, As, Cd, Cr and Pb concentrations ($\mu\text{g/L}$) in drinking water supply chain at the Langat River Basin, Malaysia (2015).

Sample	Metal	N	Range	Mean	MOH ¹	WHO ²	USEPA ³	EC ⁴
River water	Al ($\mu\text{g/L}$)	24	38.09–648.52	250.26 \pm 189.24	-	-	-	60 ⁷
	As ($\mu\text{g/L}$)	24	0.33–3.04	1.65 \pm 0.93	10	150 ⁵	-	50 ⁷
	Cd ($\mu\text{g/L}$)	24	0.39–3.43	1.22 \pm 0.88	3	0.72 ⁵	0.2 ⁶	10 ⁷
	Cr ($\mu\text{g/L}$)	24	0.12–1.22	0.47 \pm 0.27	50	11 ⁵	-	50 ⁷
	Pb ($\mu\text{g/L}$)	24	4.76–24.93	9.99 \pm 1.40	50	2.5 ⁵	1.3 ⁶	50 ⁷
Treated water	Al ($\mu\text{g/L}$)	24	51.60–197.74	131.43 \pm 48.56	200	200	200	200
	As ($\mu\text{g/L}$)	24	0.24–2.67	0.77 \pm 0.56	10	10	10	10
	Cd ($\mu\text{g/L}$)	24	0.12–0.99	0.42 \pm 0.31	3	3	5	5
	Cr ($\mu\text{g/L}$)	24	0.02–0.53	0.21 \pm 0.14	50	50	100	50
	Pb ($\mu\text{g/L}$)	24	1.69–10.83	4.78 \pm 2.50	10	10	15	10
Tap water	Al ($\mu\text{g/L}$)	45	71.87–225.04	148.31 \pm 46.56	200	200	200	200
	As ($\mu\text{g/L}$)	45	0.13–2.69	0.78 \pm 0.53	10	10	10	10
	Cd ($\mu\text{g/L}$)	45	0.13–0.77	0.42 \pm 0.19	3	3	5	5
	Cr ($\mu\text{g/L}$)	45	0.1–0.95	0.37 \pm 0.21	50	50	100	50
	Pb ($\mu\text{g/L}$)	45	1.16–8.93	4.84 \pm 1.87	10	10	15	10
HH ⁸ filtration	Al ($\mu\text{g/L}$)	45	21.1–270.57	120.68 \pm 57.47	200	200	200	200
	As ($\mu\text{g/L}$)	45	0.05–1.32	0.37 \pm 0.37	10	10	10	10
	Cd ($\mu\text{g/L}$)	45	0.03–0.74	0.31 \pm 0.21	3	3	5	5
	Cr ($\mu\text{g/L}$)	45	0.05–0.66	0.2 \pm 0.15	50	50	100	50
	Pb ($\mu\text{g/L}$)	45	0.64–14.41	4.12 \pm 2.89	10	10	15	10

Note: ¹ Drinking Water Quality Standard proposed by the Ministry of Health Malaysia [29]. ² Guidelines for Drinking Water Quality proposed by the World Health Organization [41]. ³ Regulated Drinking Water by the United States Environmental Protection Agency [49]. ⁴ Drinking Water Directive proposed by the European Commission [50]. ⁵ Criteria Continuous Concentration by the United States Environmental Protection Agency [51]. ⁶ Annual Average proposed by the European Commission [52]. ⁷ Raw water quality standard proposed by the Department of Environment Malaysia [53]. ⁸ HH filtration refers to household filtered water sample.

Similarly, the one-way ANOVA also found significant mean differences of dissolved Al ($F = 21.1$; $p = 6.1 \times 10^{-7}$) concentrations among all the eight river water sampling sites in the Langat River (Table S1). The multiple mean differences of dissolved Al concentrations through the post-hoc test of ANOVA also observed significant mean differences among all the water sampling sites between upstream and midstream areas e.g., Lolo vs. Cheras as well as Serai vs. Salak at the 0.05 confidence level. However, there was no significant difference between Pangsoon and Langat as well as Cheras stations, probably due to short distances among the water sampling sites. The mean differences of Al were also found to be non-significant among all the stations between the midstream and downstream areas because of adsorption of Al as well as the formation of authigenic aluminosilicate with the increase of salinity downstream. The Pearson correlation found a significant negative relationship between increase in salinity and decrease in Al concentration (-0.824 , $p = 0.01$) in the Langat River (Table S2).

Accordingly, the one-way ANOVA (Table S1) indicated that there were significant differences in the mean of As ($F = 3.2$; $p < 0.027$), Cd ($F = 4.4$; $p < 0.007$), Pb ($F = 29$; $p < 6.1 \times 10^{-8}$) among all the river water sampling points, except Cr (i.e., $F = 1$; $p = 0.491$). The one-way ANOVA of Cr concentrations along with the LSD post hoc test among all the river water sampling points was non-significant. It indicated that the Cr concentration in the Langat River was mostly from the natural weathering of oxisols of the serpentinite rock along the river basin. However, Cd concentrations observed significant multiple mean differences mostly in the downstream mainly because of the dissolution of Cd with increasing salinity (Pearson correlation -0.880 , $p = 0.002$) towards downstream (Table S2). Similarly, the significant mean differences of As mostly in the downstream areas of the Lan-

gat River indicated the attribution of As from anthropogenic sources, i.e., ETPs, agriculture, and as such extensively from the mid to downstream of the river. However, the multiple mean differences of dissolved Pb among almost all the sampling points from upstream to downstream of the Langat River found significant mean differences probably because of both natural and anthropogenic input of Pb into the river.

The Pearson correlations between the dissolved Al concentrations and physicochemical parameters explained the possible reason of declining Al concentration downstream of the Langat River (Table S2). A significant negative correlation was observed between the dissolved Al concentration and salinity (-0.824 , $p = 0.006$) as well as the negative correlation between Al concentration and pH from upstream to downstream in the Langat River. The significant negative correlation between Al concentration and salinity as well as conductivity might be a proper explanation for the adsorption and deposition processes of aluminosilicate formation to decrease the dissolved Al concentration in the river.

A negative correlation was also observed between As and salinity ($r = -0.402$) in the river; however, significant positive correlation is observed between As and DO ($r = 0.709$; $p = 0.024$). This indicates that the increasing salts also increased the mobility of As in the river as well as precipitation on the sediment towards downstream. Similarly, Cd has a significant negative correlation ($r = -0.880$, $p = 0.002$) with salinity in the Langat River, whereas it has a strong positive correlation with DO ($r = 0.821$, $p = 0.006$). Accordingly, DO has strong negative correlation with salinity ($r = -0.800$, $p = 0.009$) in the Langat River. Therefore, a higher Cd concentration is observed in the upstream of the Langat River than the downstream. The significant positive ($r = 0.728$, $p = 0.021$) and negative correlation ($r = -0.661$, $p = 0.037$) of Cr with DO and salinity, respectively, in the Langat River also indicated the precipitation of Cr towards downstream and higher concentration in the upstream. Moreover, the high concentration of DO in the upstream could also have affected the rate of oxidation of organic compounds and increased the release of Pb from the minerals [54] because both the Pb and DO concentrations in the upstream are higher than the downstream, and they have a significant positive correlation ($r = 0.612$, $p = 0.053$).

3.2. Mean Rank of Metal Concentrations in the River and Treated Water

The significant Mann-Whitney U mean test of dissolved Al (182; $p = 0.029$), As (151; $p = 0.004$), Cd (85; $p = 1 \times 10^{-5}$), Cr (100; $p = 5.2 \times 10^{-5}$), and Pb (82; $p = 7 \times 10^{-6}$) between river and treated water by water treatment plants (WTPs) specified the mean rank of all the WTPs in the Langat River Basin (Table 2). Although Levene's test for the equality of variance is not significant for As removal based on mean rank (2.265; $p = 0.139$; Table 2), the non-significant one-way ANOVA of absolute difference of mean for As ($F = 0.047$; $p = 0.829$; Table S3) between the river and treated water indicates the non-homogeneity of variance of As, and validates the Mann-Whitney U test for As removal by all the WTPs in the basin.

Table 2. Mean rank test of dissolved metals in river and treated water ($n = 24$).

Parameter	Location	Mean Rank	Mann-Whitney	p	Levene's Equality ¹	p
Al ($\mu\text{g/L}$)	River	28.92	182 **	0.029	29.829 ***	1.8×10^{-6}
	WTP	20.08				
As ($\mu\text{g/L}$)	River	30.21	151 ***	0.004	2.265	0.139
	WTP	18.79				
Cd ($\mu\text{g/L}$)	River	32.96	85 ***	1×10^{-5}	15.114 ***	3.2×10^{-4}
	WTP	16.04				
Cr ($\mu\text{g/L}$)	River	32.33	100 ***	5.2×10^{-5}	4.679 **	0.036
	WTP	16.67				
Pb ($\mu\text{g/L}$)	River	33.08	82 ***	7×10^{-6}	11.594 ***	0.001
	WTP	15.92				

Note: *** Significant at 0.01 level. ** Significant at 0.05 level. (two-tailed). ¹ Levene's Test for Equality of Variances.

Although the one-way ANOVA of absolute difference of mean for Al ($F = 6.614$, $p = 0.013$; Table S3) between the river and treated water was significant at 0.01 confidence level, the Levene's test for the equality of variance validates significant Al (29.829; $p = 1.8 \times 10^{-6}$; Table 2) concentration removal by the WTPs in the basin through the significant Mann-Whitney U test (182; $p = 0.029$; Table 2) at the 0.05 confidence level.

Among the investigated eight water treatment plants (WTPs) in the basin, the upstream Serai WTP had the highest Al concentration removal from treated water because of its highest mean rank 22.67 (Table 3) based on the dissolved Al concentration removal value by all the WTPs in the basin. Moreover, the dissolved Al concentration removal by all the WTP is statistically significant (Chi-Square 18.33; $p = 0.011$) at the 0.01 confidence level and the variability (i.e., effect size) accounted through the Kruskal Wallis Mean Rank Test for dissolved Al concentration removal was 79.7% (Table 3).

Table 3. Mean rank of metal concentration removal by WTPs (n = 24).

WTP	Mean Rank ¹				
	Al (µg/L)	As (µg/L)	Cd (µg/L)	Cr (µg/L)	Pb (µg/L)
Pangsoon	9.67	10.33	15.33	12.00	16.00
Lolo	20.33	8.33	9.00	18.67	22.67
Serai	22.67	19.33	22.00	21.00	20.33
Langat	15.33	9.00	16.00	4.67	13.33
Cheras	8.33	17.67	16.67	12.33	6.33
Bukit	2.67	18.33	4.67	7.00	6.00
Salak	11.67	11.00	6.67	12.33	3.33
Labu	9.33	6.00	9.67	12.00	12.00
Chi-Square	18.33 ***	11.17	14.61 **	12.15 *	20.53 ***
p value	0.011	0.131	0.041	0.096	0.005
Variability	0.797	0.486	0.635	0.528	0.893

Note: ¹ Kruskal Wallis Test. *** Significant at the 0.01 level. ** Significant at the 0.05 level. * Significant at the 0.10 level.

The one-way ANOVA of absolute difference of the dissolved Al mean in the treated water based on the value of concentration removal by the WTPs in the basin was significant ($F = 7251.1$; $p = 8.6 \times 10^{-27}$, Table S4). However, the significant differences in Al concentration removal (Chi-Square = 3.86; $p = 0.05$; Table 3) lie between the Pangsoon vs. Lolo as well as Serai WTP; Lolo vs. Langat; Lolo vs. Cheras; Lolo vs. Bukit; Lolo vs. Salak; Lolo vs. Labu; Serai vs. Langat; Serai vs. Cheras; Serai vs. Bukit; Serai vs. Salak; Serai vs. Labu; Langat vs. Cheras; Langat vs. Bukit, Cheras vs. Bukit; and Bukit vs. Salak WTPs, respectively. The effect size of Al removal variability through the Kruskal Wallis post-hoc test was also taken into account—77.2%. Therefore, it indicated that dissolved Al concentration removal from the treated water by plants in the Langat Basin is statistically different.

Similarly, the concentrations of dissolved Cd (Chi-Square 14.61, $p = 0.041$; Table 3) and Pb (Chi-Square 20.53, $p = 0.005$; Table 3) removal from treated water was statistically significant. Serai WTP in the basin has the higher mean rank 22 and 20.33 (Table 3) among the WTPs based on the concentration removal of Cd and Pb, respectively, in the basin through the Kruskal Wallis test. Besides, the effect sizes (i.e., variability) that accounted for the dissolved Cd and Pb concentration removal by WTPs were 63.5% and 89.3%, respectively (Table 3).

The one-way ANOVA of absolute difference of dissolved Cd ($F = 3.26$; $p = 0.024$; Table S4) and Pb ($F = 3.50$; $p = 0.018$; Table S4) concentrations' mean in the treated water was based on the concentration removal value by the WTPs; it was significant at the 0.05 confidence level. However, the post-hoc Kruskal Wallis test found significant dissolved Cd (Chi-Square = 3.86, $p < 0.05$; Table 4) concentration removal between the Pangsoon vs. Serai, Bukit, Salak and Labu WTPs; Serai vs. Langat, Cheras, Bukit, Salak and Labu WTPs; Langat vs. Bukit, Salak and Labu WTPs; Cheras vs. Bukit, Salak and Labu WTPs; and Bukit vs. Labu WTPs, respectively. Accordingly, significant dissolved Pb (Chi-Square = 3.86,

$p < 0.05$; Table 4) concentration removal were also observed between the Pangsoon vs. Lolo, Serai, Cheras, Bukit and Labu WTPs; Lolo vs. Langat, Cheras, Bukit, Salak and Labu WTPs; Serai vs. Langat, Cheras, Bukit, Salak, Labu WTPs; Langat vs. Salak WTPs; Cheras vs. Labu WTPs; Bukit vs. Labu WTPs; and Salak vs. Labu WTPs, respectively.

Table 4. Metals' mean rank differences based on concentration removal by the WTPs (n = 24).

(A) WTP	(B) WTP	Al ¹	As ¹	Cd ¹	Cr ¹	Pb ¹
Pangsoon	Lolo	3.86 *	0.05	0.43	0.43	3.86 *
	Serai	3.86 *	3.86 *	3.86 *	0.43	3.86 *
	Langat	0.43	0.43	0.43	0.43	0.43
	Cheras	0.05	1.19	0.05	0.05	3.86 *
	Bukit	2.33	3.86 *	3.86 *	0.43	3.86 *
	Salak	0.43	0.05	3.86 *	0.43	3.86 *
	Labu	0.05	1.19	3.86 *	0.05	3.86 *
Lolo	Serai	2.33	3.86 *	0.43	0.43	2.33
	Langat	3.86 *	0.05	0.43	3.86 *	3.86 *
	Cheras	3.86 *	2.33	0.43	0.43	3.86 *
	Bukit	3.86 *	1.19	0.43	3.86 *	3.86 *
	Salak	3.86 *	0.43	0.43	3.86 *	3.86 *
	Labu	3.86 *	0.05	0.43	2.33	3.86 *
Serai	Langat	3.86 *	1.19	3.86 *	3.86 *	3.86 *
	Cheras	3.86 *	0.43	3.86 *	3.86 *	3.86 *
	Bukit	3.86 *	0.43	3.86 *	3.86 *	3.86 *
	Salak	3.86 *	3.86 *	3.86 *	3.86 *	3.86 *
	Labu	3.86 *	3.86 *	3.86 *	3.86 *	3.86 *
Langat	Cheras	3.86 *	2.33	0.43	0.43	2.33
	Bukit	3.86 *	1.19	3.86 *	1.19	2.33
	Salak	2.33	0.43	3.86 *	3.86 *	3.86 *
	Labu	2.33	0.05	3.86 *	3.86 *	0.43
Cheras	Bukit	3.86 *	0.05	3.86 *	0.43	0.05
	Salak	1.19	0.43	3.86 *	0.43	1.19
	Labu	0.05	2.33	3.86 *	0.43	3.86 *
Bukit	Salak	3.86 *	3.86 *	1.19	3.86 *	1.19
	Labu	2.33	3.86 *	3.86 *	1.19	3.86 *
Salak	Labu	0.43	2.33	2.33	0.05	3.86 *

Note: * Significant at the 0.05 level; ¹ Chi-Square value of Mean Rank Test through Kruskal Wallis Post Hoc Test between (A) WTP vs. (B) WTP.

The dissolved As (Chi-Square = 11.17; $p = 0.131$) and Cr (Chi-square = 12.15; $p = 0.131$) concentration mean rank based on the concentration removal value by all the WTPs was not statistically significant (Table 3). However, the concentration of As removal between Pangsoon vs. Serai and Bukit WTPs; Lolo vs. Serai WTP; Serai vs. Salak and Labu WTPs; and Bukit vs. Salak and Labu WTPs was significant (Chi-Square = 3.857; $p < 0.05$; Table 4). Similarly, the dissolved concentration of Cr removal from treated water between Lolo vs. Langat, Bukit and Salak WTPs; Serai vs. Langat, Cheras, Bukit, Salak and Labu WTPs; Langat vs. Salak and Labu WTPs; and Bukit vs. Salak WTP was also statistically significant (Chi-Square = 3.857; $p < 0.05$; Table 4). The variabilities (effect size) for dissolved As and Cr based on its removal concentrations were 48.6% and 52.8%, respectively (Table 3). Hence, the different removal efficiencies as well as statistically significant differences in concentration removal of dissolved As and Cr in treated water indicated different metal removal efficiencies by the WTPs in the basin following the same traditional coagulation water treatment method.

The dissolved Al concentrations both in the treated water of Bukit and Cheras WTPs were higher than the raw water, probably because of excess application of aluminium sulphate ($Al_2(SO_4)_3$) in disinfection of treated water through coagulation due to heavily

polluted raw water, both with organic and inorganic pollutants. Both the Cheras and Bukit WTPs experienced several shutdown incidents in the last decade [9,55–59] mainly due to flash floods, along with runoff of mud, industrial effluent, etc. in the river because of colossal land clearance activities for palm oil plantations as well as industrialization in these areas [7,8]. In addition, the high pH at raw water intake points of both the Cheras 8.45 and Bukit 7.97 WTP indicate the alkaline condition of the raw water.

Therefore, the alkaline raw water along with high TDS of both the Cheras 137.63 mg/L and Bukit 106.5 mg/L WTP as well as high conductivity of both the Cheras 179.37 $\mu\text{S}/\text{cm}$ and Bukit 138.77 $\mu\text{S}/\text{cm}$ WTPs' raw water intake points compared to other stations in the Langat River indicates higher water pollution of these areas. Hence the salty raw water with low turbidity and high colour is challenging to treat [60], although in the optimum condition, the conventional method with alum coagulation can maintain Al concentration around 30 $\mu\text{g}/\text{L}$ in the treated water [61]. However, the Kruskal Wallis mean rank test specifies that Serai WTP has the highest concentration removal of dissolved Al (mean rank 22.67) in treated water, whereas Bukit WTP (mean rank 2.67) followed by the Cheras WTP (mean rank 8.33) has the lowest concentration removal of dissolved Al from the treated water in the Langat River Basin, Malaysia.

Similarly, Serai WTP observed the highest concentration removal of dissolved As, Cd, Cr, and Pb in treated water, which is supported by the highest Kruskal Wallis mean rank test (Table 3) for As (19.33), Cd (22.00), Cr (21), and Pb (20.33). Serai WTP is highest among all the WTPs in the basin and the mean ranks were based on the concentration removal values of these dissolved metals. However, the Kruskal Wallis test also observed lower mean rank based on the removal concentration values of these metals in the treated water of Lolo, Pangsoon, Langat, and Salak WTPs, i.e., As (8.33), Cd (15.33), Cr (4.67), and Pb (3.33), respectively (Table 3). The better metal concentration removal by the Serai WTP was also supported by the Kruskal Wallis mean rank test. This indicates that the Serai WTP maintained proper operation and management of conventional method to treat raw water. Therefore, the remaining WTPs in the basin requires proper management to remove these metals' concentration from treated water as long-term ingestion of these metals is an emerging health concern [1,9]. Hence, high-pressure reverse osmosis membrane technology could be used in the WTPs of the Langat River Basin, Malaysia, instead of the conventional method to remove metals because of its efficiency to remove trace metals >90% [62] from treated water.

3.3. Mean Rank of Metal Concentrations in Household Tap and Filtered Water

The Mann-Whitney U mean rank test of dissolved concentration for Al (Mann-Whitney = 682; $p = 0.004$), As (Mann-Whitney = 454; $p = 2 \times 10^{-6}$), Cd (Mann-Whitney = 668; $p = 0.003$), Cr (Mann-Whitney = 442; $p = 9.6 \times 10^{-7}$), and Pb (Mann-Whitney = 729; $p = 0.011$) found significant mean differences of these metals between household tap and filtered water (Table 5) in the Langat River Basin, Malaysia.

Table 5. Rank of trace metals in household tap and filtration water.

Parameter	Location	Mean Rank	Mann-Whitney	p	Levene's Equality ¹	p
Al ($\mu\text{g}/\text{L}$)	Tap	52.84	682 *	0.004	0.120	0.729
	Filtered	38.16				
As ($\mu\text{g}/\text{L}$)	Tap	57.91	454 *	2×10^{-6}	2.933	0.090
	Filtered	33.09				
Cd ($\mu\text{g}/\text{L}$)	Tap	53.16	668 *	0.003	0.658	0.419
	Filtered	37.84				
Cr ($\mu\text{g}/\text{L}$)	Tap	58.18	442 *	9.6×10^{-7}	9.008 *	0.003
	Filtered	32.82				
Pb ($\mu\text{g}/\text{L}$)	Tap	51.8	729 *	0.011	4.342 *	0.040
	Filtered	39.2				

Note: * Significant at 0.05 level; ¹ Levene's Test for Equality of Variances.

Moreover, the non-significant one-way ANOVA on absolute difference in means of metal concentration between household tap and filtered water for Al, As, Cd, and Cr validated the Mann-Whitney mean rank test of these metals. However, the significant ANOVA of Pb ($F = 6.412$; $p = 0.01$; Table S5) concentration removal between household tap and filtered water was justified by the Mann-Whitney test through the significant Levene's test ($t = 4.342$; $p = 0.04$; Table 5), which indicated the non-homogeneity of the variances of Pb concentration.

Carbon I ($262.54 \pm 8.03 \mu\text{g/L}$) and Carbon II ($128.13 \pm 32.17 \mu\text{g/L}$) household filtration system recorded a higher concentration of dissolved Al in the filtered water than the supply tap water $138.05 \pm 62.57 \mu\text{g/L}$ and $106.85 \pm 15.63 \mu\text{g/L}$, respectively (Figure 3). The prime reason for the high concentration of dissolved Al in the filtered water is the accumulation of Al on the cartridge of the filtration system along with lack of awareness of consumers to clean the filtration system regularly. Moreover, carbon filter (i.e., particulate filtration) can remove particle size of about $1 \mu\text{m}$, whereas Al ions may be $<0.0001 \mu\text{m}$ [63,64]. Furthermore, a relatively higher Al concentration by the Carbon I filter than the other filtration systems at household level in the Langat River Basin might be because of a broken ceramic part of the carbon filter.

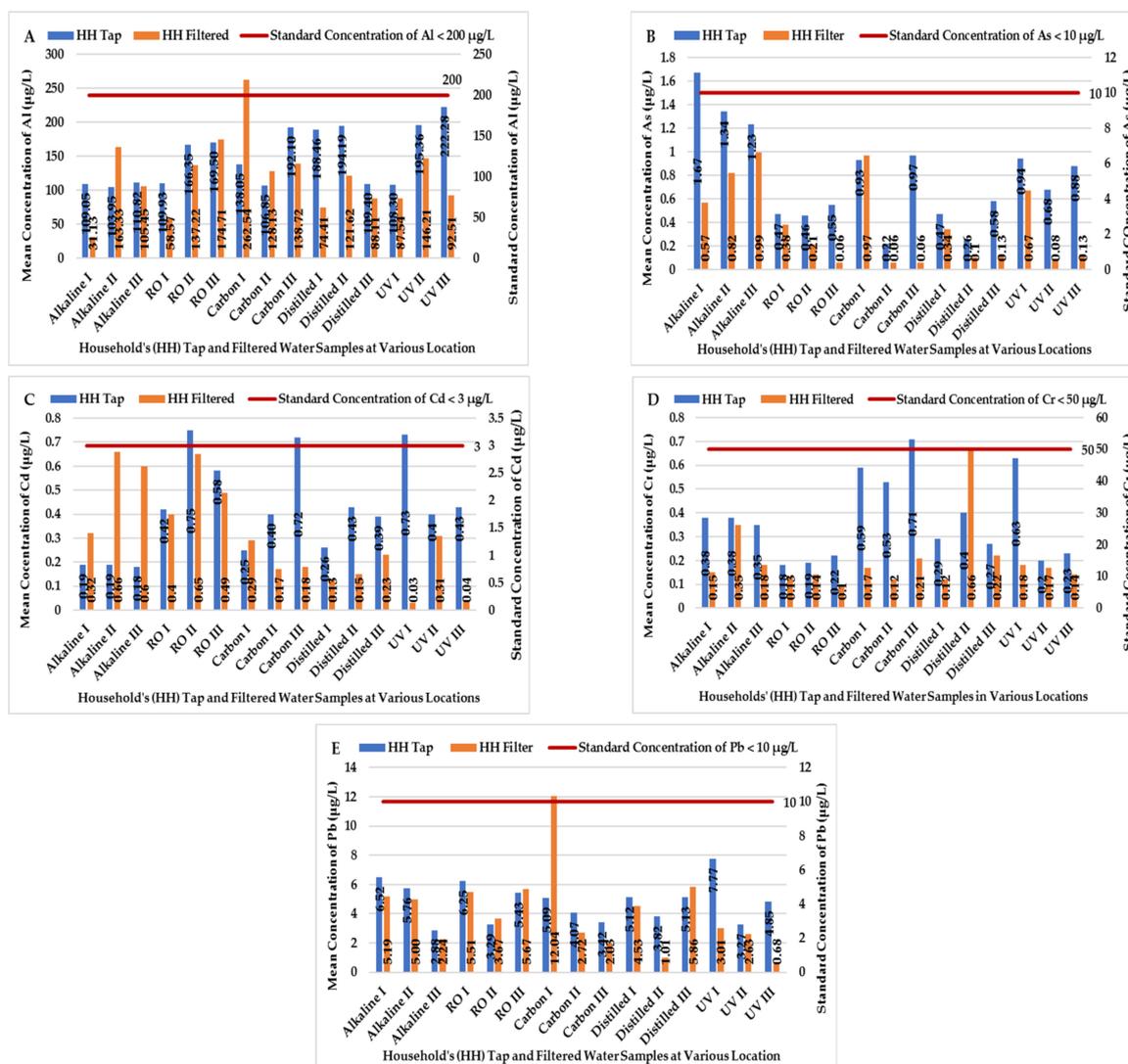


Figure 3. Al (A), As (B), Cd (C), Cr (D), and Pb (E) Concentrations ($\mu\text{g/L}$) in the household tap and filtered water at the Langat River Basin, Malaysia.

Accordingly, the concentrations of As ($0.97 \pm 0.26 \mu\text{g/L}$), Cd ($0.29 \pm 0.04 \mu\text{g/L}$), and Pb ($12.04 \pm 2.36 \mu\text{g/L}$) were also investigated to be higher in the filtered water of Carbon I filtration system than in the tap water $0.93 \pm 0.27 \mu\text{g/L}$, $0.25 \pm 0.04 \mu\text{g/L}$, and $5.09 \pm 1.71 \mu\text{g/L}$, respectively. However, Cr ($0.17 \pm 0.09 \mu\text{g/L}$) concentration removal by the Carbon I filter was better than that of other metals probably due to the adsorption of Cr in the filtration chamber, where water is stored before passing through the cartridge. Theoretically, the reverse osmosis (RO) filtration system has the highest efficiency to remove all types of metal ions, including Al ions (i.e., about $0.0001 \mu\text{m}$), but the higher concentration of Al in the filtered water by RO III ($174.71 \pm 11.86 \mu\text{g/L}$) was higher than the concentration of tap water ($169.50 \pm 7.31 \mu\text{g/L}$), probably due to the expiring of the cartridge. On the other hand, the Alkaline I filtration system showed the highest removal of dissolved Al concentration along with the distilled and UV household filtration systems. However, no removal of Al concentration by the Alkaline II filtration system might be due to the poor management of the filtration system, specifically irregular changes of the coral cartridge.

The highest concentration removal of dissolved As was investigated by the Distilled II and Distilled III filtration systems, mainly because of the volatile characteristic of As. Similarly, RO III and UV II also obtained better removal of As concentrations, probably because of particulate removal capacity by RO and UV— $0.0001 \mu\text{m}$ and $0.005 \mu\text{m}$, respectively [58]. All the five types of filtration systems (i.e., Alkaline, Reverse Osmosis (RO), Carbon, Ultraviolet (UV), and Distilled) in the Langat Basin experienced better removal of dissolved Cd and Cr concentrations mainly by the UV III and Carbon II filtration systems. However, the Alkaline II and Alkaline III filtration systems recorded slightly poor removal of dissolved Cd concentrations, which might be due to the poor cleaning of the filtration system along with the cartridge problems in manufacturing. Some studies have reported that if household water filtration systems are not cleaned properly, microorganisms can grow on the cartridges and these organisms have the capacity to retain metal ions [65–69]. Therefore, an unclean filtration system can also leach metals from the cartridge to enhance the concentration in the filtered water.

The dissolved Cr concentration removal by the Distilled II filtration system was recorded as zero, which might be due to the contamination of filtered water with the Cr through corrosion of the steel body of the distilled filter, along with the Cr attribution from the rust inside the filter. Similarly, the higher concentration of Pb in the filtered water by Distilled III over the supply water might be due to the rust inside the filtration system as well as corrosion of the galvanized iron pipe used in the filtration system. Moreover, the RO I and Alkaline I filtered water status indicates the contamination of filtered water with Pb from outside sources, apart from the leaching of Pb through cartridge, as the removal of Al, As, Cd, and Cr concentrations were better than Pb by these two filtration systems.

Even though the Kruskal-Wallis mean rank test was not found to be significant for the As (Chi-square = 22.957; $p = 0.061$; Table 6) concentration in the household filtered water among all the filtration systems at the Langat Basin, the Kruskal-Wallis post-hoc test found significant differences in mean ranks of dissolved As among the filtration systems. Similarly, the highest mean rank for the As removal was 39 by the Distilled II filter (Table 6), which was also similar with the highest As concentration removal by Distilled II, followed by RO II, along with the high mean rank 30.67. However, significant exact mean rank difference to remove As concentration exists between Alkaline III and Distilled II filtered water, RO I and Carbon II filtered water, RO I and Distilled II filtered water, RO I and UV III filtered water, Carbon I and Distilled III filtered water, Carbon I and UV II filtered water, etc. at the 0.05 confidence level with Chi-Square value 3.857. Hence, the dissolved As concentration removal by the household water filtration system may be ranked in the following order: Distilled > Alkaline > UV > RO > Carbon filter in the Langat Basin.

Table 6. Kruskal-Wallis mean rank test of metals in household filtered water in the Langat Basin (n = 45).

Filter Type	Kruskal-Wallis Mean Rank *				
	Al	As	Cd	Cr	Pb
Alkaline I	33.00	31.33	11.33	29.00	14.00
Alkaline II	16.33	26.00	3.33	31.33	21.67
Alkaline III	6.67	20.33	3.67	15.67	40.00
RO I	26.67	8.33	8.00	15.33	9.33
RO II	21.33	18.67	29.33	15.00	29.67
RO III	14.33	30.67	22.67	22.33	15.67
Carbon I	2.67	9.00	13.67	39.33	2.67
Carbon II	10.00	12.67	29.33	38.33	29.67
Carbon III	11.33	17.00	28.33	25.00	32.00
Distilled I	21.67	18.33	30.00	19.67	19.67
Distilled II	31.67	39.00	44.00	6.33	44.00
Distilled III	39.67	38.33	41.00	35.00	8.67
UV I	32.33	20.00	38.00	14.33	26.00
UV II	40.00	32.00	18.00	11.67	19.33
UV III	37.33	23.33	24.33	26.67	32.67
Chi-Square	37.52 **	22.96	41.45 **	25.01 *	34.25 **
<i>p</i>	0.001	0.06	0.0002	0.03	0.002
Variability	0.834	0.510	0.921	0.556	0.761

Note: ** Significant at the 0.01 Confidence Level. * Significant at the 0.05 Confidence Level.

The Kruskal-Wallis mean rank test (Table 6) found significant differences of mean for the dissolved concentration of Al (Chi-square = 37.52; $p = 0.001$), Cd (Chi-square = 41.45; $p = 0.0002$), Cr (Chi-square = 25.01; $p = 0.03$), and Pb (Chi-square = 34.25; $p = 0.002$) in filtered water among the household filtration systems at the basin. Moreover, the variability or effect size of the Kruskal-Wallis test mean it was well calculated—83.4% for Al, 51% for As, 92.1% for Cd, 55.6% for Cr, and 76.1% for Pb. According to the Kruskal-Wallis mean rank test, the highest mean rank was observed for the UV II (i.e., 40), followed by the Distilled III (i.e., 39.07) and UV III (i.e., 37.33) filter to remove dissolved Al concentration from the filtered water (Table 6). Moreover, the highest removal of dissolved Al concentration by Alkaline I was also supported by the high mean rank (i.e., 33, Table 6) of the Kruskal-Wallis test among the filtration systems. The Kruskal-Wallis post-hoc test also found significant mean rank differences of Al concentration in the filtration water exactly between Alkaline I and RO III, Alkaline I and Carbon I, Alkaline II and Distilled II, Alkaline II and UV II filter, and as such at the 0.05 confidence level with Chi-square 3.857. Therefore, dissolved Al concentration removal by household water filtration systems was determined by the following order: UV > Distilled > Alkaline > RO > Carbon filter.

Moreover, poor Al, As, Cd, and Pb concentrations' removal by the Carbon I filtration system was mostly due to the broken ceramic part of that filtration system and it was well determined through the comparatively lower mean rank of Al (i.e., 2.67), As (i.e., 9), Cd (i.e., 13.67), and Pb (i.e., 2.67) than the Carbon II and Carbon III filtered water (Table 6). Similarly, the highest mean ranks for dissolved Cd and Pb concentrations removal were recorded as 44 by the Distilled II filtered water, respectively. The higher mean rank of UV III, i.e., 24.33 (Table 6) also suggests better Cd concentration removal among the water filtration systems.

Additionally, the Kruskal-Wallis post-hoc test found significant mean rank differences of Cd and Pb concentrations between Alkaline I and Distilled II, Alkaline II and Carbon I, Alkaline II and Distilled II, Alkaline III and RO II, Alkaline II and UV I, Alkaline II and RO I, respectively (Table 6). Therefore, the Cd and Pb removal order by the household water filtration systems in the Langat River Basin were determined in the following order: Distilled > Alkaline > UV > Carbon > RO filtration systems, respectively (Table 7).

Table 7. Household water filtration order based on removal of metals in LRB.

Trace Metal	Household Water Filter Order ¹ Based on Removal of Trace Metals				
	1st	2nd	3rd	4th	5th
Al	UV	Distilled	Alkaline	RO	Carbon
As	Distilled	Alkaline	UV	RO	Carbon
Cd	Distilled	UV	Carbon	RO	Alkaline
Cr	Carbon	Distilled	Alkaline	UV	RO
Pb	Distilled	Alkaline	UV	Carbon	RO

Note: ¹ Based on Kruskal-Wallis Mean Rank Test.

Moreover, the Cr concentration removal was also supported by the highest mean rank for Carbon I (i.e., 39.33) followed by Carbon II (i.e., 38.33). The Kruskal-Wallis post-hoc test also found significant mean rank differences of Cr removal between the Alkaline I and RO I, Alkaline I and Distilled II, Alkaline I and UV II, Alkaline II and Carbon I filter, and as such at the 0.05 confidence level with the Chi-Square 3.857. Therefore, the Cr removal by the household filtration system may be ordered as Carbon > Distilled > Alkaline > UV > RO filter in the Langat River Basin, Malaysia.

Although theoretically reverse osmosis (RO) water filtration has the highest capacity to remove metal ions around 0.0001 μm , followed by ultraviolet (UV) around 0.005 μm and carbon filter about 1 μm ; however, metal ions can be <0.0002 μm [59]. Hence, the upgrading of household filtration system is a timely demand, observing the current contamination status of drinking water sources by development activities. Moreover, the poor removal status of the RO filtration system in the Langat Basin might be due to the irregular cleaning activities of the filtration system (i.e., not changing the expired cartridge) along with the problem of cartridge pore size during manufacturing. However, better metal removal by the distilled filter might be due to the volatile characteristics of the trace metals, e.g., arsenic, cadmium.

3.4. Metal Concentrations in Drinking Water Supply Chain

Overall, all the investigated metals' concentrations in household tap water (Al $148.31 \pm 46.56 \mu\text{g/L}$, As $0.78 \pm 0.53 \mu\text{g/L}$, Cd $0.423 \pm 0.19 \mu\text{g/L}$, Cr $0.37 \pm 0.21 \mu\text{g/L}$, Pb $4.84 \pm 1.87 \mu\text{g/L}$) were found to be a bit higher than the concentrations in treated water at the WTPs (Figure 4). This was a clear evidence of the contamination of supply water in the old long water distribution pipeline as well as unclean distribution fittings of supply water at the old buildings/apartments at the Langat River Basin. However, the determined concentrations of all the metals in the tap water were within the drinking water quality guidelines (Al < 200 $\mu\text{g/L}$, As < 10 $\mu\text{g/L}$, Cd < 3 $\mu\text{g/L}$, Cr < 50 $\mu\text{g/L}$ and Pb < 10 $\mu\text{g/L}$) set by the Ministry of Health Malaysia (MOH) and the World Health Organization (WHO).

The one-way ANOVA of the selected metals were Al ($F = 11.4$; $p = 1 \times 10^{-6}$), As ($F = 16.6$; $p = 3.02 \times 10^{-9}$), Cd ($F = 27.9$; $p = 4.62 \times 10^{-14}$), Cr ($F = 13.1$; $p = 1.56 \times 10^{-7}$), and Pb ($F = 20.2$; $p = 7.32 \times 10^{-11}$), which was found to be significant in the drinking water supply chain. The drinking water supply chain here refers to the river water, treated water by WTPs, and tap and filtered water from the same household. Moreover, the mean difference of these selected metals were significant at the 0.05 confidence level among all the four stages of drinking water supply chain at the Langat River Basin (Table S6).

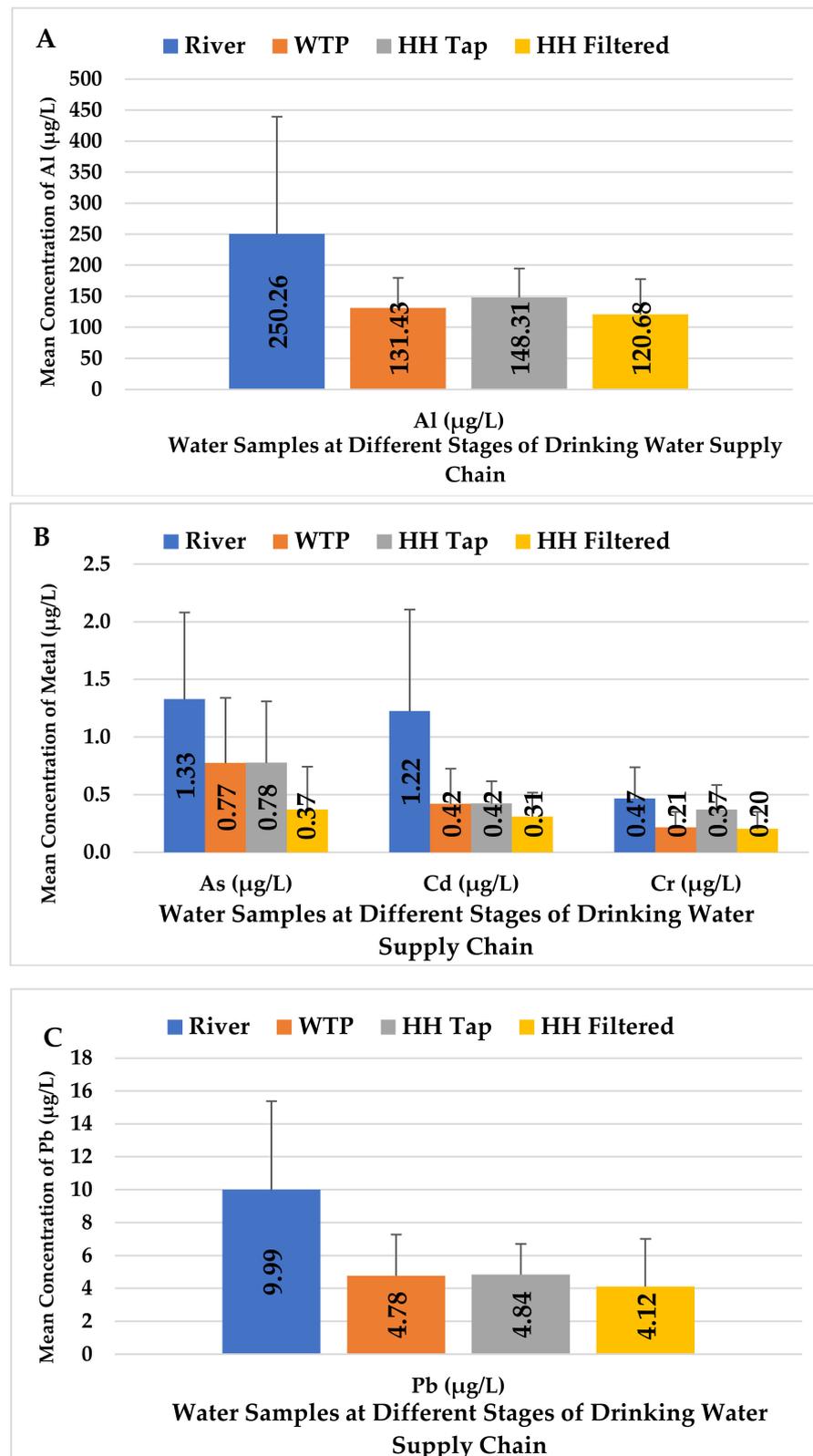


Figure 4. Al (A), As, Cd, Cr (B) and Pb (C) Concentrations (µg/L) in the drinking Water Supply Chain at the Langat River Basin Malaysia. **Note:** HH refers to household.

The Kruskal-Wallis mean rank test (Table 8) also found significant mean rank differences for dissolved Al (Chi-square = 13.991; $p = 0.003$) in the drinking water supply chain

at the basin in the following order: river water 89.79 > Household tap water 76.31 > treated water at the water treatment plant 63.88 > Household filtration water 54.87. Accordingly, the significant mean rank differences for As (Chi-square = 40.4; $p = 8.8 \times 10^{-9}$), Cd (Chi-square = 37.429; $p = 3.7 \times 10^{-8}$), Cr (Chi-square = 37.035; $p = 4.5 \times 10^{-8}$), and Pb (Chi-square = 35.064; $p = 1.2 \times 10^{-7}$) were also found in the following mean rank order of the concentration in the drinking water supply chain i.e., river > tap > WTP > household filtration system. Moreover, the variability or effect size of the Kruskal-Wallis tests were well accounted for: Al 10.2%, As 29.5%, Cd 27.3%, Cr 27% and Pb 25.6%.

Table 8. Mean rank of metal concentrations in the drinking water supply chain (n = 138).

Parameter	Water Supply Chain	Mean Rank	Chi-Square ¹	p Value	Variability
Al (µg/L)	River	89.79	13.991 *	0.003	0.102
	WTP	63.88			
	Household Tap	76.31			
	Household Filter	54.87			
As (µg/L)	River	102.88	40.4 *	8.8×10^{-9}	0.295
	WTP	73.88			
	Household Tap	77.16			
	Household Filter	41.71			
Cd (µg/L)	River	112.33	37.429 *	3.7×10^{-8}	0.273
	WTP	64.08			
	Household Tap	67.78			
	Household Filter	51.27			
Cr (µg/L)	River	98.13	37.035 *	4.5×10^{-8}	0.270
	WTP	52.50			
	Household Tap	85.18			
	Household Filter	47.62			
Pb (µg/L)	River	111.00	35.064 *	1.2×10^{-7}	0.256
	WTP	62.92			
	Household Tap	68.27			
	Household Filter	52.11			

Note: ¹ Kruskal Wallis Test; * Significant at 0.05 level.

Therefore, the Kruskal-Wallis test indicates significant mean rank differences in the concentrations of these metals among the four stages of the drinking water supply chain and any changes in the concentrations of these metals in any of the three stages, except in the household filtration system, will significantly influence the concentration in other stages of the drinking water supply chain, especially in the final filtered water by the household filtration system. The significant mean differences of all the investigated metals between the river and other stages of the drinking water supply chain indicated that the pollution of the river (i.e., mean rank is higher in every case, Table 9) was high because of both point and non-point sources of pollutions [70–72]. Therefore, the Kruskal-Wallis post-hoc test suggested management of the Langat River to reduce the water pollution. Hence, river pollution reduction will also reduce the concentration of metals in other stages of the drinking water supply chain.

Table 9. Mean rank of metals' concentrations in four stages of the drinking water supply chain (n = 138).

Parameter	Location	Mean Rank	Chi-Square ¹	p Value	Variability
Al (µg/L)	River	28.92	4.777 *	0.029	0.102
	WTP	20.08			
	Household Tap	40.71 31.96	2.979	0.084	0.044
As (µg/L)	Household Filter	45.17 29.58	9.450 *	0.002	0.139
	River WTP	30.21 18.79	7.980 *	0.005	0.170
	River Household Tap	45.08 29.62	9.296 *	0.002	0.137
Cd (µg/L)	River Household Filter	52.58 25.62	28.267 *	1.10×10^{-7}	0.416
	River WTP	32.96 16.04	17.521 *	2.80×10^{-5}	0.373
	River Household Tap	51.58 26.16	25.143 *	5.30×10^{-7}	0.370
Cr (µg/L)	River Household Filter	52.79 25.51	28.941 *	7.50×10^{-8}	0.419
	River WTP	32.33 16.67	15.027 *	1.10×10^{-4}	0.320
	River Household Tap	40.21 32.22	2.48	0.115	0.0359
Pb (µg/L)	River Household Filter	50.58 26.69	22.203 *	2.50×10^{-6}	0.3265
	River WTP	33.08 15.92	18.043 *	2.20×10^{-5}	0.3839
	River Household Tap	50.96 26.49	23.284 *	2.00×10^{-6}	0.3424
	River Household Filter	51.96 25.96	26.293 *	2.90×10^{-7}	0.3867

Note: ¹ Kruskal Wallis Test; * Significant at the 0.05 level.

Accordingly, the Kruskal-Wallis post-hoc test (Table 10) of metals' concentrations also found significant mean rank differences between river water and WTP, household tap as well as filtered water in the Langat River Basin, except river and tap water for Al and Cr, respectively, which might be due to the attribution of Al (Chi-square = 2.979; $p = 0.084$) and Cr (Chi-square = 2.48; $p = 0.115$) not only from natural sources but also from water treatment processes and drinking water distribution systems.

Table 10. Post-hoc mean rank of metals in the drinking water supply chain (n = 138).

Parameter	Location	Mean Rank	Chi-Square ¹	p Value	Variability
Al (µg/L)	WTP	30.292	2.027	0.155	0.030
	Household Tap	37.511			
	WTP	38.500	1.120	0.290	0.016
	Household Filter	33.133			
	Household Tap	52.844	7.113 *	0.008	0.105
	Household Filter	38.156			
As (µg/L)	WTP	33.833	0.124	0.724	0.002
	Household Tap	35.622			
	WTP	46.250	11.571 *	0.001	0.170
	Household Filter	29.000			
	Household Tap	57.911	20.312 *	6.6 × 10 ⁻⁶	0.299
	Household Filter	33.089			
Cd (µg/L)	WTP	33.833	0.124	0.724	0.002
	Household Tap	35.622			
	WTP	39.208	1.619	0.203	0.024
	Household Filter	32.756			
	Household Tap	52.000	5.571 *	0.018	0.082
	Household Filter	39.000			
Cr (µg/L)	WTP	24.167	10.73 *	0.001	0.158
	Household Tap	40.778			
	WTP	36.667	0.254	0.614	0.004
	Household Filter	34.111			
	Household Tap	58.178	21.195 *	4.1 × 10 ⁻⁶	0.312
	Household Filter	32.822			
Pb (µg/L)	WTP	33.167	0.307	0.579	0.005
	Household Tap	35.978			
	WTP	38.833	1.343	0.246	0.020
	Household Filter	32.956			
	Household Tap	51.800	5.234 *	0.022	0.077
	Household Filter	39.200			

Note: ¹ Kruskal Wallis Test; * Significant at the 0.05 level.

There are mean rank differences in the concentrations of trace metals between treated water and supply tap water at the household level mostly due to contamination in the water distribution system. However, the mean rank difference was not significant between WTP and tap water for the trace metals (i.e., Al, As, Cr, and Pb) except for Cr (Chi-square = 10.73; $p = 0.001$). The primary sources of Cr are natural, and the mean rank difference between WTP and supply water might be due to prolonged water stagnation periods in the drinking water pipeline system, along with the corrosion of steel pipe. Similarly, WTP vs. household filtration water was not significant for all the trace metals (i.e., Al, Cd, Cr, and Pb), except As (Chi-square = 11.571; $p = 0.001$), might be due to the adsorption tendency of As in the filtration system. However, the significant mean rank difference was observed for all the metals (i.e., Al, As, Cd, Cr, and Pb) between tap water and household filtration water at the basin. Therefore, the significant Kruskal-Wallis post-hoc test indicates both the filtration processes, i.e., at the WTP and household filtration systems, are essential to reduce the metal concentration in drinking water. Hence, the management of water treatment in WTP and filtration systems at the household level will significantly ensure safe drinking water quality.

The water treated by water treatment plants (WTPs) is transported a long distance through a pipeline in the Langat Basin to reach households. Thus, the contamination of treated water, especially with chemicals, was evident in the long transport system. Hence, better water technology is required to reduce chemical concentration in supply water at the household level. Otherwise, long-term ingestion of trace metals by drinking water could be harmful for human health, even though the dissolved concentration is within safe limits. Therefore, the management of drinking water source, i.e., reducing the pollution of the river as well as upgrading the drinking water treatment process as well as household water filtration technology, is obvious to obtain safe drinking water as well as to achieve sustainable development goals (SDGs) 6 of safe drinking water by 2030.

4. Conclusions

Safe drinking water supply at the household level in the Langat River Basin Malaysia largely depends on the management of pollution reduction in drinking water source (i.e., Langat River) that has been polluted both from point and non-point sources. Three stages of drinking water supply chain (i.e., water treatment plants, household tap and filtration system) at the basin was significantly influenced with metal concentration changes in the Langat River, such as concentration changes of Al (Chi-square = 13.991, $p = 0.003$), As (Chi-square = 40.4, $p = 8.8 \times 10^{-9}$), Cd (Chi-square = 37.429, $p = 3.7 \times 10^{-8}$), Cr (Chi-square = 37.035, $p = 4.5 \times 10^{-8}$), and Pb (Chi-square = 35.064, $p = 1.2 \times 10^{-7}$).

Significant mean differences in concentration of these metals were also determined between the river and treated water at the plants; however, each treatment plant had a different concentration removal capacity while following the same traditional coagulation method to treat raw water. Similarly, among the investigated household water filtration systems, the distilled system, followed by the alkaline and ultraviolet filtration systems, were found to be more efficient to remove metal concentration. Although theoretically, reverse osmosis (RO) is better than the other water filtration systems, at the Langat River Basin, the metals' concentration removal by RO is poor, probably due to the proper cleaning process not being maintained. Therefore, this study suggests examining the cartridge's efficiency along with pore size to identify the efficiency in removing metal concentrations. Moreover, a two-layer water filtration system could be introduced in the Langat River Basin to ensure safe drinking water supply at the household level. This two-layer filtration system should consist of a slow pond sand filtration system at the treatment plants and RO at the household kitchen tap, maintained by the same agency as the water-billing agency. However, people should use a proper distilled water filtration system at the household level for better drinking water quality in the basin until the two-layer water filtration system is implemented.

Apart from this, special measures should be taken by the agencies to have better collaboration in managing the pollution of the tropical Langat River in line with the Integrated River Basin Management. This could include the proactive and effective leadership roles of local authority through awareness raising, advocacy, and capacity building, while practising the PENTA-HELIX (i.e., government, business, academia, NGO and community) partnership model in decision-making processes.

Supplementary Materials: The following are available online at <https://www.mdpi.com/article/10.3390/w13081032/s1>. Table S1. One-way ANOVA of water quality parameters of the Langat River sampling points, Table S2. Correlations among water quality parameters in the Langat River (2015), Table S3. ANOVA of absolute mean difference of metal concentrations in river and treated water, Table S4. ANOVA of absolute mean difference of metals in treated water based on concentration removal by WTPs, Table S5. Absolute difference in metals' concentration between household tap and filtered water, Table S6. One-way ANOVA of metal concentrations in the drinking water supply chain.

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