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Effects of plumbing premise on the occurrences and variability of heavy metals in desalinated and blended tap water

Fayzul Kabir[a], Shakhawat Chowdhury[a,*], Mohammad Abu Jafar Mazumder[b], Md. Hasan Zahir[c], Khalid Alhooshani[b]

[a]*Department of Civil and Environmental Engineering, Water Research Group, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia, email: fkabirceesust@gmail.com (F. Kabir), Schowdhury@kfupm.edu.sa (S. Chowdhury),*
[b]*Department of Chemistry, King Fahd University of Petroleum and Minerals, Dhahran 31261, Saudi Arabia, email: jafar@kfupm.edu.sa (M.A.J. Mazumder), hooshani@kfupm.edu.sa (K. Alhooshani)*
[c]*Center of Research Excellence in Renewable Energy (CoRERE), Research Institute, King Fahd University of Petroleum & Minerals, (KFUPM), Dhahran 31261, Saudi Arabia, email: hzahir@kfupm.edu.sa (M.H. Zahir)*

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ABSTRACT

Chronic exposure to heavy metals in tap water has been linked to various health effects. The regulatory agencies often monitor heavy metals in water treatment plant and water distribution system (WDS) while people are exposed to water from tap in house. Stagnation of water in plumbing pipes (PP) and hot water tanks (HWT) prior to reaching the tap is likely to increase the concentrations of few heavy metals, which may exceed the regulatory guidelines. In this study, occurrences and variability of 17 heavy metals in water were investigated in WDS, PP and HWT through a 7-month sampling program. The water samples were collected and analyzed for 7 times a day on bi-weekly basis. The concentrations of heavy metals in HWT were 1.2–8.1 and 1.4–6.7 times the concentrations in WDS and PP respectively. Among the priority pollutants, concentrations of As, Cr, Cu, Pb, Hg, Ni and Zn were in the increasing order of WDS, PP and HWT. Higher concentrations of heavy metals in PP and HWT increase metal concentrations in tap water, which can pose elevated risk to humans. The concentrations of several heavy metals were seasonally variable with higher values in summer. The findings highlight the limitations of current sampling locations for regulatory compliance, exposure analysis and risk control from heavy metals in tap water.

Keywords: Desalinated and blended water; Heavy metals; Plumbing premise; Water stagnation; Water quality monitoring

1. Introduction

Heavy metals have high specific density (≥ 5g/cm^3) and long half-lives, which are also bio-accumulative [1].The residential or occupational exposure to thirty-five metals can have risk to humans while twenty-three are known as heavy metals [2]. The heavy metals have continuous impact on the environment (e.g., water, soil) and humans, which have been a concern for decades. These metals are often originated from rocks and concentrated in water, soil or in air through multiple transferring media. Drinking water is one of the potential sources for human exposure to heavy metals [3]. The atmospheric heavy metals are washed away by rainfall, which often reach to the potential sources of drinking water [4,5]. The manufacturing and industrial activities can also serve as the anthropogenic sources of heavy metals in water [6]. For example, the Kamioka Zinc Mine industry in Japan was responsible for cadmium contamination in the Jinzu River resulting in kidney problems among the surrounding populations [7]. The agricultural, industrial and mining activities around Sonora, Mexico caused elevated levels of Cu, As, Hg, Pb and Cd in groundwater [8]. Approximately 43% samples exceeded the action

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*Corresponding author.

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level for Pb (e.g., 15 µg/L) and 8.9% samples exceeded the guideline value of As [8–10]. The industrial, mining and agricultural activities in the surrounding areas polluted the drinking water sources[8].

Despite the efforts of removing heavy metals from water, several metals remain in drinking water in trace concentrations. Leaching of heavy metals from water distribution system (WDS) can increase the concentrations of these metals [11]. The diurnal and seasonal variability in water use, temperature variability and water chemistry have implications on the concentrations of these metals in drinking water. Changing the source of water supply from Lake Huron to more corrosive Flint River resulted in increased concentrations of metals (e.g., Cu and Pb) in the consumer tap in Flint, USA [12]. Application of ferric chloride (to reduce disinfectant byproduct formation) also increased the corrosivity of water. The corrosive water increased the dissolution of metals, which was responsible for higher levels of Pb than the WHO recommended value (e.g., 15 µg/L) in 20–32% homes [13].

In 2007, concentrations of Pb were reported to be higher than 10 µg/L in approximately 25% old houses in London, ON (Canada) [14]. Tamasi and Cini [15] reported higher levels of Fe, Cu, Zn and Pb at the dead end of WDS than in the origin (e.g. treatment plants) in Mount Amiata and downtown Siena), indicating leaching of metals from WDS. Concentrations of Cu were below the ICAP (inductively coupled argon plasma emission spectrometry)detection limit in treatment plants, which were increased up to 3000 µg/L at consumer taps following transportation through WDS [16]. In Dakhlia (Egypt),concentrations of Pb (0.58 ± 0.23 µg/L), As (0.29 ± 016 µg/L) and Zn (5.9 ± 2.3 µg/L) were higher in tap water than the bottled water [17]. Corrosion of plumbing pipe (PP) was the major source for contamination. Stagnation of water in WDS of Riyadh(Saudi Arabia) showed higher levels of Fe and Ni than the WHO recommended values in 2% samples [18]. Concentrations of Cu were increased up to 67% during transportation of water from first floor (Cu = 600 µg/L) to sixth floor (Cu = 1000 µg/L) of a six-story building in Dhahran (Saudi Arabia) [16]. A study on 127 randomly selected hot water tanks (HWT) in Dalmatian county (Croatia) indicated higher levels of Zn, Fe, Cu and Mn (averages: 180, 40, 10 and 4.96 µg/L respectively) in water. The mean temperature was 54°C and several samples exceeded the maximum contaminant levels [19].

The municipal water enters the plumbing premise of a house or a building from WDS. Based on the size of plumbing network, water may stay in PP and HWT for significant amount of time, which can increase the leaching of metals into water. A study on 12,000 school and workplace tap water in USA reported higher concentrations of Pb (e.g., 15 µg/L) in 17.2% first draw samples. However, flushing of water prior to use reduced Pb concentrations significantly [20]. Another study in Auckland (New Zealand) reported elevated concentrations of Pb (≥ 10 µg/L) in 90% unflushed tap water samples while more than 90% reduction was reported after flushing [21]. Lower alkalinity and lower hardness were likely to be responsible for the high aggressiveness of water[21]. In addition, water quality parameters (WQP) in plumbing premise, type of pipe materials and faucets can also affect the leaching of metals into water. Increased concentrations of few heavy metals were reported in water cooler compared to WDS [22,23].An study on 400 coolers in Riyadh reported the maximum concentrations of Fe (411.7 µg/L), Pb (59.82 µg/L) and Ni (228.68 µg/L) in cooler water while the feed water in WDS had much lower concentrations of these metals (20.43, 2.85 and 1.72 µg/L respectively) [24].

In context to Saudi Arabia, desalinated water is blended with treated groundwater, pH adjusted and chlorinated prior to supplying to the communities. The relatively acidic desalinated and blended water contains trace amount of heavy metals including Pb, Zn, Fe, Ca and Mg [16]. In addition, the desalinated and blended water is likely to increase the release of heavy metals from pipe materials and pipe coatings into water. Past studies reported increased levels of metals (e.g., Cu, Fe, Zn) from desalination plants to consumer taps where pipe lengths showed significant effects [16]. In addition, stabilization and pH adjustment of desalinated water increased the levels of As, Cd, Hg and Se in desalination plants [25]. Further, water spends significant time in plumbing premise prior to reaching the tap, which can increase heavy metal concentrations.

People consume water from the household taps, which are likely to have higher levels of heavy metals than the WDS. Some heavy metals, including Cd, As,Pb, Cr, Hg, Ni and Cu in drinking water can pose risk to humans [26,27]. The effects include mental disorder, brain damage, central nervous system failure, damage of DNA structure, skin infections, blood composition disorder, failure of lungs, heart, liver, kidneys and other major organs [27,28]. Chronic exposure to few heavy metals can induce cancer, hypertension, is chemic heart disease and allergies [29]. As an example, exposure to As in water caused cancer and skin damage, while Cd was reported to be responsible for cancer and kidney damage [27]. Some other effects, such as, liver and kidney function damage form Hg, high blood cholesterol and heart diseases from Sb, and gastrointestinal disorder from Cu were also reported [27]. In addition to health effects, higher concentrations of some metals (e.g., Ca, Fe, Mg, Al) can cause technical and aesthetic problems. For example, higher levels of Al (from fluoride additives) and iron imparted undesirable color, tastes and odors [30,31]. Higher concentrations of Zn (≥ 3 mg/L) developed the greasy films in boiled water, and imparted opalescent color and undesirable astringent taste in drinking water [32]. In general, the desalinated permeates contain lower levels of salts and the taste is less appealing, which can be improved through proper remineralization or blending. A case study in Barcelona with various proportions of blending reported radical changes in drinking water flavor where the samples with low salinity (TDS: 200–400 mg/L) were appreciated by the consumers [33–35]. The remineralization of desalinated water also showed the effects on the taste of water. Presence of cations (Na^+, Mg^{2+}, Ca^{2+}, K^+ etc.) affected the taste of drinking water while anions(Cl^-, NO_3, SO_4^{2-}, etc.)could adjust the taste intensity [34]. Alteration of mineral concentrations from<5 mg/L to 440 mg/L instigated the consumer perception from fresh to bitter, dry and metallic feelings [36]. Presence of Ca in the range of 42–117 mg/L was appreciated by the consumers while conductivity and Na showed negative outcomes [36,37].

To minimize the health risk from heavy metals in drinking water, several regulatory agencies have endorsed the maximum allowable levels for some of metals [10,27,38,39]. The regulatory agencies, health professionals and/or monitoring programs generally collect water samples from the WDS or water treatment plants to ensure regulatory compliance, and to perform exposure and risk analysis. The effects of plumbing premise have not been given much attention. In this study, implications of plumbing premise on the concentrations of heavy metals in tap water were investigated. The changes of heavy metal concentrations from WDS to PP and HWT were analyzed. The factors affecting the concentrations of heavy metals in PP and HWT were investigated. The diurnal and seasonal variability of these metals in tap water were assessed. The limitations of current regulatory compliance and sampling locations were highlighted to better protect human health.

2. **Materials and method**

2.1. Sampling program

The water samples were collected from a housing complex at King Fahd University of Petroleum and Minerals (KFUPM), Dhahran, Saudi Arabia. The samples were collected on bi-weekly basis for the period of 01/11/2015–29/05/2016. The samples were analyzed in duplicates from WDS, PP and HWT at seven different times in a day. The total number of samples collected and analyzed from WDS, PP and HWT were 42, 28 and 28 respectively. A typical plumbing premise is illustrated in Fig. 1 where water flows from WDS through PP and HWT to reach the consumer tap. The sampling strategy is presented in Fig. 2. The seven samples represented the diurnal variability of heavy metals in WDS, PP and HWT. The first sample (S1) was collected at late evening after 10 min free flow of tap water to represent WDS. The second sample (S2) was the first flush of water collected in the early morning of next day from the same tap. The S2 represented the effects of overnight (e.g., 8–12 h) stagnation of water in PP. During the night, there was no use of water from the tap. The third sample (S3) was collected in the morning from hot water tap to represent the HWT. The fourth sample (S4) was collected after 10 min free flow of cold water through the tap, which represented the water from WDS. The fifth and sixth samples (S5 and S6) were collected from the taps of cold and hot water respectively, in the afternoon following normal uses of water during the day. These samples represented PP and HWT respectively. The seventh sample (S7) was collected at afternoon after 10 min free flow through the tap to represent WDS. Each sample was collected in 100 mL glass bottle for metal analysis and 125 mL plastic bottles for analyzing the physical and chemical parameters.

2.2. *Laboratory analysis*

The heavy metals and WQP were analyzed in the laboratory. The water samples were collected and analyzed following the standard methods [40]. The total chlorine (TCl), free residual chlorine (FCl), UV absorbance at 254 nm (UV_{254}), total organic carbon (TOC), temperature, pH, turbidity and residence time were measured while temperature and pH were measured *in-situ*. The samples were transported to the laboratory in a cooler (4ºC). The TCl and FCl were measured by the HACH spectrophotometer (HACH DR 3900 model) following the HACH methods 8021 and 8167 respectively. TOC was measured using the Shimadzu TOC analyzer (Model: TOC-L-CSN) according to Standard Method 5310B [41]. The UV_{254} was measured using a spectrophotometer (Genesys 10 UV VIS model) at 254 nm with a 10-mm optical path quartz cell.

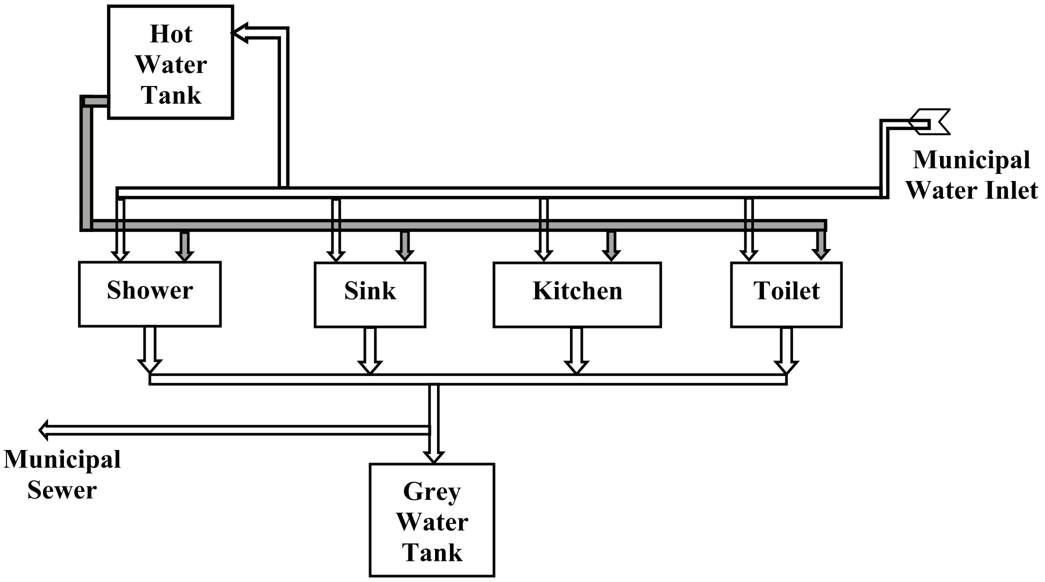

Fig. 1. Typical plumbing system of a house with water intake points.

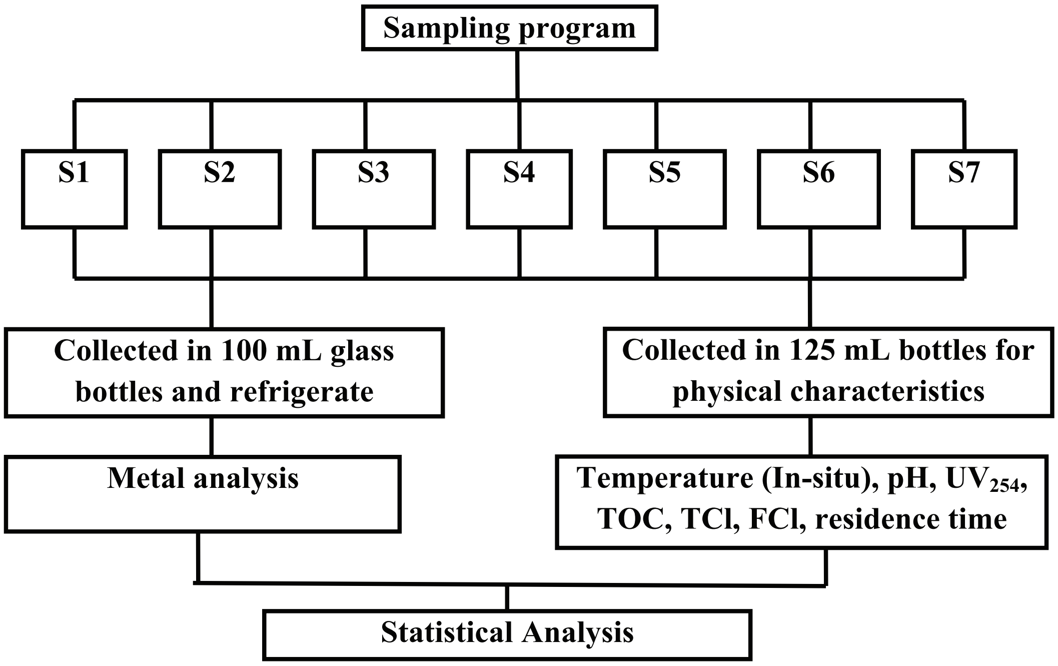

Fig. 2. Sampling program for data collection and methodology. S1: first sample collected at the late evening after 10 min of free flush in tap water; S2: first flush of water collected in the early morning of the next day from the same tap; S3: samples collected in the morning from hot water tap; S4: samples collected in the morning after 10 min of free flow of cold water through the tap; S5: samples collected from the cold water tap in the afternoon following normal uses; S6: samples collected from the hot water tap in the afternoon following normal uses; S7: samples collected at the afternoon after 10 minutes of free flow through the tap. TCl: Total chlorine; FCl: Free residual chlorine UV_{254}: UV absorbance at 254 nm; TOC: Total organic carbon.

Prior to measuring UV_{254}, samples were filtered through 0.45 µm membrane filters. The heavy metals were measured with the Inductively Coupled Plasma Mass Spectrometry(ICP-MS) (Thermo electron corporations, Model: ICP-MS XSERIES-II) following USEPA method-200.8 [42]. The data was processed by the Plasma Lab Windows platform software.

2.3. *Statistical analysis*

The experimental data for WDS, PP and HWT were analyzed using the JMP™ and Minitab™ statistical software. The data were summarized. These data were analyzed for trends, diurnal and seasonal variability, outliers and correlation structure. The data varaibility and statistical distributions were investigated. The coefficient of variation (*CV = std. dev./mean*) was determined and the outliers were identified. The data for WDS, PP and HWT were compared.

3. **Results**

3.1. *Overall statistics*

The WQP in WDS, PP and HWT for seven sampling scenarios are summarized in Table 1. The averages of temperature in WDS, PP and HWT were 27.2°C, 26.7°C and 60.4°C respectively with the ranges of 23–33°C, 22.5–32°C and 58–62°C respectively. The averages of FCl in WDS, PP and HWT were 0.04, 0.055 and 0.045 mg/L respectively, and the ranges were 0.01–0.21, 0.02–0.33and 0.01–0.17 mg/L respectively. The averages of pH in WDS, PP and HWT were 6.93, 6.83 and 6.77 respectively with the ranges of 6.37–8.2, 6.17–7.4 and 6.26–8.04 respectively. The averages of pH in these samples were below the neutral value, which were slightly acidic. The aggressivity index (AI) of water is a function of pH, alkalinity and calcium hardness, which is calculated as: AI = pH + C + D; where C = Logarithm (base 10) of calcium hardness (mg/L); and D = Logarithm (base 10) of alkalinity (mg/L). In AI, pH is likely to play the major role while the AI values <10, 10–12 and >12 indicate highly aggressive, moderately aggressive and non-aggressive water respectively [43]. The lower values of pH was likely to lower the AI and thus the corrosivity might be increased. The ranges of UV_{254} in WDS, PP and HWT were 0–0.005 cm^{-1}, 0–0.009 cm^{-1} and 0-0.009 cm^{-1} respectively, and the averages were 0.001 cm^{-1}, 0.0025 cm^{-1} and 0.002 cm^{-1} respectively. The averages of TOC in WDS, PP and HWT were 0.1, 0.19 and 0.22 mg/L respectively with the ranges of 0.01–0.56, 0.02–1.07and 0.07–0.55 mg/L respectively (Table 1).

Concentrations of heavy metals are summarized in Table 2. Few metals showed higher concentrations and higher variability in HWT than WDS and PP (e.g., Ca and Mg, Cr). The average concentrations of Cr in HWT (S3, S6), WDS (S1, S4, S7)and PP (S2, S5) were in the ranges of 0.68–0.79, 0.60–0.63 and 0.567–0.574 µg/L respectively. The corre-

Table 1
Water quality parameters at different sampling locations

Samples		Temp (°C)	FCl (mg/L)	TCl (mg/L)	pH	UV$_{254}$ (/cm)	TOC (mg/L)
S1	Mean	27.04	0.04	0.03	7.06	0.002	0.11
	Std. Dev	3.04	0.04	0.03	0.44	0.002	0.10
	Min	24.00	0.02	0.02	6.60	0.000	0.01
	Max	33.00	0.12	0.11	8.20	0.007	0.38
	CV	0.11	0.91	0.83	0.06	0.964	0.93
S2	Mean	25.91	0.06	0.05	6.81	0.003	0.20
	Std. Dev	1.81	0.09	0.08	0.38	0.002	0.10
	Min	23.00	0.02	0.02	6.17	0.000	0.12
	Max	29.00	0.33	0.29	7.40	0.009	0.44
	CV	0.07	1.52	1.50	0.06	0.847	0.50
S3	Mean	60.77	0.04	0.05	6.75	0.002	0.23
	Std. Dev	1.11	0.04	0.07	0.47	0.001	0.11
	Min	58.00	0.01	0.02	6.26	0.000	0.09
	Max	62.00	0.17	0.26	8.04	0.005	0.55
	CV	0.02	1.16	1.37	0.07	0.652	0.49
S4	Mean	27.23	0.04	0.05	6.93	0.001	0.07
	Std. Dev	2.82	0.04	0.10	0.29	0.002	0.04
	Min	24.00	0.01	0.01	6.53	0.000	0.01
	Max	32.00	0.15	0.37	7.67	0.005	0.13
	CV	0.10	1.06	1.85	0.04	1.303	0.57
S5	Mean	27.50	0.05	0.03	6.84	0.002	0.17
	Std. Dev	2.92	0.07	0.02	0.43	0.002	0.28
	Min	22.50	0.02	0.02	6.33	0.000	0.02
	Max	32.00	0.22	0.10	7.87	0.005	1.07
	CV	0.11	1.26	0.77	0.06	0.983	1.62
S6	Mean	60.05	0.05	0.07	6.79	0.002	0.21
	Std. Dev	1.09	0.03	0.07	0.47	0.003	0.09
	Min	58.00	0.01	0.01	6.31	0.000	0.07
	Max	61.00	0.09	0.25	8.02	0.009	0.40
	CV	0.02	0.58	0.96	0.07	1.188	0.45
S7	Mean	27.41	0.04	0.04	6.79	0.001	0.11
	Std. Dev	3.33	0.05	0.06	0.29	0.001	0.15
	Min	23.00	0.02	0.02	6.37	0.000	0.01
	Max	33.00	0.21	0.24	7.19	0.004	0.56
	CV	0.12	1.45	1.65	0.04	0.957	1.30

S1: first sample collected at the late evening after 10 min of free flush in tap water; S2: first flush of water collected in the early morning of the next day from the same tap; S3: samples collected in the morning from hot water tap; S4: samples collected in the morning after 10 min of free flow of cold water through the tap; S5: samples collected from the cold water tap in the afternoon following normal uses; S6: samples collected from the hot water tap in the afternoon following normal uses; S7: samples collected at the afternoon after 10 minutes of free flow through the tap; Std. Dev: Standard deviation; Min: Minimum; Max: Maximum; CV: Coefficient of variation; TCl: Total chlorine; FCl: Free residual chlorine; UV$_{254}$: UV absorbance at 254 nm; TOC: Total organic carbon .

sponding CV were in the ranges of 0.46–0.48, 0.35–0.36 and 0.40–0.42 respectively. Higher averages of Cr in HWT can be partially attributed to temperature driven reaction with the wall of HWT and deposits in the tank [44]. The average concentrations of Fe, Zn, Br, Sr, Ba and Co were higher in HWT than WDS. Average concentrations of Cu, As, Hg and Ni in PP (S2, S5) were higher than WDS (S1, S4, S7) due to overnight stagnation of water in PP. In PP, averages of Cu, Hg and Ni were 4.3, 2.6 and 28.9 μg/L respectively while in WDS, these were 0.4, 0.8 and 1.6 μg/L respectively. In PP, ranges of Cu, Hg and Ni were 0.02–38.28, 0.08–25.40 and 0.12–113.7 μg/L respectively while in WDS, these were 0.04–4.76, 0.05–14.87 and 0.15–59.22 μg/L respectively. Further details on other metals can be found in Table 2.

Table 2
Metal concentrations at different sampling points (in µg/L)

Samples		Na	Mg	Ca	V	Cr	Mn	Fe	Cu	Zn	As	Sr	Mo	Ba	Hg	Pb	Ni	Co
S1	Mean	186229	5487	9570	4.58	0.60	1.04	60.89	0.43	6.72	1.09	324.9	2.36	74	2.00	3.95	1.79	0.19
	Std. Dev	291009	10464	17290	7.38	0.22	1.22	28.83	0.69	12.22	1.48	623.3	3.94	987	4.03	3.23	2.05	0.07
	Min	248	21.51	38.65	0.03	0.23	0.02	8.01	0.04	0.07	0.09	1.7	0.01	1.52	0.09	0.29	0.18	0.04
	Max	788040	27219	45100	21.05	0.98	3.45	99.70	2.67	45.32	4.16	1649	13.16	3394	14.87	8.90	5.40	0.27
	CV	1.56	1.91	1.81	1.61	0.36	1.17	0.47	1.62	1.82	1.35	1.9	1.67	1.33	2.01	0.82	1.15	0.39
S2	Mean	109405	2764	5886	5.05	0.57	2.70	51.23	4.30	398	0.88	170.9	34.94	1241	2.64	4.08	28.89	1.09
	Std. Dev	202231	7029	11823	7.90	0.23	5.06	29.40	10.9	931	1.14	432.2	72.14	2200	6.89	3.34	46.74	1.38
	Min	266	31.68	60.11	0.03	0.18	0.03	3.83	0.04	0.72	0.06	2.5	0.01	1.17	0.08	0.13	0.12	0.06
	Max	747648	24921	40506	21.64	0.92	18.6	93.14	38.3	3032	3.88	1564	209.8	7525	25.40	10.57	113.7	3.89
	CV	1.85	2.54	2.01	1.56	0.40	1.87	0.57	2.53	2.34	1.29	2.5	2.06	1.77	2.61	0.82	1.62	1.26
S3	Mean	257644	13910	32144	5.62	0.68	1.06	68.51	0.79	201	1.69	723.4	1.78	2772	1.70	3.67	14.23	0.31
	Std. Dev	331341	19473	57468	7.56	0.33	1.37	23.86	1.14	642	1.62	949.9	2.38	4379	2.31	3.61	29.73	0.35
	Min	4000	143	493	0.04	0.12	0.03	36.24	0.01	0.22	0.22	8.2	0.01	1.42	0.12	0.16	0.12	0.01
	Max	994600	67050	213600	23.98	1.29	4.23	114.1	3.73	2334	4.91	3023	8.97	15010	7.48	9.21	87.55	0.85
	CV	1.29	1.40	1.79	1.35	0.48	1.29	0.35	1.44	3.19	0.96	1.3	1.34	1.58	1.36	0.98	2.09	1.12
S4	Mean	126848	3403	6982	5.20	0.63	0.69	54.45	0.92	9.47	0.86	193	35.90	1248	0.81	3.60	1.58	0.45
	Std. Dev	174413	6217	11957	7.21	0.23	0.88	22.19	1.31	15.87	0.96	351.6	83.60	2369	0.92	3.15	1.81	0.66
	Min	2863	41	120.2	0.04	0.33	0.02	14.81	0.05	0.57	0.08	3.7	0.02	0.76	0.05	0.18	0.16	0.01
	Max	521444	16824	30545	19.61	1.03	2.78	93.57	4.76	55.99	3.09	991.8	239.8	7960	3.10	9.02	4.51	1.99
	CV	1.37	1.83	1.71	1.39	0.36	1.28	0.41	1.42	1.68	1.11	1.8	2.33	1.90	1.14	0.87	1.15	1.45
S5	Mean	90651	1964	4258	3.61	0.57	1.12	54.57	2.00	10.42	1.05	103.5	26.04	1931	1.50	4.25	11.11	0.42
	Std. Dev	126509	4644	8248	4.31	0.24	1.01	20.26	5.05	12.49	1.61	222.8	47.39	3500	1.77	3.29	28.93	0.40
	Min	2479	44.22	119.6	0.03	0.22	0.01	28.22	0.02	0.48	0.07	3.5	0.05	2.96	0.18	0.17	0.27	0.02
	Max	471086	15915	26934	10.57	0.99	2.48	91.11	18.7	47.6	5.78	770.8	122.8	11160	4.51	10.90	82.66	1.00
	CV	1.40	2.36	1.94	1.19	0.42	0.90	0.37	2.52	1.20	1.53	2.2	1.82	1.81	1.18	0.78	2.60	0.97
S6	Mean	352681	18042	39152	9.97	0.79	0.85	75.40	1.16	3.62	2.39	969.7	19.12	3998	1.74	3.74	4.62	0.30
	Std. Dev	366156	17405	49941	9.63	0.36	1.16	25.25	1.28	4.02	1.84	842.9	44.58	6897	2.93	3.81	2.13	0.16
	Min	5482	148.61	199.9	0.15	0.19	0.01	11.40	0.06	0.10	0.16	9.2	0.02	0.59	0.03	0.14	0.12	0.05
	Max	895500	64280	195300	24.59	1.34	3.92	113.3	3.47	15.14	4.73	2726	159.6	24180	10.62	10.58	6.60	0.59
	CV	1.04	0.96	1.28	0.97	0.46	1.37	0.33	1.10	1.11	0.77	0.9	2.33	1.72	1.68	1.02	0.46	0.55
S7	Mean	145387	3287	6195	5.64	0.63	1.45	61.63	1.12	114.2	0.96	190.8	33.21	1365	1.79	3.56	8.79	0.76
	Std. Dev	207850	7503	12326	7.78	0.22	1.39	21.38	1.25	383.7	1.11	429.5	81.38	2486	2.33	3.24	20.45	1.17
	Min	3155	44	165.2	0.02	0.27	0.11	17.84	0.05	0.29	0.08	3.5	0.01	2.22	0.10	0.18	0.15	0.01
	Max	689535	23692	39462	22.28	0.99	3.22	94.19	4.04	1390	3.64	1443	272.5	8877	6.68	8.90	59.22	3.00
	CV	1.43	2.28	1.99	1.38	0.35	0.96	0.35	1.12	3.36	1.16	2.3	2.45	1.82	1.30	0.91	2.33	1.54

S1: WDS samples after last use of water in the late evening; S2: cold water samples in the early morning prior to the first water use;S3: hot water samples in the morning; S4: WDS samples in themorning; S5: samples collected from the cold water tap in the afternoon following normal uses; S6: hotwater samples in the afternoon; S7: WDS samples in the afternoon; Std. Dev: Standard deviation; Min: Minimum; Max: Maximum; CV: Coefficient of variation

3.2. *Variability in WDS, PP and HWT*

Among these metals (Table 2), elevated levels of As, Pb, Cr, Hg, Ni and Cu were reported to pose risk to humans [26,27]. The concentrations of Cr, Mn, Fe, Cu, Zn, As, Hg, Pb, Mg and Ni are shown in Fig. 3. Concentrations of Pb and Cr showed no outlier while other metals had several outliers (Fig. 3). The box plots for Fe, Ni and Zn showed 1, 3 and 4 data points, respectively, as outliers while Mg, Hg, As and Cu had more outliers, indicating higher variability. For example, averages of Cu in S1–S7 were less than 2 µg/L while the maximum was 38 µg/L. The wide interquartile ranges of Pb, Fe and Cr in S1–S7 indicated higher variability (Fig. 3). In HWT (S3), the interquartile ranges of Pb and Cr were 0.2–7.0 and 0.4–0.95 µg/L respectively (Fig. 3). Similar results were noted for Mg and As in HWT (Fig. 3). In contrast, smaller interquartile ranges of Cu, Zn and Ni indicated data closeness to averages (Fig. 3). It is to be noted that outliers are often discarded to achieve statistical consistency and modeling works. However, these are important for human exposure and risk analysis due to high concentrations and health concerns [9].

Fig. 4 shows the variability of metal concentrations in WDS, PP and HWT. Concentrations of few metals were higher in HWT than WDS and PP. Concentrations of Fe and As in WDS were 60 and 0.5 µg/L respectively, which were increased to 72 and 1.0 µg/L in HWT respectively. The averages and interquartile ranges of Cr, Mg, Cu, Hg and Ni in HWT were higher than WDS (Fig. 4). The sediments, precipitates and corroded metals were likely to be deposited in HWT, which might be partially responsible for higher concentrations in HWT [44]. In addition, higher temperature might have accelerated the release of metals during stagnation in HWT. The average concentrations of Mn, Pb, Cu and Hg were higher in PP than WDS. Average of Mn in PP (1.91 µg/L) was almost 1.8 times the average in WDS (1.05 µg/L). Average of Pb and Hg were 3.7 and 0.9 µg/Lin WDS respectively, which were increased to 4.2 and 1.1 µg/L in PP respectively (Fig. 4). Further details can be found in Fig. 4.

3.3. *Diurnal variability*

The diurnal variability of metal concentrations are presented in Fig. 5. The S1, S4 and S7 represent WDS in the evening, early morning and afternoon of the day. The S2 and S5 represent PP in early morning and afternoon respectively while S3 and S6 represent HWT in early morning and afternoon respectively. The time of sampling showed variable effects on concentrations of metals (Fig. 5). The diurnal variability was also tested for equality of medians through the Kruskal-Wallis and Mann–Whitney tests (non-parametric) due to non-Gaussian distributions of data. The a-value was set to 0.05, meaning similarity of medians when $p > 0.05$ and different when$p \leq 0.05$.

Average concentrations of Cr in WDS (S1, S4, S7) were similar (Fig. 5) with medians in the range of 0.6–0.63 µg/L (p = 0.9). In PP (S2, S5), averages of Cr were similar with medians in the range of 0.5–0.6 µg/L (p = 0.92). Averages of Cr in HWT were different with medians of 0.62 and 0.85 µg/Lin S3 and S6 respectively, indicating effects of sampling time. Average concentrations of As in WDS (S1, S4, S7) were similar with medians of 0.86–1.09 µg/L (p = 0.88). In PP (S2,S5), averages of As were also similar (medians: 0.88–1.05 µg/L; p = 0.8). However, averages of As in HWT were different with medians of 1.69 and 2.39 µg/L in S3 and S6 respectively, indicating the effects of sampling time. The possible explanation for release of Cr may include the Ni-Cr heater alloys and stainless steel in HWT from which Cr might have been released while As could have entered from the wall of HWT due to scale formation [45]. The scales were sloughed off due to use of hot water and As was released into water [46].

Averages of Pb in WDS (S1, S4, S7) were similar with medians of 3.56–3.95 µg/L (p = 0.92). In PP (S2, S5), these were similar (medians: 4.08–4.25 µg/L; p = 0.96). In HWT (S3, S6), averages of Pb were also similar with medians of 3.67–3.74 µg/L (p = 0.92). The averages of Hg in WDS (S1, S4, S7) showed variability (Fig. 5) while medians were in the range of 0.81–2 µg/L. In PP, average and median of Hg were also different (median: S2 = 2.64 µg/L; S5 = 1.50 µg/L). In HWT, average and median were consistent (medians:1.7 –1.74 µg/L; p = 0.88). The mechanisms for such variability are yet to be established.

3.4. *Seasonal variability*

Fig. 6 shows the seasonal variability of several metals in WDS, PP and HWT. The sampling duration (01/11/2015 –29/05/2016) was divided into three seasons: P1, P2 and P3 representing 01/11/2015–31/12/2015, 01/01/2016–31/03/2016 and 01/04/2016–29/05/2016 respectively. In WDS, average temperatures of water during P1, P2 and P3 were 26.31, 25.38 and 30.67°C respectively. During P3, averages of Pb, Fe, Mn and Cr were 4.78, 72.01, 1.29 and 0.71 µg/L respectively. During P2, these were 2.6, 54.18, 1.16 and 0.61 µg/L respectively. Higher concentrations of Pb, Fe, Mn and Cr during P3 might be due to higher temperature and increased water demand, which could have increased metal release from WDS [44,47]. However, averages of Mg, Cu, Zn, As and Hg were higher during P2 than P3 (Fig. 6). During P1, averages of Pb, Hg and Cu were 4.49, 3.34 and 1.31 µg/L respectively, which were 2.6, 1.05 and 0.71 µg/L during P2 respectively. The lower values in P2 might be due to low temperature. During P1, averages of Cr, Mn, Fe, Zn and As were 0.52, 0.53, 51.25, 15.76 and 0.39 µg/L respectively, which were 0.61, 1.16, 54.18, 80.68 and 1.34 µg/L respectively in P2.

In PP, averages of Mg, Cr, Fe, As and Pb were higher during P3 than P2 (Fig. 6). During P2, averages were 1542, 0.55, 46.88, 0.72 and 2.87 µg/L respectively while in P3, these were 5176, 0.67, 67.67, 0.89 and 5.72 µg/L respectively. Higher concentrations in P3 might be due to accumulation and/or release of metals in PP [46]. Higher temperature and consumption rate during P3 might have increased the dissolution of metals in stagnant water (e.g., PP). However, averages of Mn, Cu, Zn and Hg were higher in P2 than P3. During P2, the averages were 1.73, 4.87, 417.36 and 1.27 µg/L respectively while in P3, these were 0.95, 0.39, 4.37 and 0.67 µg/L respectively. The averages of Cr, Fe and Cu during P1 and P2 did not show significant variability. The averages of Mn, As, Hg and Pb were higher during P1 than P2 while the averages of Mg and Zn were lower during P1 than P2 (Fig. 6).

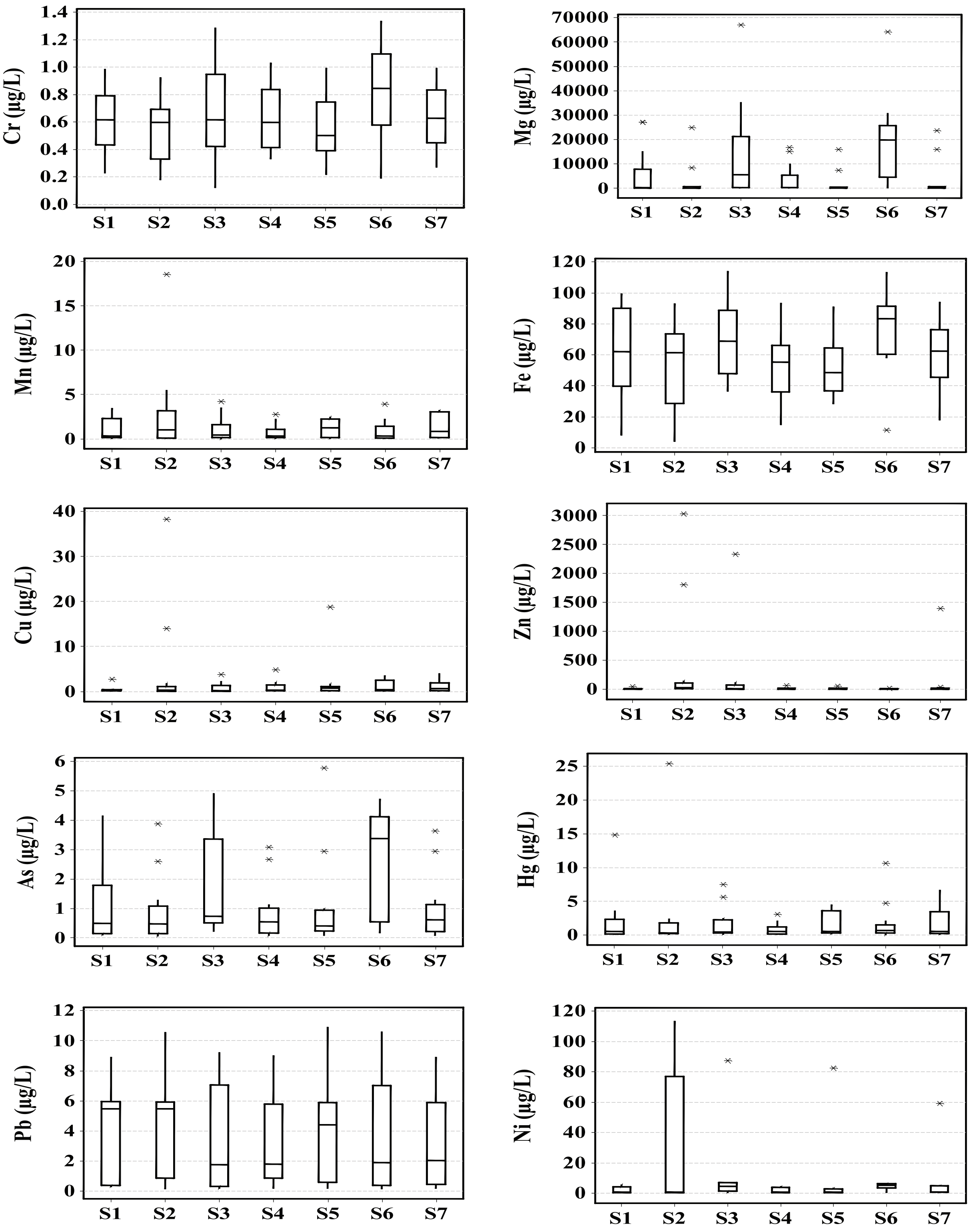

Fig. 3. Box plot of metal variability in different sampling scenarios. Bottom and top of the boxes represent the first quartile (Q1) and the third quartile (Q3) respectively. The lower whisker extends up to [Q1 – 1.5*(Q3–Q1)] and upper whisker extends up to [Q3+1.5*(Q3–Q1)]. The horizontal lines within the box represent the medians of the data. Value larger than Q3 or smaller than Q1 by at least 1.5 times the interquartile range (IQR) are outliers. S1: WDS samples after last use of water in the late evening; S2: cold water samples in the early morning prior to the first water use; S3: hot water samples in the morning; S4: WDS samples in the morning; S5: samples collected from the cold water tap in the afternoon following normal uses; S6: hot water samples in the afternoon; S7: WDS samples in the afternoon.

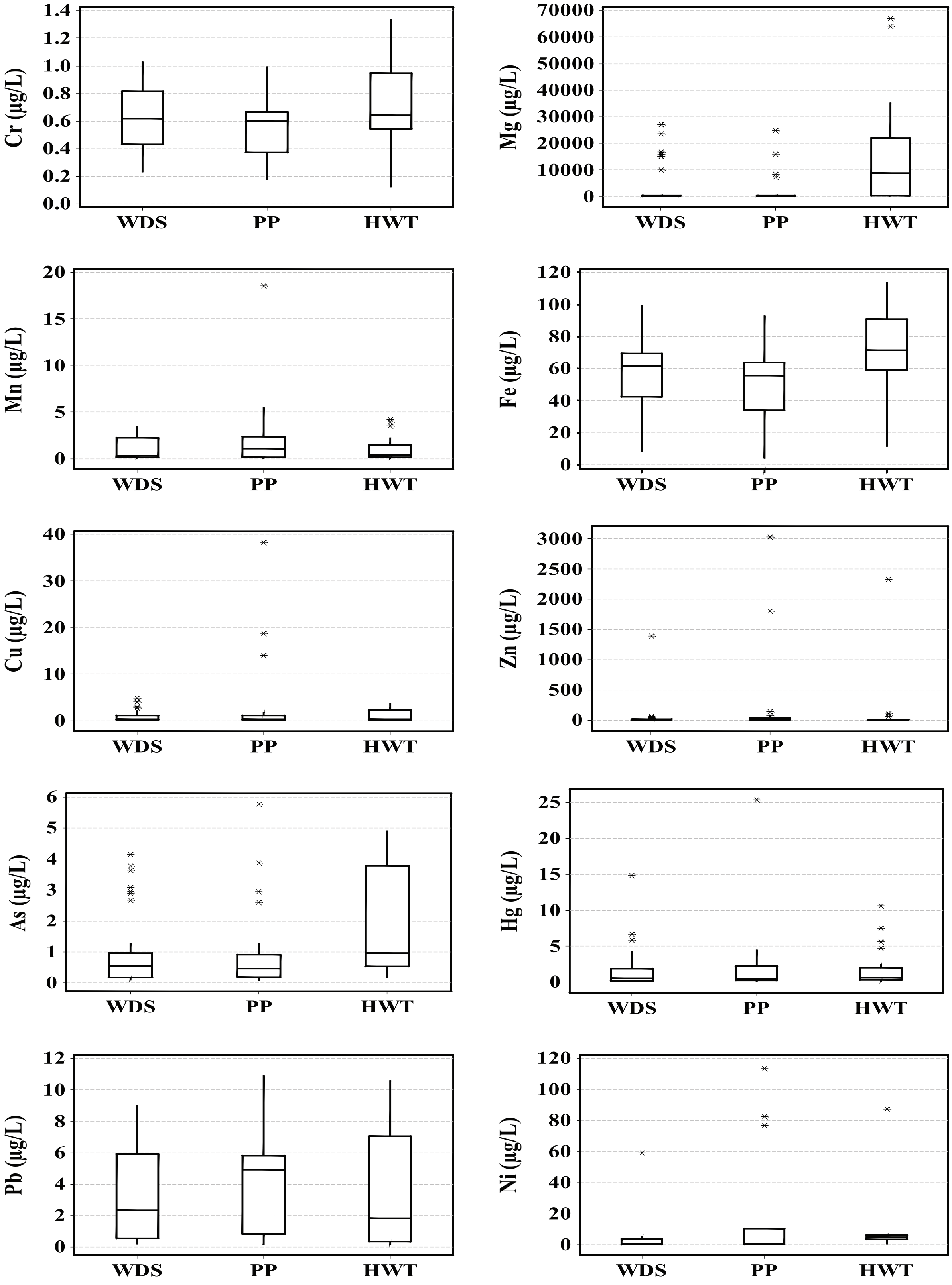

Fig. 4. Box plot of metal variability in WDS, PP and HWT. Bottom and top of the boxes represent the first quartile (Q1) and the third quartile (Q3) respectively. The lower whisker extends up to [Q1 – 1.5*(Q3–Q1)] and upper whisker extends up to [Q3+1.5*(Q3–Q1)]. The horizontal lines within the box represent the medians of the data. Value larger than Q3 or smaller than Q1 by at least 1.5 times the interquartile range (IQR) are outliers. WDS: water distribution system; PP: plumbing pipes; HWT: hot water tanks.

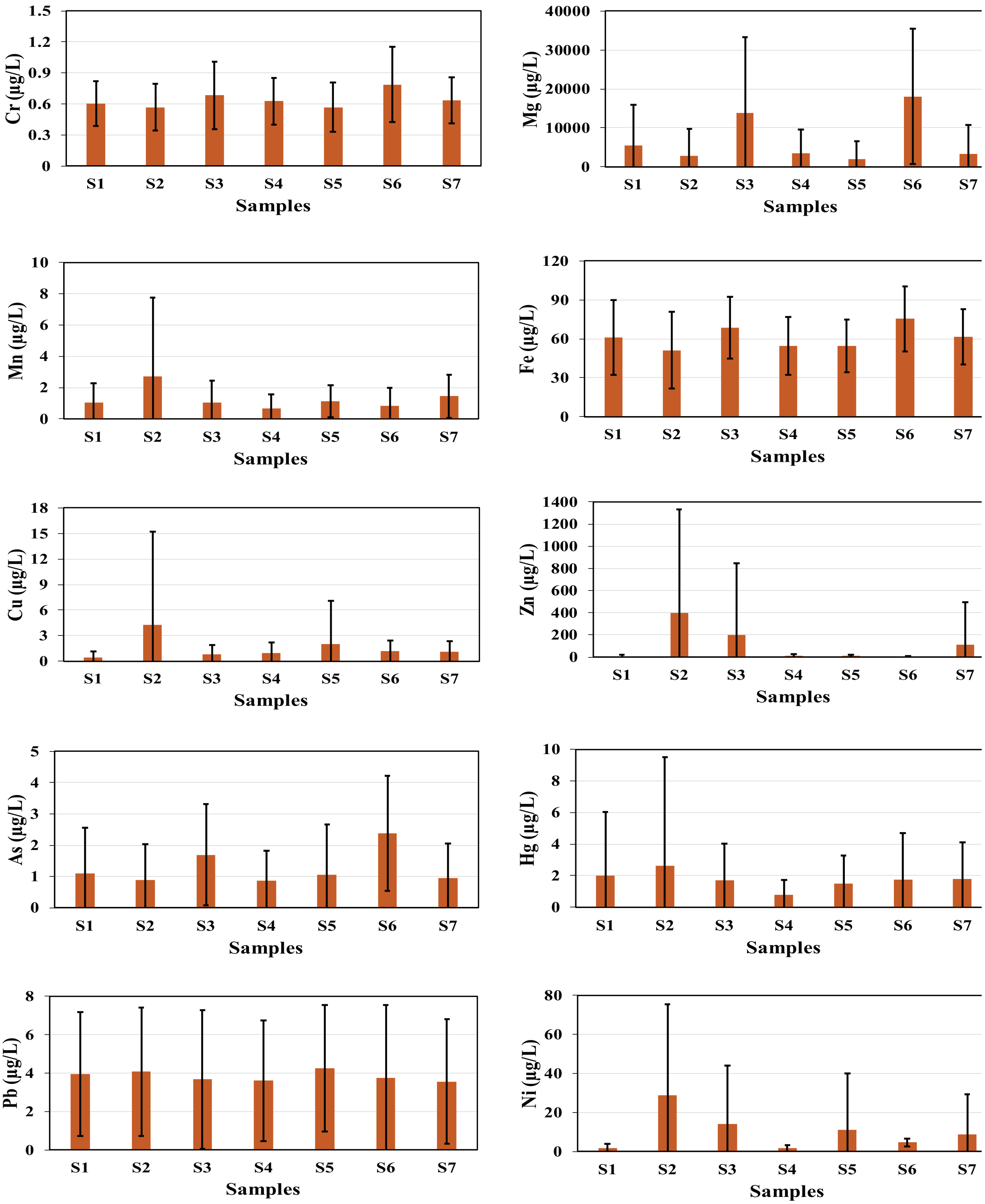

Fig. 5. Diurnal variability of some significant metals. S1: WDS samples after last use of water in the late evening; S2: cold water samples in the early morning prior to the first water use; S3: hot water samples in the morning; S4: WDS samples in the morning; S5: samples collected from the cold water tap in the afternoon following normal uses; S6: hot water samples in the afternoon; S7: WDS samples in the afternoon. The data represent the average values and the error bars represent the standard deviation.

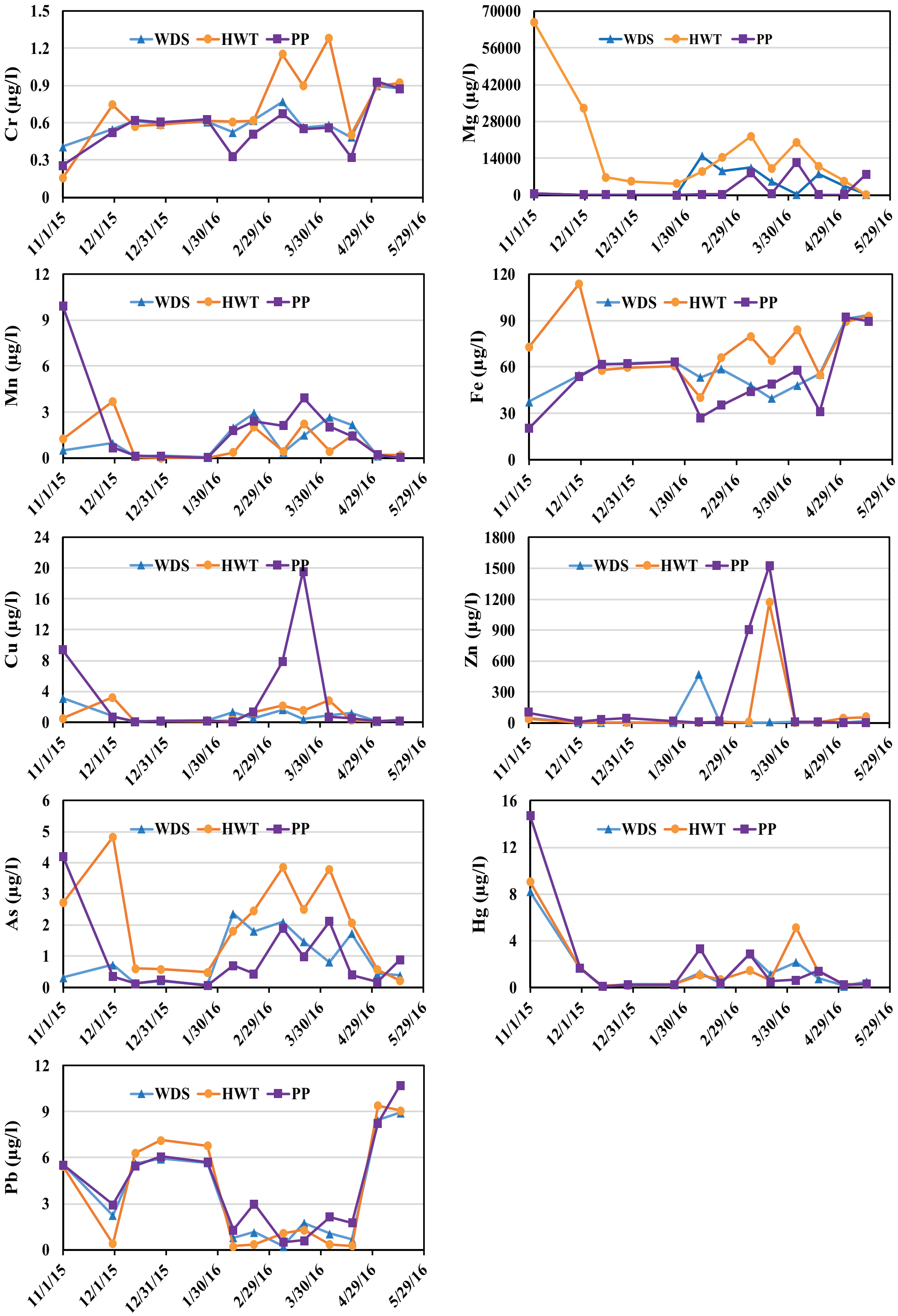

Fig. 6. Seasonal variability of some significant metals. WDS: samples from water distribution system; PP: samples from plumbing pipes; HWT: samples from hot water tanks.

In HWT, averages of Mg, Mn, Cu, Zn and As were higher during P2 than P3 (Fig. 6). The averages of these metals in P2 were 10889, 0.85, 0.91, 197.25 and 1.94 μg/L respectively. In P3, these were 9191, 0.57, 0.84, 25.79 and 1.66 μg/L respectively. The averages of Cr, Fe, Hg and Pb were lower in P2 than in P3. The averages of these metals in P2 were 0.75, 61.71, 0.73 and 2.82 μg/L respectively. In P3, these were 0.90, 80.23, 1.78 and 4.78μg/L respectively. In HWT, several metals (e.g., Mg, Mn, Fe, Cu, Zn, As, Hg and Pb) showed insignificant decrease from P1 to P2 while averages of Cr and Zn were increased from P1 to P2.

Overall, averages of Pb in WDS, PP and HWT were highest during P3 followed by P1 and P2, due to high temperature and water demand during P3 followed by P1 and P2. Averages of Zn in WDS and PP were highest during P2 and lowest during P3. Averages of Cu in PP were highest during P2 followed by P1 and P3. Both Zn and Cu showed decreasing trends with the increase of temperature. Averages of Cr in WDS, PP and HWT were highest during P3 followed by P2 and P1 while averages of Fe in WDS and PP were highest in P3 and lowest in P1. Concentrations of Mn in WDS were highest in P3 and lowest in P1. The variability of several metals (e.g., Zn, Cu, Cr, Mn) needs further analysis through cause and effect studies.

3.5. Correlation analysis

The Pearson correlation coefficients (r) are presented in Fig. 7. Temperature showed positive correlations with the concentrations of few metals (e.g., Zn, Cu, Mo, V). The correlation coefficients between temperature and Zn, Cu, Mo and V were 0.37, 0.23, 0.22 and 0.22 respectively. Higher concentrations of few metals during summer could be justified by the positive correlations with temperature. The UV$_{254}$ was positively correlated with Cu, Zn, As and V with r of 0.48, 0.41, 0.24 and 0.23 respectively while Pb and Fe were negatively correlated with UV$_{254}$ ($r = -0.32$ and -0.21 respectively). The FCl were positively correlated with Hg, Mn, Ba and Pb($r = 0.6$, 0.37, 0.35 and 0.22 respectively), and the TCl were positively correlated with Hg, Ba, Ca, Mn and Mg ($r = 0.56$, 0.42, 0.35, 0.25 and 0.22 respectively). The positive correlations of TCl and FCl indicate possible release of metals from pipe materials due to corrosive action of chlorine. In addition, few metals showed moderate to strong correlations among themselves. Concentrations of Sr showed moderate to strong correlations with Na, Mg, Ca, V, Fe and As ($r = 0.75$, 0.98, 0.89, 0.56, 0.48 and 0.79 respectively). Concentrations of Pb were negatively correlated with Na, V and As ($r = -0.63$, -0.65 and -0.48 respectively). The positive correlation of As with Mn ($r = 0.24$) indicated their co-presence and it may cause synergistic effects to health risk [3]. Further details can be found in Fig. 7.

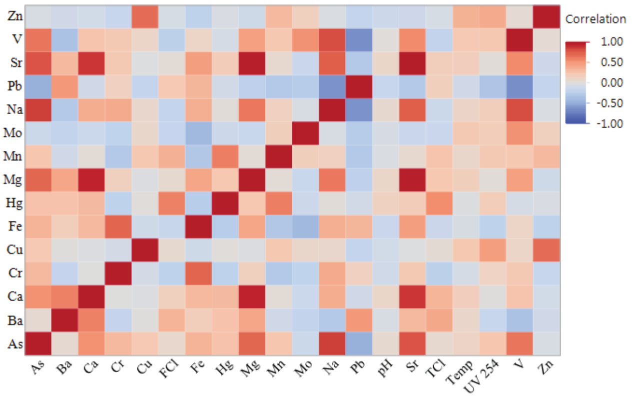

Fig. 7. Pearson correlation (r) map of heavy metals and water quality parameters (TCl: total chlorine; FCl: free residual chlorine UV$_{254}$: UV absorbance at 254 nm).

4. Discussion

Average concentrations of Pb, Ni, Co, Hg, Cu, Mn and Zn were highest in PP while the averages of Na, Mg, Ca, V, Cr, Fe, Sr, Ba and As were highest in HWT (Table 2). The WDS had the lowest averages in most cases (except Mo). Overall, concentrations of metals in HWT were 1.24–8.1 and 1.4–6.7 times the concentrations in WDS and PP respectively. In most cases, averages of As, Cr, Cu, Pb, Hg, Ni and Zn were in the increasing order of WDS, PP and HWT. Among these metals, Pb in drinking water has drawn significant attention due to its wide spread occurrences and effects to the children [26,27].

Several WQP (e.g., temperature, pH and chlorine), plumbing materials and network configuration were reported to affect the release of heavy metals from PP. The water was the mixture of desalinated and blended water, in which averages of pH were lower than the neutral value (e.g., <7.0) (Table 1), which might have increased the release of metals from PP. The extended reaction period due to stagnation of water might have increased the rate of Pb release from PP. Kim et al. [14] reported approximately 50% reduction of Pb in drinking water when pH was increased from 7.1 to 7.7 in several houses in ON, Canada. The Pb dissolution rate was increased with the increase of dissolved inorganic carbon (DIC) [48]. Further, in presence of FCl, dissolution of Pb in drinking water was reported to be constant [48,49]. However, alteration of FCl by mono-chloramine increased the concentrations of Pb significantly [50,51]. The Pb concentrations were increased from 15 μg/L (90th percentile) to 31–113 μg/L. An intermediate species, formed during mono-chloramine decay process, was responsible for Pb release from PbO$_2$ [51].

In addition, presence of iron oxide particles might have increased the particulate Pb release into water. The iron oxide/hydroxide has high affinity to Pb [52,53]. Kim et al. [14] reported higher levels of particulate Pb in water when concentrations of iron were high. The positive value of r between Pb and Fe ($r = 0.34$) is consistent to this. In addition, mechanical disturbances (e.g., repeated activation of faucet, water hammer) could have increased the release of Pb and Fe [54]. The pipe materials and coatings could have affected the leaching of metals into drinking water during stagnation in PP. The galvanized iron (GI) pipes with zinc coating were the main components in PP, which could have increased Pb in stagnant water [55]. Lasheen et al. [56] demonstrated the release of Pb and Fe into stagnant water using different pipe materials. Past study demonstrated exponential increase of Pb and Cu during 20–24 h of stagnation period [57]. When stagnation period was increased from 2 weeks to 20 weeks, concentrations of Pb and Fe were increased by 26% and 100% respectively [56]. The overnight stagnation of water in cooler increased Cr, Cu, Fe, Zn, Ni, Mn, and Pb in tap water [24], which were consistent to the current findings. Concentrations of Cu were higher in PP than WDS, due to extended reaction during overnight stagnation [16]. The higher concentrations of Zn, Fe, Cu and Mn in HWT might be due to temperature driven extended reaction [19].

The higher concentrations of heavy metals in PP and HWT might be a concern from monitoring, regulatory, human exposure and risk perspectives [9,10,26,58]. The regulatory agencies typically monitor water quality in treatment plant or in WDS. However, the plumbing premise can alter the concentrations of several metals in tap water. The regulatory agencies may further look into the strategy of water quality monitoring and regulatory compliance. Several metals in WDS, PP and HWT did not show significant diurnal variability (Fig. 3). However, the early morning and afternoon samples from HWT showed considerable variability for As and Cr. Corrosion of HWT wall, which was made of stainless steel and Ni-Cr heater alloys, might have released Cr while As could have entered from the wall of HWT due to scale formation [39]. Due to the use of hot water during the day, the scales were sloughed off and concentrations of As and Cr were increased in the afternoon samples [46]. Future study is needed to establish the mechanisms.

The concentrations of metals in WDS, PP and HWT showed seasonal variability. The averages of Pb in WDS, PP and HWT were highest during P3 and lowest during P2,indicating the effects of higher temperature (Fig. 6). The averages of Cr in WDS, PP and HWT were highest in P3 and lowest in P1, indicating variable seasonal effects (Fig. 6). Both Zn and Cu showed decreasing trends from P2 to P3, due possibly to increase in temperature. The seasonal variability of Zn, Cu, Cr, Fe and Mn needs further assessment through cause and effect studies. The interaction effects of temperature, flow rate, pH, pipe age, pipe materials and WQP need further investigation. The higher concentrations of few metals (e.g., Pb, Cr) in P3 may have elevated risk to the children [26,40,58]. Installation of appropriate household filters might reduce such effects.

5. Conclusions

A blend of desalinated and treated groundwater is supplied in Saudi Arabia. This water is relatively acidic, which can react with the pipe materials and pipe coatings during transport through WDS, resulting in the release of heavy metals into water. The diurnal and seasonal variability of temperature can affect the reactivity of desalinated/blended water with pipe materials and pipe coatings. Between WDS and tap, water spends significant amount of time in the PP and HWT. Depending on the size of plumbing premise, water quality can be degraded between WDS and the tap. During the off-peak hours(e.g., midnight to early morning), stagnation period of water in PP and HWT can be much higher, which may increase the concentrations of few heavy metals in tap water. The findings provide the basis for necessary steps to control heavy metals in tap water and to protect human health.

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