



Study on migration of phenolic and volatile organic compounds from plastic pipes used in plumbing home networks into tap water

Muhammad Mansoor Shaikh^a, Awadh O. Al Suhaimi^a, Marlia M. Hanafiah^{b,*},
Muhamad Aqeel Ashraf^c, Siti Norliyana Harun^b

^aChemistry Department, Faculty of Sciences, Taibah University, P.O. Box 30002, Al Medina Al Munawarah, Saudi Arabia

^bSchool of Environmental and Natural Resource Sciences, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, 43600 Bangi, Selangor, Malaysia, Tel. +60 3 89215865; Fax: +60 3 89253357; email: mmarlia@ukm.edu.my (M.M. Hanafiah)

^cDepartment of Environmental Science and Engineering, School of Environmental Studies, China University of Geosciences, China

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ABSTRACT

Water pipe materials can have significant effect on tap water quality. Recent reports evidenced that plastic pipes used to construct water distribution network in homes can leach potential amounts of harmful organic contaminants into tap water. In this work, the migration of phenolic and volatile organic compounds into tap water has been investigated. For this purpose, 30 samples were collected from homes within Al Medina Al Munawarah municipal residential area and analyzed for the existence of 2-butanone, 1,1,1-trichloroethane, carbon tetrachloride, 1,2-dichloropropane, 2-chloroethanol, 4-methyl-2-propanone, 1,2-dinitrophenol, pentachlorophenol and 1,2,3-trichloropropane. The most frequent compounds such as 2-butanone, 1,2,3-trichloropropane, 1,2-dinitrophenol, 4-methyl-2-propanone and 2-chloroethanol were monitored in 90%, 76%, 66%, 60% and 60% of samples, respectively. Meanwhile the levels of the compounds, 2-butanone, carbon tetrachloride and pentachlorophenol exceeded the WHO limits in 40%, 16% and 30% of samples, respectively. The migration test indicated that five of the targeted compounds occur in deionized water samples incubated in pipes in laboratory scale experiment. This implies that these pollutants are more likely to migrate from polyethylene pipes comprising home plumbing network. Although the levels of some percolated compounds were below the allowable levels, their accumulation during lengthy consumption can potentially increase the exposure to harmful constituents in water. As a prevention step, the use of sorbent filtration kits is recommended.

Keywords: Drinking water; Plastic pipes PEX; Volatile and phenolic compounds; SPE

1. Introduction

The access to pure and clean drinking water is a significant issue for human life on our planet. Currently, we rely heavily on huge distribution networks to supply water to our cities and homes. It is believed that water contamination by biospecies/chemicals can occur at several stages of its journey from source to tap. Since water can reside in the distribution system for several days before reaching the end user

[1,2], the materials in distribution system have a great impact on its chemical content. Over the last decades, water pipe materials used in the construction of distribution system and households, such as steel and copper metal pipes, have been replaced by plastic ones such as polyethylene (PE) and cross-linked polyethylene (PEX) due to their low cost and availability. Many recent reports evidenced that plastic pipes can leach significant amounts of various chemicals that deteriorate the taste and odor of drinking water and result in many harmful health effects.

Numerous organic additive compounds used during pipe manufacturing such as oxygenated compounds: ethyl

* Corresponding author.

tert-butyl ether (ETBE), methyl tert-butyl ether (MTBE) and their degradation product tert-butyl alcohol (TBA); degradation products of antioxidants, such as 2,4-di-tert-butylphenol (DTBP); and benzene, toluene, ethylbenzene and xylene solvents [3–6] have found to be trickled from polyethylene pipe; PEX and PE. Some of the easily leachable molecules, that is, ETBE, MTBE and TBA are known to alter the taste and odor of water [7]. Thus approximately 158 compounds are known to migrate from PEX pipes into water [8]. Many more chemicals are still unidentified and their consequences on humans are still not fully understood [9]. Rajasärkkä et al. [10] monitored several volatile compounds leachable from polyethylene pipes including toluene, xylene, ethylbenzene and styrene in tap water samples from homes with Helsinki area in Finland and also in water sample incubated into PEX pipes. In a similar study conducted in Denmark, the leaching of phenols and volatile organic compounds (VOCs) from four types of polyethylene pipe [11] has been investigated. The authors identified 20–30 organic compounds into water sample incubated for 7 d into two types of PEX pipes [11].

In Al Medina Al Munawarah and other Saudi cities, source water enters homes through plastic pipes branched from domestic water network and stored into large ground reservoirs (tanks), usually constructed from concrete from which water is pumped to a top tank, usually made up of fiberglass and distributed to all house/building by gravity. The tanks are often cleaned using various chemicals and disinfectant agents, thus these materials when not removed properly can contaminate water. In addition, the cleaning additives, especially bleaching ones might enhance the migration of organic molecules from plumbing systems. Presently, the most common types of plastic pipes used in water network in Saudi Arabia homes, are polyethylene (PE), and cross-linked polyethylene (PEX). The high temperature, especially in summer, is expected to rise the leaching rate of organic compounds. In a previous work, the investigators of this work, have screened several VOCs compounds, such as ethyl methacrylate, 1,2-dichloroethane, 2-picoline, dibromochloromethane, xylene, pyridine, 1,2-dichloromethane, 1,4-dichlorobenzene, benzyl chloride, 1,1,2,2-tetrachloroethane in tap water samples collected from Al Medina homes. The identified VOCs were also found in water samples incubated into PEX pipes [12]. Thus, the aim of this paper is to screen tap water samples for the occurrence of volatile and phenolic compounds and correlate them to the common plastic pipes used in plumbing networks at homes.

2. Methodology

2.1. Apparatus and materials

Sample preparation conducted making use of ASE-12 head solid phase extraction (SPE) manifold from AutoScience (Panamera's China) operated with vacuum pump (USA). A thermo gas chromatography, model 1300, equipped with flame ionization detector (FID). Mxt-5 analytical column (Rtx 30 m × 0.25 mm × 0.25 μm thickness), and integrator, Thermo model 1310 (Thermo Fisher Scientific, Mexico, USA) were used for the analysis of the targeted compounds. Acetone, methanol (HPLC grade), methylene chloride were obtained from Sigma chemicals (St. Louis, MO, 63178USA).

Hydrophilic-lipophilic-balanced 6 mL/200 mg from Starlab scientific (Xi'an City, Shaanxi Province, China). PEX ¾ inch pipes were purchased from local stores.

2.2. Stock and working standard solutions

VOCs standard Mega mix (30497) and phenolic compounds standard mixture (31029) were purchased from Restek Company (Bellefonte, PA, USA). Three-point calibration graph generated using standard solutions was applied for the quantification of the studied compounds.

2.3. Samples collection and preservation

Thirty tap water samples were collected in 1 L dark brown glass bottles from different houses within Al Medina Al Munawarah residential areas. For correlation study, 1.5-m lengths of the PEX pipes were filled with deionized water and incubated for certain period of times. Water samples were stored in fridge at 4°C until analysis.

2.4. Solid phase extraction procedure

Solid phase extraction is a rapid separation technique in which targeted analytes are retained from matrix onto solid phase (e.g., cartridge, column) and eluted with an appropriate solvent. The eluted extract is passed through sodium sulfate to remove water and concentrated as needed. In this work, SPE has been employed to extract the targeted compounds from water samples. For phenolic compounds, the cartridge was rinsed with 3 mL aliquots of methylene chloride three times, then as cleaned three times 3 mL of aliquots of methanol in such a way that methanol not go below the top of the cartridge packing. The cartridge was then rinsed three times with 0.05 N HCl for conditioning. After that, 100 ml sample of pH 2 was loaded to the cartridge at vacuum pressure 25 Psi and dried with nitrogen, and then trapped phenolic compounds were eluted with 1ml of dichloromethane and sodium sulfate. For VOCs, 100 mL sample of pH 2 was loaded to the cartridge at pressure 25 psi. Cartridge was dried with nitrogen, then trapped volatile compounds were eluted with 1 mL of dichloromethane with oven dry sodium sulphate. In the same way, another 100 mL water sample which was pre-adjusted to pH 12 was loaded to another cartridge following the same procedure. Both elute were mixed and analyzed by gas chromatography equipped with FID.

2.5. Migration study protocol

Four 1.5-m length pieces of PEX pipe (inner diameter 16 mm) were used to accommodate the required sample volume for analysis. The pipes flushed three times with MilliQ water and filled with water and left for incubation for 24, 48, 72 and 96 h. The incubated water samples were then extracted using the same procedure used for tap water samples.

2.6. GC optimization

The GC-FID system optimized for the analysis of organic compounds carried out using different temperature programs. A good separation for individual VOCs and phenolic

constituents was obtained. The chromatograms in Figs. 1 and 2 show that the components of phenols and VOCs standards were well resolved.

3. Results and discussion

3.1. Presence of volatile and phenolic contamination in tap water samples

The occurrence of VOCs in water from various sources has been evaluated in many reports [13–16]. These literatures display that more than 15 VOCs have been identified in drinking water samples. Various chlorinated by products, chlorinated solvents and different levels of oxygenated (MTBE) also have been detected in water samples from residential homes.

In the present study, seven VOCs and two phenolic contaminants were identified and quantified in 30 tap water samples collected from different houses (villas and apartment buildings) within Al Medina Al Munawarah city, Kingdom of Saudi Arabia. The results revealed that the VOCs: 2-butanone, 1,1,1-trichloroethane, carbon tetrachloride, 1,2-dichloropropane, 1,2,3-trichloropropane, 2-chloroethanol, 4-methyl-2-propanone, and the phenolic compounds 2,4-dinitrophenol, and pentachlorophenol exist in the analyzed samples at inconstant levels. The results summarized in Table 1 and graphically represented in Fig. 3. The minimum and maximum recorded concentration values for 2-butanone in tap water samples were 0.58 and 40.25 $\mu\text{g/L}$, respectively, with an average value of 13.73 $\mu\text{g/L}$. This contaminant was monitored in 90% of water samples, among them around 40% exceeding the maximum contamination level (MCL) 10 $\mu\text{g/L}$ set by the USEPA. 1,2,3-trichloropropane and 1,1,1-trichloroethane were detected in 76% and

40% of samples with concentration ranges 0.11–0.94 $\mu\text{g/L}$ and 2.14–27.17 $\mu\text{g/L}$ separately. The average values for these molecules were 0.35 and 12.31 $\mu\text{g/L}$, respectively. However, their levels were within the allowable limits stated by EPA, 5 and 200 $\mu\text{g/L}$ in the same order. The two compounds are chlorinated hydrocarbon characterized with their high chemical stability [17].

Carbon tetrachloride was found in 23% of samples collected from different locations, with concentration ranges between 2.01 and 9.32 $\mu\text{g/L}$ and average concentration of 6.78 $\mu\text{g/L}$. The level of carbon tetrachloride in 16% of samples was above permissible limit stated by EPA: 5 $\mu\text{g/L}$. This compound is widely used in plastic manufacturing and known to cause many hazardous health effects for humans such as damaging liver and lungs [18]. 1,2-dichloropropane, 2-chloroethanol and 4-methyl-2-propanone were found in 33%, 60% and 60% of the screened water samples with the concentration ranges 1.37–10.12, 0.02–0.67 and 0.29–1.20 $\mu\text{g/L}$ with average concentrations 7.31, 0.10 and 0.51 $\mu\text{g/L}$ in the same order.

The phenolic contaminants identified in tap water samples were 2,4-dinitrophenol and pentachlorophenol. The two compounds were found in 66% and 30% of the samples with concentration ranges between 2.02 and 20.64 $\mu\text{g/L}$, and 12 and 44.80 $\mu\text{g/L}$. The average concentrations were 9.96 and 24.03 $\mu\text{g/L}$, respectively. In the interim, 2,4-dinitrophenol concentration was less than the guideline limit recommended by the WHO: 40 $\mu\text{g/L}$; the pentachlorophenol level in 30% of water samples was above the EPA guideline value: 10 $\mu\text{g/L}$. Both compounds have significant health risk due to high carcinogenicity and listed on list of priority pollutants [19] due to toxicity [20]. Table 2 summarized correlation matrix Pearson (n) for VOCs and phenols in tap water samples. Apart from 1,2-DCP and

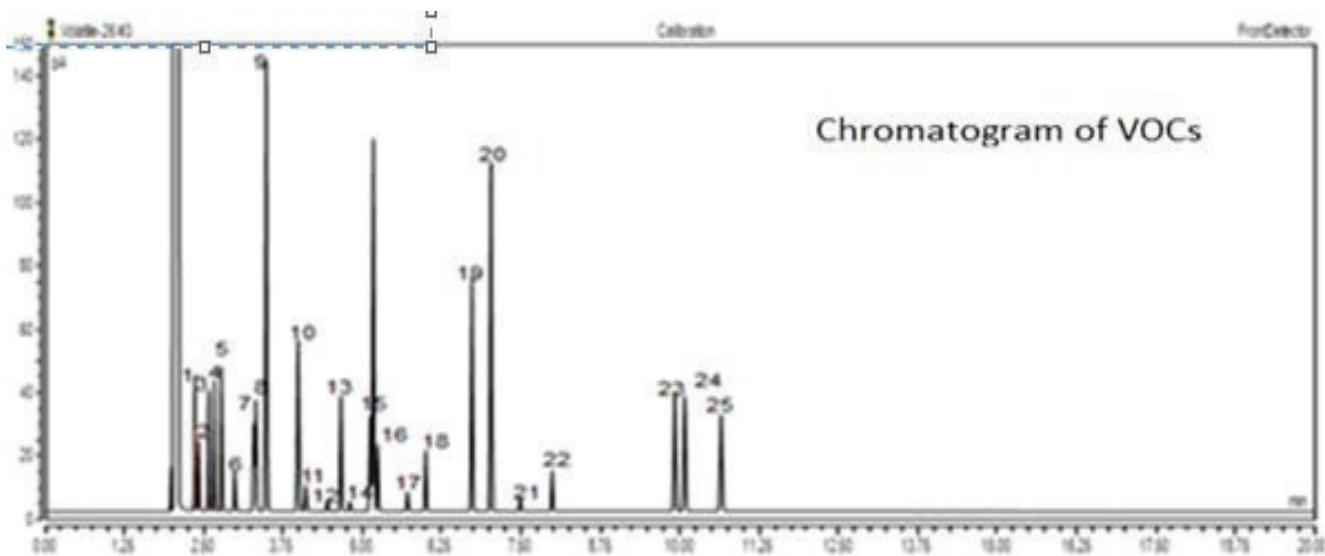


Fig. 1. Chromatogram showing 25 VOCs peaks eluted from calibrated reference standard with different retention times listed as follows: (1) benzene, (2) bromodichloromethane, (3) bromoform, (4) carbon tetrachloride, (5) chlorobenzene, (6) 2-chloroethylvinyl ether, (7) chloroform, (8) dibromochloromethane, (9) 1,2-dichlorobenzene, (10) 1,3-dichlorobenzene, (11) 1,4-dichlorobenzene, (12) 1,1-dichloroethane, (13) 1,2-dichloroethane, (14) 1,1-dichloroethene, (15) trans-1,2-dichloroethane, (16) 1,2-dichloropropane, (17) cis-1,3-dichloropropane, (18) 1,2,3-trichloropropane, (19) ethyl benzene, (20) methylene chloride, (21) 1,1,1,2-tetrachloroethane, (22) 1,1,2,2-tetrachloroethane, (23) toluene, (24) 1,1,1-trichloroethane, (25) 1,1,2-trichloroethane.

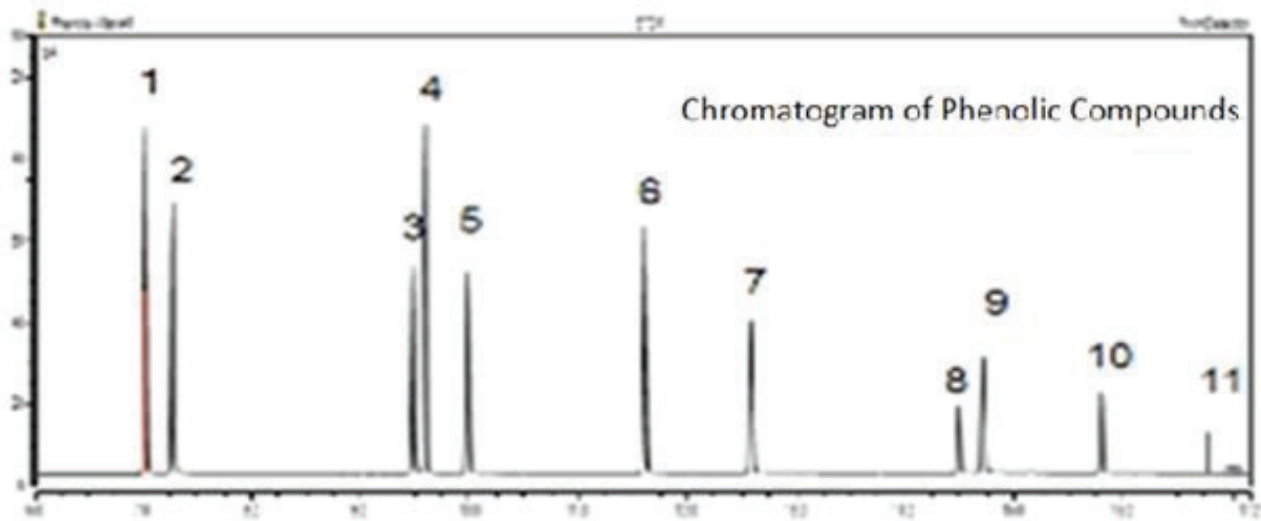


Fig. 2. Chromatogram showing 11 phenolic compound peaks eluted from calibrated reference standard with different retention times listed as follows: (1) 4-chloro-3-methylphenol, (2) 2-chlorophenol, (3) 2,4-dichlorophenol, (4) 2,4-dimethylphenol, (5) 4,6-dinitro-2-methylphenone, (6) 2,4-dinitrophenol, (7) 2-nitrophenol, (8) 4-nitrophenol, (9) pentachlorophenol, (10) phenols, (11) 2,4,6-trichlorophenols.

Table 1

Summary of concentration range, mean, standard deviation, % analytes, WHO limits and % of samples exceeding the limits

Analytes	Range	Mean	±SD	% Analytes in samples	WHO limit µg/L	% of samples above the limit
2-butanone	0.58–40.25	13.73	10.32	90	10	40
1,1,1-Trichloroethane	2.14–27.17	12.31	8.70	40	200	–
Carbon tetrachloride	2.01–9.32	6.78	3.23	23	5	16
1,2-Dichloropropane	1.37–10.12	7.31	3.95	33	40	–
2-Chloroethanol	0.02–0.67	0.10	0.12	60	–	–
4-Methyl-2-propanone	0.29–1.20	0.51	0.28	60	–	–
1,2,3-Trichloropropane	0.11–0.94	0.35	0.224	76	5	–
1,2-Dinitrophenol	2.02–20.64	9.96	7.02	66	40	–
Pentachlorophenol	12.00–44.80	24.03	13.09	30	10	30

4-M-2-P ($r = 0.539$), 2-butanone and 1,1,1-TCE ($r = 0.422$), and 1,2-DNP and PCP ($r = 0.391$) which show relatively moderate correlation, the other pollutant displays very low or no correlations.

3.2. Volatile and phenolic compounds leached from PEX pipes in the test water

The ability of volatiles (VOCs) and phenolic compounds to migrate from plastic pipes has been investigated in several studies [4,6,11,21,22]. In this work, the widely-used incubation method has been implemented to evaluate the leaching behavior of plastic pipes to comprehend to which extent the water quality can be altered due to its contact with polymeric pipes. The most popular polymeric brand, PEX pipes, has been used in this test. It has been proposed that the identified organic contaminant monitored in tap water samples might originate from polymeric pipes and other sources. However, a previous investigation by this research group shows that none of the targeted compounds identified in water samples

from source water supplied via local water network [23]. Therefore, the monitored contaminants are more likely to migrate plastic pipes. The results of the investigated compounds from migration test represented in Table 3 and demonstrated graphically in Fig. 4.

Apparently, five of the targeted nine VOCs and phenolic compounds occurred in tap water samples, 2-butanone, 1,2-dichloropropane, 1,2,3-trichloropropane, 2-nitrophenol, pentachlorophenol, have been identified in the leachability test. The other molecules, which were found in tap water samples but not monitored in migration study, might have come from other sources, for example, storage tanks, PVC connecting joints gluing resin. However, none of them exceeds the MCL recommended by WHO and USEPA. The amount of leached targeted compounds from four times incubations ranged from 1.80 ± 0.72 µg/L (for PCP) to 29.19 ± 3.03 µg/L (for 1,2,3-TCP). The order of the contaminants (lower to higher) were PCP < 1,4DCB < pyridine < TCFM < BDCM < 1, 2-DCP < 2 Butanone < 2, 4- DNP and <1, 2, 3, TCP. This finding comes in consistency with the former studies on the leaching

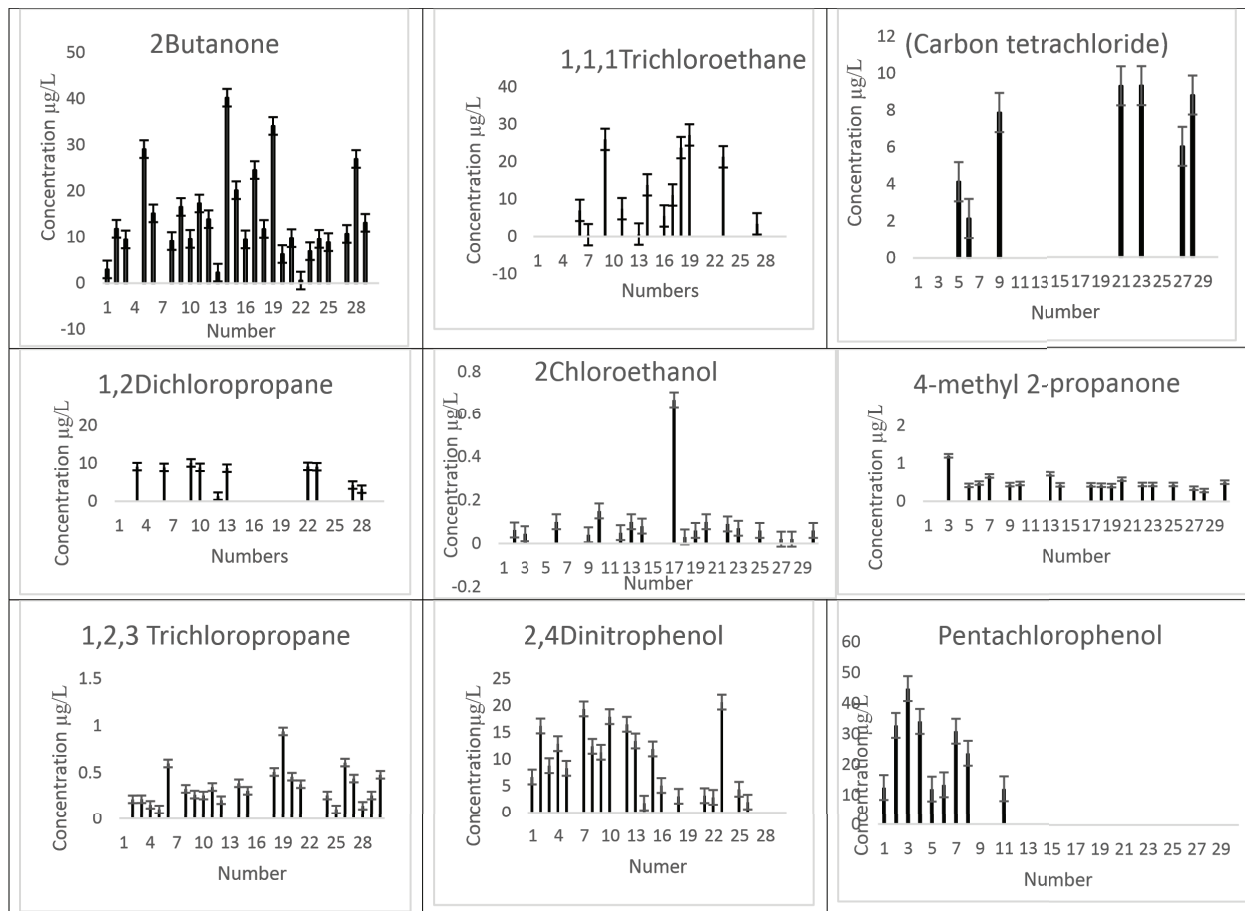


Fig. 3. Contamination levels of volatiles (VOCs) in tap water. The levels of dichloropropane in all samples were below the EPA allowable limits: 40 µg/L; meanwhile, the levels of 2-chloroethanol and 4-methyl-2-propanone are not regulated by EPA or other regulating bodies.

Table 2
Correlation matrix Pearson (n) for VOCs and phenols in tap water samples

Variable	2-Butanone	1,1,1,-TCE	CTC	1,2-DCP	2-CEthanol	4-M-2-P	1,2,3-TCP	1,2-DNP	PCP
2-Butanone	1								
1,1,1-TCE	0.422	1							
CTC	0.147	0.248	1						
1,2-DCP	-0.184	0.174	0.330	1					
2-CEthanol	0.204	0.167	-0.133	0.077	1				
4-M-2-P	-0.024	0.165	-0.003	0.539	0.249	1			
1,2,3-TCP	0.254	0.319	-0.089	-0.208	-0.207	-0.090	1		
1,2-DNP	-0.282	-0.058	0.047	0.290	-0.126	0.064	-0.468	1	
PCP	-0.209	-0.262	-0.216	0.011	-0.161	0.193	-0.153	0.391	1

1,1,1-TCE – 1,1,1-trichloroethane, 1,2-DCP – 1,2-dichloropropane, CTC – carbon tetrachloride, CEthanol – chloroethanol, M-2-P – 4-methyl-2-propanone, 1,2,3-TCP – 1,2,3-trichloropropane, 1,2-DNP – 1,2-dinitrophenol, PCP – pentachlorophenol.

of organic compounds from plastic piping, which indicated that significant concentrations of VOCs are released from PEX pipes [7]. The discharge of antioxidant degradation products such as phenols from PEX pipes also have been documented [6]. The values of Pearson correlation matrix in Table 4 reveal strong correlations between the concentrations of most of the analyzed components. This trend confirms that the migrated

compounds come from same source. Markedly, the concentration of leachable volatiles and phenols compounds increases with extended incubation time. Although the levels of the VOCs and phenols monitored in the controlled migration study as well as in some of the real samples are below the allowed limit, their accumulation over lengthy consumption of contaminated water escalate the risk on human health.

Table 3
Concentration ($\mu\text{g/L}$) of leached VOCs and phenols compounds from cross-linked polyethylene (PEX) pipes

Analytes	24 h	48 h	72 h	96 h	Total	WHO limit ($\mu\text{g/L}$)
Trichlorofluoromethane	1.5 ± 0.21	1.7 ± 0.34	1.3 ± 0.18	3.2 ± 0.52	7.7 ± 1.25	150 (California)
Bromodichloromethane	0.00 ± 0.00	1.2 ± 0.16	2.3 ± 0.47	3.6 ± 0.50	7.1 ± 1.13	60
1,2-Dichloropropane	2.20 ± 0.10	2.80 ± 0.09	3.20 ± 0.51	4.10 ± 0.68	12.30 ± 1.38	40
2-Butanone	0.00 ± 0.00	2.80 ± 0.29	3.20 ± 0.46	3.60 ± 0.38	9.60 ± 1.13	Not regulated
Pyridine	0.4 ± 0.06	1.4 ± 0.29	2.1 ± 0.32	2.1 ± 0.42	6.0 ± 1.09	Not regulated
1,4-Dichlorobenzene	0.69 ± 0.03	0.72 ± 0.07	1.40 ± 0.28	1.90 ± 0.33	4.71 ± 1.71	75
1,2,3-Trichloropropane	5.09 ± 0.65	6.80 ± 0.71	6.90 ± 0.69	10.4 ± 0.98	29.19 ± 3.03	80 (Hawaii)
1,2-Dinitrophenol	2.4 ± 0.41	4.3 ± 0.63	3.9 ± 0.43	6.7 ± 0.59	17.3 ± 2.06	40
Pentachlorophenol	0.00 ± 0.00	0.20 ± 0.02	0.40 ± 0.70	1.20 ± 0.00	1.80 ± 0.72	10

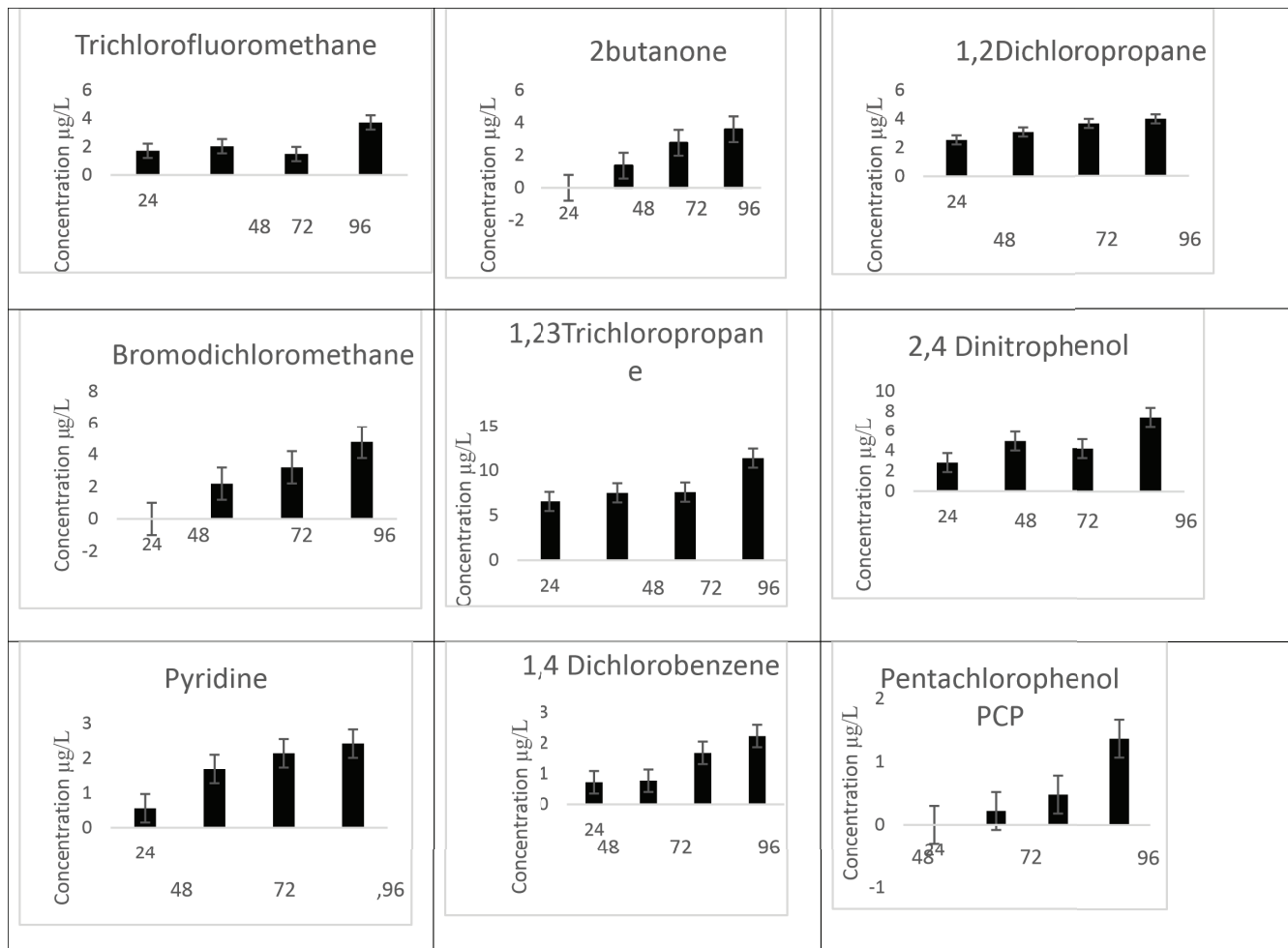


Fig. 4. Volatiles and phenolic compounds leached from plastic pipes over time (24, 48, 72 and 96 h).

4. Conclusion

The results reported in this work indicate that significant levels of VOCs and phenols occurred in tap water samples and hence they might affect water quality and esthetics significantly. Data from leaching test from the widely used

plumbing materials (PEX pipes) verify that the effect can vary depending on incubation time; however, other type of plastic pipes must be investigated in order to draw full picture on their significant impact on contaminants concentration of water exposed to the pipes.

Table 4
Correlation matrix Pearson (n) for VOCs leached from plastic pipes for 24, 48, 72 and 96 h

Variable	TCFM	BDCM	1,2-DCP	2-Butanone	Pyridine	1,4-DCB	1,2,3-TCP	2,4-DNP	PCP
TCFM	1								
BDCM	0.758	1							
1,2-DCP	-0.764	-1.000	1						
2-Butanone	-0.854	-0.987	0.988	1					
Pyridine	-0.869	-0.982	0.983	1.000	1				
1,4-DCB	0.755	1.000	-1.000	-0.986	-0.981	1			
1,2,3-TCP	0.992	0.833	-0.837	-0.911	-0.923	0.830	1		
2,4-DNP	0.999	0.734	-0.740	-0.834	-0.850	0.730	0.987	1	
PCP	0.964	0.903	-0.907	-0.961	-0.969	0.901	0.990	0.954	1

TCFM – Trichlorofluoromethane, BDCM – Bromodichloromethane, 1,2-DCP – 1,2-dichloropropane, 1,4-DCB – 1,4-dichlorobenzene, 1,2,3-TCP – 1,2,3-trichloropropanone, 2,4-DNP – 2,4-dinitrophenol, PCP – pentachlorophenol.

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