



Water quality and risk assessment of copper content in drinking water stored in copper container

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Received: 20 July 2021 / Accepted: 15 November 2021 / Published online: 7 February 2022
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Abstract

In most developing countries, drinking water is still a serious challenge. Many water purification methods exist, but they are costly and out of reach for many people. Copper is an ancient metal that has been used from several thousands of years. Because of its antibacterial properties, ancient cultures held this metal in high regard. Copper vessels are still in use today for storing drinking water due to its ability to purify water. Therefore, the objective of this study was to evaluate the copper concentration in drinking water after storing in copper containers/bottle over different time periods with respect to copper safety levels. For this study, water is stored in a copper container up to 168 h and the water samples are collected at different intervals. The concentration of copper is analyzed in water samples using ICPMS, and it ranges from 0.009 to 0.823 mg/L, which indicates as time increases simultaneously copper content also raises in the stored water. The higher copper content was detected at sample@168 h which was also well within the permissible limits of World Health Organization and United States Environmental Protection Agency.

Keywords Drinking water · Copper container/bottle · Safe limits · Copper toxicity

Abbreviations

USEPA	United States environmental protection agency
ICPMS	Inductively coupled plasma mass spectrometry
WHO	World health organization
APHA	American public health association
EPA	Environmental protection agency
EC	Electrical conductivity
TDS	Total dissolved solids
pH	Potential of hydrogen ion concentration

Introduction

Environmental pollution by various chemical elements including several toxic heavy metals is typically noticed as extremely increasing problem throughout the world (Adimalla and Qian 2021; Adimalla et al. 2020; Zhang et al. 2021). Specifically, this problem is very high in developing countries, where the drinking water quality is declining year by year. Especially, toxic chemical elements such as Cu, Hg, Cd, Pb and Cr are the main pollutants which are highly released into the environment and also greatly affecting the environment and human health (Dong et al. 2020; Subba Rao et al. 2020; Adimalla and Qian 2019; Taiwo et al. 2020; Zhang et al. 2020). It is worth noting that these pollutants are released in different ways and reach into groundwater-table, eventually poisoning the drinking water. Particularly, copper contamination in drinking water has been identified and recognized to be among the global environmental pollution concern during the 1990s due to the introduction of various health based regulations (Fitzgerald 1995; IPCS 1999; USEPA 1985 1991; WHO 1998). Importantly, copper is an essential micronutrient serves as a fundamental component of human proteins and metalloenzymes (Underwood 1977). The United States Environmental Protection Agency

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(USEPA) has fixed 1.3 mg/L as the tolerable limit of copper ions in drinking water (USEPA 1985, 1991). This presumes that intake of copper at concentrations greater than the prescribed limit can cause acute health problems namely gastrointestinal disturbance, central nervous problems, mucosal irritation, Wilson's diseases, damage of liver and kidney, wide spread capillary damage, hepatic and renal damage etc. (Bashir et al. 2021; Gomaa et al. 2021; Kulkarni and Sharma 2017). However, lower concentration of copper ion in drinking water gives health benefits (Araya et al. 2001; Buchanan et al. 1994). Typically, drinking water contains copper, which is a naturally occurring ingredient. Stagnation of water in copper and copper alloy-containing pipes during the supply and distribution process to households allows leaching of copper and increases copper levels in the water. Copper leaching can be accelerated by water characteristics such as increased acidity, increased temperature, acidic nature of water, and decreased hardness (NRC 2000; Casper 2017). Moreover, copper ions can be accumulated by various sources such as industrialization, electronic waste treatment, urban waste treatment, and natural metal erosion, dissolution and also leaching to groundwater table. Such leached copper can be an effective dietary source in situations where dietary copper consumption is poor, but most people are concerned about the acute and chronic effects of copper toxicity in drinking water (Fitzgerald 1998).

Several techniques have been developed for copper removal, water treatment, purification and disinfection (Abedi Sarvestani and Aghasi 2019; Asokan et al. 2021; Bashir et al. 2021; Gomaa et al. 2021; Lytle et al. 2019; Sharma et al. 2020) that are expensive and not easy to carry and also unavailable for rural regions. Water purification was mentioned in ancient Ayurvedic texts by storing it in copper and silver pots. Interestingly, copper as a potential biocidal agent has been utilized as a disinfectant for many years due to their antimicrobial properties. Eventually, USEPA has listed copper as the first solid antimicrobial material. Fundamentally, when water is stored in copper containers, the metal infuses into the water, providing health benefits to the drinker. Thereby, in recent years, the usage of copper water containers has largely been increased worldwide. In other words, a great number people storing water in copper containers and habituated for drinking purposes. However, to the best of our knowledge, research on how much copper is released when water is stored in copper containers has not been reported. The current study was designed to explore the possibility of copper leaching capacity when water is stored in copper containers over period of time. Therefore, the primary objective of this study was to evaluate the copper concentration in water that was stored in copper containers. To fulfil this aim, drinking water was collected from drinking water tap and stored in copper

vessels/containers. In addition, copper content of the collected water was assessed at different time intervals to test whether the quality of drinking water complies with the international guidelines for drinking purposes, and also discussed possible health risks. The findings of this study would be helpful in safeguarding the lives and also providing baseline scientific information about the water that was stored in copper containers.

Materials and methods

Sample collection and analysis

The sampling strategy, that is, initially water sample was collected from the drinking water tap and stored in 1000 mL capacity copper containers after methodically rinsed with distilled water (APHA 2012). This water sample was collected from the drinking water tap which is basically used for drinking purposes at 8.00 pm CST, on Dec-2020. And about 120 mL of first sample was taken after 12 h period from the time frame water is collected into copper container and the sample was named as sample@12 h. Similarly, rest of the five samples were taken at the time intervals of 24 h, 48 h, 72 h, 96 h, and 168 h at 8 am CST on the respective days, and they were named as sample@24 h, sample@48 h, sample@72 h, sample@96 h and sample@168 h, respectively. Finally, all these samples were stored at 4 °C and chemical analysis of copper concentrations was performed by using a Agilent 7900 inductively coupled plasma mass spectrometry (ICPMS) using EPA method 200.8. In addition, the basic water quality parameters such as pH, electrical conductivity (EC) and total dissolved solids (TDS) were performed for all collected water samples from copper container following the standard methods of American Public Health Association (APHA 2012). The pH of the collected water samples was measured by using a pH meter (Thermo Scientific ORION Star A211, USA). The pH meter was calibrated with three calibration standards (pH 4.0, 7.0, 10.0) prior to the measuring of water samples. The EC of the water samples was measured using an electrical conductivity meter ((Thermo Scientific ORION Star A215, USA). The meter was calibrated with known conductivity 3 point calibration standards. The determination of TDS in samples was performed according to the Standard method (SM 2540 C). A fixed volume of water samples was filtered through TSS fiber filter into a pre-weighed glass beaker; the filtrate was heated in oven at 180 °C until all the water was completely evaporated. The remaining mass of the residue represents the amount of TDS in a sample. Pictorial form of methodology is visibly depicted in Fig. 1 and it shows the details of sample collections, duration of time, and analysis.

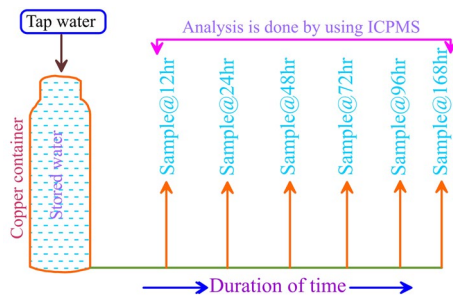


Fig. 1 Pictorial form of methodology (Raw water, stored water, samples collection, analysis)

Sample digestion

Added 2 mL of (1:1) nitric acid and 1.0 mL of (1:1) hydrochloric acid to all 100 mL digestion vials contain the 100 mL of the above collected samples. The digestion vials were placed on the hot block for sample evaporation. The hot block was adjusted earlier to a temperature of approximately but no more than 85 °C. Volume of the sample's aliquot was reduced to about 20 mL by slowly heating at 85 °C. This step took around 2 h for 100 mL aliquot with rapid increase in the rate of evaporation as the volume of the sample approached to 20 mL, then the final volume was made up to 100 mL with deionized water and then these samples were filtered and finally analysis carried out by using a Agilent 7900 ICPMS. Standards preparation, instrument specification and calibration are clearly listed in Tables S1 and S2, respectively.

Results

Hydrochemistry of the basic physicochemical parameters

The measured concentrations of the most essential physicochemical parameters including pH, EC, and TDS of the water samples are presented in Table 1. The pH values of water samples are found to be in the range between 7.02 and 7.16 with a mean of 7.07 (Table 1), which indicates that all collected water samples are in a neutral condition. Moreover, results clearly designate that pH values of all collected water samples are within the acceptable limit of 6.5–8.5 prescribed for drinking purpose (WHO 1993) and pH distribution is depicted in Fig. 2. The measured concentration of EC varies from 161 to 188.2 $\mu\text{S}/\text{cm}$ with an average of 182.67 $\mu\text{S}/\text{cm}$. The distribution of EC is shown in Fig. 2. In all collected water samples, the concentration of TDS is in between 79.1 and 94.5 mg/L, and its average is 90.01 mg/L (Table 1). It indicates that concentration of TDS is within the allowable limit of 500 mg/L (Fig. 2).

Table 1 Results of copper concentrations, pH, EC and TDS in water samples stored in copper container

Samples/time in hours	pH	EC ($\mu\text{S}/\text{cm}$)	TDS (mg/L)	Copper concentration (mg/L)
Raw water/0	7.02	161.0	79.1	0.009
Sample@12 h/12	6.97	185.4	86.7	0.327
Sample@24 h/24	7.16	185.1	94.27	0.482
Sample@48 h/48	7.09	185.9	91.05	0.519
Sample@72 h/72	7.06	186.2	91.22	0.521
Sample@96 h/96	7.12	186.9	93.2	0.710
Sam-ple@168 h/168	7.09	188.2	94.5	0.813

Concentration of copper

Before presenting the chemical analysis of copper in drinking water, using ICPMS, it is also crucial to ensure the accuracy of analysis and also calibration standards of instrument. This information typically supports into two different ways, the first is (1) to distinguish the instrument response and capability of generate the analysis results, and (2) to confirm the precision of obtained analysis using calibrated instrument. Table 2 evidently shows the calibration standards of instrument, results of obtained value and true values. Specifically, the quality of the analysis is also confirmed by initial calibration and laboratory blanks which are evidently demonstrated in Table 2. Finally, calibration curve of the copper was generated which is depicted in Fig. 3. It is worth noting from Fig. 3 that the quantified correlation coefficient and relative percentage error were 1.00 and 15.8%, respectively, and also indicates that obtained analytical results are true.

The results of copper concentration in drinking water of the present experiment are presented in Table 1. As can be seen from Table 1, the concentration of copper in raw-drinking water sample (before storing the water in copper container and named as sample@0 h) was observed at very low concentration 0.009 mg/L. Several studies reported that low copper concentration is found in public drinking water supply (Araya et al. 2003, 2001). In other words, copper concentration is very nominal in drinking water. According to WHO (1993) and USEPA (1994) standards, water is unsuitable for drinking purposes when the copper contents in water are greater than 2.0 and 1.3 mg/L, respectively. However, as discussed above, in this study the copper content was extensively examined after storing the drinking water in a copper containers/bottles at different time intervals such as 12 h, 24 h, 48 h, 72 h, 96 h and 168 h. Interestingly, the first-draw copper concentration (after 12 h's static period in copper container; sample@12 h) was recorded at lowest 0.327 mg/L (Table 1). Results also indicated that lowest (0.327 mg/L) and highest (0.813 mg/L) copper concentrations were

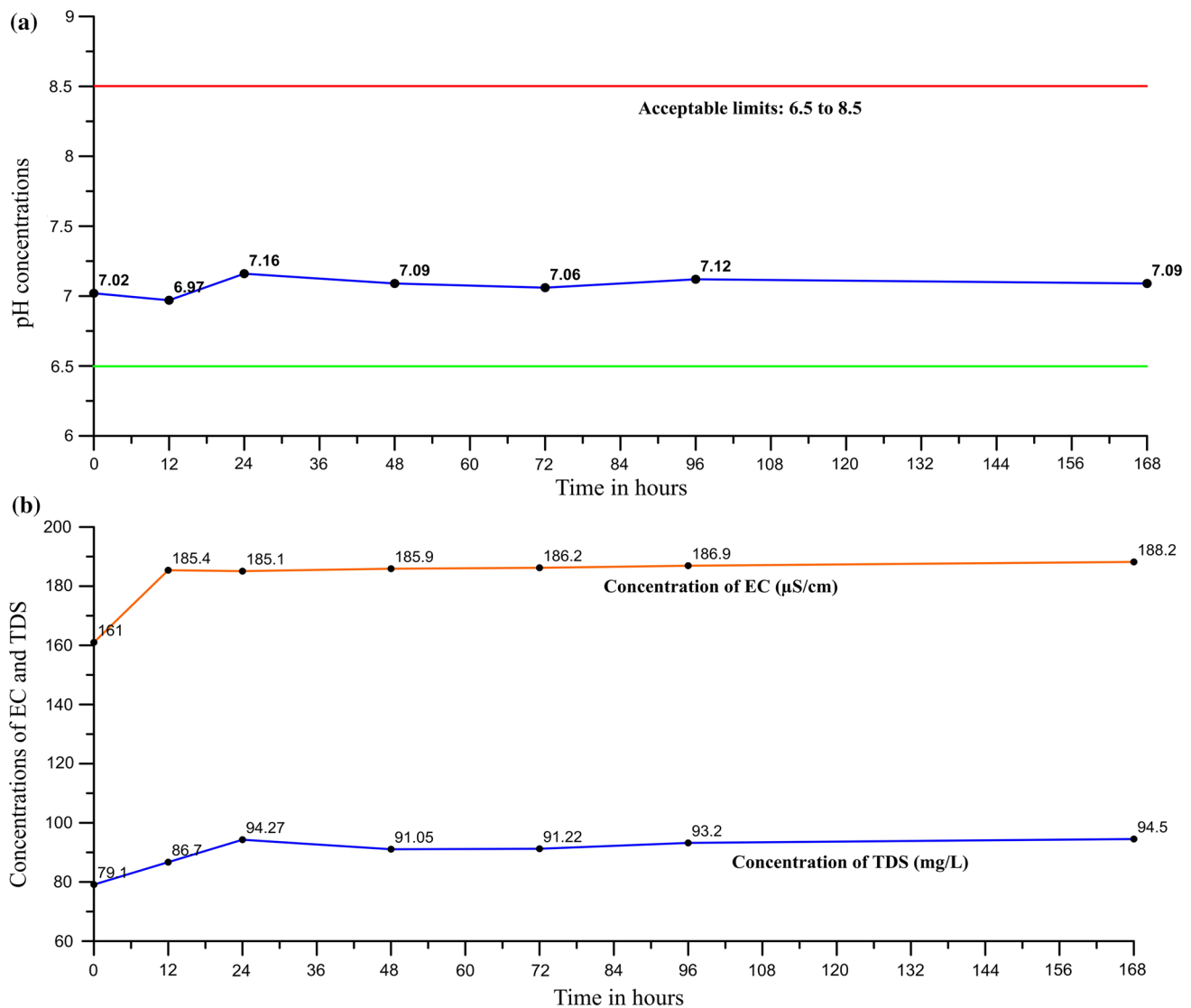


Fig. 2 Distribution of **a** pH versus time in hours, and **b** EC and TDS versus time in hours. The water samples were stored in copper containers, and measured pH, EC, and TDS in different time intervals

Table 2 Summary results of calibration standards, and quality control of analysis

Standard/sample	Obtained value (mg/L)	True value (mg/L)
ICB (initial calibration blank)	-0.00008	0.00 (1:1 HNO ₃)
Cal 2	0.001	0.002
Cal 3	0.022	0.020
Cal 4	0.501	0.500
Cal 5	0.747	0.750
Cal 6	1.002	1.000
ICV (initial calibration verification)	0.115	0.100
MB (method blank)	-0.000076	0.00 (1:1 HNO ₃)
LCS (laboratory blank)	0.0043	0.005

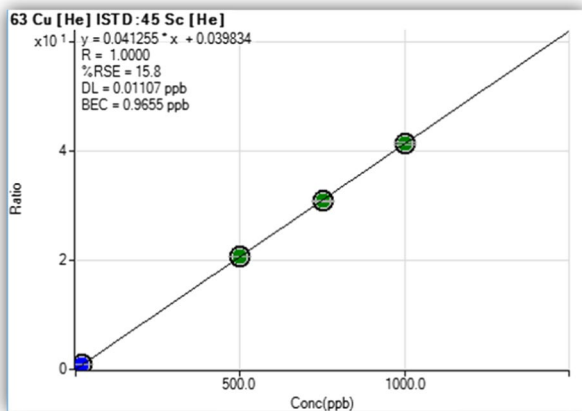


Fig. 3 Graph of copper calibration curve

observed in the sample@12 h and sample@168 h at the 12 and 168 h static periods, respectively. From this study, it is clear that trend of leaching copper concentration in water is progressively increasing as time increases, and the rising trend of copper concentration is evidently seen in Fig. 4 and Table 1. Generally speaking, the measured copper concentrations are within the guideline values recommended by both, WHO and USEPA for drinking purposes (WHO 1993; USEPA 1994). This experimental study basically suggests that the copper concentration is gradually increasing when water is stored in copper containers. Moreover, the present study evidently articulates that leaching of copper from the copper container is independent on the pH nature of the raw water. In both acidic and alkaline conditions leaching of copper from copper container increased with time. Moreover, it

is also observed from this study, concentrations of EC and TDS demonstrated no significant corrections with the copper values (Table 1). In other words, EC and TDS values have no direct influence on concentration of copper when time increases.

Discussion

Sources of copper

Through water distribution pipes

Copper is available in the environment in different states and complexes. Solubility of copper is affected by the form in which it is present. Form of the copper present in food is different from that of water. Copper is usually complexed with inorganic ligands or adsorbed to insoluble particles (Florence et al. 1992). A small amount of copper is present in drinking water (Abedi Sarvestani and Aghasi 2019; Araya et al. 2001). Copper can be released into environment due to several human activities, especially into land and mining operations (Abedi Sarvestani and Aghasi 2019; Taylor et al. 2020). In addition, release of copper into water is due to soil weathering, discharge of industrial waste into water, sewage treatment plants, and antifouling paints (IPCS 1998). Water distribution is one of the reasons for increase of copper content in water. Pipes and plumbing fixtures are mostly made of copper in many countries which are used to distribute water, which could leach copper into water in those pipelines. Copper leaching into water can be high due to the underlying conditions like acidic nature of water,

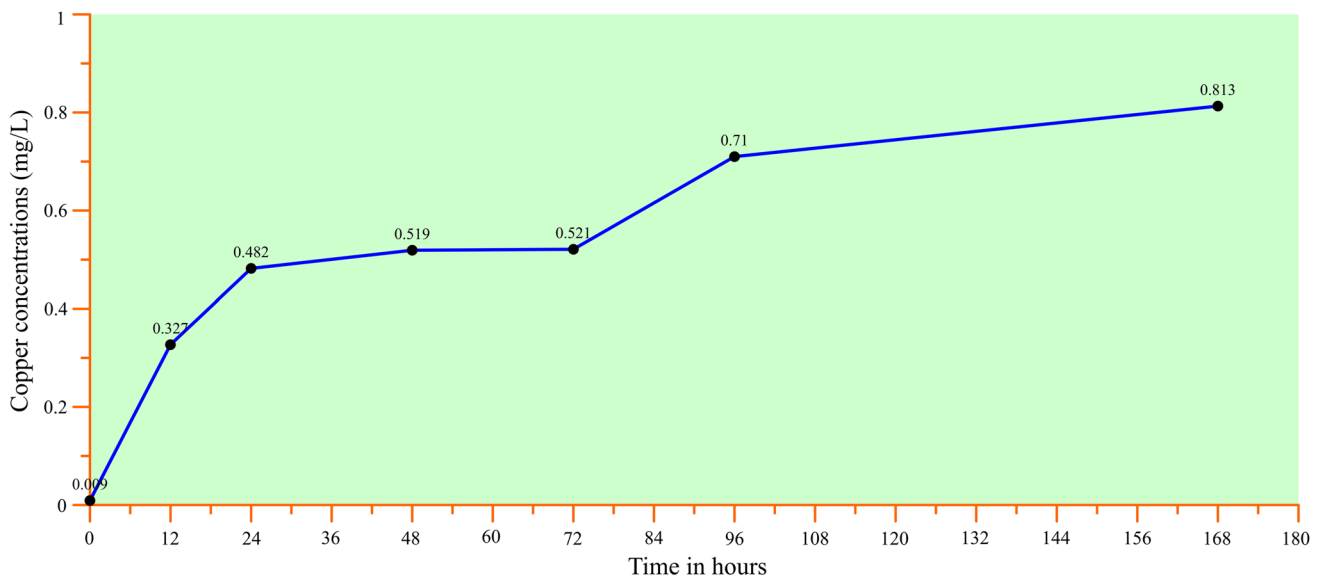


Fig. 4 Plot of copper concentration versus time in hours

hard rock terrain, high temperature, and in household with comprehensively using of copper piping systems (USEPA 1994). Another important factor is the length of the time for which water is stagnant in these pipes before it's supplied to households. The present study also indicates that as time of stagnation increases, concentration of copper also increases to several mg (milligrams) per liter in the water.

Through food and supplements

Foods rich in copper include shellfish, organ meats, whole grain products, nuts and seeds, cereals made of wheat barn and chocolate (Collins et al. 2014; Prohaska et al. 2012) (Kulkarni and Sharma 2017). The United States Food and Drug Administration (FDA) has identified a list of copper content foods which are shown in Table 3. In addition, dietary supplements also contain copper directly, and sometimes copper in combination with other ingredients is available in multimineral/multivitamin products (National Institutes of Health 2018). It is widely understood that these supplements contain copper in different forms like cupric oxide, copper gluconate, cupric sulphate etc., and generally amounts of copper form these dietary supplements range from few micrograms to 15 mg (which is 17 times more the DV for copper content) (National Institutes of Health 2018).

In the context of dietary, taking foods that contain high copper content through food and supplements, human body might be getting enough copper content already. In addition, it is apparently clear from this study that ingestion of water which was stored in copper containers for over a period of time (Table 1) can also accelerate the total copper concentration in the human body thereby higher possibility of increasing health risks. So, it is necessary to be mindful about the foods we eat, water we drink, procedures we follow for water purification which helps not to consume more amounts of copper.

Copper toxicity

It is well known, the copper is both an indispensable element and also poisonous at higher levels. As conferred above, exposure to higher levels of copper can elicit acute adverse effects to the liver and also engender the gastrointestinal symptoms like abdominal pain, cramps, nausea, diarrhea, and vomiting (Asokan et al. 2021; Buchanan et al. 1994; Fewtrell et al. 1996; Gotteland et al. 2001). Predominantly, several studies have reported ingestion of larger copper concentrations in drinking water have ensued in liver toxicosis mainly in infants and children (Abedi Sarvestani and Aghasi 2019; Gomaa et al. 2021; Taylor et al. 2020). In addition, the World Health Organization (WHO 1993) has detected

Table 3 Copper content of selected foods*

Foods	Milligrams (mg) per serving	Percent daily value
Beef, liver, pan fried (3 ounces)	12.400	1378
Oysters, eastern, wild, cooked, 3 ounces	4.850	539
Baking chocolate, unsweetened, 1 ounces	0.938	104
Potatoes, cooked, flesh and skin, 1 medium potato	0.675	75
Mushrooms, shiitake, cooked, cut pieces, ½ cup	0.650	72
Cashew nuts, dry roasted, 1 ounces	0.629	70
Crab, dungeness, cooked, 3 ounces	0.624	69
Sunflower seed kernels, toasted, ¼ cup	0.615	68
Turkey, giblets, simmered, 3 ounces	0.588	65
Chocolate, dark, 70%–85% cacao solids, 1 ounces	0.501	56
Tofu, raw, firm, ½ cup	0.476	53
Chickpeas, mature sees, ½ cup	0.289	32
Millet, cooked, 1 cup	0.280	31
Salmon, Atlantic, wild, cooked, 3 ounces	0.273	30
Pasta, whole wheat, cooked, 1 cup (not packed)	0.263	29
Avocado, raw, ½ cup	0.219	24
Figs, dried, ½ cup	0.214	24
Spinach, boiled, drained, ½ cup	0.157	17
Asparagus, cooked, drained, ½ cup	0.149	17

*This data was obtained from Food and Drug Administration of United States. The Food and Drug Administration of United States has developed Daily Values (DVs) in order to help the public compare the nutrient contents of foods and dietary supplements within the context of a total diet

nausea as the principal symptom to assess serious effects associated with consumption of higher copper containing water. However, due to the size of the copper elements, most of the copper content is present in the bone and muscle. Copper is present in ceruloplasmin which is a protein made in the liver (Adkison 2012). Therefore, ceruloplasmin transports copper from liver into the blood stream and other parts of body (Adkison 2012; Harris 2019). Around half of the copper is excreted in the bile, and the other half present in the body is excreted through gastrointestinal secretions. Copper when present in excess, the free excess copper ions cause damage to cellular components (USEPA 1994; Gotteland et al. 2001). The amount of cellular copper is determined by the intake and outflow of copper ions. Copper in excess amounts not only causes oxidative stress but also damages to DNA and reduces cell proliferation. Symptom of copper toxicity is seen when copper sulfate of more than 1 g is ingested (Asokan et al. 2021; Sharrett et al. 1982). Copper toxicosis that is resulted from metabolic defect, which is inherited, is classified as primary, and when it is resulted from high intake of copper and low excretion (due to involved pathologic process) is considered as secondary (Harada et al. 2020; Taylor et al. 2020). Copper toxicity could be caused by consumption of foods (Table 3) that are acidic and cooked in uncoated copper cookware, or when exposed to excess copper in drinking water, or through other environmental factors (USEPA 1994). Copper toxicity is unusual in healthy people without an inherited copper homeostasis deficiency (Institute of Medicine US 1988; NRC 2000).

Conclusions

This experimental study was designed to comprehensively understand the leaching potential of copper in water, after water has been stored in a copper containers over a period of time. The copper concentration in the basic raw water is very low. About low and high copper concentrations were found in the samples@12 h and sample@168 h, respectively. It is clear from the experiment study in a normal laboratory conditions, the drinking water quality is slightly deteriorated due to the presence of copper concentration when water is stored in copper containers for several hours. Therefore, immediate precautionary measures can be taken to safeguard the health of people and it is also very important to create a great awareness about water storage and consumption.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s13201-021-01542-x>.

Acknowledgements The authors are thankful to the Chemtex Environmental & Industrial Hygiene Services for providing necessary facilities to carry out this research work.

Funding No funding is provided for the preparation of manuscript.

Declarations

Conflict of interest The authors report no financial or nonfinancial conflict of interest. The authors alone are responsible for the content and writing of the paper.

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