

Original Research Article

<https://doi.org/10.20546/ijcmas.2018.712.300>

## Detection of Antimony in Drinking Water Bottles Produced in Iraq

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### ABSTRACT

#### Keywords

Antimony, PET Bottles, Atomic Absorption, Temperature, Duration

#### Article Info

Accepted:  
17 November 2018  
Available Online:  
10 December 2018

In water, antimony has no flavor, scent, or shade. It may best be detected by chemical analysis. A quick – time period exposure (over days or perhaps weeks) to antimony in drinking water at very high concentration (above 30 mg/l) can cause nausea, vomiting and diarrhea. In this study, we focus on the relationship of storage temperature and duration on the level of release of antimony (Sb) from plastic drinking water bottles produced in Iraq (seven provinces). To determine the amount of Sb release from the plastic in the water, atomic absorption technique was used. Sb release level was increased at (25, 40, 60 and -4°C), Sb release level increased with storage duration up to 2-week, indicating that Sb release from bottles may become stable under long-term storage. Human health risk was evaluated based on the worst case, i.e., storage at (60 °C) for 2-week.

### Introduction

Even though Iraq has major rivers as natural water sources, these resources are typically polluted, biologically and chemically due to lack of suitable systems for secure disposal of sewage, wastewater, commercial, agricultural and medical waste. Water treatment plants, pumping stations and water networks suffered from lack of upkeep result breakdown. (Stars Orbit Consultants and Management Development, 2010) In standard, the best of water is equally crucial as its amount consequently; water exceptional is considered as the issue to decide surroundings adjustments which can be strongly associated

with social and monetary improvement. (1) Bottled water can be defined as any potable water that is synthetic, disburied or presented for sale, sealed in food-grade bottles or other sanitary container and intended for human consumption. Assets of water in bottled water may be springs, wells, or other approved assets, the water in these bottles may be distilled, carbonated, ozonized or filtered (1, 2).

In developing nations approximately 1.8 million people, frequently kids, die each year due to water associated illnesses. Tap water can be harmful to human health because of infection with microbes. Tap water high-quality may

additionally trade due to exposure to the encompassing surroundings as open tanks or in pipes all through distances before attaining the clients, (3) the most important function of bottled water over tap water is the excellent, specifically in terms of flavor and regularity. consequently, bottled drinking water is one of the most critical assets of drinking water now not most effective in Iraq, but also within the whole global (World Health Organization (WHO), 2004) bottled water industry is one of the most thriving industries, in Iraq and global. it's miles the quickest developing drink choice where water consumption global is expanded via common 10% yearly, consequently, became the most dynamic quarter of all meals and beverage industries due to accessibility, pretty low value, better taste, and lower stage of impurities. The plastic material for packaging ingesting water differ from country to us of a, however the commonplace bundle material used is polyethylene terephthalate (PET) (4) this is because of its great fabric properties like unbreakability, exact barrier properties toward moisture, high readability, low migration trends for residual constituents and very low weight of the bottles as compared with glass bottles of the equal filling capability (5) it's far crafted from the polymerization of petroleum monomers of terephthalic acid and ethylene glycol using antimony-, titanium-, or germanium-based totally catalysts (5) over ninety percent of globally synthetic pet make use of antimony-based totally catalysts, predominantly antimony trioxide ( $Sb_2O_3$ ) with useful characteristics like excessive catalytic interest, low tendency to catalyze aspect reactions, creates no coloration in the final product and has low-value rate (5) an expected 150 billion plastic bottles are created from puppy resins yearly (6) commercialised pet resins made from using  $Sb_2O_3$  catalysts typically have residual sb ranging among 150-300mg/kg (4,7) these residual antimony amounts are very high and raise concerns,

thinking about earlier findings that, antimony was found to migrate into water and beverages stored in PET plastic packing containers over time. (4) Sb itself is a regulated consuming water contaminant and a non-critical detail for flora and animals and has no acknowledged organic or physiological characteristic and on a long-term exposure has been suspected to be carcinogenic (4,5, 6) in cases of acute intoxication, signs and symptoms such stomach and muscle aches, diarrhoea, desiccation, shocks, anaemia and uraemia may rise up. Those cause extreme myocardial inflammation, shivering, necrosis and in the end loss of life (4) other adverse ailments related to long-time period publicity to this metalloid include pneumonitis, fibrosis, bone marrow harm and carcinomas (8). numerous environmental elements have been located to affect the migration of antimony from the plastic container into the water stored internal. effects of things like temperature, daylight, period and physicochemical residences among others on migration were studied in other parts of the world (4,5,6,7).

Climatic situations in our part of the arena are very one-of-a-kind and greater intense with variations in seasons. Furthermore, the source of water for bottling and the packing fabric differs from one production agency to another. As a result, it's far imperative that puppy bottled water saved below triumphing conditions be studied to envision whether migration quotes are excessive main to infection.

## **Materials and Methods**

### **Sampling**

We collected drinking bottles water from Jan to March 2018 that produced locally in 7 different factories located in 7 provinces that covers Baghdad, middle Euphrates provinces and south provinces (Table 1), from each

factories we collected 20 different batch bottles.

### **Chemicals and reagents**

All solutions were prepared using ultrapure water. Nitric and hydrochloric acids (Merck, Darmstadt, Germany) used were of analytical grade. The antimony standard solutions were obtained by diluting a stock solution 1000 mg L<sup>-1</sup> Merck Darmstadt, Germany). Working solutions were prepared by appropriate dilution of a middle solution of 10 mg L<sup>-1</sup>. Glassware used was heated at 450 °C for at least 4 h to prevent cross contamination.

### **Sb concentration in PET bottles**

The bottles were digested to determine total Sb in PET plastic according to Hureiki and Mouneimne (7) and Westerhoffet (5). The bottles were cut into 4×4 mm<sup>2</sup> pieces. 0.5 g of plastic samples were mixed with 8 ml HNO<sub>3</sub> and 2 ml H<sub>2</sub>O<sub>2</sub>, and digested on a Hotplate for 1 h at 125°C. Sb in bottle was completely dissolved since only white powder residue remained in the digested solution. The digested solutions were diluted with deionized water, filtered through a 0.45 µm PES filter, and determined for Sb by Atomic Absorption Spectrophotometer (HVG.A.A.S),(SHIMADZU-7000).

### **Release of Sb from PET bottles**

Effects of storage temperature on Sb release from bottles was investigated at 25, 40, 60 and -4°C, simulating three storage conditions, ie.. Storage in refrigerator, room temperature, and high temperature. Bottles were stored for 7 and 14 d at 25, 40, 60, and -4°C, respectively. After 7 and 14 d, water in bottles was sampled to determine Sb. Deionized water was emptied from the bottles after 1 week of storage, and analyzed for Sb concentrations. Fresh deionized water was added again to those

bottles and stored for another week. The released concentration of Sb after 2 week of storage was accumulative of two weeks.

The concentration of Sb in water was determined by hydride vapor generator H.V.G.. To measure the fluorescence intensity of the antimony, an atomic spectrometer with hydrides generator, HG-AFS (Model SHIMADZU-7000) was used with a coupled quartz gas-liquid separator. Argon) was used as carrier gas and the flame was maintained by the C<sub>2</sub>H<sub>2</sub> generated parallel to the hydride reaction of HCl with NaBH<sub>4</sub> A high intensity hollow cathode lamp (SHIMADZU) was used as excitation source of the analyte.

### **Results and Discussion**

#### **Total Sb in PET bottles**

Total concentration of Sb (by H.V.G after HNO<sub>3</sub>-digestion) in bottles is listed in Table 1. The Sb concentrations ranged from 21.436 to 213.740 µg/g, which were similar those in previous studies (4, 9). For example, Sb concentrations in PET bottles from Hungarian, Nigeria, and Britain were 210-290 (4), 178-287, and 195-242 mg/kg (9), indicating variable quality of plastics in different countries. The total Sb released from bottle can be seen in tables, which is higher than the MCL of 5 µg/L of Sb in Iraq (Ministry of Health, 2006).

Clearly, it is unlikely that all Sb in bottles will be released into water. Therefore, releasing tests under various storage conditions are necessary to better understand release behaviors of Sb from bottles.

#### **Relationship of variable temperature and duration on Sb release from bottles**

Effect of storage temperature on Sb release from bottles was investigated by storing

bottles at -4, 25, 40 and 60 °C for 7 d and 14 d. Sb concentration ranged from (Nil) µg/L at -4 °C. Temperature increase to 25 °C enhanced Sb release from bottle, which ranged from (0.931) to (7.364) µg/L with the median at (3.218) µg/L. When comparison was based on single type, Sb release increased by (7) times if temperature increased to 25 °C, which is consistent with Reimann *et al.*, (2012) 10. For instance, for European PET bottles, 2.76-23.3 ng/L Sb was released after stored at 2°C for 7 d, and 11.7-54.1 ng/L Sb was released at 25 °C (Reimann *et al.*, 2012) 10. However, much higher Sb concentration (8.90-2570 ng/L) was reported by Shotyk and Krachler (2007) 6 based on 69 brands of PET bottles, which were stored at room temperature for 6 months. Higher Sb levels reported by (6) can be attributed to longer storage time (six months) compared with the current study and (10)(1 week). Even though higher median concentration (1.301--8.246 µg/L) of Sb in the bottled was measured at 40°C than those at 25°C (0.931-- 7.364 µ/L). When stored at 60°C, substantially higher Sb was observed across all bottles ranging from 2.034--11.015 µ/L. When compared among the three temperatures, Sb concentrations increased by 2 – 11 times when temperature increased from -4 to 60 °C, and 2 times when temperature increased from 25 to 60 °C Consistent with our study, a slight increase in Sb concentration in bottled water was reported when storage temperature increased from 2 °C to 22 °C (2.76 to 11.7 ng/L), and much higher Sb (1240 ng/L) was observed when temperature increased to 45 °C by (10).

When considering Sb release into water less than 60°C, the highest Sb at 11.015 µg/L in the first week was observed for PC plastic and at 28.116 µg/L in the second week. Since Sb release was mainly controlled by degradation of plastics (11), it was expected that PC was easier to be degraded under high temperature. The relatively low Sb release observed for

other plastic is comparable with the result of (12) who reported that Sb concentrations in most PET bottles were <1 mg/L after 70 d storage at 60 C. Nevertheless, higher concentrations were reported in other studies. For example, Sb concentrations ranged from 3.5 to 10.9 mg/L after storing PET bottles at 60 80 C for 1 week (5, 8, 10). The variable Sb release levels among different studies can be attributed to different quality of plastics materials, which may vary depending on raw materials as well as technology used for plastic production Although Sb release levels varied with different studies, the fact that elevated temperatures (e.g., 60 °C) significantly increased Sb release into water was confirmed.

To evaluate the effect of storage duration on Sb release, Sb concentration released from bottles were measured after 1, 2, week storage at 60 °C. Our data showed that Sb concentrations increased significantly with storage duration for 3 of 7 brand PET bottles.

Sb releasing rate was calculated (Equation 2) and decreased with storage duration for all brands (Table 2). It is expected that limited Sb was released from PET bottles over long storage duration as Sb release rate may gradually decreased to zero. This is consistent with (13) who showed that Sb concentration increase with storage duration gradually decreases under different temperatures.. Therefore, longer storage duration should be employed to reach the maximum release concentration of Sb in PET bottled water.

Our study shows that concentration of Sb increased with duration of 14 days for all types of bottles especially type 3 and type 7 (Table 3). Sb release from 7 brands of drinking water bottles in Iraq was investigated by storing bottles under different temperature and durations. Release of Sb increased with storage temperature, especially at 60 °C,

hence storing bottles under high temperature is similar to what happened in local warehouse of drinking water bottles especially. Meanwhile, more attention should be given to other drinks packaged using PET plastic, such as milk, coffee, and acidic juice, and other types of food containers. This is because the ingredient of these drinks and food are more complex compared to drinking

water, thus may induce more contaminants release and also more complicated speciation from the containers. For example, Sb concentrations in commercial juices are detected up to 2.7-fold above the EU limit for drinking water (5 mg/L). The more toxic inorganic Sb(III) (44%) and an Sb(V)ecitrate complex with un- known toxicity (41%) are the main species in the juices

**Table.1** Concentration of Sb in plastic

Plastic symbol no.	Local Market Brand Name	Sb (µg/g)
1	Kut	21.436
2	Thi-Qar	162.770
3	Basrah	153.129
4	Karbala	150.231
5	Maysan	31.263
6	Baghdad	52.460
7	Mousl	213.740

**Table.2** Concentration of Sb in water in the first week (µg/l)

	at 25 °C	at 40 °C	at 60 °C	at -4 °C
1	0.931	1.301	2.034	Nil
2	3.036	5.301	8.962	Nil
3	6.654	7.972	10.024	Nil
4	2.379	2.378	3.635	Nil
5	1.648	2.067	2.670	Nil
6	1.961	2.668	3.048	Nil
7	7.364	8.246	11.015	Nil

**Table.3** Concentration of Sb in water in the second week (µg/l)

	at 25 °C	at 40 °C	at 60 °C	at -4 °C
1	7.092	8.302	10.239	Nil
2	20.039	20.304	23.934	Nil
3	16.774	16.88	19.023	Nil
4	7.589	8.389	9.093	Nil
5	9.659	9.991	12.234	Nil
6	8.981	8.092	9.237	Nil
7	24.370	28.338	28.116	Nil

Our study shows that concentration of Sb increased with duration of 14 days for all types of bottles especially type 3 and type 7 (Table 3). Sb release from 7 brands of drinking water bottles in Iraq was investigated by storing bottles under different temperature and durations. Release of Sb increased with storage temperature, especially at 60 °C, hence storing bottles under high temperature is similar to what happened in local warehouse of drinking water bottles especially. Meanwhile, more attention should be given to other drinks packaged using PET plastic, such as milk, coffee, and acidic juice, and other types of food containers. This is because the ingredient of these drinks and food are more complex compared to drinking water, thus may induce more contaminants release and also more complicated speciation from the containers. For example, Sb concentrations in commercial juices are detected up to 2.7-fold above the EU limit for drinking water (5 mg/L). The more toxic inorganic Sb(III) (44%) and an Sb(V)ecitrate complex with un- known toxicity (41%) are the main species in the juices.

#### **Author contributions**

Farqad Abdulla Rashid designed the experimental scheme and did the most of the sample preparation and characterizations. Hiyam Isam Shakir analyzed the experimental data. Farqad Abdulla Rashid and Maha Salah Nawar wrote and revised the manuscript. Maha Salah Nawar and Hiyam Isam Shakir helped with the experiment. All authors reviewed the manuscript.

#### **Acknowledgment**

This work was financially supported by the Ministry of Science and Technology, Directorate of Environment and Water, Food Research Center.

#### **References**

1. Toma, J., Ahmed, R. Application of water quality index for assessment water quality in some bottled water Erbil City, Kurdistan Region, Iraq. *Journal of Advanced Laboratory Research in Biology* 2013. 4(4), 118-124.
2. Alabdula'aly, and Khan, Abdulrahman, I. and Mujahid A Khan Chemical Composition of Bottled Water in Saudi Arabia Environmental Monitoring and Assessment. 1999. 54(2):173-189 .
3. Fasano, E.; Bono-Blay, F.; Cirillo, T.; Montuori, P.; Lacorte, S. Migration of phthalates, alkylphenols, bisphenol A and di(2-ethylhexyl)adipate from food packaging. *Food Control* 2012, 27, 132-138.
4. Keresztes, S., Tata,r, E., Mihucz, V.G., Vira, G, I., Majdik, C., Zaray, G., Leaching of antimony from polyethylene terephthalate (PET) bottles into mineral water. *Sci. Total Environ.* 2009. 407, 4731-4735.
5. Westerhoff, P., Prapaipong, P., Shock, E., Hillaireau, A. Antimony leaching from polyethylene terephthalate (PET) plastic used for bottled drinking water. *Water Res.* 2008. 42, 551-556
6. Shoty, W., Krachler, M. Contamination of bottled waters with antimony leaching from polyethylene terephthalate (PET) increases upon storage. *Environ. Sci. Technol.* 2007. 41, 1560-1563
7. Hureiki, L., Mouneimne, Y. Antimony release in PET bottled natural water in Lebanon. *Water Sci. Technol. Water Supply* 2012. 12, 193-199.
8. Bach C., Dauchy, X., Severin, I., Munoz, J.F., Etienne, S., Chagnon, M.C. Effect of temperature on the release of intentionally and non-

- intentionally added substances from polyethylene terephthalate (PET) bottles into water: chemical analysis and potential toxicity. *Food Chem.* 2013. 139, 672-680
9. Tukur, A., Sharp, L., Stern, B., Tizaoui, C., Benkreira, H. PET bottle use patterns and antimony migration into bottled water and soft drinks: the case of British and Nigerian bottles. *J. Environ. Monit.* 2012. 14, 1236-1246.
  10. Reimann, C., Birke, M., Filzmoser, P. Temperature-dependent leaching of chemical elements from mineral water bottle materials. *Appl. Geochem.* 2012. 27, 1492-1498
  11. Takahashi, Y., Sakuma, K., Takaakiitai, Zheng, G.D., Mitsunobu, S. Specification of antimony in PET bottles from Japan and China by X-ray absorption fine structure spectrometry. *Environ. Sci. Technol.* 2008. 42, 9045-9050.
  12. Greifenstein, M., White, D.W., Stubner, A., Hout, J., Whelton, A.J. Impact of temperature and storage duration on the chemical and odor quality of military packaged water in polyethylene terephthalate bottles. *Sci. Total Environ.* 2013. 456-457, 376-383.
  13. Rungchang, S., Numthuam, S., Qiu, X.l., Li, Y.J., Satake, T. Diffusion coefficient of antimony leaching from polyethylene terephthalate bottles into beverages. *J. Food Eng.* 2013. 115, 322.

**How to cite this article:**

Farqad Abdulla Rashid, Hiyam Isam Shakir and Maha Salah Nawar. 2018. Detection of Antimony in Drinking Water Bottles Produced in Iraq. *Int.J.Curr.Microbiol.App.Sci.* 7(12): 2644-2650. doi: <https://doi.org/10.20546/ijcmas.2018.712.300>