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Investigating the Impact of temperature and storage time on antimony release from polyethylene terephthalate (PET) plastic used for bottled drinking water ☆

Yassin T.H. Mehdar

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Abstract

In last decade, Prosperity in contemporary Saudi Arabia and the consequent stresses on renewable freshwater resources resulted in the bottled drinking water market. Over the period, sustainable water resources planning (SWRP) has become an essential requirement for the development of places where these resources are highly constrained. To meet this market growth in Saudi Arabia is augmented by increasing demand for bottled water. This drastic increase, leads some countries to develop an alternative sources including desalinated water and treated wastewater for landscaping irrigation, industrial reuse, street cleaning, and aquifer recharge. There are claims on health concerns the possibility of chemicals migration from polyethylene terephthalate (PET) bottles into drinking water. In this paper, inductively coupled plasma -optical emission spectrometry (ICP-OES) was utilized and applied to determine the antimony (Sb) release from PET bottles into mineral water of 8 local brands at different temperature degrees within 8 months storage time.

Keywords

Polyethylene terephthalate (PET) bottles; Bottled water; Sb leachability; Health effects; Migration

1. Introduction

Plastic packages have increased tremendously in recent years due to benefit society in innumerable applications. These synthetic materials have become a multibillion-dollar business. Production of plastic for packaging purposes of beverage and food products is one of the most essential contributors to protecting from spoiling [1], [2], [3], [4], [5].

Plastics are ubiquitous in our daily lives, employed in an extensive range of applications. There are many different types of plastics, each type possessing distinct characteristics and applications. Polyethylene Terephthalate (PET) is the most plastic commonly used in bottles, such as those for soda or water, due to its durability, flexible, transparent, cost-effective, chemical and physical stability, favorable economic costs, and heat resistant [6], [7], [8], [9].

In addition, there are other plastic materials are used for packaging are Polyvinyl Chloride (PVC), High-Density Polyethylene (HDPE), Polypropylene (PP), Low-Density Polyethylene (LDPE), Polystyrene (PS), Polymethyl methacrylate (PMMA), and the recycled plastic material type (O) from the previous plastic types that, undergoes a process of cleaning, melting, and reshaping to be reusable. However, the specific properties of this material can vary depending on its original type [10], [11].

Numerous studies have been carried out to investigate the extent of Sb leaching from PET bottles. The results vary, with some studies indicating a significant amount of Sb leaching under certain conditions, while others suggest that the leaching is negligible. The variability in these results is likely due to differences in the types of PET used, and the storage conditions of the bottles [12], [13], [14], [15], [16].

It should be noted that the maximum contaminant level (MCL) for Sb in bottled water varies by region according to the World Health Organization (WHO). In Japan, the MCL is 2µg/L, while in the EU, USA, and Canada, it is 5µg/L, 6µg/L, and 6µg/L, respectively. The German

Federal Ministry of Environment also recommends 5μg/L. In contrast, Saudi Arabia has endorsed the highest WHO standard of 6μg/L [17].

One of the factors that can influence the rate of Sb leaching is temperature. Several studies have proved that leaching process from PET into drinking water is accelerated with temperature. Moreover, the storage time of bottled water also significantly impacts the amount of Sb that can be leached into water [18], [19].

Antimony trioxide (Sb_2O_3) is a white crystalline solid that is commonly used as a flame retardant in a variety of materials. In the context of PET manufacturing, Sb_2O_3 serves a dual purpose: it enhances the flame retardancy of the final product and used as a catalyst during the polymerization process of PET. It accelerates the reaction, leading to the formation of PET at lower temperatures and pressures. This results in energy savings and reduced environmental impact during the production process [8], [20], [21].

Antimony is regulated as a contaminant in drinking water due to its potential to induce health issues such as headaches, poor appetite, nausea, vomiting, stomach pain, diarrhea, dry throat, stomach ulcers, and loss of sleep [20], [22], [23]. There is evidence to suggest that Antimony is carcinogenic to humans based on animal studies [24].

In addition, the environmental Impact, particularly its release into the environment from industrial processes and its potential effects on ecosystems and wildlife [25].

This study was carried out to confirm the prevalence of PET, and to examine how various factors influence the migration of Sb into drinking water from PET bottles. The research further seeks to investigate how temperatures impact on Sb release into the water and the correlation between Sb concentrations and storage duration. To measure the concentration of Sb in bottled water, inductive coupled plasma-optical emission spectrometry (ICP-OES) was employed.

2. Materials and methods

Eight different local brands of bottled water were provided by some superstores and used throughout the investigation. All taken bottles were labeled as being PET plastic, and were colorless apart from two that had a faint blue tint. The volumes of taken bottles were 0.33L to unify the volume for the entire study. Prior to ICP-OES.

measurements, the samples were acidified with HNO_3 (Sigma-Aldrich). All solutions were prepared using ultrapure water obtained from a Millipore Direct-Q system. The laboratory equipment were soaked in a detergent solution for 24h, followed by thorough flushing with tap water and rinsing with ultrapure water to remove the detergent residue. Cleaned equipment were then allowed to dry at room temperature. In order to investigate the impact of storage conditions on the concentration of antimony (Sb), eight locally water bottles were procured from Madinah and subsequently divided into three groups. The initial concentration of Sb in the PET bottles was evaluated using inductively coupled plasma-optical emission spectrometry (ICP-OES; Thermo Scientific, ICAP 6000 Series) at the time of purchase, referred to as 'time zero'. Concurrently, the remaining two groups were set in incubator (BIOBASE, BOV-T3 incubator) for 120 and 240 days. The incubator was maintained at three different temperatures 25 °C, 35 °C, or 50 °C. The content of trace antimony Sb ranges from 0.025 to 2.17 μg/L.

3. Results and discussions

3.1. prevalence of plastic packaging materials

In this study, 120 different packaged food products were collected, and treated similar packaged food of different companies as different, even if the type of plastic used was identical. This approach was taken to assess the prevalence of plastic types in the food industry.

The findings revealed that PET plastic containers and bottles were the most frequently used packaging material as 39.84%, followed by PP, HDPE, PS as 15.63%, 14.06%, and 9.38 respectively. On the other hand, the least prevalent packaging polymeric materials were LDPE, PVC, and PMMA as 3.13%, 1.56%, and 0.78% respectively. However, a considerable number of plastic packages or bottles 7.81% lacked a code, rendering their resin composition undeterminable. Lastly, the percentage of the recycled plastic material type O was 7.81% (Fig. 1).

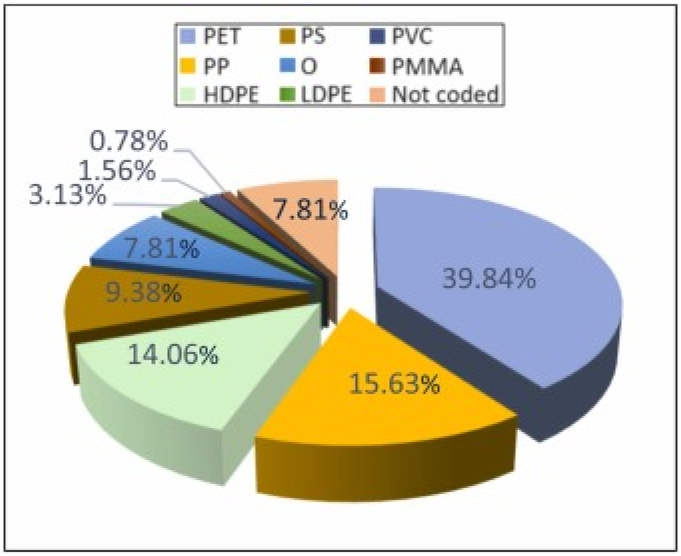

Fig. 1. The prevalence of plastic packaging materials.

3.2. Statistical analysis

All results have been processed using variance analysis (ANOVA) in Microsoft Excel, with a significance level of $p < 0.05$. One-way analysis of variance (ANOVA) revealed that the majority of the variation in Sb concentration over time and temperature is due to differences between the PET bottles (SS = 1.3076). Within PET bottles (SS = 11.5641, df = 48, MS = 0.2409), these values indicate that there is significant variation in the trace antimony concentrations within each PET bottle at each temperature (25, 35, and 50 °C) and storage time (120, 240 days). Interestingly, the p-value of 0.611 is greater than the common significance threshold of 0.05. These findings have important implications for understanding antimony migration from PET bottles and controlling its leachability to drinking water. The results show that antimony release is crucial, regardless of the type of PET bottle used. Furthermore, temperature and time are important factors that significantly influence the concentration of antimony.

3.3. Influence of temperature on antimony release

Temperature plays a critical role in the leaching of Sb from PET bottles. The impact of temperature was studied as a function of time at various degrees (25, 35, and 50 °C). The initial Sb concentration in all PET bottles, collected from Madinah in Saudi Arabia, was examined on the day of purchase (time zero). Table 1 illustrates the Sb content on purchased day were between 0.025 and 0.062 μg/L. The concentration of Sb of PET1, PET2, and PET3 were 0.033, 0.025, and 0.042 μg/L at 25 °C, which gradually increased to 0.68, 0.59, and 0.78 μg/L after 120 days respectively. The highest concentration at 25 °C was 0.97 μg/L of PET7. These results revealed a significant temperature-dependent difference in Sb leaching after 120 days. The leaching of antimony of PET6, PET7, and PET8 increased from 0.97, 1.15, and 0.93 μg/L at 25 °C to 1.30, 1.44, and 1.18 μg/L at 50 °C respectively. As clearly seen, the increased concentration of Sb with temperature of all PET bottles as directly relationship, such as PET5 rose by 40.0 %, from 0.85 μg/L at 25 °C to 1.19 μg/L at 50 °C (Fig. 3).

Table 1. Sb concentration of the 8 PET bottled water expressed in μg/L, at three different temperatures for 120, then 240 days, compared with time zero.

	Concentration of Antimony (Sb) in μg/L						
		After 120 days			**After 240 days**		
PET	**Time Zero**	**25 °C**	**35 °C**	**50 °C**	**25 °C**	**35 °C**	**50 °C**
PET1	0.033	0.68	0.85	0.97	0.92	1.05	1.22
PET2	0.025	0.59	0.7	0.88	0.8	0.95	1.15
PET3	0.042	0.78	0.92	1.07	0.98	1.16	1.46
PET4	0.038	0.76	0.88	0.94	0.91	1	1.3
PET5	0.053	0.85	0.99	1.19	1.03	1.32	1.83
PET6	0.057	0.9	0.97	1.3	1.08	1.45	2.04
PET7	0.062	0.94	1.15	1.44	1.15	1.58	2.17
PET8	0.05	0.88	0.93	1.18	1.02	1.23	1.63
$\bar{x}$	0.045	0.7975	0.92375	1.12125	0.98625	1.2175	1.6
±SD	0.01267	0.1188	0.1282	0.1920	0.1092	0.2224	0.3833

3.4. Influence of storage time on antimony release

Moreover, the rate of Sb leaching is not only dependent on the temperature at which the PET is exposed but also on the length of storage time. The results indicated that drinking water (due to longer storage time) contained higher levels of Sb. The levels of antimony released from the eight PET bottles after 240 days were all higher than the 0.90 μg/L at 25 °C. The average Sb concentration of PET botlles at 35 °C rose by 32.6 %, from 0.92 μg/L after

120 days to 1.22 μg/L after 240 days. In addition, the rate of antimony release for PET1, PET3, and PET4 at 50 °C were 0.97, 1.07, and 0.94 μg/L, and increased up to 1.22, 1.46, and 1.30 μg/L, after 240 days respectively. Sb leaching of PET2 was found to be.

the slowest, and it progressively increased from an initial concentration of 0.025 μg/L at zero time to a final concentration of 1.15 μg/L at 50 °C (Fig. 2). The overall antimony concentration in drinking water after a span of 240 days was observed to follow this order: PET7 >PET6 >PET5 >PET8 >PET3 >PET4 >PET1 >PET2. In comparison to drinking water standards, all PET bottles tested contained Sb at concentrations significantly below the recommended guidelines. The Sb concentration in water from these bottles was within the allowed limits for drinking water set by the WHO, except for PET6 and PET7, which were 2.04, and 2.17 μg/L respectively. These higher values were observed at 50 °C and surpassed the strictest Japanese limit of 2.00 μg/L.

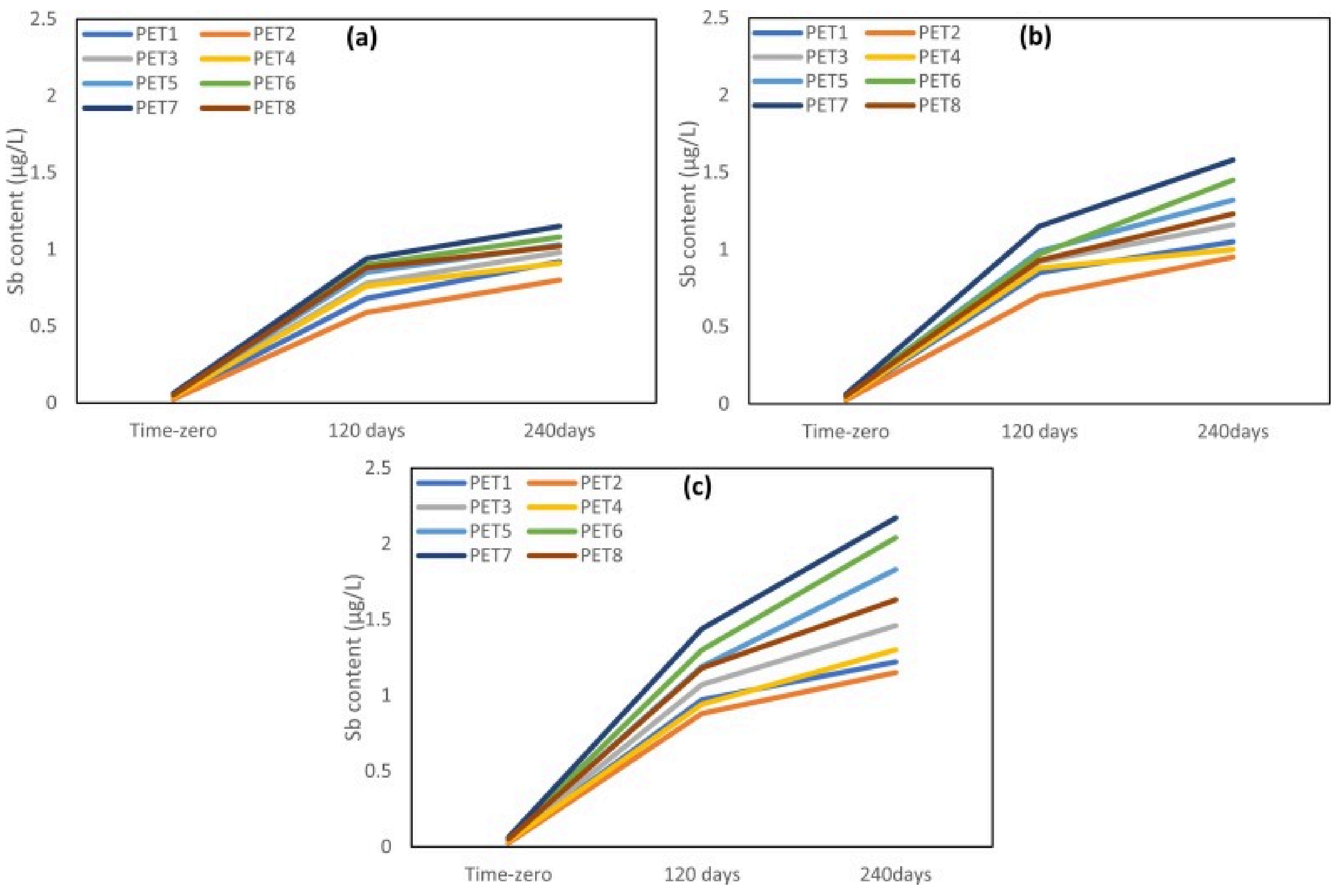

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Fig. 2. Influence of temperature on the antimony release from PET bottles as a function of time. at (a) 25 °C, (b) 35 °C, and (c) 50 °C.

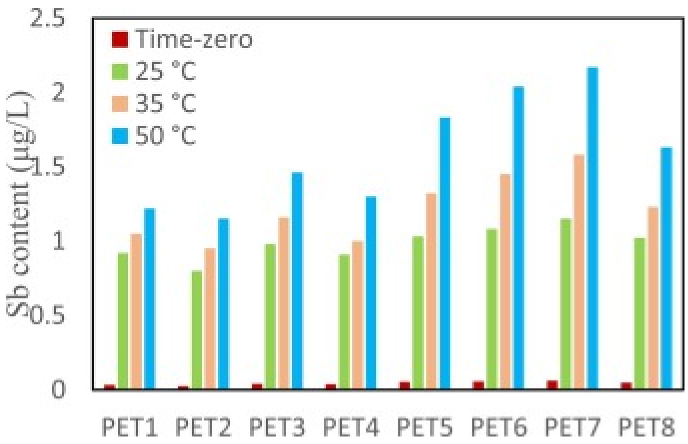

Fig. 3. Antimony (Sb) concentrations of the eight PET water bottles at different temperatures after 240 days.

Saudi Arabia has a climate that is known for its extreme heat, with temperatures often exceeding 50 °C. This high temperature can significantly impact the rate of antimony leaching. Furthermore, during the summer season, the temperatures inside closed cars, garages, and confined storage spaces can surpass 50 degrees Celsius in this city and other regions of Saudi Arabia.

4. Conclusions

The issue of antimony release from polyethylene terephthalate (PET) containers is a significant one that warrants immediate attention. The potential health risks associated with antimony, particularly its carcinogenic properties, necessitate the implementation of effective strategies to control its release.

The strategies outlined, such as material selection, optimal storage conditions, stringent regulations, and consumer knowledge, provide a comprehensive approach to tackling this issue. However, it is essential to note that these strategies are not standalone solutions but should be viewed as interconnected parts of a broader strategy.

Implementing these strategies requires the collective effort of manufacturers, regulators, and consumers. Manufacturers must prioritize the use of antimony-free materials in their production processes. Regulators, on the other hand, should enforce strict standards to limit the amount of antimony in PET containers. Lastly, consumers play a crucial role in this effort by adopting safe storage practices and being aware of the potential risks associated with antimony release. Moreover, continuous research and development in the field of material science can lead to the discovery of alternative, safer materials for PET container production.

Declaration of Competing Interest

The author declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. I declare that this study involves no conflicts of interest associated with this publication. As Corresponding author. I confirm this manuscript is original, has not been published before and is not currently being considered for publication elsewhere.

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Appendix A. Supplementary material

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Supplementary material

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Recommended articles

Data Availability

I attached everything is related to my research paper for the first time at your journal. I prefer using open access method and pay APCs to accelerate the publication processes.

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☆ I declare that this study involves no conflicts of interest associated with this publication. As Corresponding author. I confirm this manuscript is original, has not been published before and is not currently being considered for publication elsewhere.