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# Examining the Migration of Antimony (Sb) from PET Bottles into Carbonated Water: Impact of Temperature and Storage Duration

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## Abstract

Polyethylene terephthalate (PET) bottles have become a popular choice for beverage packaging. They are widely used for carbonated drinks, water, and other beverages. The clarity and stability of PET are enhanced by the addition of antimony (Sb), as a catalyst in the polymerization process. Sb is a toxic element that can cause acute and chronic health effects when ingested in high amounts. Carbonated water may be more susceptible to Sb leaching due to its slightly acidic nature. This study aims to investigate the factors influencing Sb leaching from PET bottles into carbonated water from four different brands under various temperature conditions over a storage period of 270 days. Inductively coupled plasma-optical emission spectrometry (ICP-OES) is employed for the accurate quantification of Sb concentration. The results revealed the presence of Sb in carbonated water from these PET bottles, especially that of PET3, with significantly higher ($p < 0.05$) concentrations quantified at 2.32 μg/L. Temperature and storage time were investigated as factors influencing Sb leaching over time. In this study, the values of Sb measured for bottled water at different temperatures oscillated between 0.022 and 2.32 μg/L at 30, 45, or 60 °C, over 270 days. The maximum Sb concentration reached 2.32 μg/L at 60 °C after 270 days, exceeding the Japanese limit of 2.00 μg/L, however, Sb levels were well within the range of USEPA and WHO limits. These results imply that Sb leached from PET bottles can be an active health hazard, especially in conditions of higher temperatures.

## Introduction

PET is a widely used material for food and beverage packaging. This material is derived from the condensation polymerization of terephthalic acid and ethylene glycol [1-4].

PET bottles have become an excellent choice for packaging a wide range of products due to their lightweight, cost-effectiveness, durability, and transparency, while their durability and transparency offer additional practical benefits. The prevalence of PET bottles in the packaging of carbonated beverages, water, juices, and condiments comes from their ability to protect the contents from light, oxygen, and moisture, which can degrade the quality and safety of the product. In addition, the recyclability of PET bottles contributes to their sustainability. Through the recycling process, PET bottles can be reused into new PET products, reducing the need for new raw materials and the associated environmental impact [3,5-7].

Antimony trioxide ($Sb_2O_3$) compound has a significant industrial relevance. This compound is commonly used as a catalyst within the PET resin, because it significantly enhances the material's clarity, accelerates the manufacturing reaction, and reduces its yellowing over time [8-9].

The maximum acceptable levels of Sb in drinking water vary in different international jurisdictions since each adopts its own regulatory approach in setting standards. WHO has established a limit of 20 µg/L [10], while USEPA [11-12] and the Ontario Ministry of Environment and Health Canada have set the maximum contaminant level (MCL) at 6 µg/L. The European Union and the German Federal Ministry of Environment have an even more severe threshold of 5 µg/L, whereas Japan enforces the tightest restriction at 2 µg/L. These standards differ based on the analysis of toxicological research and risk evaluations performed by each authority in order to guarantee the protection of public health [13-16].

Sb-based catalysts, primarily $Sb_2O_3$, have attracted much attention and are widely applied in the PET production process for some unique properties that provide specific benefits in performance. These catalysts have excellent stability during polymerization, which causes faster reaction rates and minimal side reactions. Current investigations strongly link Sb-based catalysts with the capability to produce quality PET with better optical qualities and mechanical strength but much lower production costs in industrial-scale procedures [3, 17].

Many studies have reported a general trend of increased Sb concentrations with increasing temperatures [5, 18-23]. For example, Sánchez-Martínez [5] reported an average Sb concentration of 0.04 µg/L, while another study in 2019 found concentrations ranging between 0.37-0.57 µg/L at 6 °C. At a higher temperature of 40 ºC, higher Sb concentrations were reported, ranging between 0.70-1.12 µg/L by several studies [18].

Recently, Al-Otoum [19] reported that at 24 °C, the levels of Sb detected were well below the WHO guideline of 5 µg/L, but at 50 °C, the concentration exceeded this limit and reached up to 6.11 µg/L.

In 2017, a study found that water stored in PET bottles had higher concentrations of Sb than water stored in glass bottles, with the maximum levels in tested samples reaching ($0.721 \pm 0.040$) µg/L [20].

The use of $Sb_2O_3$ in PET production has some health and environmental concerns associated with it [8-9,21]. Recent research reported the potential health risks associated with Sb leaching from PET bottles into the water. The contaminated drinking water with Sb can lead to more serious health issues. For instance, it has been associated with headaches, poor appetite, nausea, vomiting, dry throat, and diarrhea [24-25]. In addition, it can cause respiratory problems, including coughing, shortness of breath, and chest pain. There are also concerns about the carcinogenic potential of Sb trioxide, based on animal studies over a long period [26-28].

It should be noted that carbonated water is typically slightly acidic which increases the possibility of Sb leaching from PET bottles, based on several studies. This is attributed to the carbon dioxide gas that is dissolved in the water, which forms carbonic acid when it reacts with water [29-30].

This research was carried out to investigate how various factors influence the migration of Sb into carbonated water from PET bottles. The research also aimed to investigate how temperatures impact Sb

release into the carbonated water and the relationship between Sb concentrations and storage duration. To quantify the amount of Sb in bottled water, inductive coupled plasma-optical emission spectrometry (ICP-OES) was employed.

## Materials and Methods
### Chemicals Reagents and Equipment

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 (OKAY, EP20). The laboratory equipment was flushed with a detergent solution (Hepburn Bio Clean), followed by thorough flushing with tap water and rinsing with ultrapure water to remove the detergent residue, and then dried at room temperature. In addition, PET bottles were placed in a temperature-controlled incubator (BIOBASE, BOV-T3 incubator)

### PET Bottles and Conditions

Overall, 16 bottles of carbonated water from four different brands (Perrier, Chaudfontaine, Nestle Pure Life, and Aquafina) were supplied by some superstores and used throughout the investigation. These collected bottles were identified as PET plastic and were colorless apart from four bottles that had a faint blue tint. The thickness was measured using an electronic digital vernier caliper and its accuracy was up to 0.02 mm (0.001 inches). The inner diameter of these PET bottles was between 0.28 and 0.31 mm. The properties and specifications of the bottles used in the research are presented in Table 1.

All PET bottles were classified into four groups bottles based on their brand as PET1, PET2, PET3, and PET4. Subsequently, these PET bottles were placed in a

temperature-controlled incubator for 90, 180, and 270 days. The incubator was maintained at three different temperatures of 30 °C, 45 °C, or 60 °C. The fourth bottle of each brand was examined on the day of purchase (time zero).

*Table 1.* **The specifications of PET bottles.**

| PET bottle | PET1 | PET2 | PET3 | PET4 |
|---|---|---|---|---|
| Color | Faint blue | Colorless | Colorless | Colorless |
| Volume (L) | 0.5 | 0.5 | 1.0 | 0.5 |
| Thickness (mm) | 0.28 | 0.29 | 0.28 | 0.31 |
| Weight (g) | 28.0 | 26.5 | 37.0 | 27.0 |
| pH value | 5.4 | 5.5 | 6.5 | 5.5 |

### Instruments and Analytical Conditions

The concentration of Sb in the PET bottles was quantitatively analyzed using ICP-OES (Thermo Scientific, ICAP 6000 Series). The instrument operated at a radio frequency power of 1.2 kW, an argon coolant gas flow rate of 18 L/min, and a gas pressure was 30 psi. Before ICP-OES measurements, the samples were acidified by Nitric acid $HNO_3$ (Sigma-Aldrich). The solutions were prepared using ultrapure water obtained from a Millipore Direct-Q system. All glassware was soaked in a detergent solution for a day, then thoroughly washed with tap water and rinsed with ultrapure water to remove the detergent. Cleaned PET bottles were dried at room temperature overnight.

## Results and Discussion
### Statistical Analysis

The findings were analyzed using one-way analysis of variance (ANOVA) within Microsoft Excel, employing a significance threshold of ($p < 0.05$). The ANOVA results indicated that a significant proportion of the variance in Sb concentrations among the four PET bottle types over varying temperatures

and durations can be attributed to differences between the PET bottle types (SS = 0.575235). Notably, within the PET bottle classifications, substantial variability in trace Sb concentrations was detected, as evidenced by the within-groups sum of squares (SS = 14.90289, MS = 0.413969). These findings underscore considerable variability in Sb levels within each bottle type across different temperatures (30 °C, 45 °C, and 60 °C) and storage periods (0, 90, 180, and 270 days).

The F-value of 0.46319, when compared to the critical value of 2.86626, corroborates the conclusion that the observed discrepancies in Sb concentrations are predominantly due to random variation rather than systematic differences between the bottle types. The within-group variance being markedly higher than the between-group variance reinforces the insignificance of Sb concentration differences across the PET bottle types. Specifically, PET1 exhibited an average Sb concentration of 0.9432 µg/L, PET2 had 1.0328 µg/L, PET3 recorded 1.261 µg/L, and PET4 showed 1.1504 µg/L. Despite these variations in mean concentrations, the ANOVA test yielded a p-value of 0.71, exceeding the significance level of 0.05. This outcome indicates that the differences in Sb leaching among the various PET bottle types are not statistically significant. The results emphasize the crucial role of Sb release regardless of the specific PET bottle type, with both temperature and storage duration being significant factors influencing Sb concentration levels.
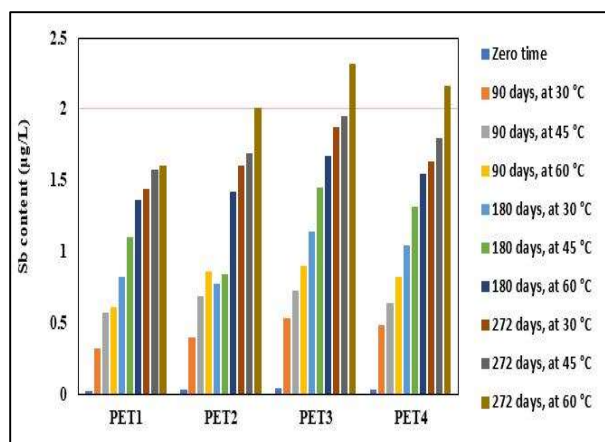
## *Influence of Temperature on Sb Release*

The investigation focused on the leachability of Sb from PET bottles into carbonated water under various temperature conditions over a period of 270 days. This study found a significant increase in Sb leaching with rising temperatures. Initial Sb concentrations in PET1 were 0.32 µg/L at 30 °C after 90 days, rising to 0.61 µg/L at 60 °C. Similarly, PET2, PET3, and PET4 showed initial concentrations of 0.4, 0.53, and 0.49 µg/L at 30 °C, which increased to 0.86, 0.90, and 0.82 µg/L at 60 °C, respectively. This trend indicates that higher temperatures result in more significant Sb release, with PET3 showing the highest increase to 2.32 µg/L at 60 °C after 270 days (Table 2).

Comparing these results with the mentioned studies reveals some consistencies and differences. For instance, the study by Aghaee [27] highlighted temperature as a crucial factor impacting Sb release. The study observed that Sb concentrations stayed below the MCL of 6 µg/L at 40 °C, outdoors, and at room temperature conditions. However, Sb levels exceeded the MCL at 65 °C and 80 °C over longer storage times. This is consistent with the current study, where higher temperatures accelerated Sb leaching, resulting in substantial increases starting at 45 °C and peaking at 60 °C (Fig. 1).

*Table 2.* **Influence of temperature on Sb release from PET carbonated bottled water after 90, 180, and 270 days, compared with time zero.**

| PET | Time Zero | Sb Concentration (µg/L) | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | After 90 days | | | After 180 days | | | After 270 days | | |
| | | 30 °C | 45 °C | 60 °C | 30 °C | 45 °C | 60 °C | 30 °C | 45 °C | 60 °C |
| PET1 | 0.022 | 0.32 | 0.57 | 0.61 | 0.82 | 1.1 | 1.36 | 1.44 | 1.58 | 1.61 |
| PET2 | 0.028 | 0.4 | 0.69 | 0.86 | 0.78 | 0.84 | 1.42 | 1.61 | 1.69 | 2.01 |
| PET3 | 0.040 | 0.53 | 0.73 | 0.90 | 1.14 | 1.45 | 1.67 | 1.88 | 1.95 | 2.32 |
| PET4 | 0.034 | 0.49 | 0.64 | 0.82 | 1.05 | 1.32 | 1.55 | 1.63 | 1.8 | 2.17 |
| x̄ | 0.031 | 0.435 | 0.657 | 0.797 | 0.947 | 1.177 | 1.5 | 1.64 | 1.755 | 2.027 |
| ± SD | 0.0077 | 0.0939 | 0.0689 | 0.1291 | 0.175 | 0.2673 | 0.1383 | 0.1812 | 0.1580 | 0.3057 |

**Figure 1.** Influence of temperature on the leaching of Sb from PET bottles as a function of time. The red line represents Japanese limit of 2.0 µg/L

**Table 3.** Sb concentration over time at different temperatures.

| Temperature (°C) | Storage Duration (days) | Sb Concentration (µg/L) | Reference |
|---|---|---|---|
| 6 - 40 | Up to 365 | 0.37 –1.12 | [18] |
| 24 - 50 | Up to 226 | 0.168 - 6.1 | [19] |
| 22 - 70 | Up to 950 | 1 - 700 | [23] |
| 40 - 80 | Up to 56 | 0.44 - 22.6 | [27] |
| 25 - 75 | Up to 15 | 0.28 - 2.30 | [29] |
| 30 - 60 | Up to 270 | 0.022 - 2.32 | Current Study |

The results of the present study agree with those obtained by Al-Otoum [19] and Zmit [18], confirming that temperature is a factor affecting Sb leaching from PET containers. This agrees well with findings by Al-Otoum, who indicates that Sb concentrations in PET bottles increase up to 6.11 µg/L when maintained at a high temperature of 50 °C, a confirmation that Sb leaching increases with heat. This study also determined that Sb levels reach 1.755 µg/L, which is 1.5 times higher than the 1.12 µg/L reported by Zmit in samples stored between 40-45 °C. In a study by Keresztes [23], the effect of temperature on Sb leaching was also evident. The study noted that at temperatures above 50 °C, Sb concentrations in still and sparkling water rose significantly. For example, Sb levels in samples stored at 60 °C ranged from 1 µg/L to 0.7 ng/mL, with even higher concentrations observed at 70 °C. Furthermore, a study by Chapa-Martínez [29] confirmed the temperature dependency of Sb leaching. The study found that Sb concentrations ranged from 0.28 to 2.30 µg/L initially, with the highest leaching occurring at 75 °C and pH 7 within 15 days (Table 3). This rapid increase at elevated temperatures parallels the findings in the current study.

## Influence of Time on Sb Release

The duration of storage significantly influenced Sb release. The initial mean concentration of Sb in carbonated water in PET bottles was 0.031 µg/L on the day of purchase. Over time, Sb leaching increased with temperature. For instance, PET1 showed Sb concentrations of 0.32 µg/L after 90 days at 30 °C, rising to 0.61 µg/L at 60 °C. After 180 days, Sb levels in PET3 increased to 1.67 µg/L at 60 °C and further to 2.32 µg/L after 270 days. This indicates a significant increase in Sb concentration ($p < 0.05$) over time, with higher storage temperatures exacerbating the effect.

A study by Aghaee [27] similarly noted time as a critical factor in Sb release and observed that Sb concentrations increased over the storage period at various temperatures. For example, Sb levels remained below the MCL at 40°C, outdoors, and at room temperature even after 8 weeks. However, significant Sb increases were observed at 65°C and 80°C after extended storage times, consistent with the prolonged Sb leaching observed in the current study.

Moreover, a study by Keresztes [23] showed that Sb concentration in one brand of still mineral water did not exceed 1 µg/L even after 3 years of storage at room temperature. However, under elevated temperatures, Sb levels increased significantly over time. In

addition, Sb concentrations in samples stored at 60 °C for 24 h ranged from 1.2 to 1.6 ng/mL, indicating a time-dependent increase similar to the findings of this study, where prolonged storage led to higher Sb levels.

Interestingly, the mean concentration of Sb in carbonated drinking water on day 90 at 45 °C was 0.657 μg/L, which increased significantly ($p < 0.05$) by 79.1% then to 49.1% after 180 and 270 days, respectively as presented in Fig. 2. Additionally, the variability in the results is evident, as the standard deviation for the PET1, PET2, PET3, and PET4 bottles after 270 days is ±3057 μg/L, which is the highest among all conditions. The leachability level of Sb in carbonated water after 270 days followed this order: PET3 > PET4 > PET2 > PET1. Despite the results of this study showing lower Sb levels than other standards, it is important to note that the Sb levels in the samples were still above the Japanese limit.

The consistent observation across all studies is that both temperature and time are pivotal factors influencing Sb release from PET bottles into the water. This investigation discusses the influencing factors and quantification of leachable Sb amounts from PET bottles into carbonated water over extended periods and varying temperatures, revealing substantial increases in Sb concentrations, particularly at higher temperatures and longer storage times. These findings align with previously reported studies, although differences in specific experimental conditions and absolute values highlight the variability in Sb leaching behavior across different studies and PET bottle materials. The consistent trend of increased Sb release with rising temperatures and extended storage emphasizes the imperative of meticulous consideration of storage parameters to minimize potential health risks associated with Sb leaching from PET containers. As PET is a popularly used material in the bottling of carbonated beverages, this issue warrants paramount attention due to the potential for Sb ingestion by consumers.
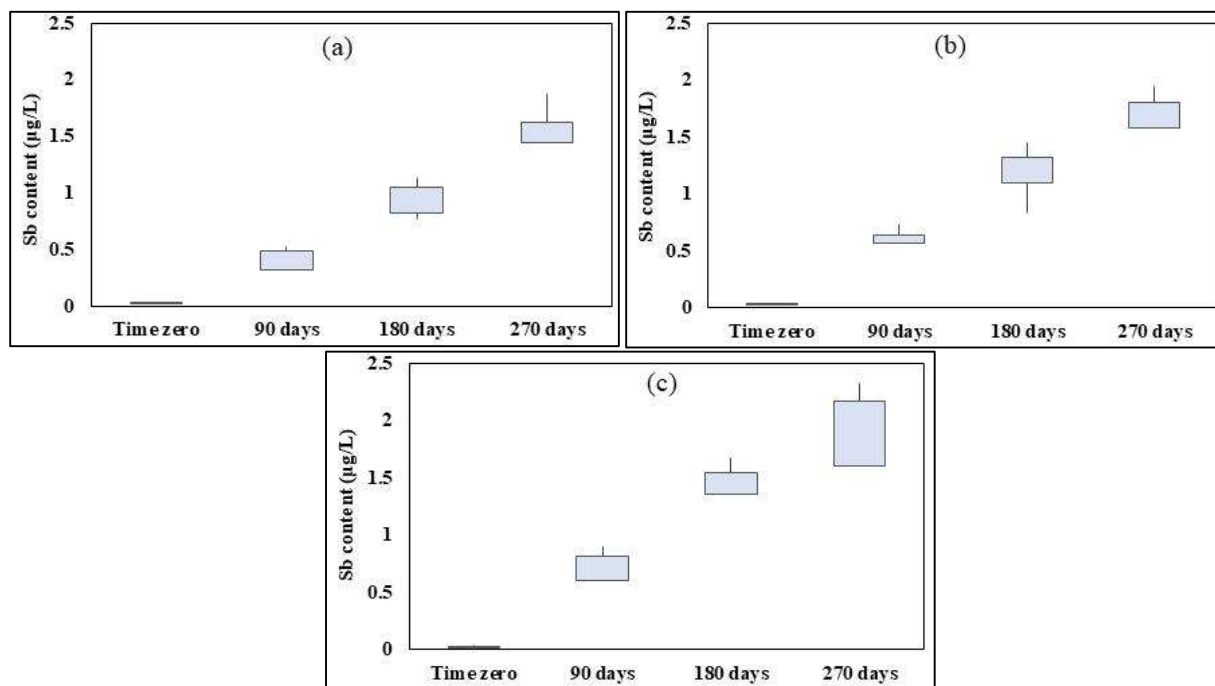


*Figure 2.* **Sb concentrations of the four PET carbonated water bottles at (a) 30 °C, (b) 45 °C, and (c) 60 °C**

## Conclusion

This study has focused on the current issue of Sb leaching from PET bottles into carbonated water, considering various temperatures and periods of storage time. Variation of Sb levels was found from different brands (PET1: 1.61 µg/L, PET2: 2.01 µg/L, PET3: 2.32 µg/L, PET4: 2.17 µg/L). These results accentuate the potential health risks related to Sb leaching, especially under conditions of high temperature.

The findings of this study also underscore the imperative of consumer awareness and more restrictive regulation to better ensure public safety. These results can inform policy and industrial practices by encouraging manufacturers to explore innovative production processes or alternative packaging materials that reduce Sb leaching.

In this respect, the use of glass instead of a PET package would be relevant, representing a point of view showing both its benefits and challenges. While glass does not pose the same risk of Sb leaching, its production is energy-intensive, and the heavier weight of glass increases the energy consumption and costs of transportation. On the other hand, glass is highly recyclable and can be reused multiple times without losing quality, which presents an opportunity for industries to develop more sustainable glass manufacturing practices that offset its environmental drawbacks.

Public awareness campaigns will go further in causing consumers to act more responsibly in the case of safe storage and the environmental implications of their choices. We will be in a position to reduce the risks of Sb exposure by facilitating informed choices and laying down strict standards for ensuring safer and more sustainable packaging in the future.

## Conflicts of Interest

As a Corresponding author, I declare that this study involves no conflicts of interest associated with this publication.

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