

Leaching of Antimony from Bottle Water

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Abstract— Bottled water has become part of everyone's daily life. Therefore, it is crucial to investigate its influence on the human health and the environment. Polyethylene terephthalate (PET) is the major type of material to be used for water bottles. The PET bottles can in fact affect the quality of water it holds. One major concern is leaching of antimony from PET water bottles. As such, this paper investigated the existence of antimony in the bottled water. Experiments were conducted on exposing water with bottles from different brands at different times and temperatures. After the experiments were conducted, water was tested for antimony using the Inductively Coupled Plasma Mass Spectrometry (ICPMS) machine. The results indicated that antimony was leaching from the PET bottles. Longer exposure indicated higher antimony leaching, as was the case for higher temperatures in most samples.

Keywords— PET, Antimony, bottle water, leaching, exposure

II. Introduction

Plastic water bottles have become an integral part of our life due to its portability and requirement for small area. Polyethylene terephthalate (PET) is the most common material used for water bottles. Glass bottles are also used in some places due to lack of health concerns from average consumers [1]. The materials of the glass can dissolve different components in the water. So, there are concerns regarding leaching of chemicals from the bottle into the water, making the material of the bottle into an important consideration. Researchers investigated on the potential leaching from a wide range of plastic water bottles [2-3]. Concerns of leaching was investigated for metal, bisphenol-A, aldehyde, phthalates [4-7]. Previous studies found Antimony (Sb) to be leaching from PET water bottles in to mineral water [8-12]. However geographical locations in addition the weather condition may play a role in the leaching of antimony.

Antimony is commonly used during polycondensation of PET [13]. PET can contain high (>200 mg/kg) concentrations of antimony [14-15]. Antimony can cause stomach ache, diarrhea, desiccation, muscle aches, shocks, anemic uremia, serious myocardial inflammation, shivering, liver necrosis [16]. Due to this, both the Environmental Protection Agency (EPA) in USA and the Council of the European Communities classify antimony and its compounds as serious contaminants. EPA classify antimony as a carcinogen. Due to concerns of health, there are regulations around the world controlling the leaching of contaminants in bottle water. The USEPA has set a maximum contaminant level goal (MCLG) for antimony 6ppb based on the best available science to prevent potential associated health problems [17]. However, the World Health Organization has a maximum level of 20ppb. Health Canada set the same standard as the US [10]. The German Federal

Ministry of Environment has set a limit of 5 ppb of antimony, whereas the Japanese drinking water standard requires levels to be less than 2 ppb [10].

There are different factors that affect the leachability from plastic bottles, in particular, PET bottles. Antimony diffusion is known as a thermally activated process [10]. Higher temperatures are more likely to enhance antimony diffusion. Different studies observed lower pH levels to be capable of increasing the antimony leaching [7-8]. However, sunlight appears to be less significant than other factors on antimony migration. Duration of the exposure also played a role in enhanced leaching for a period of six months tested on 48 different brands of bottle water [10]. Contradictory conclusions have been observed on the effect of color on the antimony migration [2, 11].

The objective of this study was to identify the presence of antimony in the different bottle water used in United Arab Emirates. Another objective of this study was to assess the factors that affect the leaching of antimony. Seven different brands of bottle water were used in this study. Standard laboratory procedures were adopted to achieve the objectives.

III. Background

Seven different brands of bottle water used in United Arab Emirates were examined for their water quality analysis. pH of the bottle water ranged from 6.8 (Bottle B) to 7.8 (Bottle A) (Fig. 1). It indicated mildly basic water to be more common than mildly acidic water. Total dissolved solids were as high as 770 ppm for bottle G. It was the water samples in a glass bottle.

There were cations to be less than 20 ppm for most bottle waters (Fig. 2). Calcium and magnesium concentrations were higher than 200 ppm for bottle F and G. Both of these were exported brands. This pattern is common for surface water sources. Sodium and potassium concentrations were relatively low, compared to calcium and magnesium.

Anion concentrations in the bottle water were mostly less than 60 ppm (Fig. 3). Sulfate concentrations reached up to 400 ppm for bottle G. Chloride concentrations were high for most other samples. It was probably due to the use of chlorine as a disinfectant during the treatment process.

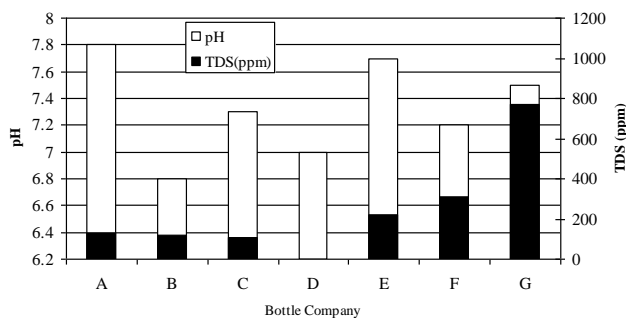


Figure 1. pH and total dissolved solids in bottle water

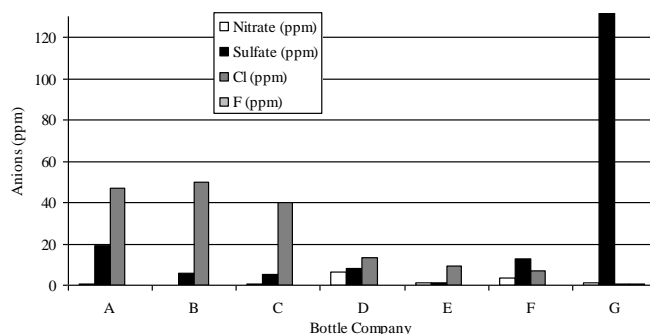


Figure 3. Anions in bottle water

IV. Materials and Methods

A. Materials

To achieve the objectives, seven different brands of bottle waters were collected from the local market, immediately the day the bottles were distributed. Three of the companies were local producers and the other four were international companies that were importing their bottle water. Bottle companies were chosen based on the popular brands that represent the typical bottle water from both local and international producers. The bottles were named A to H. The bottles A to C were local producers and the D to H were the international bottle waters. The bottles from A to G were made of PET and the bottle H were made of glass.

There were not enough information obtained regarding the source of the water and the type of treatment these water has gone through. Based on the available information, apparently, Bottle B and C were desalinated water and the rest were mineral water extracted from groundwater.

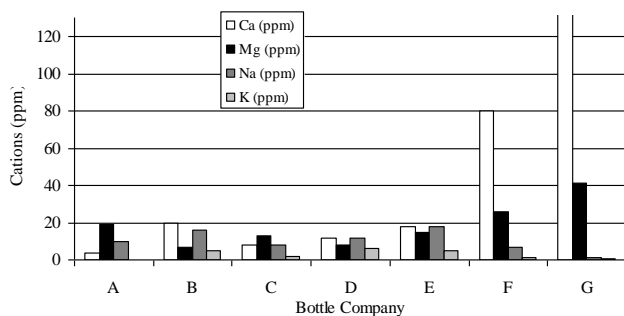


Figure 2. Cations in bottle water

Having bottle water would not necessarily protect the water from contamination.

B. Experimental Approach

All the bottles were collected from local stores from the first day of the distribution. Even though the international bottles had around two months earlier than the water produced locally. This could not be alleviated since, the local brands do not keep two month old bottles. So, the experiments could not be conducted with the same production date. The tests were conducted in two separate categories, used and unused. Used water was generated by opening the cap and then consuming small amount of water. The unused bottles were unopened and unused as well.

All the bottles were stored at three different temperatures (0°C, 24.5°C and 50°C). These three temperatures were chosen to represent water in the freeze, water inside AC controlled room environment and the last represent the high temperature during summer months.

For bottle B and C, experiments were conducted at zero days (immediately after the bottles were procured) to represent immediate water quality (blank). To investigate the effect of duration of exposure, used bottles were controlled at those temperatures for 18 days and the unused bottles went through the experiments until 10 days. Once the experiments were conducted, the samples were collected and tested for antimony concentrations in a certified laboratory.

C. Analytical Technique

Samples were analyzed for antimony based on standard testing procedures for low range concentrations. Tests were conducted on inductively coupled plasma mass spectrometer (ICP-MS). The samples were digested first using HNO₃. Standard RF power and gas flow rate was used during operation of ICP-MS.

V. Results and Discussions

Experimental results indicated the presence of antimony in almost all the samples for all the bottles tested (Fig. 4 and 5). Compared to the results of antimony concentrations at zero days, the results did not indicate any specific pattern for antimony leaching (Fig. 6). In general, the bottle C had the

lowest and the bottle F had the highest antimony concentrations among all the bottles tested.

There were no specific pattern among the local and exported brands of water even though exported brands had longer days of exposure compared to the local brands. The antimony was potentially leaching from the PET bottles. The conclusion was consistent with previous studies conducted on antimony leaching from PET bottles [8-11]. However, the concentrations of antimony in this study were higher than the concentrations of antimony observed in the previous studies. The concentrations were also higher than many regulations, set around the world for antimony. The glass bottle G did not indicate any specific advantage over the other plastic bottles. The sample size was small (one brand of glass bottle) and therefore, the authors could not draw any conclusion regarding this.

A. The Effect of Temperature on Leaching

Based on the experimental results (Fig. 4 and 5), increasing temperatures increased antimony concentrations in most of the bottle waters tested, except for samples related to unused water in bottle G. For the case of unused bottle G, the samples at 0°C were contaminated and therefore, the pattern for those samples were inconclusive. Otherwise, for both used and unused bottles, an increase in temperature showed an increase in antimony leaching in the water. At a temperature increase from 0°C to 24.5°C, maximum increase of 12.5% and 14.3% were observed for used and unused bottles consecutively. For a temperature increase of 0°C to 50°C, maximum increase of 18.8% and 14.3% were observed for used and unused bottles consecutively. The results were consistent with previous studies [8, 11]. However, comparison with the results in those studies revealed that temperatures up to 60°C in those studies had lower antimony concentrations than this study. In addition duration of exposures were much shorter than the current study. It could be the reason for the deviation of the results. It could also be due to the different qualities of PET being used in the middle east.

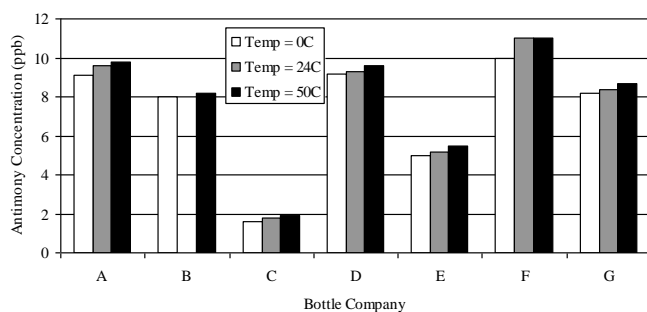


Figure 4. The effect of used water on leaching at different temperatures

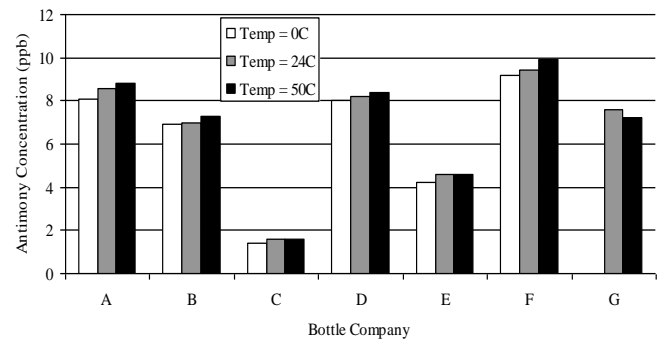


Figure 5. The effect of unused water on leaching at different temperatures

B. The Effect of Used Water on Leaching

Based on the experimental results (Fig. 4 and 5), used bottles consistently leached more antimony than the unused bottles. At 0°C, used bottles contained on an average 14.3% more antimony concentrations than the unused bottles. Similar patterns continued for other temperatures as well. At 24.5°C, antimony concentrations in used bottles were 13% higher than the concentrations in unused bottles. For the case of bottles at 50°C, the increase in concentrations were 15.5%. Previous studies did not explore the potential of using the bottle water on antimony leaching. However, since the experimentations were not uniform for all the bottles, further experimentations are needed to explore this aspect further.

C. The Effect of Exposure Time on Leaching

The analysis was limited since only two bottles (Bottles B and C) were examined at zero days, hence giving at least three dataset to examine the effect of duration of exposure. Antimony concentrations in the bottle appeared to have increased with an increasing duration of exposure for the bottle B (Fig. 6). However, for bottle C, duration of exposure reduced the antimony leaching (Fig. 6). It is unusual and contrary to common perception that increasing duration of exposure would increase the antimony concentration. The concentrations in bottle C were low and it falls within the percentage of error that the test was conducted. This is contradictory to previous studies as well [8, 11]. Those studies consistently observed an increase in antimony concentrations with an increase in duration of exposure. However, looking at all the bottles (including bottle C) for the two timeframes (10 and 18 days), antimony concentrations were higher in 18 days than in 10 days. It is consistent with previous literatures [8, 11]. Both the used and unused bottles showed similar trend on the effect of duration of exposure on the antimony leaching.

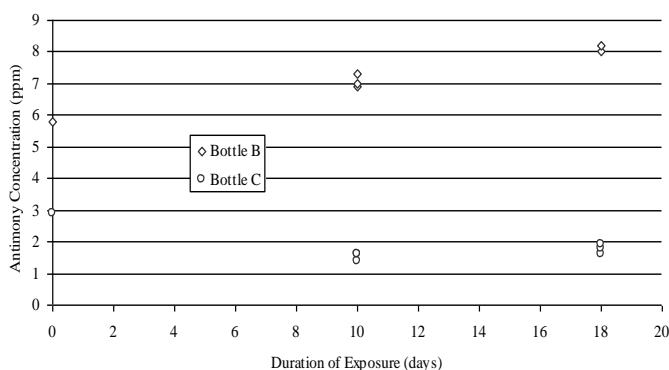


Figure 6. The effect of duration of exposure on leaching

VI. Conclusions

The study concluded that the PET bottles leach antimony to the water that it holds. Antimony concentrations were higher than the studies conducted in Europe and North America. Based on the experimental results, increase in temperature increased the antimony leaching in bottle water. The effect of using the bottle water on the antimony leaching was not conclusive. But, it showed a tendency to increase antimony leaching with used bottles than unused bottles. The effect of duration of exposure were mixed due to lack of enough data points.

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References

- [1] K. Stambaugh, "What type of Water Bottle should we be using?", Bottled Water, 2010, Page 3 – 6, (<http://www.back2tap.com/wp-content/uploads/2010/11/Bottled-Water-Facts-Updated-2010.pdf>)
- [2] C. Reimann, M. Birke, and P. Filzmoser, "Bottled drinking water: Water contamination from bottle materials (glass, hard PET, soft PET), the influence of colour and acidification", Applied Geochemistry, vol. 25, 2010, pp. 1030-1046.
- [3] P. Schmid, M. Kohler, R. Meierhofer, S. Luzi, and M. Wegelin, "Does the reuse of PET bottles during solar water disinfection pose a health risk due to the migration of plasticisers and other chemicals into the water?", Water Research, vol. 42, 2008, pp. 5054-5060.
- [4] D. Amiridou, and D. Voutsas, "Alkylphenols and phthalates in bottled waters", Journal of Hazardous Materials, vol. 185, 2011, pp. 281-286.
- [5] J. Bošnjir, D. Puntari, A. Gali, I. Škes, T. Dijani, M. Klari, M. Grgi, M. urkovi, and Z. Šmit, "Migration of phthalates from plastic containers into soft drinks and mineral water", Food Technology and Biotechnology, vol. 45, 2007, pp. 91-95.
- [6] A. Dabrowska, A. Borcz, and J. Nawrocki, "Aldehyde contamination of mineral water stored in PET bottles", Food Additives and Contaminant, vol. 20, 2003, pp. 1170-1177.

- [7] X. Cheng, H. Shi, C.D. Adams, and Y. Ma, "Assessment of metal contaminations leaching out from recycling plastic bottles upon treatments", Environmental Science and Pollution Research, vol. 17, 2010, pp. 1323-1330.
- [8] S. Keresztes, E. Tatár, V. G. Mihucz, I. Virág, C. Majdik, and G. Záray, "Leaching of antimony from polyethylene terephthalate (PET) bottles into mineral water", Science of The Total Environment, Vol. 407, Issue 16, 1 August 2009, pp. 4731-4735.
- [9] S. S. Andra, K. C. Makris, J. P. Shine, and C. Lu, "Co-leaching of brominated compounds and antimony from bottled water", Environment International, Vol. 38, Issue 1, January 2012, pp. 45-53.
- [10] W. Shoty, M. Krachler, and B. Chen, "Contamination of Canadian and European bottled waters with antimony from PET containers", Journal of Environmental Monitoring, vol. 8, 2006, pp. 288-292.
- [11] P. Westerhoff, P. Prapaipong, E. Shock, and A. Hillaireau, "Antimony leaching from polyethylene terephthalate (PET) plastic used for bottled drinking water", Water Research, vol. 42, 2008, pp. 551-556.
- [12] L. Sax, "Polyethylen Terephthalate May Yield Endocrine Disruptors." Environmental Health Perspectives, Vol. 118, No. 4, 2010, pp. 445-448
- [13] K. Pang, R. Kotek, and A. Tonelli. "Review of conventional and novel polymerization processes for polyesters". Prog Polm Sci Vol. 31, Issue 11, 2006, pp. 1009-1037.
- [14] B. Duh. "Effect of antimony catalyst on solid-state polycondensation of poly(ethylene terephthalate)." Polymer Vol. 43, Issue 11, 2002, pp. 3147-3154.
- [15] K. Nishioka, A. Hirahara, and E. Iwamoto, "Determination of antimony in polyethylene terephthalate bottles by graphite furnace atomic absorption spectrometry using micro-wave sample preparation." Bull Fac Hum Life Environ Sci Hiroshima Women's Univ Vol. 8, Issue 1, 2002, pp. 35-42.
- [16] K.L. Stemmer, "Pharmacology and toxicology of heavy metals: antimony" Pharmacol Therapeut A, Vol. 1, 1976, pp. 157-160.
- [17] United States Environmental Protection Agency. "Basic Information about Antimony in Drinking Water." 2011, (<http://water.epa.gov/drink/contaminants/basicinformation/antimony.cfm>)

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