Leaching of Trace Elements, Heavy Metals and Antimony from Polyethylene Terephthalate Bottled Drinking Water in Sri Lanka

Asanka Vishwanath[1, 2] and A. C. A. Jayasundera[1,2,3] *

Postgraduate Institute of Science, University of Peradeniya, Peradeniya, Sri Lanka [1]
Department of Chemistry, Faculty of Science, University of Peradeniya, Peradeniya, Sri Lanka [2]
Division of Mathematics and Science, Missouri Valley College, Marshall, MO, 65340, US [3]

**acaj@sci.pdn.ac.lk*

Abstract - Polyethylene terephthalate (PET) bottles used in the bottled drinking water (BDW) industry possess a high potential for leaching trace elements (LTM) into the water. There is a dearth of research on LTM composition from PET resins in Sri Lanka. Thus, scientific surveillance had qualitatively and quantitatively carried out to analyze bottle-grade PET samples. Frequently imported three resin brands designated as A, B, and C were frozen and finely grounded, then studied by X-ray fluorescence spectroscopy (XRF) and microwave digestion followed by inductively coupled plasma mass spectroscopy (ICP-MS). Outcomes were found to be Sb > Ti > Ge > Mn > Zn > Co while Cu and Ca were only recorded in resin C and A respectively. A high percentage of Sb was detected due to its leading catalytic ability, and the range can be present as 174.84 to 144.78 μg g^{-1}. Yet, Ca appeared five times higher than Sb in Resin A. Considering the results and regulations, PET-bottle and BDW parameters demonstrate contradictions. In the "Food Act No. 26 of 1980", Sb is regulated at a 0.005 mg l^{-1} for the mineral drinking water category but not for BDW though both are packed in PET bottles. Similarly, Cu, Mn, and Zn were considered water quality parameters in "SLS 894" but there is no reasonable consideration of LTM from the PET resins in "SLS 1336". In addition, migration, toxic and maximum permeability limits of the above species are strongly recommended for future research concerns. This will reflect an upgraded qualitative and quantitative study of BDW in Sri Lanka.

Keywords: Polyethylene terephthalate, bottled drinking water, catalytic metals (Sb)

I. INTRODUCTION

Polyethylene terephthalate (PET) is an extensively used packaging material for the manufacturing of bottled drinking water (BDW). However, the reliability of PET-made bottles vitally depends on health and safety concerns. To that end, LTM contained in the PET including Zn, Mn, Co, Cu, Ge, Ti, and Sb may potentially be leached into the water at BDW usage. During the synthesis of PET, these elements were anxiously chosen to catalyze the polymerization process [1]. Therefore, manufacturer-wise element composition may vary. Still, Sb appeared to be an abundant choice. As a consequence, a strong regulatory background is essential to control consumer safety. In contrast, the Sri Lankan regulatory framework, "Food Act No. 26 of 1980" [2], "SLS 894" (BDW water standard) [3], and "SLS 1336" (PET packaging standard) [4] are the key guidelines for the BDW industry. However, there is no adequate coverage and recognition of some of these trace elements in the above standards.

A. Problem statement

National PET requirement accounts for hundreds of thousands of US dollars' worth of annual imports yet there is a dearth of investigations regarding the trace element composition of PET resins and the credibility of our current regulatory framework against them. Thus, conducting surveillance study on common trace elements and Sb in frequently imported PET resin brands help to ensure the reliability of local BDW safety as well as decision-making purposes on future amendments to current regulations.

B. Objectives

Qualitative and quantitative analysis of possible trace metal species in frequently imported PET resin brands followed by determination of default Sb amounts, comparison studies of current Sri Lankan act, and standards versus analysis outcome to ensure credibility are the objectives of this study.

II. METHODOLOGY

Frequently import, three resin brands were designated as A, B, and C. Then treated with liquid nitrogen (LN) for 20 minutes and finely ground using "DJ-04 series 10A" laboratory grinder. Grounded resins were analyzed by "Fischerscope" XAn G.2.19.1 model XRF analyzer. Microwave digestion was carried out by "ETHOS EASY" milestone advanced system using 0.3g of sample with 10 mL "LOBE CHEMIE" Analytical Reagent grade 69 – 70% HNO_3. Digested solution and washings of remaining inside the vessel by ultra-pure water were filtered and directly collected into a 100.0 ml volumetric flask. The content was diluted up to the mark with the same water. 10 ml of prepared filtered solution was transferred into a 50.0 ml volumetric flask and diluted with ultra-pure water up to the mark to get 2% acidity. The analyzable sample was further filtered using a 0.45 μm cartridge and vigorously swirled using the VORTEX instrument. The prepared sample was analyzed for Sb using the “Thermo Scientific iCAP Q” ICP-MS. Blank analysis was prepared by1 ml HNO_3 acid with 49 ml ultra-pure water mixed in a 50.0 ml volumetric flask. To obtain the result following formulations were used. If the instrumental response was X μg l-1;

Sb concentration = $\frac{X\ \mu g\ /\ l}{0.3\ g}\ \times 0.05L\ \times 10$ (2.1)

Blank Sb concentration = $X\ \mu g\ l^{-1}\ \times\ \frac{50}{49}$ (2.2)

III. Results and discussion

Resin samples were treated with LN to reduce the thermoplastic effect and introduced brittle nature during the grinding process. Regards to, table 3.1; all three resin samples were positive for selected trace elements and the sequence can be represented as Sb > Ti > Ge > Mn > Zn. while Cu and Ca were detected only in resin A and C respectively. This reflects manufacturer-wise catalytic variation and thus demands specific regulations to control LTM. The amount of Sb presence in all three resins appeared to be generally higher than other elements. Thereby, it reveals the effectiveness of Sb as a catalyst in PET synthesis. Still, in Resin A; the Ca amount is approximately five times higher than the Sb amount. Default Sb level was found to be 147.62 $\mu g\ g^{-1}$, 174.84 $\mu g\ g^{-1}$, and 144.78 $\mu g\ g^{-1}$ respectively for resin A, B, and C. where values are within the mid-levels of the literature range of 100 to 300 µg g^{-1} [5]. However, considering both PET-bottle and bottled water, the quality guidelines given in regulations demonstrate contradictions and a less informative nature. Withal, Sb content in the two subcategories of bottled water explained in the "Food Act No.16 of 1980” is regulated at a 0.005 mg l^{-1} for the mineral drinking water category but not for BDW though both are packed in PET bottles. Similarly, only Cu, Mn, and Zn were regulated in the "SLS 894" as water quality parameters, but attention was not given to any LTM in the "SLS 1336" which is intended to control the quality of PET-bottle.

IV. Conclusion

Referring to the “Food Act " and “SLS 894”, except for Cu, Mn, and Zn all other trace elements were either less informative or completely absent. Forbye, it reveals the unawareness of the leaching potential of these metals since both the “Food Act " and the “SLS 894” represents the water quality of BDW. Moreover, “SLS 1336” intended to regulate PET bottle quality, observed to be in absence of any detail on trace metals. Thus, the parameters indicated in the three documents were not reflecting the true requirement. Furthermore, Sb regulated in the NMBDW section of the “Food Act " is absent in the BDW section of the same document. Hence, the “Food Act" is unable to identify PET bottles as a potential source of metal contamination. Based on a future study regarding migration levels of the above catalytic metals and their toxic levels, maximum permeability levels have to be set. Finally, a proper amendment is required to cover major trace elements in BDW standards to sustain continuous quality management of the industry.

References

[1] E. Kishi *et al.*, “Determination of various constituent elements of polyethylene terephthalate bottles used for beverages in Japan,” *Packag. Technol. Sci.*, vol. 33, no. 4–5, pp. 183–193, 2020.

[2] Food Advisory Committee, *Food Act No 26 of 1980 (Bottled or Packaged Water)*, vol. 63, no. 5. Sri Lanka: Minisry Of Health, Sri Lanaka, 2005.

[3] Sri Lanka Standard Institution, *Specification for Bottled(Packaged) Drinking Water SLS 894*. Sri Lanka, 2003.

[4] Sri Lanka Standard Institution, “SLS 1336 : 2017 Single use containers made of polymeric materials for packaging of drinking water.” SLSI, pp. 1–25, 2017.

[5] E. M. Aghaee *et al.*, “Effects of storage time and temperature on the antimony and some trace element release from polyethylene terephthalate (PET) into the bottled drinking water,” *J. Environ. Heal. Sci. Eng.*, vol. 12, no. 1, pp. 1–7, 2014.

Table 3.1: Comparison of trace elements with regards to acts and standards of Sri Lanka

Parameter	Food act No.26 (mg l^{-1} max)		SLS 894:2003 (mg l^{-1} max)	SLS 1336:2017 (mg l^{-1} max)	Abundance (%) by XRF			ICP-MS results (µg g^{-1})		
	BNMDW	BDW			Resin A	Resin B	Resin C	Resin A	Resin B	Resin C
Sb	0.005	N/A	N/A	N/A	0.76	0.92	0.85	147.62	174.84	144.78
Mn	0.5	0.5	0.5	N/A	0.31	0.42	0.39	-	-	-
Zn	N/A	3.0	3.0	N/A	0.09	0.17	0.08	-	-	-
Ti	N/A	N/A	N/A	N/A	0.44	0.77	0.59	-	-	-
Ge	N/A	N/A	N/A	N/A	0.42	0.46	0.50	-	-	-
Co	N/A	N/A	N/A	N/A	0.38	0.41	0.34	-	-	-
Cu	1.0	1.0	1.0	N/A	N/D	N/D	0.78	-	-	-
Ca	N/A	N/A	N/A	N/A	3.74	N/D	N/D	-	-	-

N/D = Not detect, N/A = Not available, BNMDW = Bottled Natural Mineral Drinking Water, BDW = Bottle Drinking Water, SLS = Sri Lankan Standards.