

Migration of Phthalic Acid Esters from PET Water Bottles: Implications of Reusing Empty Bottles for Storing Ambient and Warm Water for Subsequent Consumption**Perera P.H.G.^{1*}, Bandara N.J.G.J.¹, Danushika U.A.A.G.², Jayaweera M.W.^{2*}, Somaratne M.C.W.³, Thathsara S.K.T.³, Gunawardana B.², Manatunge J.M.A.²**¹*Department of Forestry and Environmental Science, University of Sri Jayewardenepura, Sri Lanka*²*Department of Civil Engineering, University of Moratuwa, Sri Lanka*³*Department of Polymer and Chemical Engineering, University of Moratuwa, Sri Lanka***hasini.perera.hp@gmail.com***Abstract**

Phthalic Acid Esters (PAEs) are a group of emerging contaminants that are widely used as a plasticizer in manufacturing polyethylene terephthalate (PET) bottles to improve softness, flexibility, durability, longevity and workability. Some of PAEs have, however, been identified to be carcinogenic. People in Sri Lanka often use the PET bottles for storing ambient and warm water repeatedly for subsequent consumption. With the reuse of bottles, PAEs are easily detached and migrated to the water, because of weak chemical bonding between the main polymer structure and PAEs. However, no comprehensive studies on the migration of PAEs from the material of the bottle to water in the context of different reuse efforts of storing ambient and warm water have been reported; hence, this study. Ten different commercial brands of PET bottles were selected. The initial storage temperature of selected PET bottles was measured and they were under room temperature conditions. It was ensured that they were not exposed to the direct sunlight under storage condition. Three bottles of each brand were filled with warm water (initial temperature $60\pm 2^\circ\text{C}$) and another three with the same water at room temperature ($27\pm 2^\circ\text{C}$) and kept for 12 hours. The PAE levels of the water in the bottles were analysed by Gas Chromatography-Mass Spectrometer (GC-MS) using EPA method 8061A. Six PAEs, bis (2-ethylhexyl) phthalate (DEHP), dibutyl phthalate (DBP), diethyl phthalate (DEP), benzylbutyl phthalate (BBP), dimethyl phthalate (DMP) and di-n-octyl phthalate (DnOP) were quantified. The same bottles were subject to PAE analyses for six consecutive reuse events. Characterisation of PET bottles of two brands was done to investigate the thermally induced structural changes using the Fourier-Transform Infrared spectroscopy, Scanning Electron Microscopy, Differential Scanning Calorimetry and X-Ray powder Diffraction. In general, DMP, DEP, DnBP, BBP, and, DnOP were not detected in the water under room temperature ($27\pm 2^\circ\text{C}$) and $60\pm 2^\circ\text{C}$. Among the six different PAEs tested, DEHP was the only PAE that has leached out at room temperature and at 60°C for different brands. Results manifested the maximum and minimum levels of DEHP being migrated after each event of reuse at room temperature to be $82\pm 4\ \mu\text{gL}^{-1}$ and $17\pm 2\ \mu\text{gL}^{-1}$, respectively, and the maximum and minimum levels of DEHP leached out at $60\pm 2^\circ\text{C}$ were $133\pm 3\ \mu\text{gL}^{-1}$ and $26\pm 2\ \mu\text{gL}^{-1}$, respectively. Characterisation studies indicate that there were structural changes with each reuse event at $60\pm 2^\circ\text{C}$ in comparison to those at room temperature. In conclusion, this study provides evidence of migration of DEHP from the material of the bottle to water in the reuse efforts for storing warm water for subsequent consumption. Hence, regulating the quantities of DEHP to be added during the manufacturing process of PET bottles and the reuse efforts of empty PET bottles is of utmost importance to minimize the potential health and environmental impacts.

Keywords: DEHP, PAE, Polyethylene terephthalate, Reuse, Temperature