

## The Effect of Different Storage Conditions for Refilled Plastic Drink Bottles on the Concentration of Microplastic Release in Water

Mustafa Dheyaa Mohamed Hadeed<sup>1</sup> and Kossay Kamaldeen Al-Ahmady<sup>2</sup>

<sup>1</sup>M.Sc. Student, College of Environment Sciences and Technology, University of Mosul, IRAQ.

<sup>2</sup>Professor of Environmental Engineering, University of Mosul, IRAQ.

<sup>1</sup>Correspondence: mustafa.20evp20@student.uomosul.edu.iq



www.jrasb.com || Vol. 1 No. 4 (2022): October Issue

Received: 03-09-2022

Revised: 24-09-2022

Accepted: 04-10-2022

### ABSTRACT

The demand for water consumption in plastic bottles has increased in recent years. Most consumers of this type of water think that it is well sterilized. This is due to the high level of propaganda that accompanies its production regarding the extent of its validity. It is also common to reuse these bottles for several times by filling it frequently for different uses, and their use for preserving and storing different types of liquids.

This study is conducted for the purpose of detecting and evaluating the pollution caused by microplastics in plastic bottles filled with water for one time, as well as water bottles reused more than once. Moreover, it evaluates the effect of storage period and conditions on the abundance of microplastics when bottles are stored for different times and under the influence of different conditions (shade, cooling and freezing). The results of the study show the presence of fine microplastics in open bottled water exposed to different storage conditions. The highest concentration of microplastics is obtained in storage conditions under the influence of shade with a limit of (1050) microplastics / liter, while the lowest concentration is obtained at around (20) microplastics / liter. The highest concentration of microplastics is obtained in storage conditions under the influence of cooling, with a limit of (850) microplastics / liter, whereas, the lowest concentration is obtained in the range of (16) microplastics / liter. Under the influence of freezing, the highest concentration of microplastics is obtained in the range of (648) microplastics / liter, while the lowest concentration is obtained in the range of (20) microplastics / liter.

Through the results, it is noted that there is a noticeable gradient in the concentration of microplastics for the samples that have been studied. It is also noted that there is an increase in the concentration of microplastics with repeated reuse during continuous periods of time. It is recommended that more studies and research be conducted indicating the extent of the impact of microplastics on human health in addition to other environmental components with emphasis on finding treatment means to get rid of microplastics.

**Keywords-** microplastics, plastics bottled water, various exposure conditions.

### I. INTRODUCTION

The consumption of fresh water for drinking purposes is an essential and vital part of human life. It works to increase the global demand for bottled water in recent years, which has made it the fastest growing element in the beverage economy around the world (Doria, 2006). The massive consumption of plastic bottled water results in great pollution due to plastic bottles that become a burden on the ecosystem, and that do not decompose easily if left untreated. The Environmental Protection Agency (EPA, 2013) reports that a large

proportion of Americans throw away 28 billion plastic bottles each year, which has led to an environmental disaster that requires spending huge amounts of money to get rid of. This is what has prompted those concerned to spread the culture of recycling which works to reduce financial cost and reduce the damage to the environment. The technology of recycling waste and especially plastic waste is one of the environmentally friendly and harmless techniques, in addition to its low economic cost. Despite this, it has been observed that the behavior of most consumers of plastic water bottles is that when reusing it, they do not make any effort to clean, which may result in

microbial pollution that harms human health. Due to its light weight and durability, the use of plastic has increased in various manufacturing processes, including the production of water bottles, but it is noted that the properties of molecular plastic change slightly if exposed to certain conditions, which leads to its decomposition into small and minute particles that spread in the water (Kawecki et al., 2018; Meng et al., 2020). Various types of microplastics have been detected in the environment, as well as in foods and drinking water (Crawford and Quinn, 2017; Mintenig et al., 2019; Sharma and Chatterjee, 2017; Van Cauwenberghe and Janssen, 2014; Yang et al., 2015).

Picheta (2018) confirms by his study of the presence of plastic in human stool. Moreover, microplastics can act as a carrier of many toxic pollutants, including heavy elements that threaten human health (Wang et al., 2017; Tang et al., 2020 and Barboza et al., 2018). Several studies and researches have shown that the composition of many microplastics in bottled water is similar in composition to the material of the plastic bottle and its lid (Bergmann et al. 2015). These microplastics are produced and released into the water due to exposure to photo-oxidation processes by sunlight when exposed which is transmitted to the digestive system of humans when they use and drink this water (Julienne et al, 2019). In 2019, the World Health Organization (WHO) called on scientists from all countries to strengthen research for the impact of microplastics on human health (Laskar and Kumar, 2019; WHO, 2019). Therefore, with the increasing inputs of microplastics into the environment, microplastic pollution has become a global problem and concern (de Souza Machado et al., 2018). As microplastics have attracted increasing attention worldwide, previous reports have focused on the source, distribution, fate and toxicity of microplastics. However, many of these studies and reviews are not entirely comprehensive, and most have focused solely on marine environments. Microplastics come in a variety of sizes, shapes, and colors with some being spherical, fibrous, or random. According to Crawford and Quinn, (2017), a microplastic is generally defined as any piece of plastic in size over its longest dimension length, whose standard size can be classified into large plastic ( $\geq 25$  mm), intermediate plastic ( $< 25$  mm to 5 mm), elastomers ( $< 5$  mm), microplastics in addition to small microplastics (smaller than 5 mm to 1  $\mu$ m), and finally nanoplastics (smaller than 1  $\mu$ m). On the other hand, microplastics can also be categorized as (granules, microbeads, fragments, fibers, film, and foam). Fragments are solid particles of angular and irregular shape, and some microplastics are thick with sharp, twisted edges. (Wang, et.al, 2020). The current study aims at verifying the presence of microplastics in water-filled plastic bottles used for the first time, and to verify the levels of microplastics at each reuse of the plastic bottle and under the influence of different storage conditions.

#### **Collection and preparation of samples:**

Samples of bottled water are randomly collected starting from November 2021 until the end of March 2022 from the local markets of Mosul city within the production validity period to avoid the possibility of changing some water properties with the expiration date of the water plastic bottles. The most commonly used plastic bottles are selected with a volume of 0.5 liters. The plastic bottle samples are divided into two groups that included the first group, which consists of four samples that are reused plastic bottles filled with filtered water (0.45 micron) to be purified from microplastics for the purpose of ascertaining the source of microplastics generation for the reused bottles. The second group consists of four standard samples (bottled water) as the standard control group, for the purpose of comparing with samples of reused bottles (Duwiejuah et al., 2013). The two groups are placed under the influence of three different storage conditions (under the influence of shade (15-23) C, the influence of cooling in the refrigerator at a degree of (4) C, finally under the influence of freezing at a temperature of (-15) C (Alak, et. al, 2021). The plastic bottles are re-used during different periods of time, where the first re-use occurred after three days. The first reuse of plastic bottles starts at 3 days of storage, up to the 10th reuse at 30 days of storage, in all conditions, in addition to the standard control group (Chisupakitsin, et al. 2019).

#### **Sample Analysis**

The outer surface of the bottled water samples is cleaned to avoid plastic contamination and to eliminate the possibility of contamination from the outside (Kankanige & Babel, 2020). This indicates the degree of prevention through the use of a cotton lab coat and surface wiping steps as confirmed by Wesch et al. (2017). The laboratory preparations include clothing, work surface, utensils should be free of microplastics to avoid contamination, and cotton coats are worn throughout the microplastics analysis and during sample analysis to prevent sample contamination. Lab coats are used as a single protection measure to avoid fabric fibers from seeping out of clothing and contaminating samples since cotton fibers are abundant in indoor air, including in the laboratory. Hence, it is highly recommended that microplastics analysis work surfaces are cleaned intensively which indicates the presence of airborne particles from the lab coat and the indoor environment and that the glass Petri dishes are washed thoroughly between experiments (Vianello et al., 2019). A certain volume of the sample is filtered by a vacuum filtering device using filter papers of the type (Grid Membrane, Diameter 47 mm, Size (0.45)  $\mu$ m). The filter papers are placed in sterile glass petri dishes to dry at room temperature, taking into account their tightness to avoid laboratory contamination. The optical analysis is performed using a Wild m7a Microscope with an eyepiece magnification (10X) and an objective lens of (40X) magnification (Kosuth et al., 2017; Hidalgo- Ruz et al., 2012).

## II. RESULTS AND DISCUSSION

The results of the plastic bottles stored under the influence of the shade show that the number of plastic particles begin to increase upon re-use six times, reaching (140) microplastics / liter and for the sample used once by (75) microplastics / liter. The number of microplastics continued to multiply by a large amount during the seventh, eighth and ninth reuse with the number of microplastics reaching (640,410,200) particles / liter, respectively. In contrast, the number of microplastics for the one-time sample reached (100,100,90) microplastics / liter. As for the tenth use, the number of microplastics reached (1050) microplastics/ liter, compared to (120) microplastics / liter for that used for one time, as shown in Figure (1). The reason for the increase in the values of the microplastics of the sample during storage in the shade can be attributed to the effect of chlorine, as it is an

effective element in scratching the plastic bottle over time, which may lead to the deterioration of the polymers that come into contact with these media due to oxidation, as two main factors act: The first is the analysis of the outer layer that deals with water, and this is observed as a remarkable behavior during the use of plastic bottles that are used and stored for a long period of time, as a weakly bonded emulsion layer is formed with the basic substance of the plastic material, which is one of the first items to decompose plastics in water. As for the second factor, a decomposition process may arise (a high probability) in that the permeability is to be inward, which leads to the opening of channels within the structure of the plastic material. This process is very similar to the process of creating porous holes, which after a while will lead to a kind of swelling in the structure of the plastic material, resulting in the generating of internal pressure that increases the release of microplastics from outside its structure (Samarth, & Mahanwar, 2021).

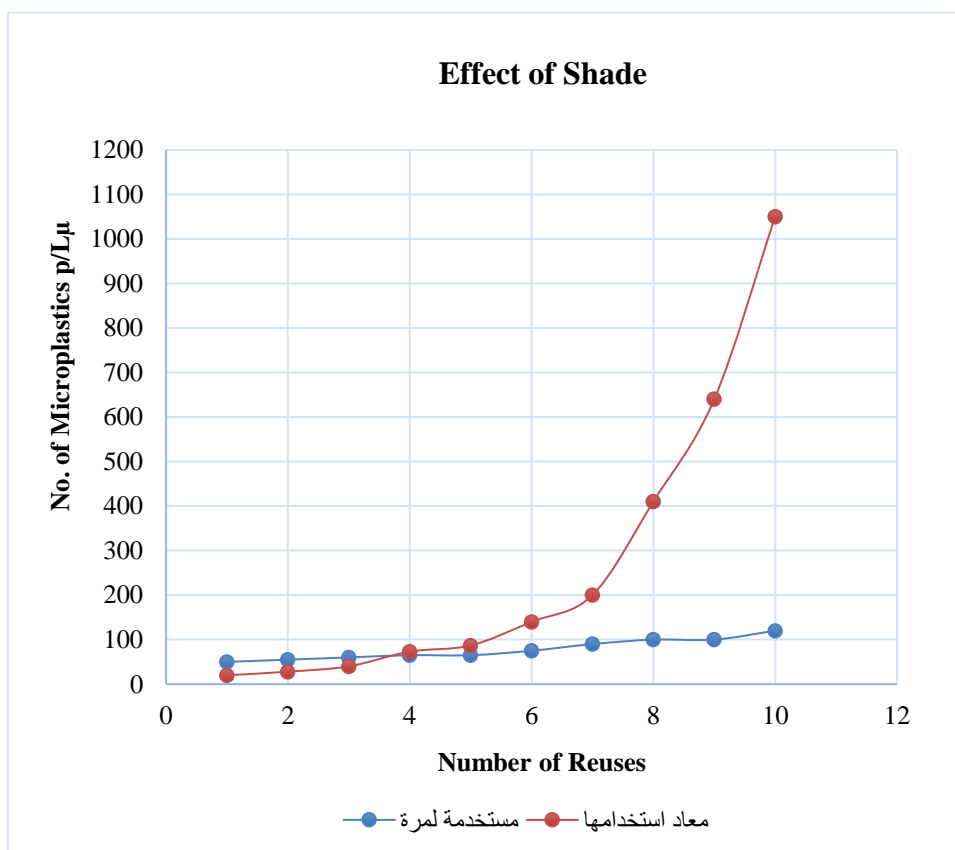


Figure 1: Effect of reusing plastic water bottles on the concentration of plastic particles when storing in the shade

The results show that the number of microplastics under the influence of cooling begins to rise significantly upon re-use 6 times, reaching (150) microplastics / liter, compared to (50) microplastics / liter for the sample used for one time. It is noted that the number of microplastics doubled significantly when the plastic bottle is re-used for seven times, reaching (320) microplastics / liter, and for the one-time sample reaching

(100) microplastics / liter. The increase continues clearly until it reaches its highest value when the tenth reuse amounts to (850) microplastics / liter compared to (120) microplastics / liter for the single-use sample (See Figure 2). This indicates the deterioration of the plastic bottle during repeated use, which has had a clear effect through the process of emptying, filling and exposure to mechanical stress.

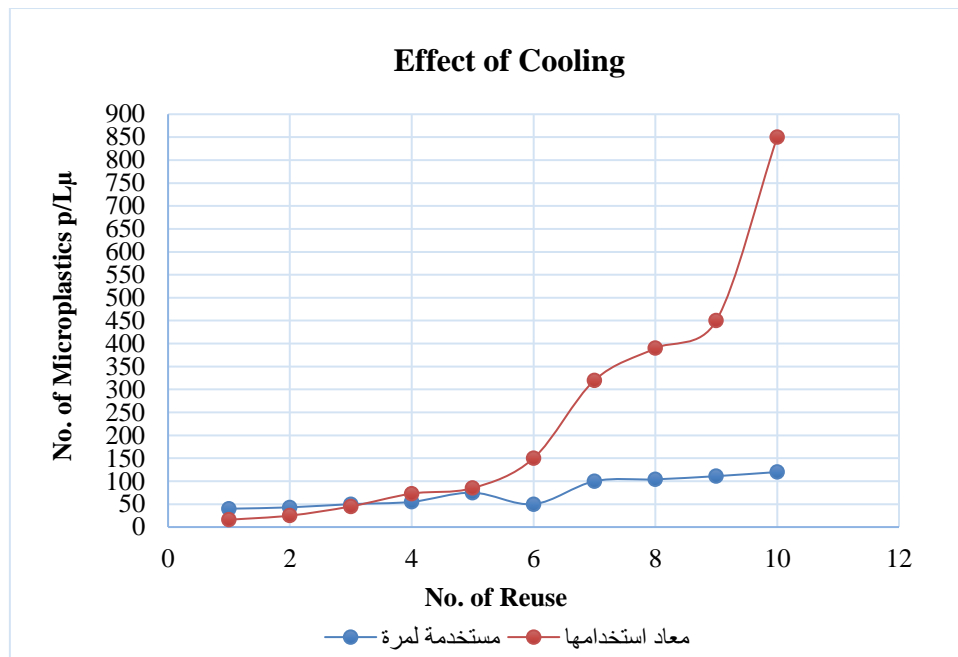


Figure 2: The effect of reusing plastic water bottles on the concentration of microplastics when stored in cooling

As for storage under freezing conditions, the results show that there is an increase in the number of microplastics when reusing plastic bottles. An increase is noted when using for (5) times, as the number of microplastics reached (110) microplastics / liter, while the number of microplastics for the sample used for one time reached (56) microplastics / liter. The number of microplastics continued to increase exponentially upon reuse by (8) times, as the number of microplastics reached (350) microplastics / liter, and for the one-time sample it amounted to (120) microplastics / liter. The number of microplastics upon the ninth use of the plastic bottle reached (405) microplastics / liter compared to (119) microplastics / liter for the sample that is used once. The bottle began to collapse upon the reuse of (10) times, and the number of microplastics increased dramatically reaching (648) microplastics/l, as shown in Figure (4-8).

The reason for the increase in the release of microplastics from the process of freezing and melting leads to an increase in the mass of the bottle during freezing and its return to its normal size upon melting. The continuation of the process and repeated re-use creates pressure on the bottle body, which may lead to cracks in the polymeric matrix and the loosening of the bonds that are connected with each other. This leads to its deterioration and the release of microplastics. When looking at the process of cooling plastic materials accurately, we have to prove the following points. Cooling is divided into two main parts: the first is normal cooling, and the second is called deep cooling (freezing). The basic difference between the two types of cooling can be explained as follows: when plastic materials are cooled to a temperature not exceeding zero degrees Celsius, this stage leads to an effect only on the surface layer of the plastic materials, because the thermal conductivity of

plastic materials in general is weak conduction and the temperature within this range is limited to parts of a micrometer of the surface of the plastic material that is being dealt with. Thus, the amount of influence in such a case is a superficial one (Ferrari, et al.2020). As for exposure to deep cooling (freezing), the process causes an impact on most of the depth of the plastic material, which makes all the molecules and polymeric chains in the material being affected (Kuo et al., 2020). In order to explain why the microplastics observed in the case of cooling are higher than those in the case of freezing (deep cooling), the main reason for this is due to the fact that in the case of deep cooling, all the body of the plastic material is subject to the same influence, which makes the absence of a difference between layers of the interior with the surface layers, which in turn leads to the material maintaining its relative cohesion (Pourzamani., et al. 2017). In the case of (normal cooling), there is a difference in the temperatures of the different layers of the plastic material, which makes a thermal gap between the different layers.

This gap leads to a weakening of the bonding of microplastics with each other between the different layers and leads to an increase in the number of microplastics ejected outside the structure of the plastic material (Munno et al., 2018). When comparing the shape of microplastics resulting from single-use of plastic bottles with those of bottles used for several times, it is observed that microplastics in the form of fibers and blue colored fragments of single-used bottles, which can belong to the cap of the plastic bottle, as shown in Figure (4). The majority of microplastics in reused bottles are in the form of flakes, transparent and white pellets, fibers of different sizes, and foam often generated from the body and cap of the samples.

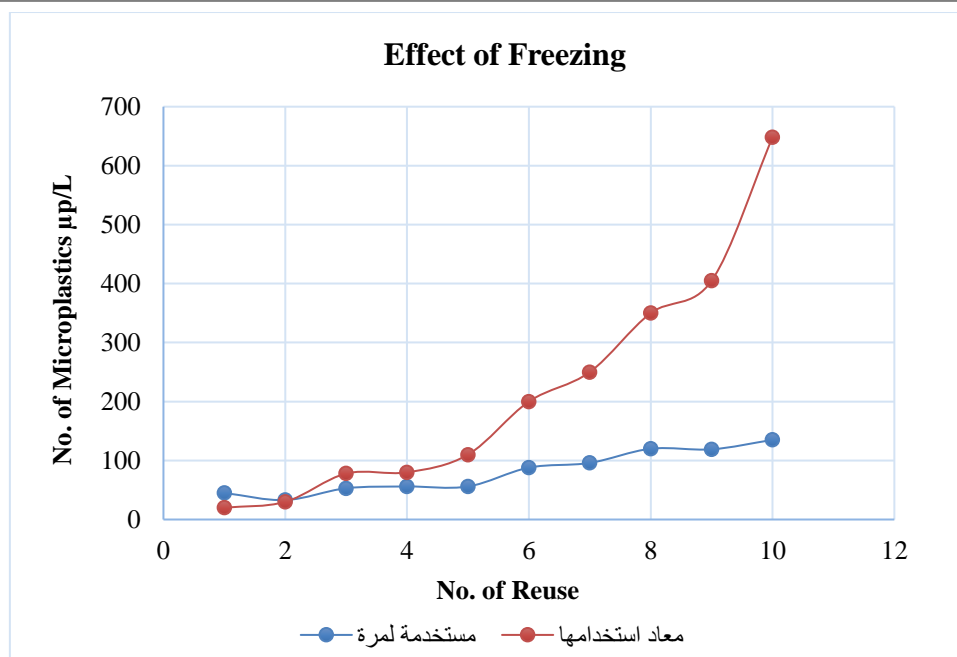


Figure (3): The effect of reusing plastic water bottles on the concentration of microplastics when stored in the freezer

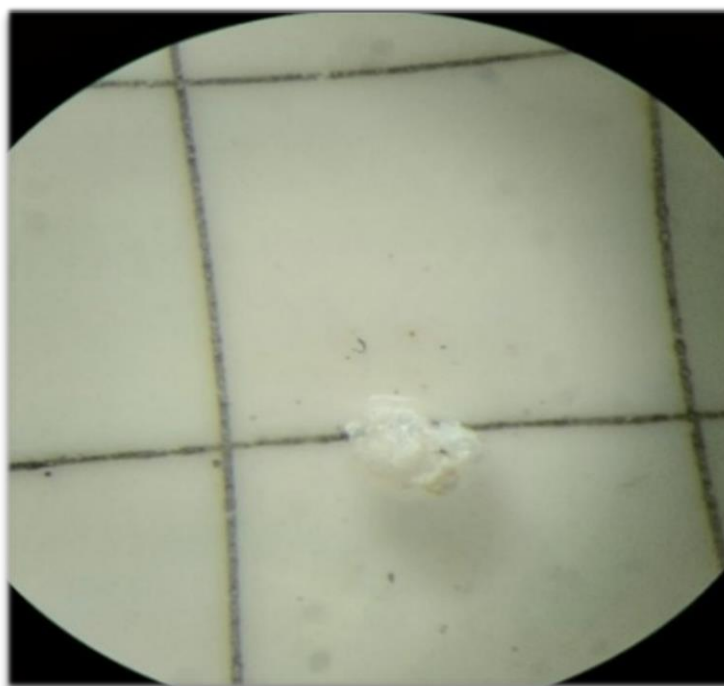


Figure (4): Shapes of microplastics of reused samples on filter paper under a microscope

When comparing Figures (1), (2) and (3), it is noted that the values of the number of microplastics during reuse of (1-5) times are very close and for all conditions. This convergence can be explained on the basis that the plastic material is still able to maintain its properties, and has the ability to resist external influences, but when plastic bottles are re-used from (6-8) times, the beginning of the collapse of the plastic material is noticed by increasing the release of microplastics. This change or

collapse is irregular depending on the nature of the influence. For some influences, the plastic material has had the ability to resist it, as opposed to not indicating its ability to resist in harsher conditions. After re-use for (8) times, rising up to (9-10) of re-use, the plastic bottles have reached the stage of collapse and aging under most of the storage conditions used in the research. The result is consistent with what is reported by Oßmann, et al (2018).

### III. CONCLUSIONS

Microplastics are found in water-filled plastic bottles, single-use and reused. The number of microplastics for plastic bottles increases at each reuse and under the influence of storage conditions (shade, cooling, and freezing). It is recommended that further studies be conducted to find out how dangerous these plastic particles are to the health of humans and environmental components.

### REFERENCES

[1] Doria, M. (2006). Bottled water versus tap water: understanding consumers preferences. *J. Water Health* 4:271–276

[2] United States Environmental Protection Agency (EPA) (2013). 10 Fast facts on recycling. Available at: <http://www.epa.gov/reg3wcmd/solidwasterecyclingfacts.htm>. Accessed 4 September 2015.

[3] Kawecki, D., Scheeder, P.R., Nowack, B. (2018). Probabilistic material flow analysis of seven commodity plastics in Europe. *Environ. Sci. Technol.* 52, 9874-9888.

[4] Meng, Y., Kelly, F.J., Wright, S.L. (2020). Advances and challenges of microplastic pollution in freshwater ecosystems: A UK perspective. *Environ. Pollut.* 256, 113445.

[5] Crawford, C., Quinn, B. (2017). *Microplastic pollutants*. Elsevier.

[6] Sharma, S., Chatterjee, S. (2017). Microplastic pollution, a threat to marine ecosystem and human health: a short review. *Environ. Sci. Pollut. R.* 24, 21530-21547.

[7] Van Cauwenberghe, L., Janssen, C.R. (2014). Microplastics in bivalves cultured for human consumption. *Environ. Pollut.* 193, 65-70.

[8] Yang, D., Shi, H., Li, L., Li, J., Jabeen, K., Kolandhasamy, P. (2015). Microplastic pollution in table salts from China. *Environ. Sci. Technol.* 49, 13622-13627.

[9] Picheta, R., 2018. Microplastics found in human stools, research finds. CNN. <https://edition.cnn.com/2018/10/23/health/microplastics-human-stoolpollution-intl/index.html>.

[10] Wang, C., Zhou, S., He, Y., Wang, J., Wang, F., Wu, S. (2017a). Developing a black carbon-substituted multimedia model for simulating the PAH distributions in urban environments. *Sci. Rep.* 7, 1-9.

[11] Tang, S., Lin, L., Wang, X., Feng, A., Yu, A. (2020). Pb (II) uptake onto nylon microplastics: Interaction mechanism and adsorption performance. *J. Hazard. Mater.* 386, 121960.

[12] Barboza, L.G.A., Vethaak, A.D., Lavorante, B.R., Lundebye, A.K., Guilhermino, L. (2018). Marine microplastic debris: An emerging issue for food security, food safety and human health. *Mar. Pollut. Bull.* 133, 336-348.

[13] Bergmann, M., Gutow, L. and Klages, M. (2015) Marine anthropogenic litter. Springer Open. doi: 10.1007/978-3-319-16510-3.

[14] Julienne, F., Delorme, N. and Lagarde, F. (2019) 'From macroplastics to microplastics: Role of water in the fragmentation of polyethylene', *Chemosphere*. Elsevier Ltd, 236. doi: 10.1016/j.chemosphere.2019.124409.

[15] Laskar, N., Kumar, U. (2019). Plastics and microplastics: A threat to environment. *Environ. Technol. Inno.* 14, 100352.

[16] World Health Organization (WHO) (2019). *Microplastics in Drinking-water* (Geneva. License: CCBY-NC-SA 3.0 IGO).

[17] De Souza Machado, A.A., Kloas, W., Zarfl, C., Hempel, S., Rillig, M.C. (2018a). Microplastics as an emerging threat to terrestrial ecosystems. *Global Change Biol.* 24, 1405-1416.

[18] Z. Wang, T. Lin, W. Chen, Occurrence and removal of microplastics in an advanced drinking water treatment plant (ADWTP), *Science of The Total Environment*. 700 (2020) 134520

[19] Duwiejuah, A.B., Cobbina, S.J. and Akrong, M.O. (2013). "Effect of Storage on the Quality of Sachet-Vended water in the Tamale Metropolis, Ghana". *Journal of Environmental Protection*, (4): 629-637.

[20] Alak, G., Köktürk, M., & Atamanalp, M. (2021). Evaluation of different packaging methods and storage temperature on MPs abundance and fillet quality of rainbow trout. *Journal of Hazardous Materials*, 420, 126573.

[21] Chaisupakitsin, M., Chairat-utai, P., & Jarusiripot, C. (2019). Degradation of polyethylene terephthalate bottles after long sunlight exposure. *Songklanakarinn Journal of Science & Technology*, 41(2)

[22] Kankanige, D., & Babel, S. (2020). Smaller-sized micro-plastics (MPs) contamination in single-use PET-bottled water in Thailand. *The Science of the Total Environment*, 717, 137232. <https://doi.org/10.1016/j.scitotenv.2020.137232>

[23] Wesch, C., Elert, A. M., Wörner, M., Braun, U., Klein, R., & Paulus, M. (2017). Assuring quality in microplastic monitoring: About the value of clean-air devices as essentials for verified data. *Scientific Reports*, 7(1), 1–8. <https://doi.org/10.1038/s41598-017-05838-4>

[24] Vianello, A., Jensen, R. L., Liu, L., & Vollertsen, J. (2019). Simulating human exposure to indoor airborne microplastics using a Breathing Thermal Manikin. *Scientific Reports*, 9, 8670. <https://doi.org/10.1038/s41598-019-45054-w>

[25] Kosuth, M., Wattenberg, E.V., Mason, S.A., Tyree, C., and Morrison, D. (2017). "Synthetic polymer contamination in global drinking water". *Orb media*.

[26] Hidalgo-Ruz, V., Gutow, L., Thompson, R. C., & Thiel, M. (2012). "Microplastics in the marine environment: a review of the methods used for identification and quantification". *Environmental science & technology*, 46(6): 3060-3075.

[27] Samarth, N. B., & Mahanwar, P. A. (2021). Degradation of Polymer & Elastomer Exposed to Chlorinated Water—A Review. *Open Journal of Organic Polymer Materials*, 11(1), pp.1-50.

[28] Ferrari, F., Esposito Corcione, C., Montagna, F., & Maffezzoli, A. (2020). 3D printing of polymer waste for improving people's awareness about marine litter. *Polymers*, 12(8), 1738.

[29] Kuo, C.C.; Chen, W.H.; Lin, Y.X.; Gao, Q.; Gian, S.J.; Xiao, C.X (2020). Effects of different fillers on the silicone rubber mold with conformal cooling channels. *Int. J. Adv. Manuf. Technology*, 108, pp.1509–1525.

[30] Pourzamani, H., Falahati, M., Rastegari, F., & Ebrahim, K. (2017). Freeze–melting process significantly

decreases phthalate ester plasticizer levels in drinking water stored in polyethylene terephthalate (PET) bottles. *Water Science and Technology: Water Supply*, 17(3), 745-751.

[31] Munno, K., et al. (2018). Impacts of temperature and selected chemical digestion methods on microplastic particles. *Environ. Toxicol. Chem.* 37 (1), 91–98.

[32] Möller, J.N., et al., 2021. Tackling the challenge of extracting microplastics from soils: a protocol to purify soil samples for spectroscopic analysis. *Environ. Toxicol. Chem.* 41 (4), pp.844–857.

[33] Oßmann, B., Sarau, G., Holtmannspotter, H., Pischetsrieder, M., Christiansen, S.H. and Dicke, W. (2018). "Small-sized microplastics and pigmented particle in bottled mineral". *Water Research*, (4): pp.307-16.