PLASTIC RESIDUES IN SEA SALT OBTAINED FROM THE LEBANESE MARKET

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Abstract

Salt is one of the most used food additives not only to improve taste, but also for preservation and different functional properties. In recent years, there is a notable growing body of evidence demonstrating that salt is contaminated by extraneous substances such as microplastics. Many substances with adverse health effect such as heavy metal and endocrine disruptors can adhere to microplastics or enter as inherent part of the plastic end product. Plastics end up in body of water such as oceans and seas. This is the case in Lebanon where very little portion of plastics are recycled and the rest is thrown in dump sides near the Mediterranean Sea. Within this scope, few studies have been conducted to assess the contamination of salts with microplastics in the Mediterranean Sea. To the best of our knowledge no similar study has been conducted in Lebanon. Therefore, the aim of the study was to assess the presence of microplastics in sea salt either sold or produced in Lebanon. To meet this objective, twenty-seven salt brands from the Lebanese market were collected and analyzed between September 2019 and December 2020. Determination of the presence of different types of plastic polymers was performed using Fourier-Transform Infrared (FTIR) techniques with three replicates for each sample. This study showed seven types of microplastic polymers including polypropylene, thermoplastic elastomer, gomma plastificata, polyester, polyethylene, plasticizer and BBP 12-4. Accordingly, this study highlights the presence of microplastics and their implications of the human health.

Keywords: sea salt, microplastics, Lebanon, polymers

Chapter I: Salt function and plastic use, and its adverse effect on environment and human health

I.1. Physiological, nutritive importance, and use of salt

Salt can be extracted from mines or from sea through evaporation. The focus will be in this study on sea salt. Salt is composed of sodium chloride, a compound that helps regulate fluid balance and blood pressure in the body (Streit, 2019); therefore, its consumption in moderate quantities is important for human health and good body functioning. Salt is a highly consumable ingredient worldwide, so the World Health Organization recommends a salt intake of 5 g per day for a healthy adult (World Health Organization, 2012). However, the amount consumed is 8–11 g per day for Europe and 10 g per day globally. it is used for seasoning and preservation purposes in food (Mozaffarian *et al.*, 2014).

I.2. Contamination of salt with foreign substances

Oceans and seas as many body of water are important sources for sea salt but also subject to heavy sources of pollution like non-sorted solid waste, hazardous waste, etc. One of these main contaminants are plastics that will decompose and degrade into smaller particle of micro and nano-size. The origin of plastics can be the different sources such as packaging, clothing, construction, automotive, electronics (EU, 2018). One important item is biodegradable plastic that will not be dissolved entirely rather than they will disintegrate into microscopic plastic beads (Parta *et al.*, 2019).

They will enter marine ecosystem and reach through the food chain the human beings. Plastics will transfer with them other pollutants such as persistent organic pollutants in the ecosystem and to humans' beings (Sharma *et al.,* 2017).

I.3. Types of polymers of plastic polymers

Plastic is the term commonly used to describe a wide range of synthetic or semi-synthetic materials that are used in a huge and growing range of applications such as wood, paper or wool (Association of Plastics Manufacturers, N.A). Also, it is an essential component of many items, including water bottles, combs, and beverage containers. There are seven different types of plastics: polyethylene terephthalate (PET), high density polyethylene (HDPE), polyvinyl chloride (PVC), low-density polyethylene (LDPE), polypropylene (PP), polystyrene or Styrofoam (PS) and miscellaneous plastics (includes: polycarbonate, polylactide, acrylic, acrylonitrile butadiene, styrene, fiberglass, and nylon). Each type has specific chemical composition, structure, characteristics and criteria which helps formation of different products. Since 1950 the number of plastics dumped into ocean has increased dramatically (UNEP, 2016), there are between 4.8 and 12.7 million metric tons entering the oceans every year (Jambeck *et al.*, 2015). Softened rubber or "Gomma Plasitificata" that may be made from different polymers such as polyvinylchloride, polystyrene as fire retardants (Luna *et al*., 2018), and it enters in its composition plastic softer such as phthalates (Corea-Téllez *et al.,* 2008). These two latter are classified as endocrine disruptors with adverse effect on the ecosystem and the human health (Benjamin *et al.*, 2017).

I.4. Plastic waste in the Mediterranean and Lebanon

The Mediterranean Sea is ranked the 4th after North Pacific, Indian and North Atlantic oceans with quantity of plastic waste floating on surface (Ritchie *et al*., 2018). Three Mediterranean countries account for two-thirds of plastic leaked into nature which are Egypt with 42.5% from which Lebanon imports large amounts of salts, Turkey with 18.9% and Italy with 7.5%. According to the

European Parliament, plastics in Western Mediterranean come from Spain and in Eastern Mediterranean from Turkey (European Parliament, 2019). In addition to that, a recent study has found that the shores in Cyprus have second highest amount of microplastics after Hong Kong (Duncan *et al.*, 2018). Microplastic particles detected in the study were found to be from highest to lowest: Cyprus, Turkey, Lebanon, Libya, Palestine, Syria, Greece, Egypt, Italy, Tunisia and Malta. The presence of microplastics on shores of Cyprus from Lebanon is due to the 2015 garbage crisis that led to leakage of high amounts to the sea. These amounts migrated to different countries such as Cyprus and Italy. Therefore, populations in Lebanon are at high risk of being exposed to microplastics that have migrated from the Mediterranean Sea into salt.

Lebanon is a small Mediterranean country exposed to multi-point source contaminants. There are many marine landfills and dumpsites on the Lebanese coast where more than 90% of plastics are dumped without sorting or being recycled (GIZ, 2014). In Lebanon, plastics generated from domestic solid waste account for approximately 500 tons/day (approximately 182000 tons/annum), only 8% of them are sorted out for recycling. The rest is dumped into the nature and they end up in body of water and will be brought downstream to the Mediterranean Sea or the are dumped directly in coastal landfill in Tyr, Saida, Costa Brava, Bourj Hammoud, and Tripoli (GIZ, 2014).

I.5. Definition of Microplastics

Microplastics are small plastic pieces less than five millimeters while nanoplastics are 1 to 1000 nm (NOAA, 2018; Gigault *et al.*, 2018). Plastic waste is littering our oceans and threatening the marine life (World Animal Protection, 2017). One plastic bag has an average usable life of just 15 minutes. In the United States alone, more than 60 million plastic bottles are thrown away every day (Plastic Soup Foundation, 2014). Seals, whales, dolphins, seabirds, fish, crabs and many other sea animals are dying and becoming sick because of this deadly environmental concern which a major part of it is micro plastics. Over time the plastic degrades into smaller pieces generating micro plastic particles (Urbanek *et al*., 2018). Sea animals often eat microplastics because of their small size. Moreover, micro plastics are becoming carriers of other toxic chemicals, which can affect the reproduction and sea animal may suffer for months before they die. In 2014, an estimated 15 to 51 trillion microplastic particles were floating in the world's oceans, weighing between 93,000 and 236,000 tons (World Animal Protection, 2017). Plastic cannot be easily removed from water; it accumulates in organisms and sediments, and persists much longer than on land.

There is a growing body of evidence indicating a complex toxicology of plastic micro- and nanoparticles on marine life, as these plastics contain or adhere some toxic materials such as heavy metals and endocrine disruptors. These latter toxic materials can have adverse effect on human health leading to cancer in some cases. These microplastics reach human beings through the food chain (Worm *et al.*, 2017).

I.6. Adverse Health Effects of Microplastic

Microplastics and human health is an emerging field, complementary existing fields indicate potential particle, chemical and microbial hazards. If inhaled or ingested, micro plastics may accumulate and exert localized particle toxicity by inducing or enhancing an immune response. Chemical toxicity could occur due to the localized leaching of component monomers, endogenous additives, and adsorbed environmental pollutants. Chronic exposure is anticipated to be of greater concern due to the accumulative effect that could occur (Wright *et al*., 2017).

I.7. Presence of Microplastic in Sea Salt

Few studies worldwide showed that sea salt was contaminated with plastic (Mohamad *et al*., 2013; Donggi *et al*., 2015; Karami *et al*., 2017; Gündoğdu, 2018; Iñiguez *et al*., 2017; Seth *et al*., 2018). To the best of our knowledge, few efforts have been deployed in this field in Lebanon although its international importance is gaining an incremental momentum at the international level.

Plastic pollution is incrementally having a negative impact on the marine ecosystems, especially in oceanic habitats. Environmental pollution with microplastic particles is also causing food consumed by humans to be increasingly polluted, including sea salts. A study conducted in China tested the presence of micro plastics in 15 different brands of sea salts, lake salts and rock/well salts (Donggi *et al.*, 2015). The micro plastics content was 550–681 particles/kg in sea salts, 43–364 particles/kg in lake salts, and 7–204 particles/kg in rock/well salts. These results indicated that sea products, such as sea salts, are more contaminated by micro plastics than salts from other sources. Similar results were obtained in Turkey. The assessment of different brands (n= 16) of rock and sea salts obtained from the Turkish market, showed that sea salt brands contained higher amount of micro plastics (16–84 item/kg) compared to other salt types such as lake salt (8–102 item/kg) and rock salt (9–16 item/kg) (Gündoğdu, 2018). Another study was done in India reported contamination of Indian sea salts with different micro plastic particles, as a consequence of using contaminated sea water (Seth *et al.*, 2018). In addition to that, a study done on 21 salt brands found on the Spanish market found that micro plastic content was of 50–280 MPs/kg salt with no significant differences among all the samples. These results indicate that even though the micro-particles might originate from multiple sources, there is a background presence of micro plastics in the environment (Iñiguez *et al*., 2017).

I.8. Techniques Used to Assess Microplastics in Sea Salt

Two main techniques can be used for the identification of microplastics in sea salt. The most commonly used one is Fourier-transform infrared spectroscopy (FTIR) (Seth *et al*., 2018; Selvam *et al*., 2020). Raman Microscopy System (Renishaw Plc., New Mills, Wotton-under-Edge Gloucestershire, UK) equipped with a 785 nm diode and a 514 nm Ar+ laser was also used in a study by Gündoğdu (2017).

Chapter II: Assessment of the contamination of sea salt available in Lebanon with microplastics and others contaminants

II. 1. Introduction

Plastic waste is littering our oceans and threatening the marine life (World Animal Protection, 2017). Over time plastic degrade into smaller pieces generating microplastic particles (Urbanek *et al.,* 2018). Microplastics are small plastic pieces of less than five millimeters dimension and nanoplastics range between 1 to 1000 nm (NOAA, 2018; Gigault *et al.*, 2018). Plastic cannot be easily removed from water; it accumulates in organisms and sediments, and persists much longer than on land. There are different plastic polymers such as low-density polyethylene (LDPE), polypropylene (PP), polystyrene (PS), and polyethylene terephthalate (PET) where each have certain characteristics. Different formulations behave differently in the environment, which means different plastics break down in the ocean at different rates. One plastic bag has an average usable life of just 15 minutes and takes 50 years to degrade in water (Whiting, 2017). In the United States alone, more than 60 million plastic bottles are thrown away every day and each takes 450 years to degrade in water (Plastic Soup Foundation, 2014). While a large amount of microplastics result from the fragmentation of larger pieces of plastic waste, significant quantities also enter the environment directly, making it more challenging to track and prevent them. In 2014, an estimated 15 to 51 trillion microplastic particles were floating in the world's oceans, weighing between 93,000 and 236,000 tons (World Animal Protection, 2017). Ingestion of water contaminated with microplastics is the main exposure route for several marine and freshwater species. In addition to that, sea animals often eat microplastics because of their small size. Seals, whales, dolphins, seabirds, fish, crabs and many other sea animals are dying and becoming sick because of this deadly environmental concern which is due mainly to contamination with microplastics (World Animal Protection, 2017).

Microplastics and human health is an emerging field, complementary existing fields indicate potential particle, chemical and microbial hazards. If inhaled or ingested, microplastics may accumulate and exert localized particle toxicity by inducing or enhancing an immune response. Chemical toxicity could occur due to the localized leaching of component monomers, endogenous additives, and adsorbed environmental pollutants. Chronic exposure is anticipated to be of greater concern due to the accumulative effect that could occur (Wright *et al.*, 2017).

Salt or sodium chloride (NaCl) is a mineral used as food additive. It is classified as generally recognized as safe (GRAS) by the FDA. There are different types of salt depending on its original such as rock salt, sea salt, table salt and lake salt among others. Rock salt is from mines and mountains such as Himalayan salt. Table salt consists of tiny cubes tightly bound together through ionic bonding of the sodium and chloride ions. The most obvious difference between the two is that rock salt consists of larger grains than table salt, which is usually finely ground. Lake salt are salts produce by evaporation of lake water. Similarly, sea salt is produced by evaporation of sea water. Salt is composed of sodium chloride, a compound that helps regulate fluid balance and blood pressure in the body (Streit, 2019). Therefore, it's necessary to consume it to maintain optimal health. It is one of the world's most important cooking ingredients; it is used as a seasoning in cooking and for preserving food. Salt is effective as a preservative because it reduces the water activity of foods. The water activity of a food is the amount of unbound water available for microbial growth and chemical reactions. Today, few foods are preserved solely by the addition of salt (Henney *et al*., 2010). Sea salt around the world has been contaminated therefore, its safety is being a rising issue. The main contaminate to consider are plastics. This is because they are highly found in water due to its pollution from factory dumping and domestic waste in general. They degrade in sea water and become an ingredient of sea salt when being evaporated or accumulate in sea salt. Sea salt is a highly consumable ingredient worldwide, so the World Health Organization recommends a salt intake of 2g of sodium in salt, European Food Safety Authority (EFSA) recommends 2g of sodium in salt and Food and Drug Association (FDA) recommends 2.3g of sodium in salt per day for a healthy adult (World Health Organization, 2012; EFSA, 2019; FDA, 2020). However, the amount consumed is 10g per day globally, 8–11g per day for Europe and 3.4g (Mozaffarian *et al*., 2014). There is no maximum residue limit (MRL) for the presence of microplastics in sea water nor in sea salt.

Among 192 countries of the world, only 44 (23%) of the countries have carried out research regarding microplastics (Ajith *et al.*, 2020). There is a growing body of evidence indicating a complex toxicology of micro- and nanoparticles on marine life. Once swallowed, plastic fills the stomach and surprisingly this reduces the feeling of hunger (Plastic Soup Foundation, 2014). Therefore, animals eat less, obtain less energy, and weaken. Larger pieces of plastic can also block their gastrointestinal tract so that the plastic can no longer be excreted. In other cases, plastic is ground into small pieces in the stomach and then scattered everywhere. In this way, the northern fulmar grinds and spreads millions of pieces every year. Some of it is left at abandoned nesting sites. Also, these plastics contain or adhere some toxic materials such as heavy metals and endocrine disruptors (Cole *et al*., 2011). Bisphenol A and Phthalates are two disruptors that are used in manufacturing of plastics (NIH, 2015). These latter toxic materials can have adverse effect on human health leading to cancer in some cases (Schung *et al*., 2014). These microplastics reach human beings through the food chain (Worm *et al.*, 2017). There are several ways by which these plastic particles can be ingested: orally through water, consumption of marine products which contain microplastics, through the skin via cosmetics (identified as highly unlikely but possible), or inhalation of particles in the air (Revel *et al*., 2018). Several studies assessed the presence of microplastic in food such as fish, shellfish, honey, sugar, beer and salt (Rainieri *et al.*, 2019; De la Torre, 2019; Barboza *et al.,* 2018; Carbery *et al.*, 2018; EFSA CONTAM, 2016; Gutow *et al.*, 2015; Van Cauwenberghe *et al.*, 2014).

Few studies worldwide showed that sea salt was contaminated with plastic (Mohamad *et al.*, 2013; Donggi *et al*., 2015; Karami *et al.*, 2017; Gündoğdu, 2018; Iñiguez *et al*., 2017: Seth *et al.*, 2018). China produces the largest quantity of plastic waste, at nearly 60 million tons. This is followed by the United States at 38 million, Germany at 14.5 million and Brazil at 12 million tons (Ritchie *et al*., 2018). Twenty-two countries that are comprising the Mediterranean region produce 10% of all plastic goods, making it the world's 4^{th} largest plastic producer (WWF, 2019).

The average salt intake is 3130 mg of sodium in Lebanon with up to 60% of individuals consuming more than the recommendation of 2000 mg (Alley *et al.*, 2014). Lebanese sea salt is produced through the following steps: (1) seawater is drawn into meter-deep concrete ponds via pumps powered by small windmills, (2) the water sits in the ponds of up to 20 square meters for at least 20 days in order to evaporate and leave a salty liquid residue, (3) salty water is then swept into shallower concrete pans, and left to concentrate further for another 10 days, producers sweep the seawater across the pan to ensure it dries evenly every day, (4) as the liquid disappears, white salt crystals emerge in lines, (5) salt are extracted and ready to be packed (Chalhoub, 2017). However, Lebanese production of sea salt does not meet huge demands of the local market and thus Lebanon imports salts from different countries. According to the ministry of trade and economy and in 2018, salt was imported to Lebanon mainly from Egypt (70%), and France (13%) (Ministry of Economy and Trade, 2018).

One very recent study has documented the presence of microplastics in marine sediments and marine biota (Kazour *et al*., 2019); this is suggestive of high probability to find microplastics in sea salt produced in Lebanon. The aim of this study was to detect the presence of microplastics in sea salt originating from the Lebanese coastal and found on the Lebanese market.

II.2. Materials and Methods

II.2.1. Sample Collection

A total of 27 sea salt brands were collected from the Lebanese market in packed and bulk forms between September 2019 and September 2020. Three replicate samples from each brand were assessed.

II.2.2. Confidentiality

Confidentiality means that any identifying information is not made available to, or accessed by anyone but the project team. In addition to that, ethical issues will be taken into consideration based on the NU- IRB Guidebook. However, informed consent forms will not be used in this study since questionnaire and interviews will not be used.

II.2.3. Microplastics Extraction

The analyses of microplastics were conducted following the procedure used by Renzi *et al.,* (2018). For each experimental replicate, 180 g of sea salt was completely dissolved in 500 ml of deionized water mixed for 20 min in a glass beaker and filtered using a filtrating vacuum pump apparatus on a filter fiber (Fisher Brand, Qualitative Filter Paper, Size: 90mm, Stk: 100, thickness: 0.45 mm, pore size: 0.7 μm) disks successively dried in oven at 40°C. Total suspended solids were determined in a solution of 200 g/L of salt samples in statistic replicated to evaluate salt contamination by total solid materials. Classifications were performed according to type, shape, size, and color following criteria reported by the literature (Hidalgo-Ruz *et al.*, 2012; Galgani *et* *al.*, 2013; Alomar *et al.*, 2016). Experimental blanks were performed to evaluate possible crossover contamination during air exposure in laboratory as detailed reported by previous studies performed by the research group (Fastelli *et al*., 2016; Blašković *et al*., 2017). The collected particles (n=208) were analyzed with Fourier Transform infrared spectroscopy (FTIR) (Perkin Elmer Spectrum Two FT-IR Spectroscopy). FTIR spectroscopy offers the possibility of accurate identification of plastic polymer particles according to their characteristic IR spectra (Thompson *et al.*, 2004; Reddy *et al.*, 2006; Frias *et al.*, 2010; Harrison *et al.*, 2012; Vianello *et al.*, 2013). Also, FTIR spectroscopy can provide further information on physio-chemical weathering of sampled plastic particles by detecting the intensity of oxidation (Corcoran *et al.*, 2009). The FTIR was used in attenuated total reflectance (ATR) mode. The spectra were recorded as 64 scans in the spectral range 400-4000 cm^{-1}. Plastic polymer types were identified by matching the sample's IF spectrum with the spectra stored in an integrated FTIR polymer spectrum library.

II.3. Results

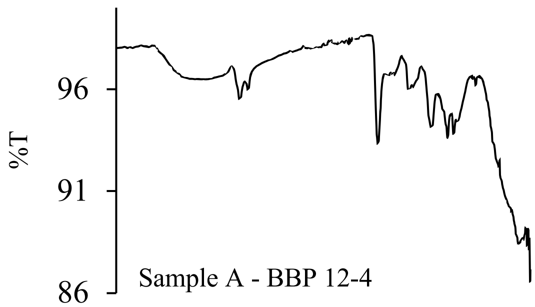
In our study, 27 brands of Lebanese commercial salt were examined for the presence of microplastics. Each brand was analyzed in triplicate, resulting with 81 samples. The microplastic particles were found in 16 samples. After the drying process, most of the particles observed on the filter papers were colorless and transparent, but few of them were in diverse colors such as yellow, blue, and black (Fig. 1 and Fig. 2). The extracted particles had irregular fragments and fibers shape. The distribution of these fiber particles where skewed towards smaller size although some large fibers were also found.

Figure 1. Petri Dish containing Salt Sample

(A) **(B)**

Figure 2. Images of some of the extracted particles: (A) Polypropylene & (B) polyethylene

The FTIR analysis revealed the presence of seven types of polymers: polypropylene, thermoplastic elastomer, gomma plastificata, polyester, polyethylene, plasticizer and BBP 12-4 (Fig. 4). Polypropylene was detected in one replicate of samples R, S and Y. BBP 12-4 was detected in one replicate of samples A and C. While, polyester was detected in one replicate of samples I and V. Thermoplastic elastomer was found in replicate 1 of sample C. Gomma plastificata or softener rubber was found in one replicate of samples K, P, Z and 2 replicates of sample D. Polyethylene was found in 1 replicate of samples B, U and all 3 replicates of sample T. Plasticizer was detected in one replicate of AA.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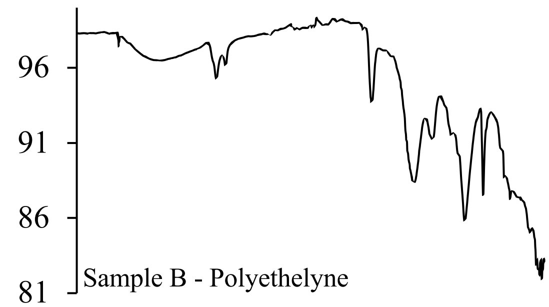
91
86
Sample A - BBP 12-4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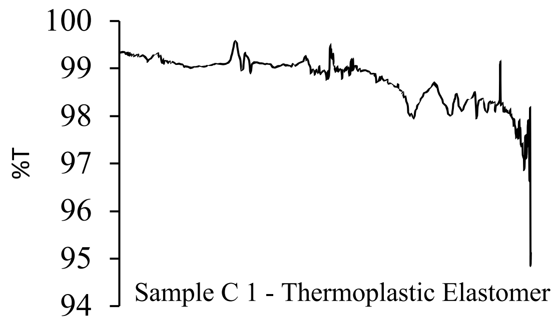
Sample B - Polyethelyne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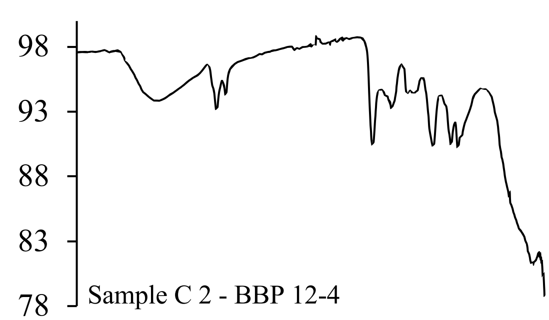
Sample C 1 - Thermoplastic Elastomer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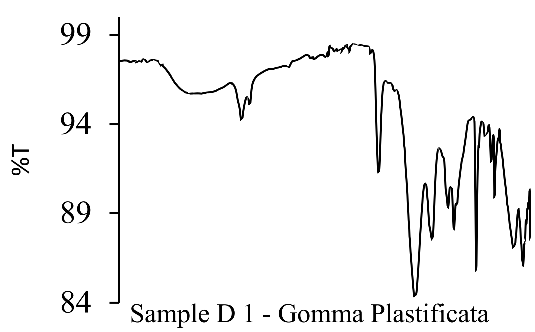
Sample C 2 - BBP 12-4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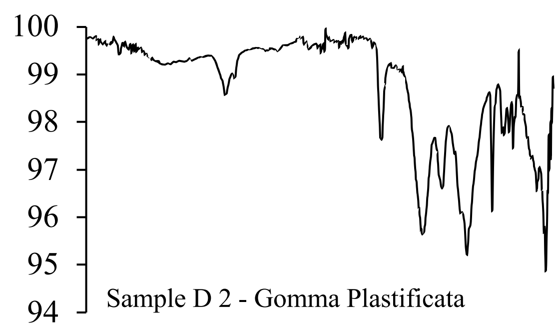
Sample D 1 - Gomma Plastificata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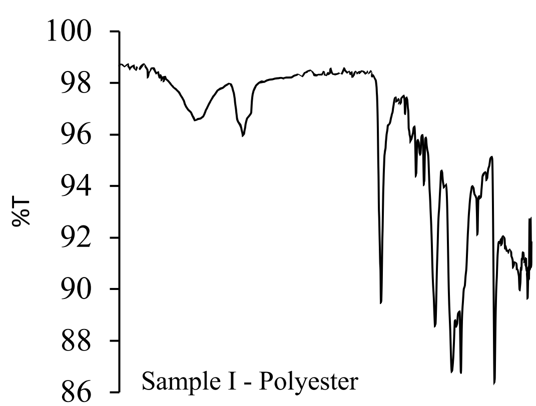
Sample D 2 - Gomma Plastificata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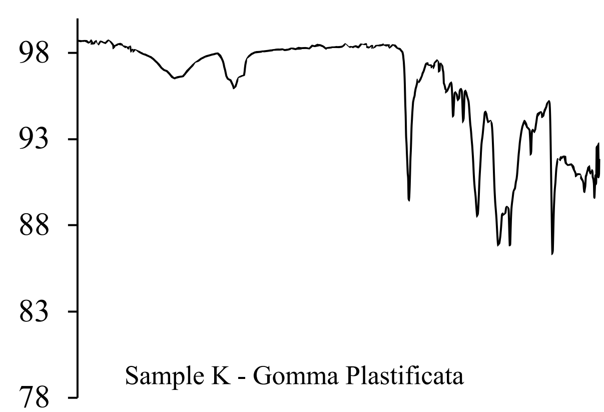
Sample I - Polyester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Sample K - Gomma Plastificata

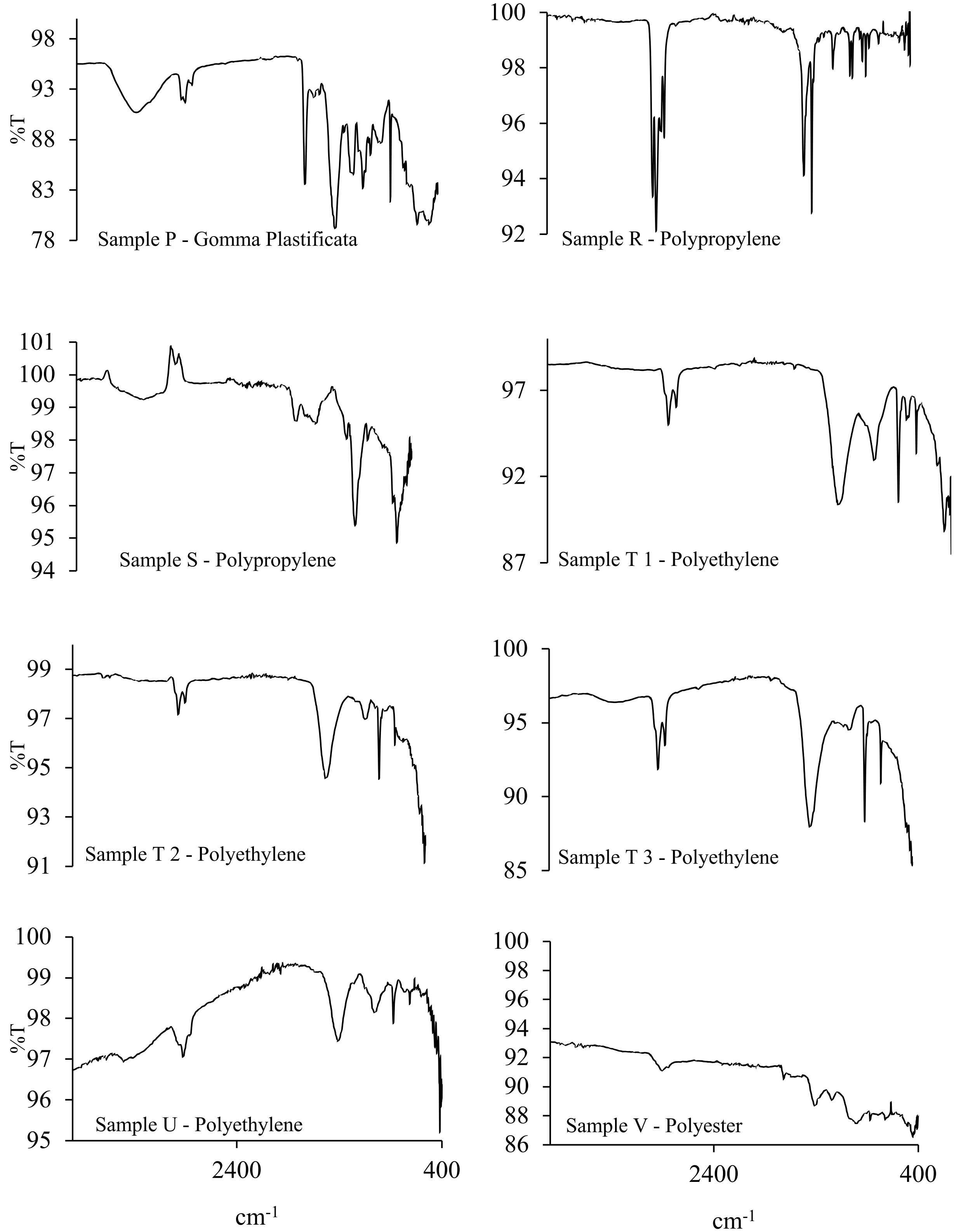

Figure 3. Identification of the polymer type of extracted particles: FTIR spectra of the particle.

Overall composition of all extracted particles was dominated by polyethylene and gomma plastificata with 26.31%, followed by polypropylene with 15.97%, BBP 12-4 and polyester with 10.53% and thermoplastic elastomer and plasticizer with 5.26 % (Fig. 4).

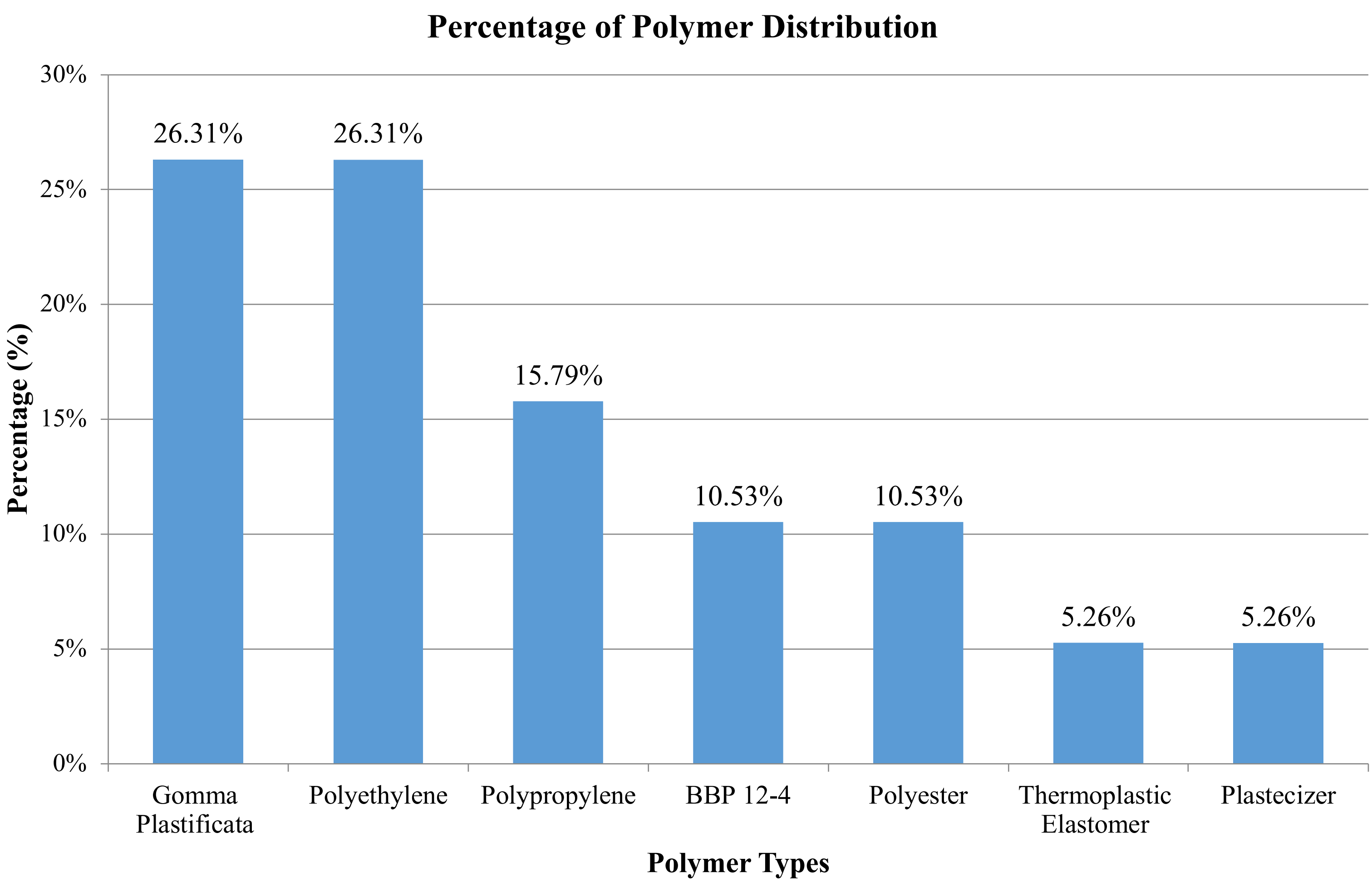

Figure 4. Percentage distribution of different types of microplastic polymers

Studies have been done on the presence on microplastics in the Mediterranean region. In Mallorca Island and Cabrera Island (Balearic Islands, western Mediterranean) of Spain, microplastic sediments were found in a study done by Alomar *et al*., (2016). A study done also in Spain on 16 samples of sea salt of which 9 were from the Mediterranean Sea of Barcelona, Gerona, Valencia, Alicante, Murcia and Menorca (Iñiguez *et al*., 2017). Several types of microplastics were identified, being the most common one's polyethylene terephthalate with 83.3%, polyethylene with 3.3% and polypropylene with 3.3%. Moreover, in a study done to determine the load of microplastics in the Western Mediterranean Sea, polyethylene, polypropylene and polystyrene were detected with polyethylene found to be having highest abundance (De Haan *et al*., 2019). Another study done in Turkey determined that many types of plastics such as polyethylene, polyethylene terephthalate, polyurethane, polypropylene, polymethyl-methacrylate, polyamide-6 and polyvinylchloride were present in 15 salt brands (Gündoğdu, 2018).

In Lebanon, a study was done to investigate the quantity and types of microplastics found in three matrices (sea water, sediments and marine biota) along the Lebanese coast (Kazour *et al*., 2019). Three sampling sites were chosen along the coast according to their degree of anthropogenic pressures making them pollution hotspots (Shaban, 2008). The first site was Tripoli, located in North of Lebanon. The second site was Beirut, located in the middle of the country and is the capital. Finally, the third site is Sidon (Saida), located in South of Lebanon. All located sites showed the presence of microplastics in marine sediments and marine biota.

Many ocean and sea pollutants are released into the environment far upstream from coastlines. These pollutants can be divided into chemical and physical pollutants. All samples that were tested in our study where positive for contaminants detection as shown in Table 1. These contaminants were gelatin, cellulose, Arabic gum, calcium sulfate, quartz, wood, glass, iron oxide and lubricants.

Table 1. Distribution of contaminates detection between samples

Label	Origin	Microplastics (MP)	Type of MP	Other Contaminants
A	Lebanon	Positive	BBP 12-4	Positive
B	Lebanon	Positive	Polyethylene	Positive
C	Lebanon	Positive	Thermoplastic Elastomer/ BBP 12-4	Positive
D	Lebanon	Positive	Gomma Plastificata	Positive
E	Lebanon	Negative		Positive
F	Lebanon	Negative		Positive
G	Lebanon	Negative		Positive
H	Lebanon	Negative		Positive
I	Lebanon	Positive	Polyester	Positive
J	Lebanon	Negative		Positive
K	Lebanon	Positive	Gomma Plastificata	Positive
L	Lebanon	Negative		Positive
M	Lebanon	Negative		Positive
N	Lebanon	Negative		Positive
O	Lebanon	Negative		Positive
P	Lebanon	Positive	Gomma Plastificata	Positive
Q	Lebanon	Negative		Positive
R	Lebanon	Positive	Polypropylene	Positive
S	Lebanon	Positive	Polypropylene	Positive
T	Lebanon	Positive	Polyethylene	Positive
U	Lebanon	Positive	Polyethylene	Positive
V	Lebanon	Positive	Polyester	Positive
W	Lebanon	Negative		Positive
X	Lebanon	Negative		Positive
Y	Lebanon	Positive	Polypropylene	Positive
Z	Lebanon	Positive	Gomma Plastificata	Positive
AA	Lebanon	Positive	Plastecizer	Positive

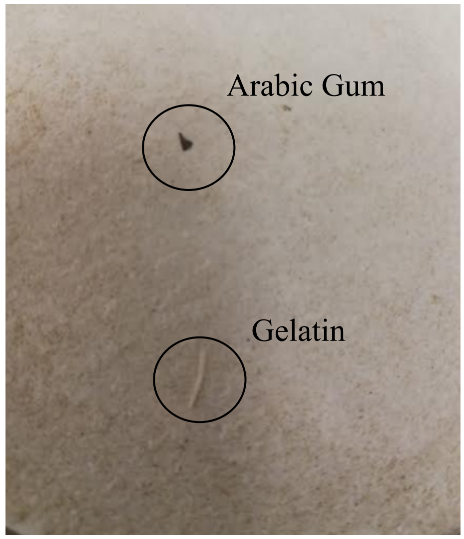

Figure 5. Arabic gum and Gelatin found in Sample N

II.4. Discussion

Out of the 27 samples tested, 59.26% were contaminated with microplastics. The detected microplastics related back to seven different types synthetic polymers. Distribution of these different microplastics particles was at its highest, three particles, in sample T with polyethylene as shown in Fig. 6. In addition to that, samples C and D contained the second highest particle count which is two particles. Thermoplastic elastomer and BBP 12-4 were the particles detected in C and two gomma plastificata (softened rubber) in D that may raise a serious health issue as most probably in its composition an endocrine disruptor such as phthalates is present. While the remaining samples contained only one particle of certain microplastics polymer contamination.

In India, four types of polymers: polyesters, polystyrene, polyamide, and polyethylene (Seth *et al*., 2018). Two of these polymers, polyester and polyethylene, are consistent with our study where they were also detected. However, polyester was the dominant polymer in Seth, et al., 2018 study. On the other hand, in another most recent study done in India, presence of four major polymers such as cellophane, polystyrene, polyamide, and polyether with cellophane maximum percentage of in salts (Sivagami *et al*., 2021). In addition to these two studies, nylon, cellulose, polyethylene, and polypropylene were identified in another study done in India in 2020 (Selvam *et al*., 2020). Similar to our study, polyethylene was the dominant between the identified polymers. A study was done on the presence of microplastic in 14 brands of commercialized sea salt from different countries: Australia, France, Iran, Japan, Malaysia, New Zealand and Portugal (Karami *et al*., 2017). Major plastic polymers detected were polypropylene at 40% of the total plastic polymers followed by polyethylene 33.3%, polyethylene terephthalate 6.66%, polyisoprene/polystyrene 6.66%, polyacrylonitrile 10%, and polyamide-6 3.33%. This shows that the presence of microplastics in different regions of the world.

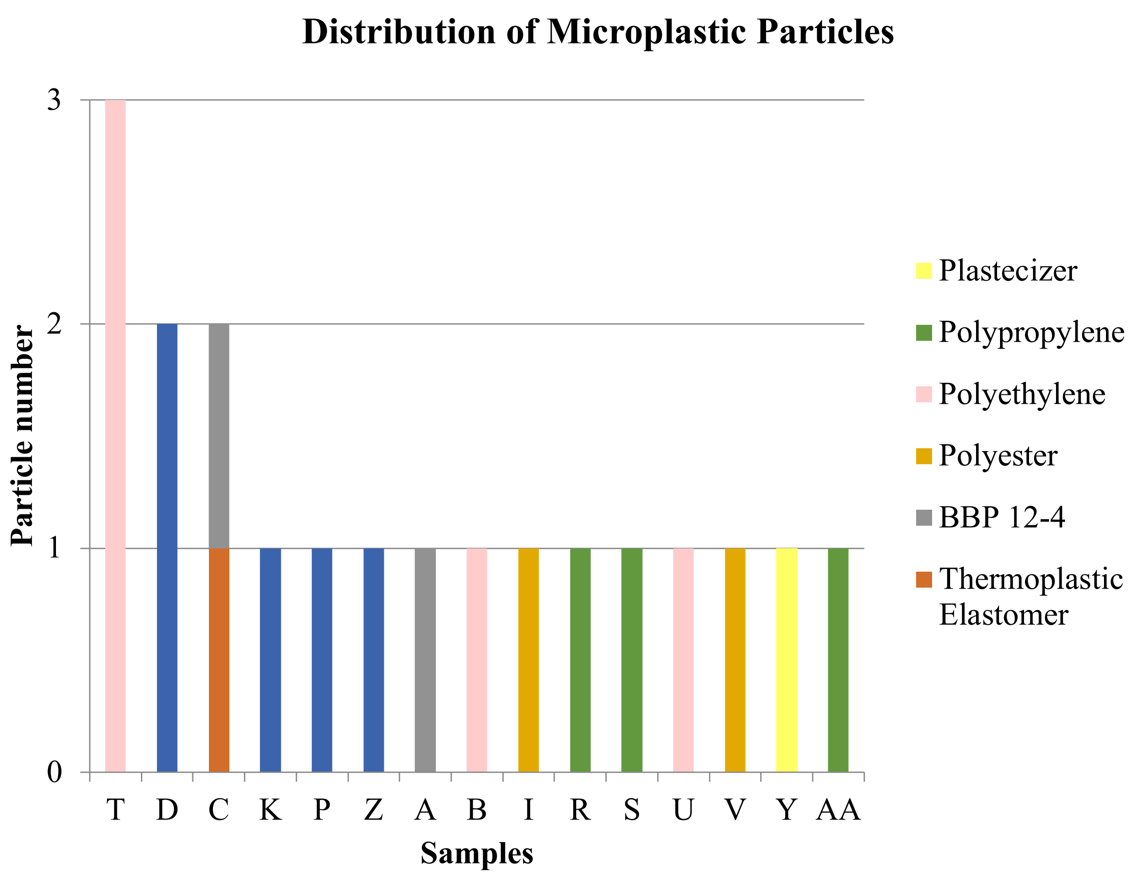

Figure 6. Distribution of microplastics particles among contaminated samples

The results in this study further confirm the global presence of microplastics in the sea salts as reported from different parts of the world (Yang *et al.*, 2015; Iñiguez *et al.,* 2017; Karami, *et al.,* 2017; Gündoğdu, 2018). It is expected that contaminated sea water is the primary source for the presence of these particles in sea salts. However, some contaminations may also have reached the salts during different stages of production process through airborne microplastics as well as packaging (Yang *et al.,* 2015). A study done in China tested the presence of micro plastics in 15 different brands of sea salts, lake salts and rock/well salts. The micro plastics content was 550–681 particles/kg in sea salts, 43–364 particles/kg in lake salts, and 7–204 particles/kg in rock/well salts. These results indicated that sea products, such as sea salts, are more contaminated by micro plastics than salts from other sources (Donggi *et al.,* 2015). The assessment of different brands (n=16) of rock and sea salts obtained from the Turkish market, showed that sea salt brands contained higher number of micro plastics (16–84 item/kg) compared to other salt types such as lake salt (8–102 item/kg) and rock salt (9–16 item/kg) (Gündoğdu, 2018). Another study was done in India reported contamination of Indian sea salts with different micro plastic particles, as a consequence of using contaminated sea water (Seth *et al.,* 2018). In addition to that, a study done on 21 salt brands found in the Spanish market found that micro plastic content was of 50–280 item/kg salt with no significant differences among all the samples. These results indicate that even though the micro-particles might originate from multiple sources, there is a background presence of micro plastics in the environment (Iñiguez *et al*., 2017).

208 particles of contaminates were detected in sea salt in this study such as glass fibers and lubricants among many others shown in Fig. 7. Calcium carbonate, calcium sulfate and quartz are the most abundant contaminants. Possible reason for the two spikes in calcium sulfate and calcium carbonate are due to the fact that they are used as dessicants. Other types of pollution such as water

pollution occurs when toxic substances from farms, towns, and factories readily dissolve into and mix with it. This type of pollution is the major

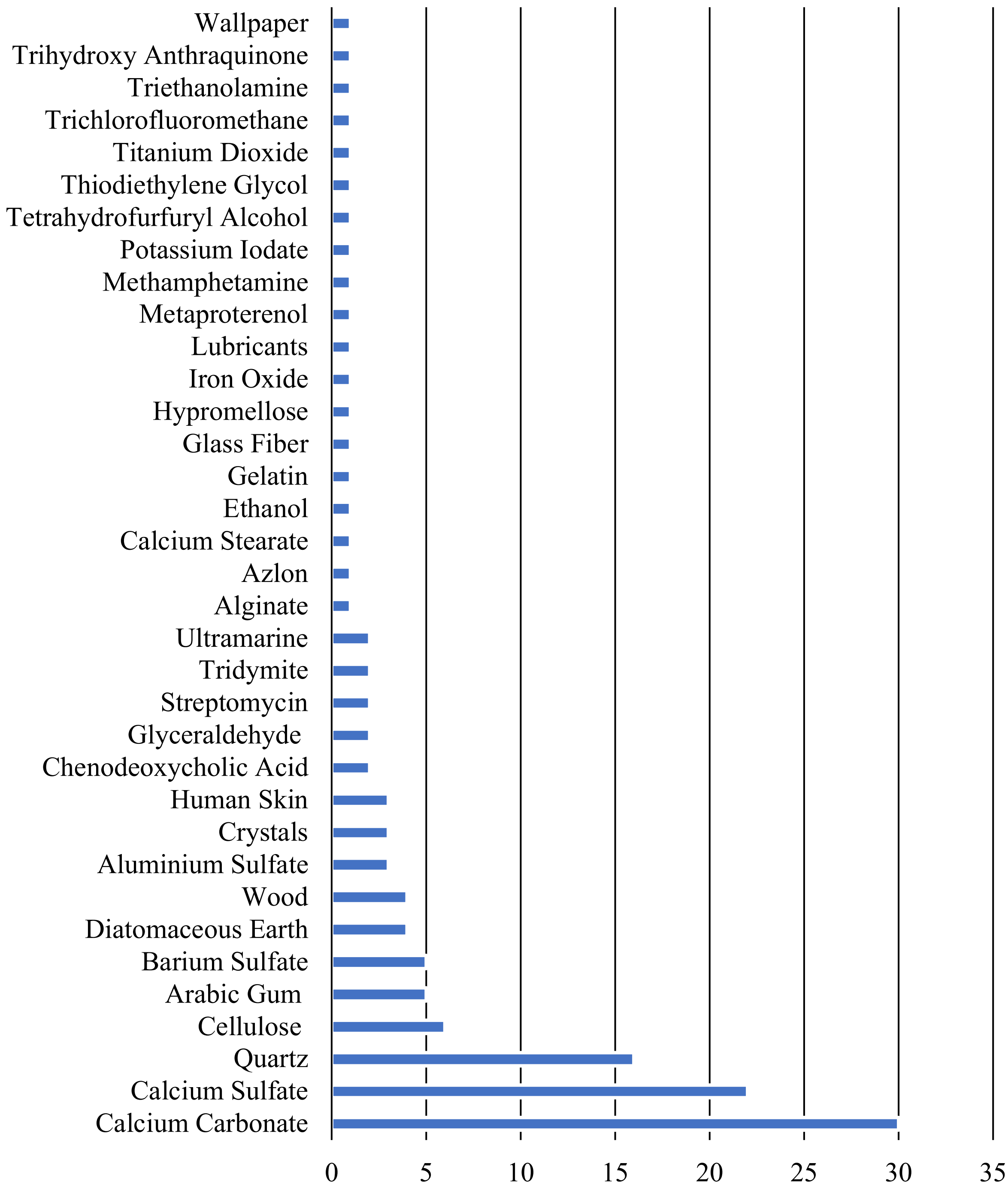

Figure 7. Contaminants present in all samples

contributor to seafood and sea salt contamination especially with absence of governmental control in Lebanon.

This study showed the presence of microplastics in sea salt produced in Lebanon. Polyethylene was the most detected microplastic. This is because polyethylene is a polyester that is widely used in plastic containers, bottles and bags. Packaging is a one of the major fields of application for polyethylene and polystyrene polymers (Andrady, 2011). Various studies have reported the presence this contaminant in sea salt. Polyethylene have been observed in salts from other countries also (Yang *et al*., 2015; Iñiguez *et al*., 2017; Karami *et al*., 2017; Gündoğdu, 2018), while polyamide was observed in Turkish salts (Gündoğdu, 2018). In addition to that, a recent study done on the Lebanese coast in order to test for microplastics, all of the sampled Lebanese beaches showed high level of contamination from microplastics (Kazour *et al*., 2019). Contamination of the Lebanese coast contributes majorly to the presence of micro plastics in sea salt.

Salt evaporation ponds used where spread on all shores of Lebanon from south to north. Over time the Lebanese government has enforced high taxes for the owners in order to push them to sell their land and build resorts. Unfortunately, the government's plan was a success and there are only two remaining salt evaporation ponds left located in Anfeh. The owners of these ponds are not able to produce sufficient amount to supply the market. As a result, Lebanon imports huge amounts of salt from other countries such as Egypt. These imported salts are being imported in bulk forms then packed in Lebanon under different brand names and labeled made in Lebanon due to lack of regulations and governmental control. In addition to that, there is no specific limit set by any governmental body regarding the presence of microplastic sea salt nor there is an international limit that Lebanon can refer to as well.

Plastic waste is one of the major pollutants of the solid waste throughout the world. Every day around 8 million pieces of plastic makes their way into our oceans (Condor Ferries, 2020). It is estimated that plastic accounts for over 80% of marine litter. Plastic debris is then transported by marine currents, sometimes over very long distances (IUCN, 2018). The slow degradation rate of plastic waste results in death of billions of living organisms in marine and terrestrial environments (Shahnawaz *et al.,* 2019). Once these plastics get into the water, they migrate into marine animals and sea salts. According to International Union for Conservation of Nature (IUCN), seven major sources are contributing to presence of microplastic in water which are tires, synthetic textiles, marine coatings, road markings, personal care products, plastic pellets and city dust (IUCN, 2018). These sources influence the level of presence of different polymers. As means to contribute to solving plastic pollution, biodegradable, oxo-biodegradable and compostable bags were introduced to market. These bags are capable of being decomposed under a shorter period of time (6 to 36 months). However, a study examined biodegradable, oxo-biodegradable, compostable, and high-density polyethylene (conventional plastic carrier bag) materials over a 3 years period (Napper *et al*., 2019). These materials were exposed in 3 natural environments: open-air, buried in soil and submersed in seawater, as well as in controlled laboratory conditions. In the marine environment, the compostable bag completely disappeared within 3 months. However, the same compostable bag type was still present in the soil environment after 27 months but could no longer hold weight without tearing. After 9 months exposure in the open-air, all bag materials had disintegrated into fragments. Therefore, it is not clear that the oxo-biodegradable or biodegradable formulations provide sufficiently advanced rates of deterioration to be advantageous in the context of reducing marine litter, compared to conventional bags. In December of 2020, the European

Commission (EU) has recommended limiting the use of biodegradable plastics in the open environment (EU, 2020).

In addition to harming the environment, marine litter causes economic damage to activities such as tourism, fisheries and shipping. In industries such as fisheries and tourism the costs of marine litter are beginning to be quantified and are considerable. The costs of these marine litter are most often used to cover removing debris or recovering from the damage which they have caused, this expenditure represents treatment rather than cure, and although probably cheaper than inaction do not present a strategy for cost reduction (Newman *et al.*, 2015). The Northwestern Pacific Plan (NOWPAP) report has stated that marine litter are impacting the interruption of fishing operation, damage to aquaculture facilities, breakdown or repair of fishing gear, breakdown of vessels and cost of marine litter clean up (NOWPAP, 2013). Also, according to a report done by Kommunernes International Miljøorganisation (KIMO), municipalities throughout the Northeast Atlantic region continue to face high costs associated with the removal of beach litter. United Kingdom (UK) municipalities spend approximately €18 million each year removing beach litter, which represents a 37% increase in cost over the past 10 years. Similarly, removing beach litter costs municipalities in the Netherlands and Belgium approximately €10.4 million per year (Mouat *et al*., 2010). Economic instruments, such as taxes and charges addressing the drivers of waste, for instance those being developed for plastic bags, could be used to reduce the production of marine litter and minimize its impacts.

The United Nations (UN) have 17 developed sustainable development goals (SDGs) to be achieved by 2030. Under each goal there are targets that need to be reached. Goal number 14 is about life below water and target 14.1 is to reduce marine debris and nutrient pollution. Plastic is one of these marine debris being present into the oceans. In addition to that, the Our Ocean conference in 2016 stated that nutrient pollution comes from diverse sources, including agricultural runoff and sewage and wastewater discharges. It overloads marine environments with high concentrations of nitrogen, phosphorous, and other nutrients, which can produce large algal blooms (Our Ocean, 2016). The Convention for the Protection of the Mediterranean Sea Against Pollution (Barcelona Convention) was adopted on 16 February 1976 in Barcelona and entered into force in 1978 (UNEP, 2020). The Barcelona convention and its 7 protocols adopted in the framework of the Mediterranean Action Plan (MAP) constitute the principal regional legally binding Multilateral Environmental Agreement (MEA) in the Mediterranean. To date, 22 countries including Lebanon have adopted these protocols. The agreement consists of the following protocols: dumping (from ships and aircraft), prevention and emergency (pollution from ships and emergency situations), land-based sources, specially protected areas and biological diversity, offshore (pollution from exploration and exploitation), hazardous wastes and integrated coastal zone management (ICZM). Euro-Mediterranean governments aim to tackle the top sources of Mediterranean pollution by the year 2020 through the Horizon 2020 initiative. It is an initiative aimed to reduce the most significant marine pollution sources, focus on capacity building measurements and develop indicators to monitor its success.

In addition to presence of microplastics in water, air pollution is another route for this contaminant. Carried by the wind, dust particles from places such as the Sahara Desert can float halfway around the world before settling to the ground (Hershberger, 2020). To date, and to the best of our knowledge, only two studies have demonstrated the presence of fibrous microplastics in the atmospheric compartment, thus suggesting potential human exposure (Daris *et al*., 2016; Daris *et al*., 2017). Also, microplastics can reach air through incineration of garbage. Burning plastic and other wastes releases dangerous substances such as heavy metals, persistent organic pollutants, and other toxics into the air and ash waste residues (GAIA, 2020). Out of the 2.55 million ton of waste produced per year in Lebanon, 12% is being incinerated (AUB, 2018). Another way of oceans and seas pollution with microplastics is from rivers. Rivers carry trash over long distances and connect nearly all land surfaces with the oceans. The Yangtze of China alone pours up to an estimated 1.5 million metric tons into the Yellow Sea (Patel, 2018). In order to reduce the exposure to this contaminate, reduction in usage of single use plastic can help. Canada, Kenya, New York, Hawaii, California, China and European Union have banned the single-use plastic bags (World Economic Forum, 2020). India has enforced nationwide ban on plastic bags, cups and straws. While Zimbabwe has introduced a ban on polystyrene food containers in 2017. While 170 nations pledged to "significantly reduce" use of plastic by 2030. Also, recycling helps keep plastics out of the ocean and reduces the amount of new plastic in circulation. Applying appropriate waste sorting and disposing techniques through segregation at source will reduce amount of plastic and other contaminates transfer to the oceans and seas. Other step can be to introduce tax on plastic bags such as in UK. In addition to that, filtering sea water will help in reducing the amounts of contaminants especially plastics in sea salt (Werft, 2016).

The presence of human skin in the sample from the slat produced in Lebanon raise the issue of lack of implementation of good manufacturing practices in these production site. This may require at least wearing of protective personal equipment such gown and gloves during handling sea salt.

II. 5. Conclusion

The results of this study suggest that the Lebanese sea salt is contaminated with microplastics. The contamination was found to be independent of the salt brand and with dependence on the background contamination of sea water. Extracted particles ranged between different sizes and shapes belonging to different varieties of plastic. These results should be considered as a preliminary analysis on microplastics in sea salt present in the Lebanese market. The level of microplastics found were high, thus, extensive dietary consumption of these sea salts exposes the population to associated health effects of microplastic ingestion. This study is the first of its kind from Lebanon and the Arab region since no previous studies have tackled the issue of the effect of plastics residues in sea salt on human health. This makes a genuinely missed opportunity for leading cross-national comparisons within the Middle East region. Further investigation is in need in order to assess the potential health risk of microplastic consumption on Lebanese population.

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