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Microplastic pollution in table salt and sugar: Occurrence, qualification and quantification and risk assessment

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ABSTRACT

Microplastics (MPs) have recently been observed in many products and environmental samples, ranging from drinking water to food products. In this study, the contamination of several popular brands of Iranian table salts and sugars by MPs was investigated. By using an optical and staining stereomicroscopic method the samples were counted. Results showed MPs were detected in all samples of salts and sugars. The average amounts of MPs in different brands of salt and sugar observed were 55.2 ± 43.7 MP/kg and 57.7 ± 20.6 particles/kg, respectively. Based on Nile red staining the average microplastics in salts and sugars were 151.3 ± 61.8 and 226 ± 99.5 particles/kg. Most MPs were present in the shape of fragments (78.57 % and 95.8 %), while only a few were in the fiber shape (21.43 % and 4.1 %) in salts and sugars, respectively. The qualifying of plastic particles was conducted with Fourier-transform infrared spectrometry (FTIR) and scanning electron microscopy (SEM). Polyethylene (PE) and polypropylene (PP) were identified as the most likely polymers. The maximum annual exposure of MPs was approximately 15,540 and 23,660 particles/kg/bw/year for adults and 3552 and 5408 particles/kg/bw/year for children through salts and sugars, respectively. Based on these findings, it can be concluded that MPs can enter the food chain through additives (such as heavy metals, toxic organic matter and pathogens), and can affect human health.

1. Introduction

The plastic materials that have the highest production and utilization in the world include polyethylene (PE), polypropylene (PP), polystyrene (PS), polyethylene terephthalate (PET), and polyvinyl chloride (PVC), and many fragments of these polymers have been found in the environment (Ribic et al., 2010). Together, these types of plastic account for approximately 90 % of total global plastic production (Engler, 2012; Ribic et al., 2010). In recent years, microplastics (MPs) with a diameter < 5 mm have received worldwide attention as emerging environmental pollutants and are one of the four major global environmental threats with health hazards to humans (Katsnelson, 2015). MPs are responsible for negative biological responses in humans, such as inflammation, genotoxicity, oxidative stress, cell apoptosis and tissue necrosis, localized cell and tissue damage, fibrosis and potentially carcinogenesis (Deng et al., 2017a; Nel et al., 2006; Wright and Kelly, 2017). Also, MPs can cause inflammatory response, cytotoxicity, and genotoxicity in lung tissue (Donaldson et al., 2000). In addition, MP exposure leads to lung diseases, including asthma and pneumoconiosis (Prata, 2018). MPs can adsorb organic and inorganic nutrients from the water environment (Frère et al., 2018).

MPs can be divided into two groups: primary and secondary microplastics. Primary MPs from industrial sources produced in microscopic proportions include plastic pellets (plastic beads), fibers, films, granules, and powders used in cosmetics (e.g., sunscreens), industrial materials, and cleaning products (Fendall and Sewell, 2009; Zitko and Hanlon, 1991). Secondary MPs are the result of physical, chemical, and biological degradation of larger plastic debris. Secondary MPs are formed by light degradation, UV radiation, embrittlement, and biological degradation (Browne et al., 2007; Ryan et al., 2009).

In the 1970s, scientists reported the existence of small plastic particles in the ocean (Schymanski et al., 2018). In 2014, Richard Thompson reported the distribution of MPs in the ocean for the first time (Law and Thompson, 2014). In recent years, many studies have been done on

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distribution and environmental behavior of MPs in terrestrial and freshwater systems (Eerkes-Medrano et al., 2015). Also, different types of MPs have been detected in foods and drinking water (Mintenig et al., 2019). They have been found in human stools (Picheta, 2018). Furthermore, various studies were done on MPs as carriers for toxic pollutants such as heavy metals (Tang et al., 2020), polycyclic aromatic hydrocarbons (PAHs) (Lo et al., 2019), polychlorinated biphenyls (PCBs) (Velzeboer et al., 2014), dichlorodiphenyltrichloroethane (DDT) (Wang et al., 2018), and polybrominated diphenyl ethers (PBDEs) (Chua et al., 2014). In 2019, the World Health Organization (WHO) called on scholars to research the impact of microplastics on human health (Organization, 2019).

Due to the small size and similarity of MPs to the food of many aquatic organisms, these particles are bioavailable to organisms and can accumulate through the food chain (Teuten et al., 2009). The entry of MPs into the human body occurs from different pathways. They are ingested by various aquatic organisms, accumulate in specific tissues, and are transmitted through the food chain (Deng et al., 2017b; Setälä et al., 2014). It should be noted that the organic and inorganic components (e.g., bisphenol A, tributyltin, Zn, Pb) are released into the environment, and the coexisting contaminants (e.g., PAHs, PCBs) can adsorb on MPs, leading to combined toxicity. Therefore, the indirect toxicity due to MPs cannot be neglected (Wang et al., 2021). Also, MPs have been detected in various food products such as beer, honey, canned sardines (Karami et al., 2018), and drinking water (Makhdoumi et al., 2021a). Finally, MPs come to the dining table of humans (Kosuth et al., 2018). The contamination of sugar (Possatto et al., 2011), and table salts (Karami et al., 2017a) consumed by humans with MPs has been demonstrated in previous studies.

However, the importance of contamination comes when the amount of consumption is considered. The commercial salts (of different origins) that are consumed by humans include sea salt (fine or coarse), rock salt, lake salt, rock/well salt, and well salt (Peixoto et al., 2019). The WHO has set a standard salt intake of 5 g per adult per day (<2 g/day sodium), and the maximum intake of 2 g/day sodium in adults should be adjusted downward based on the energy requirements of children relative to those of adults (Organization, 2012). However, Iranians consume about two or three times more than the recommended amount of salt (Brown et al., 2009; Layeghiasl et al., 2018). In addition, the per capita consumption of sugar in Iran is up to 30 kg/year (FAOSTAT, 2021), while the per capita consumption of sugar in the world is only 23 kg/year (Singh et al., 2020).

According to the culture of salt and sugar consumption in Iran, it is necessary to make sufficient and clear assessments regarding the contamination of these products with MPs. So far, only one study has been conducted abroad that reported low abundances of MPs in sea salt produced in Iran (Karami et al., 2017a). However, the chemical composition of MPs was not determined. When we began this study in 2021, no study had evaluated the presence of MPs in commercial salt and sugar in package form in Iran. Indeed, relevant studies on human health risk assessment by MPs are unclear due to limited information on its exposure routes, health effects, and biological fate. This study aims to investigate the most common brands of commercial sugar and salt consumed in Iran for MPs contamination. We investigated whether there is a potential health risk from the consumption of MPs in commercial salt and sugar sold in Iran, and what is the level of the contamination.

2. Method and materials

2.1. Purchase of edible table salts and sugar

After identifying the types of salt and sugar brands used in Iran, the four common brands were purchased from different supermarkets. These salts were all rock salt. For each brand, five packages were selected randomly. Due to privacy reasons, the commercial names of the purchased products cannot be made public.

2.2. Quality assurance and quality control

To eliminate the possibility of contamination, all apparatuses used during the laboratory analysis were washed three times with distilled water in advance and placed under a fume hood for drying (Lusher et al., 2015). They were immediately covered with aluminum caps when not in use. All lab personnel wore cotton (100 %) lab coats and nitrile gloves during all laboratory phases. Before and after each procedure, all work surfaces and instruments were wiped down with ethanol (70 %). The application of plastic material was forbidden during the whole sampling process. The inside of the jars was washed with filtered water before the end of the vacuum process to eliminate the loss of plastic particles. The whole procedure of filtering was performed in an air flow cabinet while an air cleaner ran routinely to avoid airborne plastic contaminants. We also conducted two controls:

1. Control water: The same amount of distilled water used in sample solution preparation was filtered through a new filter fiberglass paper (Johnson GC/50, 1.2 mm thick and pore size 1.1 μm) to check for probable water and/or filter paper contamination.
2. Control air: 100 mL of distilled water were poured into an open wide-mouth glass jar and placed next to the laboratory equipment in order to catch airborne particles. The water from the container was filtered through a new filter of fiberglass paper after time passed for the run of each laboratory dissolving and filtering one of the total salt products.

2.3. Extraction of MPs

The salt and sugar samples in packages were transferred directly to a 1-liter glass jar using a metal spoon. After weighing 10 g of salt and sugar and transferring to the flask, 100 mL distilled water were added, and the flask's open top was covered immediately with aluminum foil. The combination of 10 g of salt and sugar and 100 mL of distilled water, separately, was considered as the best solution with suitable density for filtering. We then stirred the flask using a metal spoon to accelerate the salt's dissolution. After removing the cap, the solution was directly filtered through a new 0.45 μm pore size filter fiberglass paper (Johnson GC/50, 1.2 mm thick and pore size 1.1 μm) which took less than one minute. The filter papers were then left in clean and covered petri dishes in order to dry completely at room temperature (21 ± 5 °C), and sealed it with tape prior to observation of plastic particles.

2.4. MPs inspection

After the preparation of salt and sugar samples, visual identification of MPs was performed using an Olympus SZX16 stereomicroscope (Olympus K.K., Japan) for the quantitative and qualitative detection of MPs. MPs were categorized into three main groups based on definitions established by Karami et al. (2017a): fibers/filaments, fragments, and pellets. The amount, shape, color, and size of MPs, which we could visibly detect for each filter paper, were noted. Also, all particles were photographed.

For more accuracy in MPs counting, Nile Red (NR) dye (9-diethylamino-5*H*-benzo(α)phenoxazinone-5-one) staining was performed, and each filter was treated with a small amount of NR (10 mg/L) solution, separately. The stock concentration was set to 1 mg NR (Sigma-Aldrich, St Louis, MO) in 1 mL acetone (23) and kept at − 20 °C until later use; 100 μL of this solution were diluted with acetone to prepare a 10 mL working solution (i.e., 10 μg in 1 mL) for NR staining. NR with its fluorescence properties has been known to stain synthetic polymers and (or) neutral lipids found in biological samples using a quantitative differential staining approach (Jee et al., 2009). Considering the hydrophobic nature of plastics because of their construction made of hydrocarbons derived from natural gas, biomass, or petroleum, NR is especially interesting for their analysis. About 2–3 drops of NR solution

were added to each dry filter.

Then, the fluorescently tagged filters were transferred to a fluorescence microscope (Axio Scope A1; Zeiss, Jena, Germany) with blue filter (excitation wavelength (ex): 365 nm; emission wavelength (em): 445 nm) and green-yellow filter (ex: 450–490 nm; em: 515–565 nm). All samples were photographed using a 20 × magnification or higher lens.

2.5. Characterization of the plastics

2.5.1. SEM-EDS

To identify the kinds of MPs in salt and sugar samples and confirm the accuracy of the results obtained by the visual inspection methods, scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDS) analysis were used on each sample of MPs selected by stereomicroscope. SEM-EDX allowed for the reasonable separation of potential MPs in a comparatively short time. To capture high-resolution images and examine the MP morphology, MPs were mounted on carbon paste and coated with a 10 nm layer of gold using a desktop magnetron spray. The analysis was accomplished using a Hitachi SU3500 scanning electron microscope (Bruker Nano GmbH Berlin, Germany) with a voltage of 20 kV. EDX (Bruker, Germany) characterizes the signature of the main composition of MP levels.

2.5.2. ATR-FTIR microscope

To identify the nature of MPs, attenuated total reflection Fourier transform infrared (ATR-FTIR) microscopy analyses were performed with a spectrum GX ATR-FTIR microscope (64 scans, resolution 4 cm^{-1} and 450 cm^{-1} to 4000 cm^{-1}; Perkin Elmer, Waltham, MA).

2.6. Estimated annual intake (EAI)

The EAI of MPs concentrations in salt and sugar samples is estimated by using Eq.1 related to annual salt or sugar consumption (Zuccarello et al., 2019). Here the ingestion rate (IR) was determined according to Iranian people consumption of salt and sugar (6 and 60 kg per year). C is the number of MPs (as particles/L), and the body weight (bw) was considered for adult and children to be 70 kg and 16 kg, respectively.

EAI (MP/kg/day) = (C × IR)/bw × 365 (1).

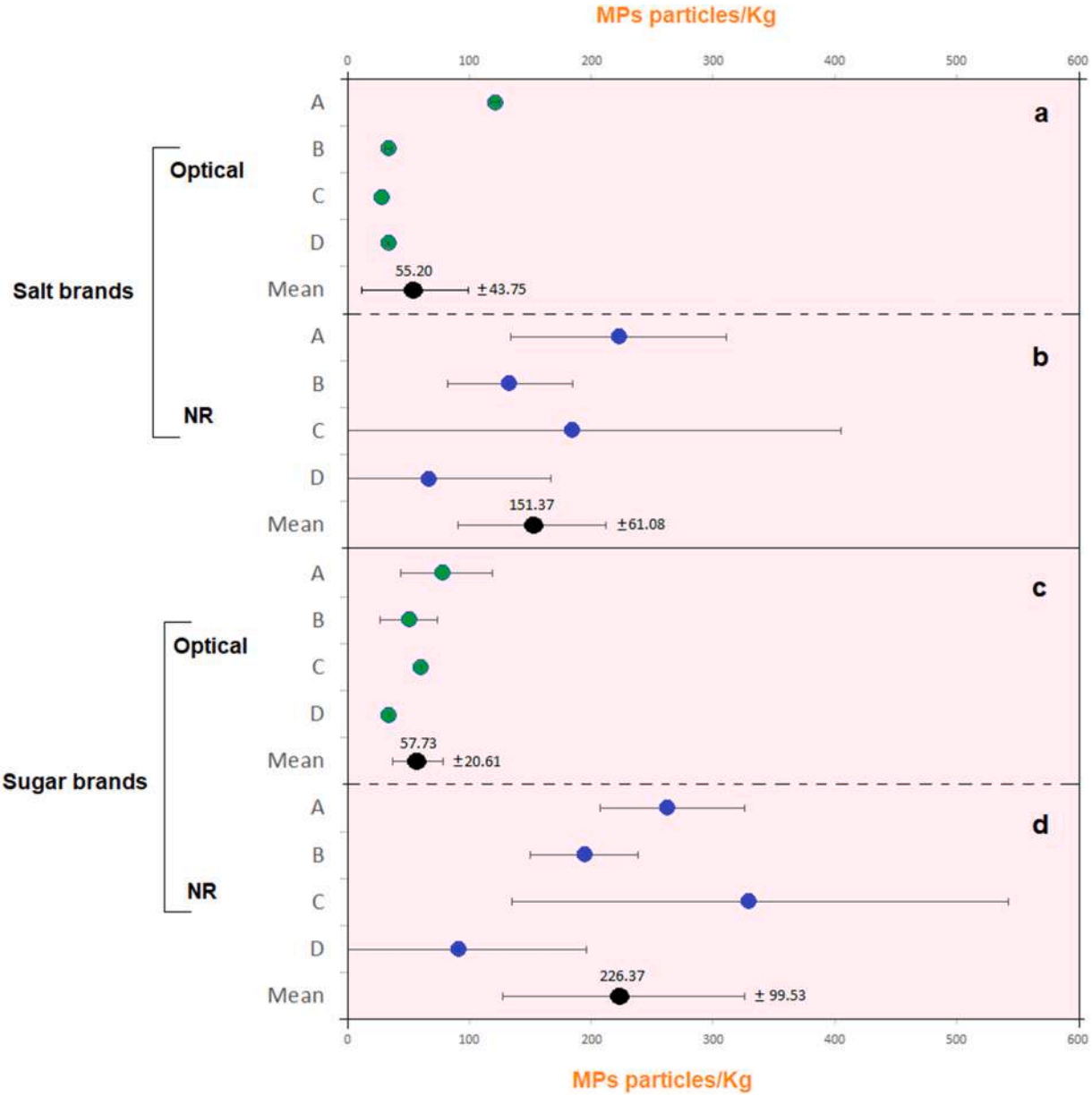

Fig. 1. Histogram of the number of MPs in the different samples of table salt (a and b) and sugar (c and d).

IR is the annual consumption rate of salt (per capita), and C is the MP number (particles/L).

3. Results and discussion

3.1. Quantification of MPs

Fig. 1(a–d) presents a histogram of the total number of MPs projected to 20 samples from 4 brands of table salt and sugar. MPs were observed in 100 % of samples. The results of stereomicroscope analysis (visual method) showed different numbers of MPs in all samples. As depicted in Fig. 1a, various amounts of MPs between 33 ± 4.16 and 120 ± 10.6 MP/kg, with an average of 55.2 ± 43.7 MP/kg were observed in different salt brands, optically. The amounts of MPs in different sugar brands were also in a wide range between 33 ± 4.16 and 80 ± 4.16 with an average of 57.7 ± 20.6 particles/kg (Fig. 1c). The number of observed MPs here is higher than what was reported in sea salt (46 item/kg) by Gundogdu (2018).

Unlike stereomicroscope analysis, the fluorescence staining with NR represents a lot of MP content (Fig. 1b, d). According to the result of the fluorescence microscope, MP content varied from the lowest (77 ± 7.77 MP/kg) to the highest amount (222 ± 36.6 MP/kg), while a mean value of 151 ± 61.8 MP/kg was calculated for salt from all samples (Fig. 1b). The abundance of MPs/kg was even more in sugar, with a range from 105 ± 36.6–338 ± 36.6 and an average of 226 ± 99.5 (Fig. 1d). Moreover, no plastic particles were observed in the two filters arising from procedural blanks and control Petri dishes. This result strongly suggest that the MPs identified originated from the salt and sugar products themselves.

Regarding to the difference in amounts of MPs observed by optical and fluorescence microscopes, it can be concluded that the lower occurrence of MP may be caused by limitation of optical analyzing for fine particles as well as transparency of many MPs. Significant differences between the results of the optical and fluorescence methods are due to the specific affinity of NR to the plastics. Quantification of NR-stained MPs is demonstrated by Prata et al. (2020), as a fast, reliable, and stable method with a good recovery rate for environmental samples. In addition, the results of (Stanton et al., 2019) showed that the lone use of NR staining could lead to the overestimation of MP particles. According to Fig. 1, it is clear that there is a coordination between data of optical and fluorescence methods which affects the data accuracy.

MPs in food containers have been extensively addressed. Contamination of table salt by MPs has been detected in more than 100 brands all over the world with a wide variation of abundances (Gündoğdu, 2018; Iñiguez et al., 2017; Lee et al., 2019; Renzi et al., 2019; Seth and Shriwastav, 2018; Tahir et al., 2019). The highest abundance was reported in Croatia (1.4×10^4 –2.0×10^4 particles·kg^{-1}) (Renzi and Blašković, 2018), followed by Indonesia (1.4×10^4 particles·kg^{-1}) (Kim et al., 2018), Italy (1.6×10^3 –8.2×10^3 particles·kg^{-1}) (Renzi and Blašković, 2018), the United States (0.5×10^2 –8.0×10^2 particles·kg^{-1}) (Kosuth et al., 2018), and China (5.5×10^2 –6.8×10^2 particles·kg^{-1}) (Yang et al., 2015).

Parallel to our study, MPs were detected in all of the salt samples analyzed from Chinese supermarkets by Yang et al. (2015). The highest abundance of MPs (550–681 particles·kg^{-1}) was found in sea salts, then in lake salts (43–364 particles·kg^{-1}) and rock/well salts (7–204 particles·kg^{-1}). Seth and Shriwastav (2018) also reported MP contamination in all eight commercial sea salt brands from India. A total of 626 MP particles were extracted from the studied samples.

The presence of MPs in all of the analyzed Spanish salt samples (sea salt and well salt) from different locations in the country has been reported by Iñiguez et al. (2017). The concentration of MPs ranged from 50 to 280 particles/kg in sea salt samples, and 120–185 particles/kg in well salts.

Although, the MP contamination rate is different in various regions, they cannot be directly compared with each other. One reason may be due to the differently used analytical methods (Zhang et al., 2020). Another reason can be related to manufacturing the salt. Zhang et al. (2020) reported a relatively low level of MPs found in the table salts from several countries, including Iran, Malaysia, Japan, Australia, Portugal, Zealand, France, and some regions of Africa (Zhang et al., 2020).

3.2. Qualification of MPs

3.2.1. Shape of MPs

Fig. 2a, b displays the main shapes of detected MPs in commercial table salt and sugar samples. Two types of MPs were dominant in the samples. With regards to the particle morphology (Fig. 2a), fragments were the predominant type (78.57 %), while fiber structures (21.43 %) were also present in salt samples. The same result was obtained in sugar samples with fragment as the main type of MPs (95.8 %) and fiber at a lower percentage (4.1 %) (Fig. 2b). No film, bead, and other types were isolated from the samples.

Consistent with the findings of this study, others have reported fragments and filaments (fiber) as the main shapes of MPs (Blašković et al., 2017; Karami et al., 2017b). Yang et al. (2015) have reported that fragments and fibers were the predominant types of polymer found in sea salts rather than the other types like pellets and sheets (less than 6 %). Seth and Shriwastav (2018) found both fibers (37 %) and fragments (63 %) as the most abundant plastic particles with large variations in size.

Fig. 3 represents the stereo- and fluorescence microscope images of the MPs with different shapes and sizes. Accordingly, the real color and type of particles could be observed by stereomicroscope. As can be seen in Fig. 3, fragment and fiber are obviously present. In addition, the impurity matter with salt crystals is seen in Fig. 3a, b, d, and f. (Fig. 3b) by optical microscopy. Fig. 3g, h, and i, show the stained microplastics in blue. Previous research demonstrated that some particles separated from the salt samples had a similar composition to their packaging (Karami et al., 2017b). These findings can imply that the contamination of salt products may happen due to the degradation of the packaging materials. The process of producing the table salt may be a reason for the increase in the number of fragments than fibers for existing MPs, including salt rock milling from a mine, centrifuging, and packing. For sugar, some unit processes can affect the size and abundance of fragments. These are washing, preparation of beet slices or shredding the sugarcane, drying the pulp by pressing, centrifuging, and packing. Washing is improved by rubbing the beet slices together which leads to breaking the MPs into fragment particles.

In Iran, table salt and sugar are packaged in plastic containers, and bags. So, after unpacking the bags separation of MP fragments can occur. Also, opening and closing the container may cause release of MPs into samples. Ambient air (indoor and outdoor) in a production unit can be a source for fragments and fibrous MPs (Gasperi et al., 2018).

The importance of the shape of MPs is due to their fate and toxic effect on human health. For example, the migration of fragment particle into cells. Stock et al. (2020) disclosed that the physicochemical conditions of MPs such as shape and size, can affect the passage of MP particles into the gastrointestinal tract (Stock et al., 2020). Gray and Weinstein (2017), reported that the shape of MPs has a great effect on uptaking polymers in grass shrimp. Also, they found that the accumulation of fragments is much higher than spheres and fibers particles in the gut of shrimp. Makhdoumi et al. (2021) reported the evidence of MPs accumulation in the muscle of fish. They found that the fragment particles were about 96 %, while only 4 % of particles was fiber (Makhdoumi et al., 2021b). Qiao et al. (2019) stated that shape influenced the effects of MPs, and should not be disregarded in the health risk evaluation of MPs. They also found that the accumulation of MPs can be conducive to numerous toxic effects in the intestine of zebrafish, which include mucosal damage, inflammation, and metabolism disruption, with fibers possessing greater toxicity than fragments or beads (Qiao

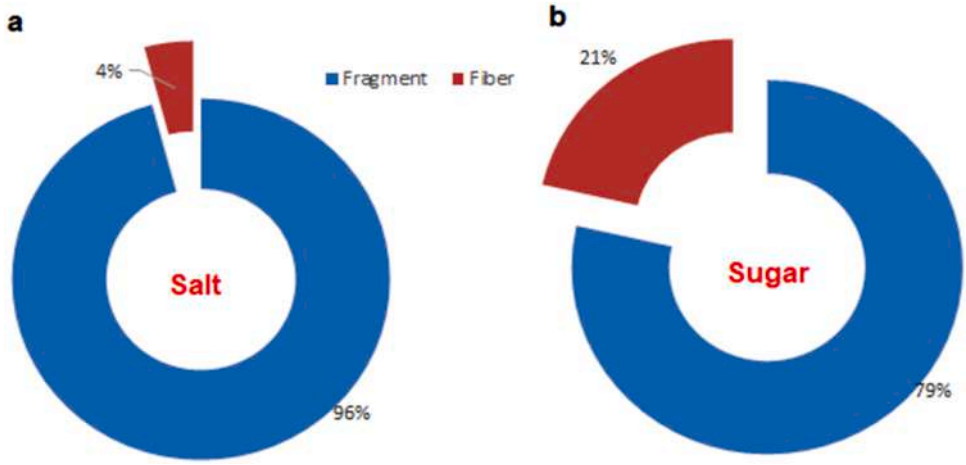

Fig. 2. Major shapes of detected MPs in commercial table salt and sugar samples.

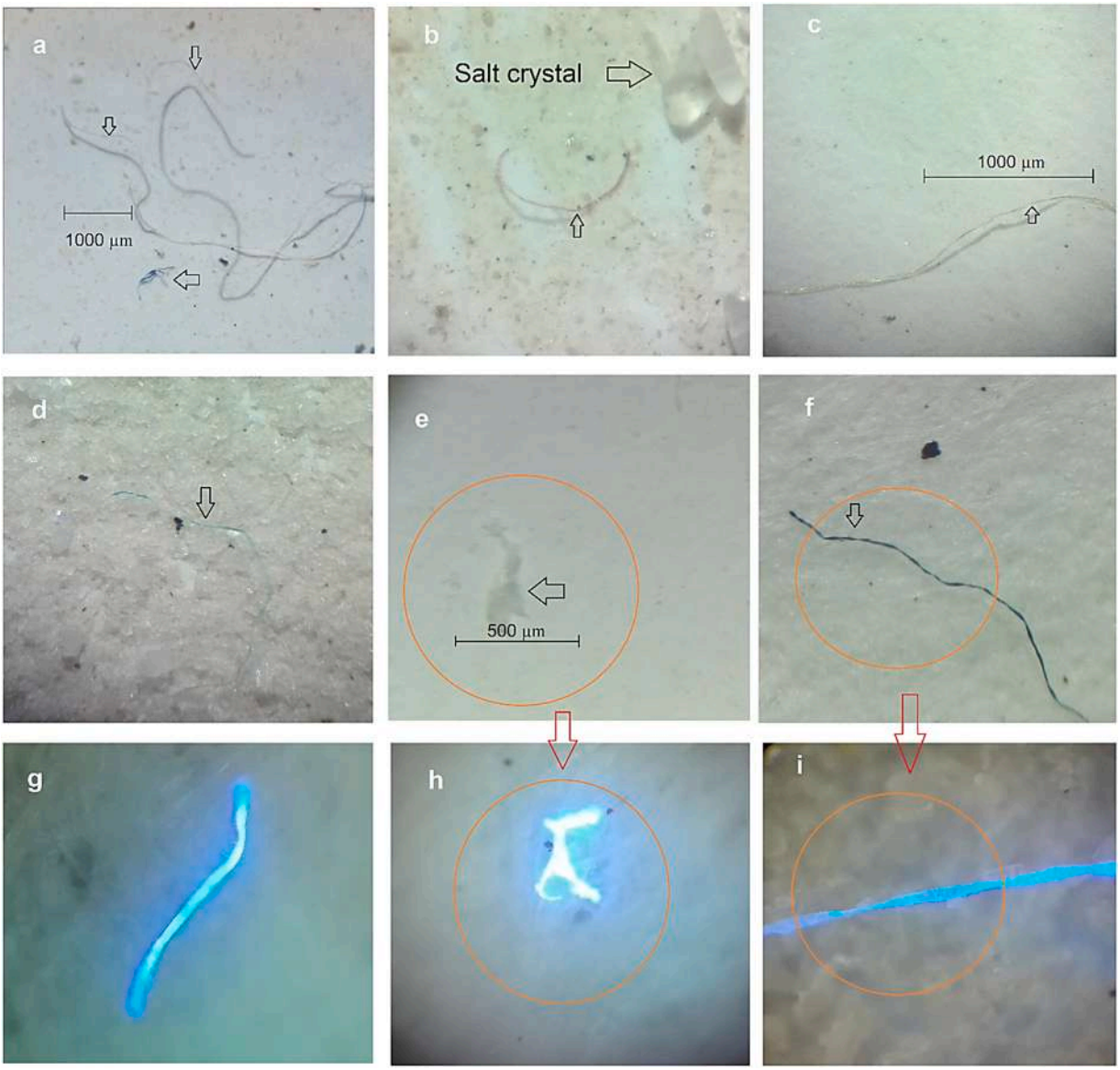

Fig. 3. Photographs of some microplastics under stereo- and fluorescence microscope µm.

et al., 2019).

3.2.1.1. SEM-EDS. SEM can show high-magnification and three-dimensional images of samples to better observe their morphology. Fig. 4 shows some separated fibers, and fragments of MP particles with a magnification of 50–100 μm. In Fig. 4a, b, c, and d, MPs have a smooth surface with no scratches, but some deposited material on the surface is seen. In contrast, other SEM images (Fig. 4, e and f) show breaking and tearing with formation of some deposit materials.

3.2.1.2. EDS. Regarding the contamination of food products like salts, it was suggested that some contamination did not derive from the salt rocks, but originated from sources when products are processed and packaged (Gündoğdu, 2018). Fig. 5 represents some EDS analyses from the surface of MPs. Fig. 5a and b show the MPs from salt brands. Correspondingly, many heavy metals can leak onto the surface of MPs. Here, some toxic metals like Pb, As, Cd, Mg, Si and I were seen. The sharp peaks of carbon and oxygen revealed the composition of the plastic. Fig. 5c shows the sharp peak of sulfur with carbon and oxygen.

3.2.2. Size distribution of MPs

MPs of 1000–5000 μm were the most abundant MPs (59 %) in the salt samples (Fig. 6a); 18 % and 23 % of the remaining MPs were in the size ranges of 500–1000 and < 500 μm, respectively. The size pattern distribution of MPs in the sugar samples showed that a higher number (38 %) of detected MPs had a size of 500–1000 μm (Fig. 6b), while 37 % of MPs were in the 1000–5000 μm size range, with lower amounts (25 %) being < 500 μm. Fig. 6c and d shows the size distribution of MPs based on the shape. Accordingly, a large part of MPs in the forms of fiber and fragment were determined in the 500–5000 μm range in the salt brands. In contrast, in the sugar brands fiber was found only in the size of < 500 μm. In the study of Yang et al. (2015) the size of the MP particles in all of the salt samples ranged from 45 μm to 4.3 mm. They observed the majority of MPs were less than 200 μm, accounting for 55 % of the total number of MPs in samples (Yang et al., 2015). In another study, Seth and Shriwastav (2018) observed that 80 % of the fibers were smaller than 2000 μm, while the fragments were less than 500 μm. The size of fibers was determined from 30 μm to 3.5 mm by Iñiguez et al. (2017) in 21 samples of Spanish table salt (Iñiguez et al., 2017). They also reported no particles smaller than 30 μm. Stock et al. (2019) believed that the larger size of MPs (from 0.1 and 5 mm) could be considered an environmental issue, while particles with size of less than < 150 μm can cause concern due to their bioavailability to humans. In addition, MPs < 4 μm can be adsorbed through intestinal cells (Stock et al., 2019).

3.2.3. Color of MPs

The results showed that the majority of plastic particles in the samples were white (Fig. 7a, b). In salt brands white is the most frequent color, followed by black, red, blue, and green with no colorless particle (Fig. 7a). The frequency of color distribution of MPs in sugar samples was white, red, black, and colorless, respectively, along with blue and green in the same and lower amounts (Fig. 7a).

According to the findings of color patterns, the most release of MPs has happened from the white body of the bottle. Also, white color, red and black were detected more than other types in both salt and sugar brands. According to the type of salt and sugar packaging in Iran, part of the color of MPs may be caused by the containers. The most dominant colors of MPs in the samples of table salts reported by Iñiguez et al. (2017) were black, white, transparent, red and blue (Iñiguez et al., 2017). Kosuth et al. (2018) reported a wide range of colors comprising blue, red/pink, and transparent. Also, dominant colors of black, white, transparent, gray, blue and green were reported by Kim et al. (2018). The wide range of colors in the samples implies that the particles originated from different sources (Lee et al., 2019).

3.2.4. The type of MPs

The FTIR analysis was used to distinguish the main polymer types of identified MPs in salt and sugar. Also, to determine the most likely polymer type of MPs found in the samples, the fingerprints of the functional groups were considered (Fig. 8). According to the literature, in the samples of salts and honey, all types of fabricated polymers have been found, such as polyethylene (PE), polystyrene (PS), polyamide (PA), polypropylene (PP), polystyrene (PS), polyethylene terephthalate

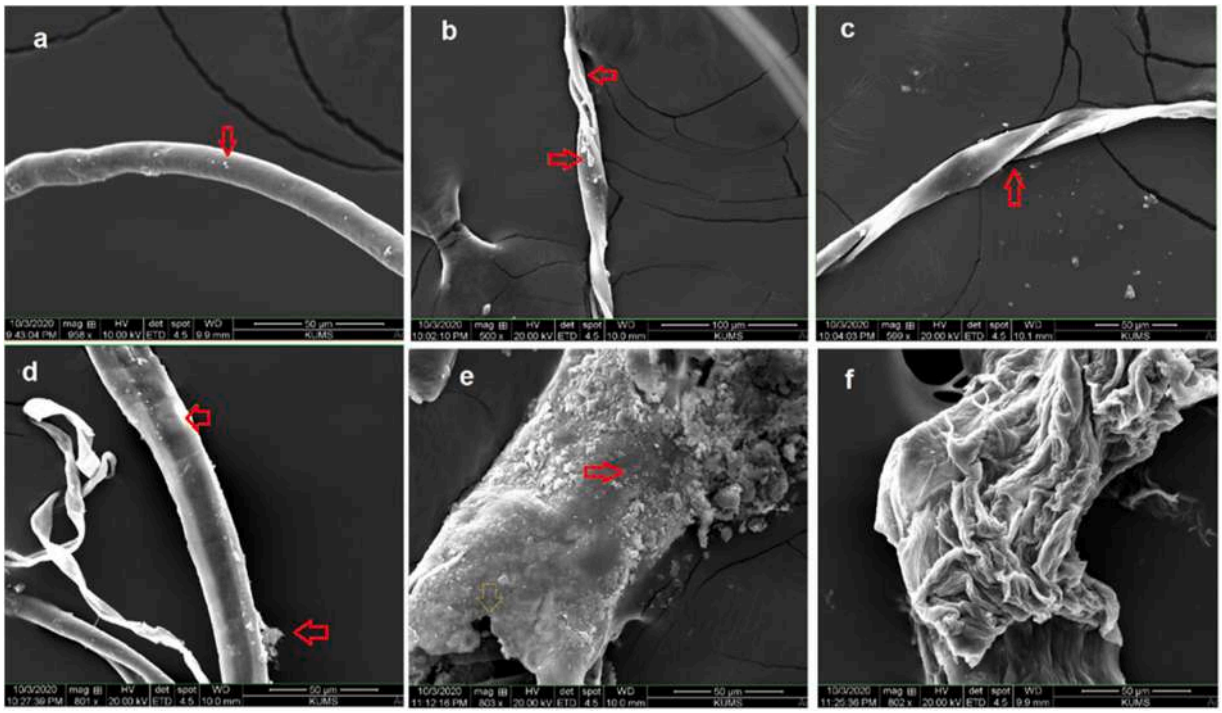

Fig. 4. SEM images of separated MPs.

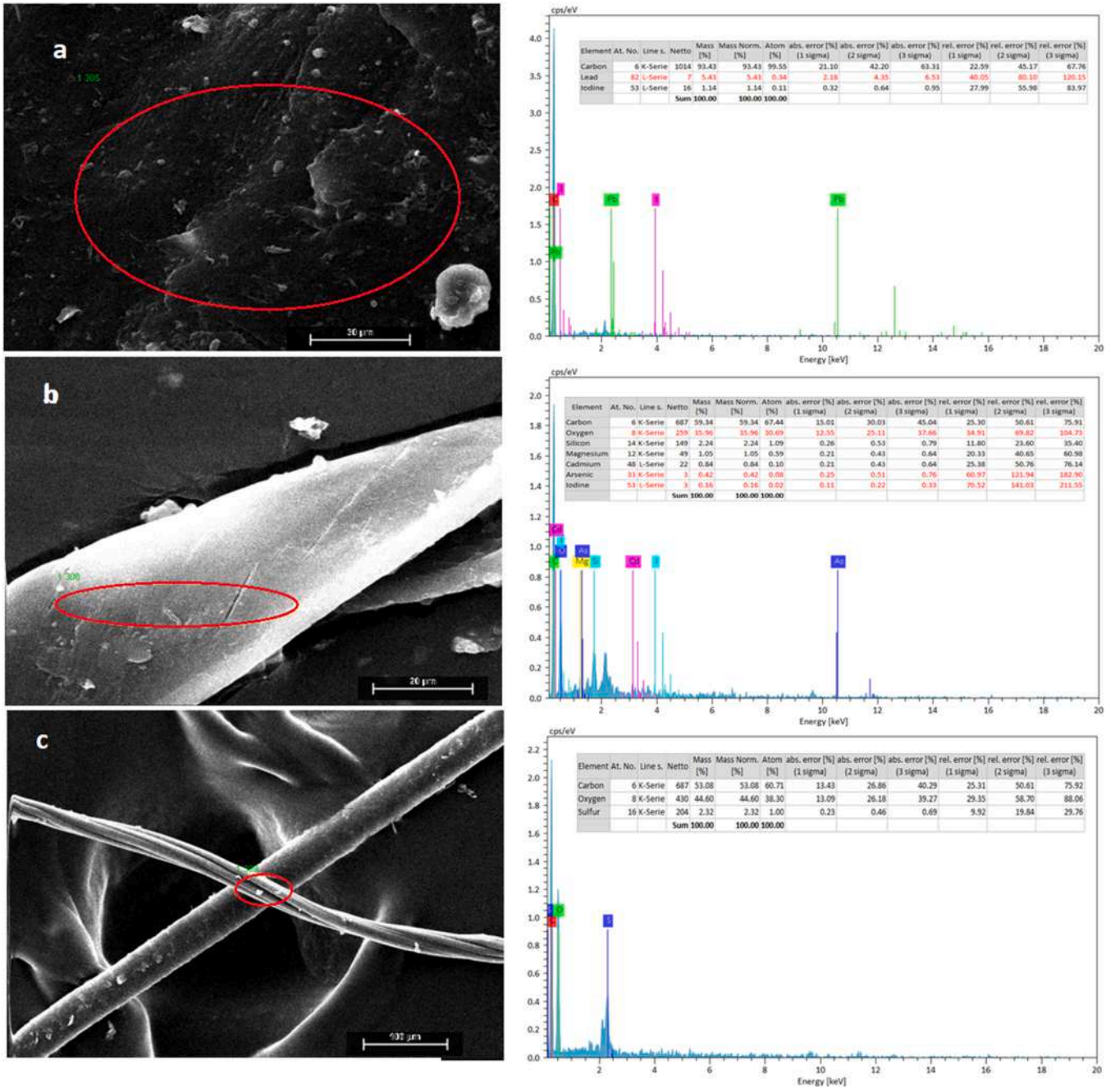

Fig. 5. EXS analysis of MPs.

(PET), polyvinyl chloride (PVC), and nylon (Renzi et al., 2019; Yang et al., 2015). In this study PE (1332, 1438, 2814, 2856, 2964 cm^{-1}) and PP (1124, 1386, 1463, 2856, 2924 cm^{-1}) were identified in salt brands. PE only was found in sugar brands. PE is commonly used as the raw material in plastic bags and in fabricating numerous products. Polymer type can be a reason for its toxicity, bioavailability and affinity for absorbing pollutants, such as organic matter, heavy metal and pathogens. Hwang et al. (2019) found that PP particles can cause cytotoxicity, interfere in the immune system, and enhance potential hypersensitivity. Also, PE can create several problems such as change in lipid profile, alter the gut microbiota, interfere with metabolism and cause liver damage (Kannan and Vimalkumar, 2021).

3.3. Human intake

Iranian salt and sugar consumption is about 6 and 30 kg/year, respectively, greater than the global average. According to the guidelines of the WHO, adults should consume $\leq$ 1.8 kg/year of salt (WHO, 2020), so, high consumption equals a high intake of MPs. The EAI result for Iranian populations is shown in Table 1. The EAI was estimated based on both methods of MPs detection and three contamination levels, minimum, average and maximum of MPs concentration. The results indicate that the maximum annual exposure to MPs is approximately 15, 540 (salt) particles/kg/bw/day and 23,660 (sugar) particles/kg/bw/-year for adults and approximately 3552 (salt) particles/kg/bw/day and 5408 (sugar) particles/kg/bw/year for children.

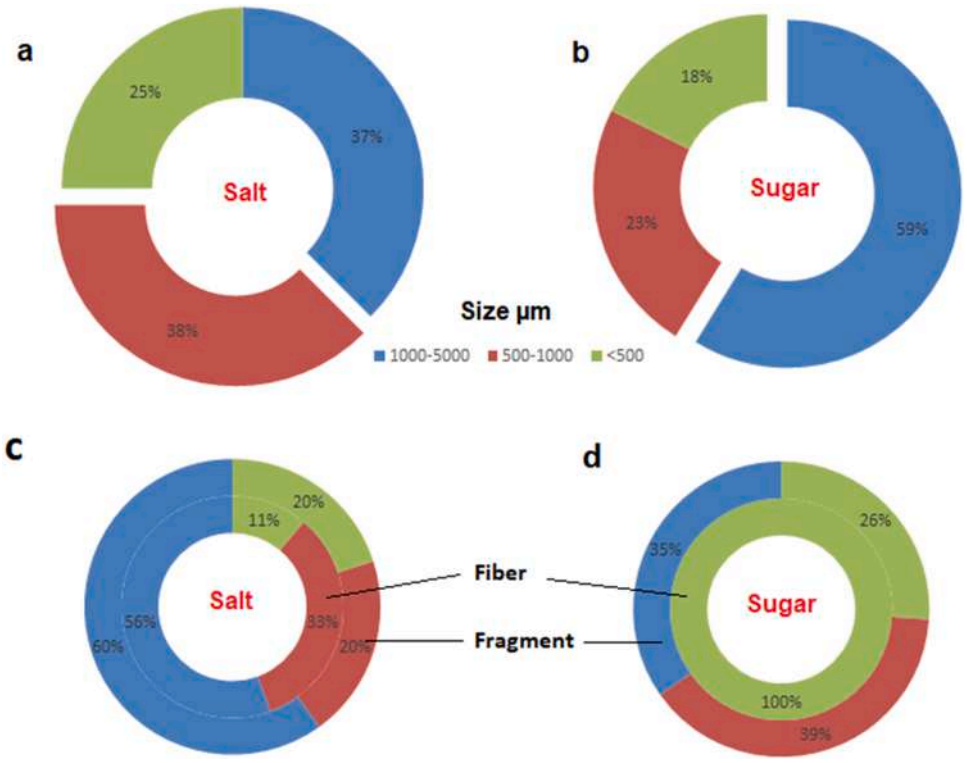

Fig. 6. Major sizes of detected MPs in commercial table salt and sugar samples (a), size distribution based of shape of MPs (b).

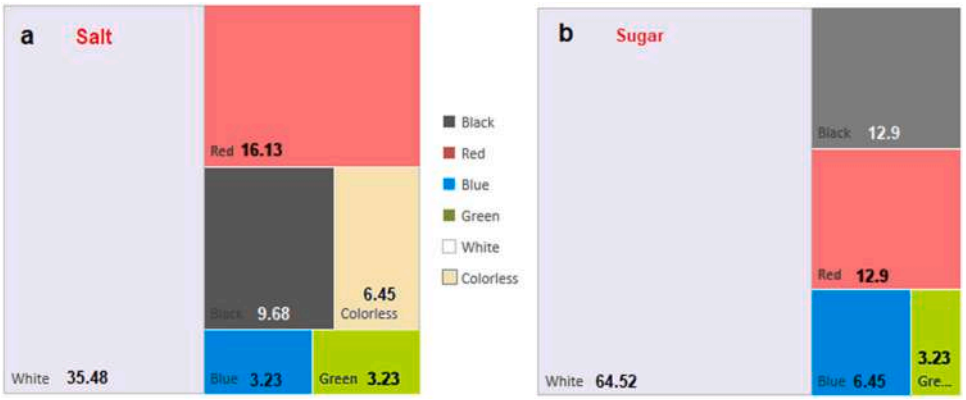

Fig. 7. Major colors of detected MPs in commercial table salt and sugar samples.

3.4. Statistical analysis

Regarding MPs data, statistical analysis was performed based on the expiry dates of salts and sugars samples, and MPs characteristics, such as number, color, size, and type. No significant difference was observed according to the Mann-Whitney, Kruskal- Wallis and Spearman correlation tests.

4. Conclusion

In the present study, the occurrence, quantity, quality, and human exposure of MP contamination of Iranian table salt and sugar were analyzed for the first time. The results showed that all product samples were polluted by MPs, and most fractions were composed of fragment and fiber structures. A large part of MPs found in both products was related to fiber particles with sizes larger than 500 µm. Also, MPs in the salt and sugar samples were mainly white in color, followed by red and black. Polyethylene (PE) and polypropylene (PP) were identified as the most likely polymers. The EAI estimation showed a maximum exposure to humans of about 15,540 (salt) particles/kg/bw/day and 23,660 particles/kg/bw/day (sugar) for adults, based on fluorescence detection data.

CRediT authorship contribution statement

Pouran Makhdoumi: Writing, Methodology, Validation, Editing. **Meghdad Pirsaheb:** Concept, Methodology, Validation. **Abdulfattah Ahmad Amin:** Writing – review & editing. **Sara Kianpour:** Writing – review & editing. **Hooshyar Hossini:** Validation, Writing – review & editing, Software.

Ethical consideration

The authors have observed all the ethical issues throughout the experiment (Including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission,

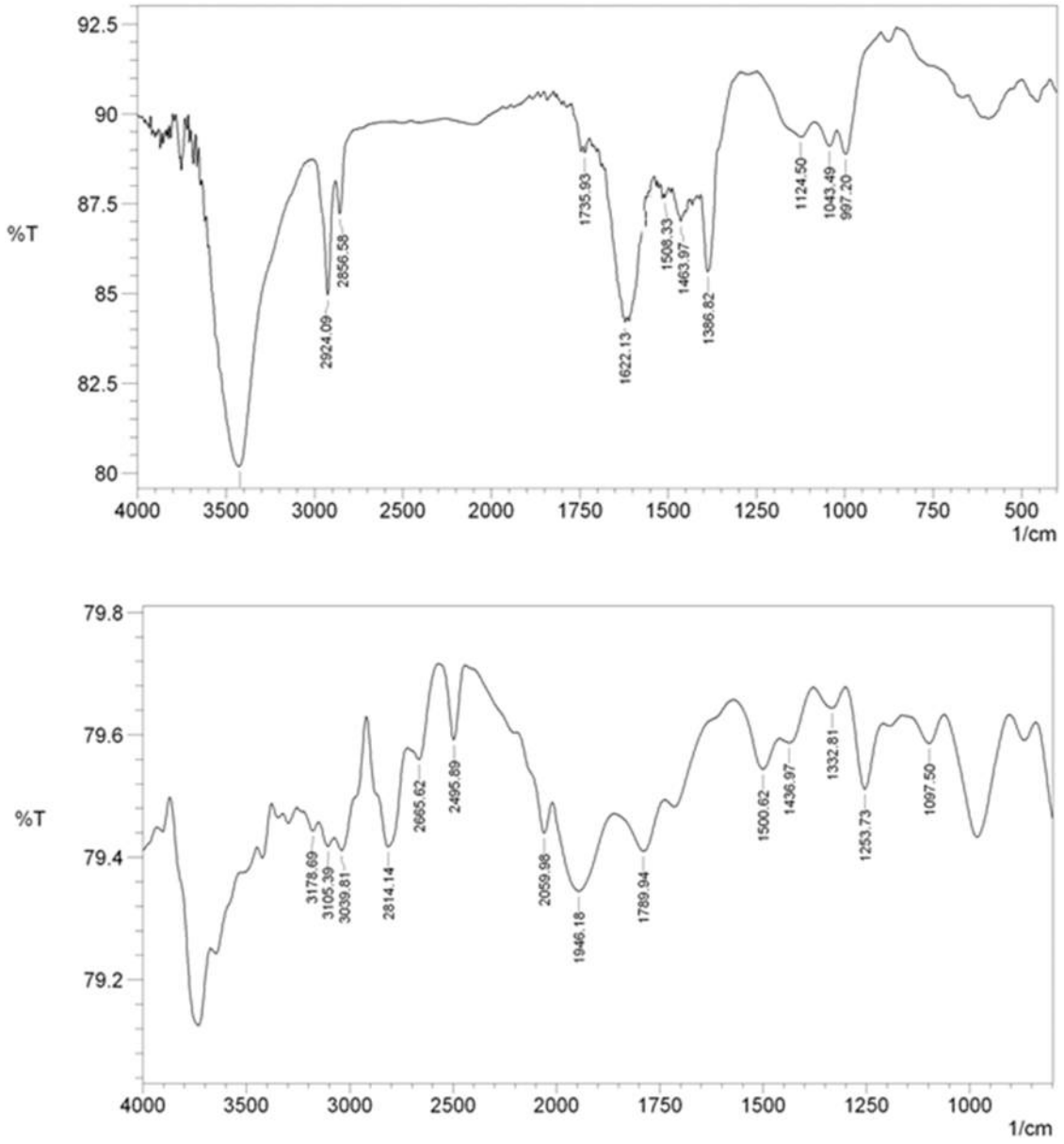

Fig. 8. FTIR spectra of some MPs detected in table salt and sugar samples.

Table 1
EAI for adults and children.

Method of detection	Product	MPs occurrence in salt and sugar (particles/kg)	EAI for Adults (item/kg/bw/year)	EAI for Children (item/kg/bw/year
Optical	Salt	33	2310	528
		55	3850	880
		120	8400	1920
	Sugar	33	2310	528
		57	3990	912
		80	5600	1280
Florescence	Salt	77	5390	1232
		151	10,570	2416
		222	**15,540**	**3552**
	Sugar	105	7350	1680
		226	15,820	3616
		338	**23,660**	**5408**

redundancy, etc.).

Ethical consideration

Ethical issues (Including plagiarism, Informed Consent, misconduct, data fabrication and/or falsification, double publication and/or submission, redundancy, etc) have been completely observed by the authors.

Declaration of interest

The authors report no conflicts of interest. The authors alone are responsible for the content and writing of this article.

Data Availability

Data will be made available on request.

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References

Blašković, A., Fastelli, P., Čižmek, H., Guerranti, C., Renzi, M., 2017. Plastic litter in sediments from the Croatian marine protected area of the natural park of Telaščica bay (Adriatic Sea). Mar. Pollut. Bull. 114 (1), 583–586.

Brown, I.J., Tzoulaki, I., Candeias, V., Elliott, P., 2009. Salt intakes around the world: implications for public health. Int. J. Epidemiol. 38 (3), 791–813.

Browne, M.A., Galloway, T., Thompson, R., 2007. Microplastic—an emerging contaminant of potential concern? Integr. Environ. Assess. Manag.: Int. J. 3 (4), 559–561.

Chua, E.M., Shimeta, J., Nugegoda, D., Morrison, P.D., Clarke, B.O., 2014. Assimilation of polybrominated diphenyl ethers from microplastics by the marine amphipod, Allorchestes compressa. Environ. Sci. Technol. 48 (14), 8127–8134.

Deng, Y., Zhang, Y., Lemos, B., Ren, H., 2017a. Tissue accumulation of microplastics in mice and biomarker responses suggest widespread health risks of exposure. Sci. Rep. 7 (1), 46687.

Deng, Y., Zhang, Y., Lemos, B., Ren, H., 2017b. Tissue accumulation of microplastics in mice and biomarker responses suggest widespread health risks of exposure. Sci. Rep. 7 (1), 1–10.

Donaldson, K., Stone, V., Gilmour, P., Brown, D., MacNee, W., 2000. Ultrafine particles: mechanisms of lung injury. Philos. Trans. R. Soc. Lond. Ser. A: Math., Phys. Eng. Sci. 358 (1775), 2741–2749.

Eerkes-Medrano, D., Thompson, R.C., Aldridge, D.C., 2015. Microplastics in freshwater systems: a review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. Water Res. 75, 63–82.

Engler, R.E., 2012. The complex interaction between marine debris and toxic chemicals in the ocean. Environ. Sci. Technol. 46 (22), 12302–12315.

FAOSTAT, 2021. Sugar Consumption Per Capita in Iran.

Fendall, L.S., Sewell, M.A., 2009. Contributing to marine pollution by washing your face: microplastics in facial cleansers. Mar. Pollut. Bull. 58 (8), 1225–1228.

Frère, L., Maignien, L., Chalopin, M., Huvet, A., Rinnert, E., Morrison, H., Kerninon, S., Cassone, A.-L., Lambert, C., Reveillaud, J., Paul-Pont, I., 2018. Microplastic bacterial communities in the Bay of Brest: Influence of polymer type and size. Environ. Pollut. 242, 614–625.

Gasperi, J., Wright, S.L., Dris, R., Collard, F., Mandin, C., Guerrouache, M., Langlois, V., Kelly, F.J., Tassin, B., 2018. Microplastics in air: are we breathing it in? Curr. Opin. Environ. Sci. Health 1, 1–5.

Gray, A.D., Weinstein, J.E., 2017. Size- and shape-dependent effects of microplastic particles on adult daggerblade grass shrimp (Palaemonetes pugio). Environ. Toxicol. Chem. 36 (11), 3074–3080.

Gündoğdu, S., 2018. Contamination of table salts from Turkey with microplastics. Food Addit. Contam.: Part A 35 (5), 1006–1014.

Hwang, J., Choi, D., Han, S., Choi, J., Hong, J., 2019. An assessment of the toxicity of polypropylene microplastics in human derived cells. Sci. Total Environ. 684, 657–669.

Iñiguez, M.E., Conesa, J.A., Fullana, A., 2017. Microplastics in Spanish table salt. Sci. Rep. 7 (1), 1–7.

Jee, A.-Y., Park, S., Kwon, H., Lee, M., 2009. Excited state dynamics of Nile Red in polymers. Chem. Phys. Lett. 477 (1), 112–115.

Kannan, K., Vimalkumar, K., 2021. A review of human exposure to microplastics and insights into microplastics as obesogens. Front. Endocrinol. 12.

Karami, A., Golieskardi, A., Choo, C.K., Larat, V., Galloway, T.S., Salamatinia, B., 2017a. The presence of microplastics in commercial salts from different countries. Sci. Rep. 7, 46173.

Karami, A., Golieskardi, A., Choo, C.K., Larat, V., Galloway, T.S., Salamatinia, B., 2017b. The presence of microplastics in commercial salts from different countries. Sci. Rep. 7 (1), 1–11.

Karami, A., Golieskardi, A., Choo, C.K., Larat, V., Karbalaei, S., Salamatinia, B., 2018. Microplastic and mesoplastic contamination in canned sardines and sprats. Sci. Total Environ. 612, 1380–1386.

Katsnelson, A., 2015. News Feature: Microplastics present pollution puzzle. Proceedings of the National Academy of Sciences 112(18), pp. 5547–5549.

Kim, J.-S., Lee, H.-J., Kim, S.-K., Kim, H.-J., 2018. Global pattern of microplastics (MPs) in commercial food-grade salts: sea salt as an indicator of seawater MP pollution. Environ. Sci. Technol. 52 (21), 12819–12828.

Kosuth, M., Mason, S.A., Wattenberg, E.V., 2018. Anthropogenic contamination of tap water, beer, and sea salt. PLoS One 13 (4), e0194970.

Law, K.L., Thompson, R.C., 2014. Microplastics in the seas. Science 345 (6193), 144–145.

Layeghiasl, M., Malekzadeh, J., Shams, M., Maleki, M., 2018. Opinions of yasuj people about salt consumption: a qualitative study with content analysis approach. Armaghane danesh 23 (4), 459–475.

Lee, H., Kunz, A., Shim, W.J., Walther, B.A., 2019. Microplastic contamination of table salts from Taiwan, including a global review. Sci. Rep. 9 (1), 1–9.

Lo, H.-S., Wong, C.-Y., Tam, N.F.-Y., Cheung, S.-G., 2019. Spatial distribution and source identification of hydrophobic organic compounds (HOCs) on sedimentary microplastic in Hong Kong. Chemosphere 219, 418–426.

Lusher, A.L., Tirelli, V., O'Connor, I., Officer, R., 2015. Microplastics in Arctic polar waters: the first reported values of particles in surface and sub-surface samples. Sci. Rep. 5, 14947.

Makhdoumi, P., Amin, A.A., Karimi, H., Pirsaheb, M., Kim, H., Hossini, H., 2021a. Occurrence of microplastic particles in the most popular Iranian bottled mineral water brands and an assessment of human exposure. J. Water Process Eng. 39, 101708.

Makhdoumi, P., Hossini, H., Nazmara, Z., Mansouri, K., Pirsaheb, M., 2021b. Occurrence and exposure analysis of microplastic in the gut and muscle tissue of riverine fish in Kermanshah province of Iran. Mar. Pollut. Bull. 173, 112915.

Mintenig, S.M., Löder, M.G.J., Primpke, S., Gerdts, G., 2019. Low numbers of microplastics detected in drinking water from ground water sources. Sci. Total Environ. 648, 631–635.

Nel, A., Xia, T., Madler, L., Li, N., 2006. Toxic potential of materials at the nanolevel. science 311 (5761), 622–627.

Organization, W.H., 2012. Guideline: Sodium Intake for Adults and Children. World Health Organization.

Organization, W.H., 2019. Microplastics in drinking-water.

Peixoto, D., Pinheiro, C., Amorim, J., Oliva-Teles, L., Guilhermino, L., Vieira, M.N., 2019. Microplastic pollution in commercial salt for human consumption: a review. Estuar., Coast. Shelf Sci. 219, 161–168.

Picheta, R., 2018. Microplastics found in human stools, research finds. CNN. CNN.

Possatto, F.E., Barletta, M., Costa, M.F., do Sul, J.A.I., Dantas, D.V., 2011. Plastic debris ingestion by marine catfish: an unexpected fisheries impact. Mar. Pollut. Bull. 62 (5), 1098–1102.

Prata, J.C., 2018. Airborne microplastics: consequences to human health? Environ. Pollut. 234, 115–126.

Prata, J.C., Alves, J.R., da Costa, J.P., Duarte, A.C., Rocha-Santos, T., 2020. Major factors influencing the quantification of Nile Red stained microplastics and improved automatic quantification (MP-VAT 2.0). Sci. Total Environ. 719, 137498.

Qiao, R., Deng, Y., Zhang, S., Wolosker, M.B., Zhu, Q., Ren, H., Zhang, Y., 2019. Accumulation of different shapes of microplastics initiates intestinal injury and gut microbiota dysbiosis in the gut of zebrafish. Chemosphere 236, 124334.

Renzi, M., Blašković, A., 2018. Litter & microplastics features in table salts from marine origin: Italian versus Croatian brands. Mar. Pollut. Bull. 135, 62–68.

Renzi, M., Grazioli, E., Bertacchini, E., Blašković, A., 2019. Microparticles in table salt: Levels and chemical composition of the smallest dimensional fraction. J. Mar. Sci. Eng. 7 (9), 310.

Ribic, C.A., Sheavly, S.B., Rugg, D.J., Erdmann, E.S., 2010. Trends and drivers of marine debris on the Atlantic coast of the United States 1997–2007. Mar. Pollut. Bull. 60 (8), 1231–1242.

Ryan, P.G., Moore, C.J., Van Franeker, J.A., Moloney, C.L., 2009. Monitoring the abundance of plastic debris in the marine environment. Philos. Trans. R. Soc. B: Biol. Sci. 364 (1526), 1999–2012.

Schymanski, D., Goldbeck, C., Humpf, H.-U., Fürst, P., 2018. Analysis of microplastics in water by micro-Raman spectroscopy: release of plastic particles from different packaging into mineral water. Water Res. 129, 154–162.

Setälä, O., Fleming-Lehtinen, V., Lehtiniemi, M., 2014. Ingestion and transfer of microplastics in the planktonic food web. Environ. Pollut. 185, 77–83.

Seth, C.K., Shriwastav, A., 2018. Contamination of Indian sea salts with microplastics and a potential prevention strategy. Environ. Sci. Pollut. Res. 25 (30), 30122–30131.

Singh, P., Ban, Y., Kashyap, L., Siraree, A., Singh, J., 2020. Sugar and sugar substitutes: recent developments and future prospects. Sugar Sugar Deriv.: Chang. Consum. Prefer. 39–75.

Stanton, T., Johnson, M., Nathanail, P., Gomes, R.L., Needham, T., Burson, A., 2019. Exploring the efficacy of Nile Red in microplastic quantification: a costaining approach. Environ. Sci. Technol. Lett. 6 (10), 606–611.

Stock, V., Böhmert, L., Lisicki, E., Block, R., Cara-Carmona, J., Pack, L.K., Selb, R., Lichtenstein, D., Voss, L., Henderson, C.J., 2019. Uptake and effects of orally ingested polystyrene microplastic particles in vitro and in vivo. Arch. Toxicol. 93 (7), 1817–1833.

Stock, V., Fahrenson, C., Thuenemann, A., Dönmez, M.H., Voss, L., Böhmert, L., Braeuning, A., Lampen, A., Sieg, H., 2020. Impact of artificial digestion on the sizes and shapes of microplastic particles. Food Chem. Toxicol. 135, 111010.

Tahir, A., Taba, P., Samawi, M., Werorilangi, S., 2019. Microplastics in water, sediment and salts from traditional salt producing ponds. Glob. J. Environ. Sci. Manag. 5 (4), 431–440.

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.

Teuten, E.L., Saquing, J.M., Knappe, D.R., Barlaz, M.A., Jonsson, S., Björn, A., Rowland, S.J., Thompson, R.C., Galloway, T.S., Yamashita, R., 2009. Transport and release of chemicals from plastics to the environment and to wildlife. Philos. Trans. R. Soc. B: Biol. Sci. 364 (1526), 2027–2045.

Velzeboer, I., Kwadijk, C.J.A.F., Koelmans, A.A., 2014. Strong sorption of PCBs to nanoplastics, microplastics, carbon nanotubes, and fullerenes. Environ. Sci. Technol. 48 (9), 4869–4876.

Wang, C., Zhao, J., Xing, B., 2021. Environmental source, fate, and toxicity of microplastics. J. Hazard. Mater. 407, 124357.

Wang, F., Wong, C.S., Chen, D., Lu, X., Wang, F., Zeng, E.Y., 2018. Interaction of toxic chemicals with microplastics: a critical review. Water Res. 139, 208–219.

WHO, 2020. Salt reduction.

Wright, S.L., Kelly, F.J., 2017. Plastic and human health: a micro issue? Environ. Sci. Technol. 51 (12), 6634–6647.

Yang, D., Shi, H., Li, L., Li, J., Jabeen, K., Kolandhasamy, P., 2015. Microplastic pollution in table salts from China. Environ. Sci. Technol. 49 (22), 13622–13627.

Zhang, Q., Xu, E.G., Li, J., Chen, Q., Ma, L., Zeng, E.Y., Shi, H., 2020. A review of microplastics in table salt, drinking water, and air: direct human exposure. Environ. Sci. Technol. 54 (7), 3740–3751.
Zitko, V., Hanlon, M., 1991. Another source of pollution by plastics skin cleaners with plastic scrubbers. Mar. Pollut. Bull. 22, 41–42.
Zuccarello, P., Ferrante, M., Cristaldi, A., Copat, C., Grasso, A., Sangregorio, D., Fiore, M., Oliveri Conti, G., 2019. Exposure to microplastics (<10 μm) associated to plastic bottles mineral water consumption: The first quantitative study. Water Res. 157, 365–371.