PRESENCE OF MICROPLASTIC PARTICLES IN EDIBLE SALTS IN BANGLADESH

Md Tasneem Zafar¹, Md Wahidul Haque¹, S M Shamsul Huda¹ and Mohammad Mosharraf Hossain^{*1}

¹Institute of Forestry and Environmental Sciences, University of Chittagong, Chittagong 4331, Bangladesh

*Corresponding Author

ABSTRACT

Ubiquitous contamination of the marine environment by microplastic is a global environmental disaster. Occurrence of microplastics in human excreta indicates their entry into the human food chain indicating the urgent need to explore candidate foods and food additives for microplastic contamination from public health perspective. The number of publications on microplastics contamination of various matrices is on a rise globally unlike in Bangladesh where it has yet to get attention. This study aimed to address this gap by evaluating the occurrence of microplastic in edible salts – the most common food additives in the country. Salt is traditionally produced by solar drying of microplastic contaminated sea water without refinement. Hence, the occurrence of microplastic is highly probable in raw sea salts which may gradually decrease under different intensities of refinement. In this study, raw salt samples (21) were collected from salt farms in Cox's Bazar and refined salt samples (8) were collected from local salt refineries while super refined salt samples (3) were collected from the market. Microplastics extraction was based on NOAA protocol that combined density separation technique with wet peroxide oxidation to remove any non-plastic floating particles. Isolated microplastic particles (MPP) were then observed under a microscope, photographed and analyzed by image analysis software to get their size, shape and, type color. Raw (unrefined) salt contained 2105 MPP/kg compared to 283 MPP/kg in refined salts. In contrast, super refined salt contained only 4 MPP in 5 kg. Unlike most other studies which focus on 0.33-5mm MPPs - this study has also looked for smaller particles, as smaller particles are more harmful to health. Worrisomely, minute MPPs, as small as 0.058mm, were the most dominant in salt. The results indicate a high level of microplastic contamination in raw salts and arrested the need to make mass people aware about the microplastic contamination in refined soil and to promote the use of super-refined salt for human consumption.

Keywords: Microplastics, Sea salt, Salt, Bangladesh, Salt refining.

1. INTRODUCTION

Plastics are predominantly synthetic organic polymers of high molecular mass, most commonly derived from petrochemicals. When other materials became scarce during World War II, researchers looked to synthetic polymers to fill the gaps, as they being synthetic their stock depends on the hands of producers, and are also easy and cheap to make, not saying requiring a very short time comparing to any natural alternatives. Until very recent times, typically the fate of the product after the end initial lifetime not considered in designing a plastic or its product. This resulted in huge amount of plastic debris, which by dint of the very properties that made them noble material have become a burden on the environment, pervading all the spheres. Plastic marine debris has been an environmental concern for decades. Despite the increased international attention, the build-up of these materials in the environment is considered problematic due to an increasing global plastic production and the continuing improper disposal of plastic waste. These mismanaged discarded plastics; going through different media gets shredded and fragmented repeatedly by various weathering agents like the mechanical forces exerted by wave action, the influence of UV radiation, the oxidative properties of the atmosphere and hydrolytic properties of seawater etc. and gets converted to smaller particles (Carson, Colbert, Kaylor, & McDermid, 2011). Microplastics are plastic particles smaller than 5.0 mm in size(Arthur, Baker, & Bamford, 2008). Particles< 5 mm, are of especial concern mainly due to their long environmental persistence, small size, high surface/volume ratio, and their capability of entry into the cells and induce adverse effects(Iñiguez, Conesa, & Fullana, 2017). There are two main ways microplastics are formed and enter a body of water: primary and secondary microplastics. Primary microplastics consist of plastic material, which are, from the very production stage, are micro, for example, micro beads used in personal care products such as face scrubs. Secondary microplastic introductions occur when larger plastic items enter a beach or ocean and undergo mechanical, photo (oxidative) or biological degradation. This degradation breaks the larger pieces into progressively smaller plastic fragments which eventually become undetectable to the naked eye. Owing to their size, Microplastics have totally different properties and hazards, compared to plastics in general. It has been stressed that more investigation is needed to understand the impacts of microplastics on marine organisms and food web (Wright, Thompson, & Galloway, 2013). Furthermore, marine microplastics are far more difficult to trace back to its origin than macro-debris, which presents an additional obstacle in managing this type of pollution. Very recently microplastics have been found in human excreta, which indicate their entrance in out digestive tracts, through edibles (Liebmann, Bucsics, Königshofer, & Köppel, 2018). Since these are not our target intake, hence we may conclude that our intakes have been contaminated by micro plastics. Microplastics have the capacity to translocate across living cells to the lymphatic and circulatory system in humans and other mammals, possibly via the intestine (Rieux et al., 2005). Provided that they are considered nondegradable, Microplastics have the potential to bioaccumulate in secondary organs, with possible impacts in the immune system and cell health (Smith, Love, Rochman, & Neff, 2018). Moreover, these particles can adsorb and concentrate high levels of hydrophobic organic contaminants (HOCs) (e.g., PAHs, organochlorine pesticides and polychlorinated biphenyls), metals (e.g., cadmium, lead, selenium, chromium), non-metals, and additives/monomers (e.g., bisphenol- A, polybrominated diphenvl ethers, nonvlphenols and octvlphenol) (Smith et al., 2018) The question that will than arise is what items do carry these contaminants to our guts. Fish are known to ingest microplastic, making them a potential vector of toxic chemicals through food chains and into human diets. Due to typically high concentrations of microplastic along urbanized coasts, there is a need for more research in these areas to quantify and assess the extent of microplastic ingestion by fishes and other marine biota specially the species humans consume. Similarly, since dehydration of near coast sea-water is the main mode of salt production, so, the status of microplastics contamination salt, which is consumed by everyone have become essential to investigate (Kim, Lee, Kim, & Kim, 2018). In all, the objectives of this study were to evaluate microplastic contamination in salt, to characterize the microplastics available in salt samples and to check the impact of salt refinement on microplastic removal.

2. METHODOLOGY

2.1. Sample Collection

Raw salt samples were collected from field as well as during transporting to the milling area of Islampur. Raw salt collected were from Mognama, Ilishia, Kutubdia, Khutakhali, Gumatali, Moheshkhali, and Badarkhali, as well as a batch of imported salt, and a batch of wash salt, (which is obtained in the milling area, by drying the brine that have been discarded after being repeatedly used for refining). Salt refineries of Cox's Bazar are all located in the Islampur industrial area under Eidgah upazilla and sample were collected from 8 of 24 operational mills, and samples were collected. Since most of the mills at a time process salts of one batch entirely, so a total 8 batches of refined salts were collected. Packed refined and super refined salts were collected from the market.

2.2. Laboratory Analysis

The laboratory processing of any microplastics study is isolation and separation from the matrix. In this study, the protocol developed by NOAA for microplastics study was adopted. According to the NOAA protocol, microplastics are plastic particles that can stand oxidation by a standard Hydrogen peroxide solution, and shows floatation in a 5M NaCl solution. The usual size range of microplastics in most studies is between 5-0.33mm, however, in this study, all plastics <5mm are considered.

First the collected salts are sundried to remove moisture. Then 20gm of each sample was dissolved in previously prepared 5M brine, made by dissolving super-refined table salt, vacuum filtered to remove any particles present. After thorough mixing in a magnetic stirrer, it was let to rest, so that the heavy undissolved particles to settle down. Then the top portion containing any floating debris was removed. Equal amount of 30% H2O2 solution, (lab grade) was mixed, along with acidified Fe(II) solution (0.05 M) (Prepared by adding 7.5 g of FeSO4.7H20 to 500 mL of water and 3 mL of concentrated sulfuric acid) catalyst. It was again placed on the magnetic stirrer at 600c until a fume was seen. After cooling the mixture, again density separation was done, and top portion was pipetted out on a petri dish, and dried and observed under an Omron Microscope with 10X zooming.

Images were taken and analyzed using image processing software, and the physical characters of the particles identified as plastics under microscope or the image was recorded.

3. RESULTS

Characterization was done based on physical appearance, as photographed under the microscope. The size, shape, type and color of the objects were the parameters observed. The shapes found commonly were angular, bar-shaped, elongated, oval, irregular and round. All the obtained particles were classified on the basis of their appearance into plastic dust, filaments, foam, broken plastic fragment and polythene fragment. Only 4 particles were found in 5kg of super refined salt, which suggests that the process of super refining practically removes any microplastics from the salt. Accounting for the very minimal abundance, super refined salt is included under the refined category in this study.

3.1 Particles Distribution

The percentage ratio of the particles as they fall under several categories of different parameters was found to be as follows:

3.1.1 Distribution of Different Types

Microplastics from both refined and unrefined samples were categorized as dust, filament, foam, broken fragment and shredded polythene fragments, based on their appearance under microscope. The distributions of both the types are shown in the figure 1. In unrefined salt, fragment was found to be most dominant, followed by dust. The order of dominance was found to be fragment, dust, filament, foam and finally polythene in raw salt. Fragment was most dominant, in refined salt, too. The order of the other types was dust, polythene, filament and foam.

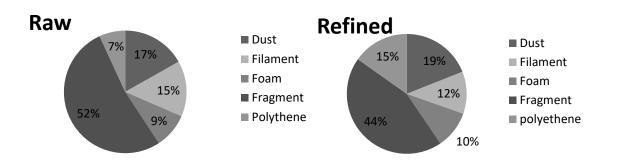


Figure 1: Distribution of plastic types in raw and refined salt

3.1.2 Distribution of Different Colors

In raw salt, black plastic particles were found to be most dominant (24%). The abundance of other colors was in the following order: brown, transparent, orange, red, yellow, ash, greenish, blackish, green, brownish, white and blue. Black was also found to be the dominant color in refined salt. The other colors, was sorted as orange brown, green, red, ash, white, yellow, blue and transparent.

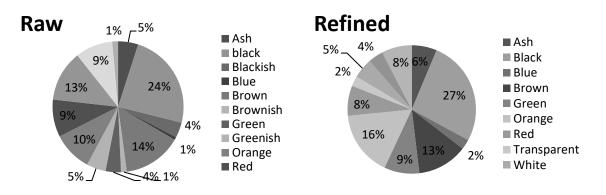


Figure 2: Color distribution of microplastic in raw and refined salt

3.1.3 Distribution of Different Shapes

The order of dominance of shapes of plastic fragments, found in raw salt was Irregular>Elongated>Angular>Bar>Round>Oval. In refined salt, the order of the shapes were Irregular>Elongated>Angular>Bar>Round>Oval>others. Thus the shape distribution of plastics in refined and unrefined salts was found to be same, indicating that the refining process works irrespective of shapes of particles.

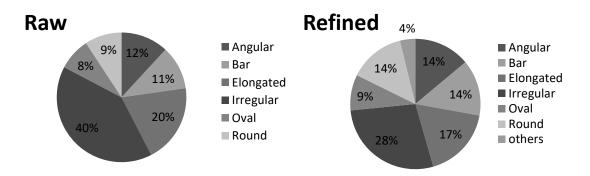


Figure 3: Distribution of shapes of microplastics in raw and refined salt

ICCESD-2020-5118-4

3.1.4 Distribution of Different Sizes

From the figure, it is apparent that very small particles, below 0.33mm, which are often ignored from microplastics studies, including Eerkes-Medrano, Thompson, & Aldridge, 2015; Kim et al., 2018; Lam et al., 2018 etc. are the most dominant in raw sea salt. Hence care should be taken for these too, as these are also not our target intakes. The share of other size ranges were 0.33-1mm>1-2mm>2-3mm>3-4mm> 4-5mm, i.e abundance reduces as size increases. In refined salt, as we can see from the above figure 4.8, the most dominant was the particles of very small size (<0.33mm), followed by the next 2 groups, covering particles of 0.33-1 mm and 1-2mm respectively. It is however notable, that the larger particles of size above 2mm was not significantly present at all, which might be the effect of refining.

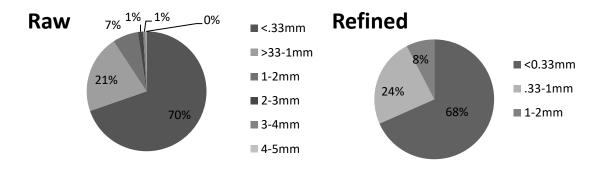


Figure 4: Distribution of size of microplastics in raw and refined salt

3.2 Particles Association

3.2.1 Association of Size and Type

Figure 5 shows, is the abundance of different types of particles from various classes. It is significant that in raw salt filaments of size between 0.33 and 1mm are as abundant as fragments of the same category, which is the dominant type. Apart from the figure, only 1 fragment particle of size class 4-5mm was found in raw salt.

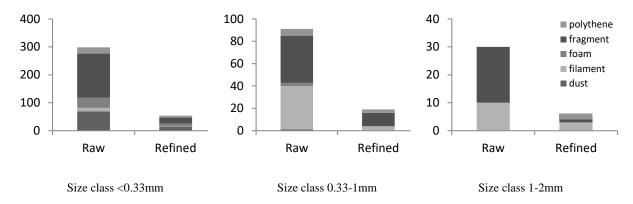


Figure 5: Size-wise distribution of types of particles in raw and refined salt

3.2.2 Association of Shape and Type

Figure 6 shows us a complete comparative picture among raw and refined salt, along with types variation. As can be seen from this graph, the abundance of microplastic greatly varies between raw and refined salt. In both the types, fragments are dominant, and irregular shaped fragments are most abundant. Filaments are another dominant type in raw salt, but not in refined salt.

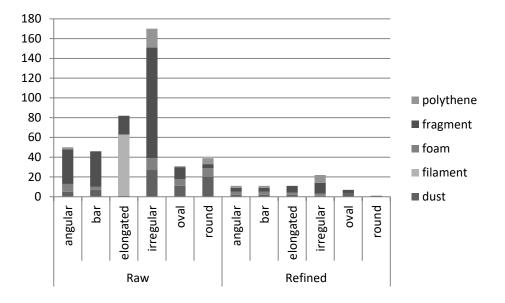


Figure 6: Shape-wise distribution of types of particles in raw and refined salt

3.2.3 Association of Size and Shape

Irregular particles are most dominant in both raw and refined salt, however the abundance varies significantly. Besides, the abundance varies with size, larger particles being less frequent. Especially in refined salt, larger particles are very few, suggesting the refining process can remove bigger particles more efficiently.

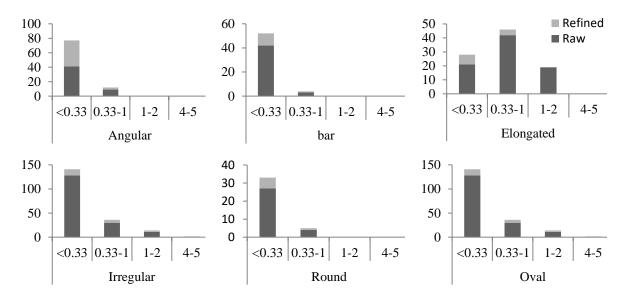


Figure 7: Distribution of Size of particles, according to shape

3.3 Distribution of Particles in Raw and Refined Salt (Sorting According to Color)

Like figure 6, figure 8 also displays the large variation of microplastics abundance in refined and raw salt. Black particles are dominant in both the types followed by brown and transparent in raw, and orange and brown in refined salt.

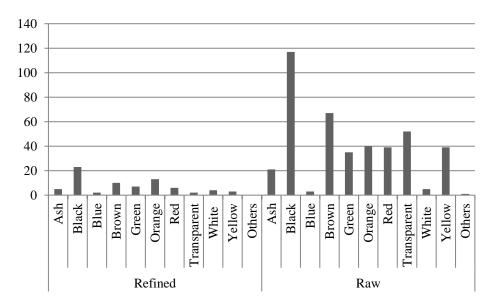


Figure 8: Distribution of Particles of various colors in Raw and Refined salt.

3.4 Discussion

Similar studies have been conducted around the world looking into salts from various places, and the findings altogether shows a wide deviation range. The presence of MPs in 15 brands of commercial salt in Chinese supermarkets was found in a study conducted, during October and November 2014. MPs were present in all of the samples analyzed. Higher concentrations of MPs were found (550-681 particles/kg) in sea salts than the other salt types(Yang et al., 2015rios). Another study analyzed eight commercial sea salt brands from India. They found MPs in all samples with concentrations ranging from 56 to 103 particles/kg (Seth & Shriwastav, 2018). The presence of MPs analyzed in 21 Spanish salt samples from different locations in the country. MPs were found in all of the analyzed samples. In sea salt samples, the concentrations ranged from 50 to 280 particles/kg (Iñiguez et al., 2017). The findings of the present study is almost in line with this one, reporting 283 particles/kg refined salt. However, in raw salt, the figure is as high as 2105/kg. Another research, studying 11 salt brands from Italy and Croatia, found that, all the sampled lots contained MPs. The concentrations of MPs ranged from 22 to 594 particles/kg in Italian brands, which again confirms the validity of our finding, and from 13,500 to 19,800 particles in per kg Croatian salt. According to this study, the concentrations recorded in their work could be related to an overestimation, given the proximity of sea salt production sites to highly populated urban settlements and the water received from polluted rivers (Renzi & Blašković, 2018).

In another study, investigation of the presence and concentration of MPs in 17 different brands of salt, from eight countries (Australia, France, Iran, Japan, Malaysia, New Zeeland, Portugal, and South Africa). MPs were present in 88% of the salts. In the contaminated samples, the concentrations of the particles ranged from zero in French sea salt to 10 particles/kg in Portuguese sea salt (Karami et al., 2017) The concentration of MPs assessed in sea, rock and lake salts from Turkey. MPs were present in 100% of the salts analyzed. MP concentration ranged from 16 to 84 particles/kg in sea salts (Gündoğdu, 2018).In another study, MPs also quantified in 12 salt brands from different world regions (Atlantic Ocean, Celtic Sea, Himalayan region, Mediterranean Sea, Mexico, North Sea, Pacific Ocean, Sicilian Sea, USA, and Utah Salt Lake). MPs were present in all of the salts analyzed. The concentrations of MPs found ranged from 47 to 806 particle/kg being fibers the most frequent plastic type in their samples and not particles (Kosuth, Mason, & Wattenberg, 2018).

4. CONCLUSION

This study was the first time in Bangladesh to look into microplastics contamination scenario in salts of Bangladesh. The abundance and distribution of microplastics in salt, along with their physical characterization was studied. It was found that the abundance varied largely between refined and unrefined salt. Considering physical characteristics, black colored particles was found to dominate in both the types, also irregular shaped ones if we consider the shape. As it was seen that the dominant size range in both the salt types were below 0.33mm, it should be focused in further studies, especially in similar consumable media, as plastics are assumed to have toxic effects conveying other toxic substances within our body(Rios, Moore, & Jones, 2007). Besides refining process should be developed, to eliminate the hazards of microplastics contamination through salt. Unlike the other possible food items, salt is universally consumed by all, and have no exception. So it should be considered seriously.

REFERENCES

- Arthur, C., Baker, J., & Bamford, H. (2008). *Proceedings of the international research workshop on the occurrence, effects, and fate of microplastic marine debris.* Silver Spring, MD 20910: NOAA.
- Carson, H. S., Colbert, S. L., Kaylor, M. J., & McDermid, K. J. (2011). Small plastic debris changes water movement and heat transfer through beach sediments. *Marine Pollution Bulletin*, 62(8), 1708– 1713. https://doi.org/10.1016/j.marpolbul.2011.05.032
- 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 Research*, 75, 63–82. https://doi.org/10.1016/j.watres.2015.02.012
- Gündoğdu, S. (2018). Contamination of table salts from Turkey with microplastics. *Food Additives & Contaminants: Part A*, 35(5), 1006–1014. https://doi.org/10.1080/19440049.2018.1447694
- Iñiguez, M. E., Conesa, J. A., & Fullana, A. (2017). Microplastics in Spanish Table Salt. Scientific Reports, 7(1). https://doi.org/10.1038/s41598-017-09128-x
- Karami, A., Golieskardi, A., Choo, C. K., Larat, V., Galloway, T. S., & Salamatinia, B. (2017). The presence of microplastics in commercial salts from different countries. *Scientific Reports*, 7(46173), 1–9.
- 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. *Environmental Science & Technology*, 52(21), 12819–12828. https://doi.org/10.1021/acs.est.8b04180
- Kosuth, M., Mason, S. A., & Wattenberg, E. V. (2018). Anthropogenic contamination of tap water, beer, and sea salt. *PLOS ONE*, *13*(4), e0194970. https://doi.org/10.1371/journal.pone.0194970
- Lam, C.-S., Ramanathan, S., Carbery, M., Gray, K., Vanka, K. S., Maurin, C., ... Palanisami, T. (2018). A Comprehensive Analysis of Plastics and Microplastic Legislation Worldwide. *Water, Air, & Soil Pollution, 229*(11). https://doi.org/10.1007/s11270-018-4002-z
- Liebmann, B., Bucsics, T., Königshofer, P., & Köppel, S. (2018, October 28). Assessment of *microplastic concentrations in human stool final results of a prospective study*. Presented at the Conference on Nano and microplastics in technical and freshwater systems, Monte Verità, Ascona, Switzerland.
- Renzi, M., & Blašković, A. (2018). Litter & microplastics features in table salts from marine origin: Italian versus Croatian brands. *Marine Pollution Bulletin*, (135), 62–68.
- Rieux, A. des, Ragnarsson, E. G. E., Gullberg, E., Pr'eat, V., Schneider, Y.-J., & Artursson, P. (2005). Transport of nanoparticles across an in vitro model of the human intestinal follicle associated epithelium. *European Journal of Pharmaceutical Sciences*, (25), 455–465.
- Rios, L. M., Moore, C., & Jones, P. R. (2007). Persistent organic pollutants carried by synthetic polymers in the ocean environment. *Marine Pollution Bulletin*, 54(8), 1230–1237. https://doi.org/10.1016/j.marpolbul.2007.03.022
- Seth, C. K., & Shriwastav1, A. (2018). Contamination of Indian sea salts with microplastics and a potential prevention strategy. *Environmental Science and Pollution Research*.
- Smith, M., Love, D. C., Rochman, C. M., & Neff, R. A. (2018). Microplastics in Seafood and the Implications for Human Health. *Current Environmental Health Reports*.

- Wright, S. L., Thompson, R. C., & Galloway, T. S. (2013). The physical impacts of microplastics on marine organisms: A review. *Environmental Pollution*, (178), 483–492.
- Yang, D., Shi, H., Li, L., Li, J., Jabeen, K., & Kolandhasamy, P. (2015). Microplastic Pollution in Table Salts from China. *Environmental Science & Technology*, 49(22), 13622–13627. https://doi.org/10.1021/acs.est.5b03163