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TABLE SALT AND COARSE SALT: A SOURCE OF CONTAMINATION BY MICROPLASTIC

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ABSTRACT

Pollution from plastic waste has attracted attention due to the long-term durability of plastic waste in the environment and its adverse effects on marine fauna as well as humans. Despite the occurrence of microplastics in sea salt consumed by humans in several countries, there is no available information on the presence of microplastic in commercial salts produced in Brazil. This study analyzed five different brands of table salts and three brands of coarse salts from two main salt producing regions in Brazil (sea salts from Areia Branca City of Rio Grande do Norte State and lake salts from the Araruama lagoon, in Cabo Frio City of Rio de Janeiro State January). Samples were analyzed under a light microscope to verify the grain size and under a stereo microscope to visualize the plastic fragments. In addition, ATR-FTIR analysis was used to identify the type of microplastic. The results indicated that Brazilian commercial salts contained polyethylene and polypropylene. As table salts are used as condiment and food preservative, people are continually exposed to plastics. Therefore, more effective rules for regulating the salt refinement process are needed.

KEY WORDS : Microplastic, Sea salt, Lake salt, Brazil, ATR-FTIR analysis

INTRODUCTION

Plastics have been used worldwide and benefited contemporary human society in several ways; however, their durability and wide use in various environments make them a persistent pollutant (Villarrubia-Gómez *et al*., 2018 and Hitchcock and Mitrovic, 2019).

In the environment, plastics deteriorate and fragment so that it is found in various dimensions and categorized as macroplastic (particles > 25 mm), mesoplastic (particles > 5 mm and < 25mm), microplastic (particles < 5mm), and nanoplastic (1 to 100 nm) (Zhao *et al*., 2018 and Li *et al.,* 2018).

Most plastics in the environment occur as microplastics (MP) that can be originally produced in a specific size (primary microplastic) or result from the fragmentation of larger pieces (Hitchcock and Mitrovic, 2019 and Zhao *et al*., 2018). The fragmentation of plastics can occur through photodegradation, physical or chemical degradation, or as a result of biological interactions (secondary microplastic) (Li *et al.*, 2018). MPs are discarded in the environment as debris from different sources related to human activities (Eerkes-Medrano *et al*., 2015) such as industrial residues and consumer products (e.g., cosmetics) (Moos *et al*., 2012).

Since MPs can act as a carrier of toxic substances to various internal organs, they can represent a human health problem (Batel *et al*., 2016). For Yang and coworkers (2015), plastic could absorb contaminants from the seawater and transfer them to sea products, and this way, salt may pose a risk to food safety. Despite the lack of consistent reports that MPs can directly affect human health, they have already been found in human stool (Liebmann *et al*., 2018), indicating that the ingested MPs transit through the gastrointestinal tract and thus carry toxic substances to the human body (see Peixoto *et al*., 2019).

Notwithstanding MPs have been reported in commercial salts from different countries, salt is undoubtedly the most ancient known ingredient, performing numerous functions, such as condiment and food preservative (Elias *et al*., 2019). Commercial salts from seawater and lake water are mainly produced through a crystallization process as a result of evaporation under the combined effects of heat from sunlight and wind (Karami *et al*., 2017).

Although the refinement and distribution of salt is carried out by different refineries in Brazil, large-scale commercial salt extraction occurs mainly in Areia Branca (Rio Grande do Norte State, RN) and Araruama Lagoon (Rio de Janeiro State, RJ), making salt the main export product in 2018 (MDIC, 2018). Furthermore, salt consumption in the country is exaggerated. Mill and coworkers (2019) estimate that the Brazilian population consumes approximately twice the amount of salt recommended by the World Health Organization (5g per day).

The aim of this study was to identify MPs in commercial salts, since there are no reports on MPs in commercial salts of both salt producing regions in Brazil (Areia Branca and Araruama).

Fig. 1. The two major producing regions of table salt: Areia Branca (RN) in red; Araruama Lagoon in Cabo Frio (RJ) in yellow, Google Maps. Bar = 100,000 Km.

MATERIALS AND METHODS

Five brands of table salt and three brands of coarse salt commercially available in supermarkets during June and July 2019 were purchased. From each brand of salt, three packs of 500g of sea salt from Areia Branca city [Rio Grande do Norte State (RN)] and lake salt from Araruama Lagoon in Cabo Frio city [Rio de Janeiro State (RJ)] were used (Fig. 1).

The size of the Table salt grains was analyzed under a light microscope (Leica microscope DM750) to verify if they were in accordance with the standards established by the Brazilian legislation (Law decree nº 75.697/75), that is, particles larger than 0.840 mm must be retained by industrial sieves such as large microplastics, mesoplastics and macroplastics (Li *et al*., 2016). The size of the coarse salt grains was analyzed under a stereomicroscope (Leica EZ4HD).

The sample preparation was conducted in triplicate and according to the protocol established by Yang and coworkers (2015). For this, for each 250g of each commercial salt was placed in a 2 L glass beaker and 100 mL of 30% H_2O_2 (hydrogen peroxide) was added to digest the organic matter. The glass beaker containing the solution was covered with a glass cover and the solution kept at 65 °C under 80 rpm stirring for 24 hours. After that, the solution was cooled to room temperature and kept without stirring for 48 hours. After that 800 mL of ultrapure water was added the solution kept stirring for 1h. Each solution was filtered using a cellulose nitrate membrane filter with a pore size of 3 µm and a 2 mm radius with a glass syringe. The cellulose nitrate membrane was placed in a glass Petri dish and dried at room temperature for further analysis. To verify if there was environmental contamination, one blank extraction group without salt was tested simultaneously to correct the potential contamination and the control group was made using pure NaCl (CAS 7647-14-5, Isofar Indústria e Comércio de Produtos Químicos).

After filtering procedures, the filters were observed under a Leica EZ4 HD stereo microscope coupled to the image capture LAS system to visualize the plastic fragments.

To analyze and identify the type of microplastic, Attenuated Total Reflectance with 105 Fourier Transformation Infrared Spectroscopy (ART/FTIR) was used and performed at the Brazilian Center for Physical Research using a Shimadzu IR Prestige-21, coupled with the accessory ATR-8200H. The spectrum range was 4000-700 cm^{-1} with 4.0 cm^{-1} of resolution and 128 scans. Before analyzing each sample, the ATR diamond crystal was cleaned with 70% acetone and a background scan was performed. The procedures for identification of microplastics followed the criteria cited in the literature for acceptable identification, i.e., the samples must have at least four absorption bands. Data were compared to the polymer spectrum library provided by Jung and coworkers (2018).

RESULTS AND DISCUSSION

During the all procedure process, microplastic contamination by air was avoided. Although sand grains were detected in all samples, multiples types of MP particles of diverse dimensions and colors were visualized, including small fragments and fibers (Fig. 2). After filtering, the filter of sea salt was slightly darker when compared to the filter of lake salt.

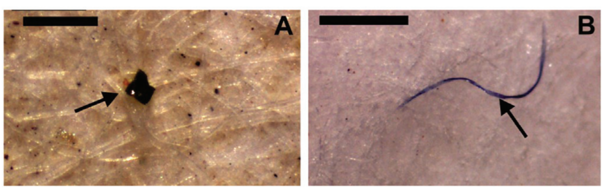

Fig. 2. Fragment and fiber (arrows) in table salts. A: sea salt (RN); B: lake salt (RJ). Bar = 50mm.

Notwithstanding the absence of law regulating the grain diameter of coarse salt, the maximum size of both sea and lake salt grains indicated that the dimensions were in accordance with the Brazilian law, which states that the grains should range from 0.105 to 0.840 mm (Table 1).

After ART/FTIR analysis, the control and blank samples did not show spectrum for microplastics, confirming that there was no environmental contamination, besides revealing the efficiency of the methodology used.

Table 1. Average salt grain size

Salt	Maximum size*
Sample A (from Cabo Frio, RJ)	0.547 mm
Sample B (from Areia Branca, RN)	0.710 mm
Sample C (from Areia Branca, RN)	0.801 mm
Sample D (from Areia Branca, RN)	0.770 mm
Sample E (from Areia Branca, RN)	0.698 mm
Sample F (from Areia Branca, RN)	3.4 mm
Sample G (from Areia Branca, RN)	2.0 mm
Sample H (from Areia Branca, RN)	2.4 mm

*Maximum size according to Brazilian law for table salt is 0.840 mm.

The samples A and B revealed the spectral appearance and wavenumber nearly identical for both polyethylene (PE) (2914 cm^{-1}, 2845 cm^{-1}, 1460 cm^{-1}, 1396 cm^{-1}, 719 cm^{-1}) and 135 polypropylene (PP) spectrum database (2954 cm^{-1}, 2914 cm^{-1}, 1454 cm^{-1}, 1396 cm^{-1}). 136 Furthermore, sample from the salts, C and H (table salts and coarse salts of both lake and sea) exhibited the spectrum and the wavenumber similar to PE (2914 cm^{-1}, 2845 cm^{-1}, 1475 cm^{-1}, 719 cm^{-1}). Based on the criteria proposed by Jung and coworkers (2018) for identification microplastic, it was not possible to guarantee the presence of microplastic in samples D since the transmittance signal was low and only three bands were identified. Nevertheless, Nor and Obbard (2014) confirm the occurrence of three bands to confirm the presence of MP.

The samples E (Table salt), F and G (both coarse salts) did not show any spectrum for MPs (Fig. 3).

As sodium is essential for the maintenance of homeostasis of the human body, it is mainly consumed from common salt. Notwithstanding MPs reveal the ability to absorb organic pollutants (Engler, 2012), salt containing MPs can affect human health. Microplastic (MP) in commercial salt from Brazil was visualized through microscopic observation. Moreover, the identification of the MP type was confirmed with ATR- FTIR, which is the highly recommended method for its chemical characterization (Shim *et al*., 2017).

MP fragments were found in different brands of commercial salt in different countries (Yang *et al*., 2015; Karami *et al*., 2017; Kim *et al*., 2018; Gündogdu, 2018; Seth and Shriwastav, 2018 and Peixoto *et al*., 2019). However, they did not comment if the size of salt grain is in accordance to the legislation of each salt producing country, especially regarding the size of the sieves, which are fundamental for the retention of MPs from environmental. Among all polymer types, PE and PP are the most abundant in aquatic environments, mainly in the sea surface (Erni-Cassola *et al*., 2019).

Human activities in the marine environment contribute 20% of total plastic debris (Andrady, 2011), while MP from terrestrial sources contribute to the remaining 80% since they are released into the natural water system, the most of which are transported into the oceans by rivers (Browne *et al*., 2010 and Free *et al*., 2014).

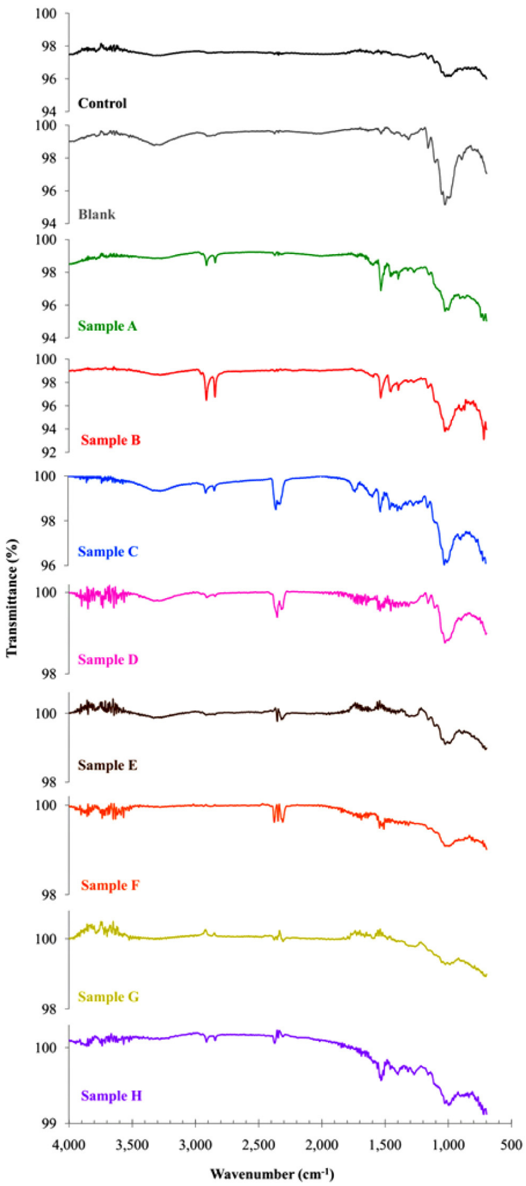

Fig. 3. MP spectra of five table salt brands and one coarse salt brand. ATR-FTIR spectrum was assigned as a mixture of polyethylene and polypropylene (sample A and B) and polyethylene (samples C and H). Sample D revealed a spectrum with only three bands similar to a mixture of PE and PP, making it impossible to confirm the presence of MP in the sample. Samples E, F, and G did not show the presence of MP. Control = NaCl. Blank = Ultrapure water without salt. Sample A = Lake table salt. Sample B-E = Sea table salt. Sample F = Lake coarse salt. Sample G-H= Sea coarse salt.

Morphometric analysis revealed that the size of grains of both sea and lake table salt is in accordance with Brazilian legislation which determines the maximum retention of 5% in 840 µm mesh and 90% retention in 105 µm mesh. Consequently, the sieve mesh allowed the passage of small grains of salts and smaller plastic fragments in Brazilian salts.

PP and PE were detected in most table salt samples, and PE was identified in a coarse salt sample. Yang and coworkers (2015) suggested that there are several routes for table salt contamination by MPs, such as atmospheric transport. As a result, the presence of MPs may be due to contamination during different stages of salt production (drying in the wind and collecting salt), in addition to packaging in plastic bags made of PE.

CONCLUSION

Despite the lack of reports on the presence of microplastics in the marine environment of the Brazilian coast or in water of Lagoon, it is not possible to confirm that the microplastics came from sea and lake water or if they were introduced into the salt during the production process. Thus, the risk to consumer health is high since commercial salt is used in food preparation. Without any intervention by institutions around the world, it is possible for microplastics to increase considerably in the aquatic environment, causing effects on human health and aquatic organisms.

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Conflict of Interest

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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