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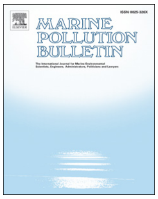

Litter & microplastics features in table salts from marine origin: Italian versus Croatian brands

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A B S T R A C T

This study estimates litter content, including microplastics, mesoplastic, and macroplastic in marine table salts coming from Italy and Croatia. Both high (HC) and low (LC) costs commercial brands easily found at the supermarket were analysed. Any macroplastic or mesoplastic were recovered while microplastics and other litter impurities significantly affect table salts of all tested brands. Average microplastic values ranged within 1.57 (HC) – 8.23 (LC) (Italy) and 27.13 (HC) – 31.68 (LC) items/g (Croatia). Microplastics sizes (min-max) ranged within 4–2100 μm (Italy) and 15–4628 μm (Croatia). In samples from both Nations, a significant general positive correlation between the average number of items/g recorded and the total amount of general impurities was recorded. Concerning microplastic shapes, in Italy, fragments dominated even if fibres, granules, films, and foams are frequently recorded. On the contrary, clear PP fibres dominated in Croatian brands even if also other shape classes were recorded.

1. Introduction

Plastic debris is currently a huge problem of global concern (Hollman et al., 2013) that affects the environment entirely. From sediments (Blašković et al., 2017; Cannas et al., 2017; Renzi et al., 2018a, 2018b) and water (Eriksen et al., 2013; Zettler et al., 2013) to the animals (Fossi et al., 2016) and humans (EFSA, 2016). Is widely established the transmission of the microplastic through the food chain (Avio et al., 2015). Now more than ever we are aware of the possible action of the plastic litter on human's health. Litter transfer from marine ecosystems to humans represent an important task that should be better described and clarified by the literature to achieve Horizon 2020 targets concerning the Marine Strategy Framework Directive principal purposes (2008/56/EC).

Humans are exposed by diet to plastic litter intake (EFSA, 2016) and seafood represent the most explored source of pollution coming from marine trophic web (Avio et al., 2015).

In coastal area, salt flats represent important coastal areas of great ecological and economical interest at the interface between land and sea and exposed, as coastal transitional ecosystems (Renzi et al., 2012; Renzi et al., 2013), to different kinds of human pressure. That activity could produce significant effects on plastic litter accumulation both in local trophic webs and commercial products. In spite of that, recent literature focuses their research on plastic pollution risks related to natural seafood and maricultured products (Avio et al., 2015; Dehaut et al., 2016; Pellini et al., 2018) and shows that seafood, as well as pelagic and benthonic fish species is affected by microplastics and, in particular, by fibres (Neves et al., 2015). Mussels and other filter feeding bivalves show a large number of individuals affected by the plastic litter (Karlsson et al., 2017; Van Cauwenberghe et al., 2015; Renzi et al., 2018a, 2018b). There are different types of table salts related to the salt origin as well as sea salt, lake salt, rock salt, river salt, well salt. Production processes are different for different salt types; sea and lake salts are obtained by water evaporation, rocky salt is obtained by mining, while river and well salt is obtained from wells in non-coastal zones (Iñiguez et al., 2017).

Very few studies are performed on table salt from marine origin that represent another important commercial product coming from the sea and potentially affected by marine pollution by plastic litter. Two studies performed recently evaluated levels of plastic litter in table sea salts (Iñiguez et al., 2017; Karami et al., 2017). Karami et al. (2017) performed plastic litter determinations on 17 salt brands from 8 different countries and evidenced that microplastics (171–515 μm) were present in almost all brands at concentrations included between 1 and 10 MPs/kg of salt. Related to shape, authors evidenced that fragments were dominant (63.8%) followed by fibres (25.6%) and films (10.6%). A research performed in Spain by Iñiguez et al. (2017) on 21 different samples of commercial table salts, evidenced ranges within 50–280 MPs/kg salt with any differences between samples due to the origin (sea salts and well salts) and treatments (before and after packaging).

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Nevertheless, researches on this field are far to be exhaustive.

The aim of this study was to evaluate litter content, including microplastics, in eleven different commercial brands of iodate fine table salts produced in by six different Italian brands and five Croatian brands of sea salt produced locally. Analysed salt samples were easily found at the supermarket and sold in carton (Italian; Croatian) and plastic (Croatian) packs. Total amounts, principal features of recovered litter (including microplastic, mesoplastic, macroplastic), and possible differences among high cost (HC) and low cost (LC) brands were checked.

2. Materials and methods

Eleven different brands of table salt of marine origin were selected in this study. All selected brands were fine sea iodate salt. Commercial name of products cannot be made public for privacy reasons nevertheless commonly used brands were selected to improve representativeness. We sampled six Italian and five Croatian brands available at the supermarkets coming from different production plants. Geographical locations of the production site of sea salt were not always indicated on packaging but sampled sea salts were always declared to be national produced. Italian brands were all sold in carton packs; only one brand from Croatia was sold in carton while the other four brands were sold in clear plastic envelopes. Selection was performed on the basis of the final cost: three high cost (0.45–0.50 Euro, named HC1, HC2, HC3) and three low cost (0.35–0.37 Euro, named LC1, LC2, LC3) brands were selected in Italy. Also Croatian brands were grouped concerning costs (high cost HC4, HC5, HC6; low cost LC4, LC5). Salts were sampled statistically to ensure representativeness.

For each experimental replicates, 360 g of sea salt was completely dissolved in 1 L of deionized water mixed for 20 min in a glass backer and filtered using a filtrating vacuum pump apparatus on a filter fibre (0.45 μm) disks successively dried in oven at 40 °C. Total suspended solids (TSS, method APAT CNR IRSA 2090) were determined on a solution of 200 g/L of salt samples in statistic replicated to evaluate salt contamination by total solid materials. In this case, fibre filters were weighted before and after filtration of the salt solution and successively dried in oven at 60 °C till constant weight. Observations were performed by stereomicroscopy (Nikon, SMZ-800 N). A rigid protocol of identification was applied to minimize operator classification mistakes as reported: (1) no structures of organic origin should be visible; (2) fibres should be equally thick and have a three-dimensional bending to exclude a biological origin; (3) particles should be clear and homogeneously coloured; (4) transparent or whitish particles must be examined under high magnification to exclude a biological origin (Löder and Gerdts, 2015).

Classifications were performed according to type, shape, size, and colour following criteria reported by the literature (Hidalgo-Ruz et al., 2012; JRC EU, 2013; Galgani et al., 2013; Alomar et al., 2016). Multicolour and Violet colour classes were added to the classification reported by classification criteria adopted by the literature (Galgani et al., 2013; Fastelli et al., 2016). Coloured images were collected by high-resolution digital camera linked to the microscope (Nikon, DS-Fi2). Nikon's software for the imaging analysis was applied to the litter dimensional measurements (Nikon ACT-1). 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). Confirmations were performed by μFT-IR (Nicolet iN10 MX, Thermo Scientific) technique.

3. Results

Plastic litter recovered during this study are represented in Fig. 1. In particular, black foam (Fig. 1a); tubular unrecognized black (Fig. 1b); multicolour fragment (Fig. 1c); violet fibre (Fig. 1d); various granules (Fig. 1e, f); clear fibres (Fig. 1g); plastic knot (Fig. 1h) of determined microplastics are reported.

General features of tested samples are reported in Table 1. In particular, in Table 1, two price-sizes are reported grouping samples in two classes: High Cost (HC) and Low Cost (LC) brands. HC1 and HC3 are produced in the same geographical area (North-Adriatic Sea) but are sell as different brands. On the contrary, Italian LC table salt is produced from different regions (Southern Tyrrhenian, Southern-Adriatic, and Northern-Adriatic seas). As regards as Croatian marine analysed salts, originated from two different areas. Any macro- or meso- plastics are recovered. All recovered plastic litter are microplastics ranging within 4–2100 μm (Italian salts) and 15–4628 μm (Croatian salts). Statistics performed evidenced a significant difference ($p < 0.01$) among HC and LC brands concerning n. items/g. Total Suspended Solids (TSS, μg/g of salt) is considered as indicator of the total amount of impurities. A comparison among average levels of TSS and microplastic average items/g of salt measured is reported in Fig. 2. Data evidence a general positive correlation between the average amount of MPs items and TSS recorded (Test T; $p < 0.001$) in both Nations.

Average (SD) shape frequencies of microplastics recorded are reported in Table 2 as percentage of each shape versus the total amount recorded. Concerning Italian salts, fragments are the dominant percentage of microplastic recovered followed by fibre, films, foam, and granules. The percentage of unrecognized shape class is closed to 2% in almost all LC brands. Percentages are quite similar in tested brands evidencing a common structure of the shape distribution even if considering number of items/g LC brands evidenced numbers generally higher than HC ones. On the contrary, Croatian salts show a percentage of fibres exceeding 80% in almost all analysed samples. Films, fragments and granules are also recovered. In Fig. 3, average colour fingerprint of tested table salts is reported for both LC and HC brands and expressed as average % (SD). As observed by the figure, colour fingerprints are similar on a National basis even if percentages of items/g of salt are generally higher in LC compared to HC brands. In Italian brands, black is the dominant colour followed by grey and blue. Orange, brown, green, pink, yellow and violet are represented in each brands. High frequencies of multicolour microplastics (Fig. 1) are recorded in both price-sizes even if higher levels are recorded in LC ones. As regard as Croatian salts, the clear colour is the principal observed in recovered microplastics followed by blue, black, white, and yellow colours. Concerning microplastic size, a wide range of dimension is recorded in both Croatian and Italian salts (Table 1) even if the size class < 100 μm of maximum diameter is the most represented for both HC and LC prize-size of table salt tested.

4. Discussion

Sea salt production is performed in salt flats by pumping marine water into evaporation ponds where wind and sun increase evaporation rates and determine the precipitation of salt crystal on the bottom of ponds. Crystallized salt is cut and collected and subjected to different physical processes before packaging (Iñiguez et al., 2017). On the basis of the results obtained by this study, all Italian and Croatian sea salts tested resulted polluted by microplastic and other impurities. Total average measured by this study in tested samples of different brands ranged within 1.6–8.2 (Italian) and 13.5–19.8 (Croatian) items per gram of salt. These levels are significantly higher values compared to data reported by literature (Karami et al., 2017; Iñiguez et al., 2017). Observed higher levels should probably be related to different factors. First of all, possible overestimations of plastic numbers could occur for the particles with a diameter minor than 500 μm during the first step of visual identification of plastic items. Literature considers visual determinations as a valid method for particle dimension larger than 500 μm (Hidalgo-Ruz et al., 2012) while reports a percentage of classification mistakes by visual identification ranging within 20% (Eriksen et al., 2013) - 70% (Hidalgo-Ruz et al., 2012) that increases with the