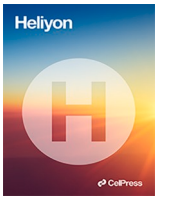
Heliyon 10 (2024) e31609

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon



Research article

An exploratory study of fibre microplastics pollution in different process stages of salt production by solar evaporation in Spain

M.M. Cledera-Castro [a,b,c,*], K. Hueso-Kortekaas [a,b,d], C. Sanchez-Mata [a], C. Morales-Polo [a,b,c], J. Calzada-Funes [a,b], N. Delgado-Mellado [a,b], R. Caro-Carretero [a,e]

[a] ICAI School of Engineering. Universidad Pontificia Comillas, Alberto, Aguilera, 25, 28015 Madrid, Spain
[b] Group on Emerging Pollutants and Resource Recovery. Universidad Pontificia Comillas, Alberto Aguilera, 25, 28015, Madrid, Spain
[c] Institute for Research in Technology, ICAI, Universidad Pontificia Comillas, Alberto Aguilera, 25, 28015, Madrid, Spain
[d] IPAISAL Network, Apartado de Correos 50, 28450, Collado Mediano, Madrid, Spain
[e] Cátedra de Catástrofes Fundación AON España, Calle Alberto Aguilera, 23, 28015, Madrid, Spain

ARTICLE INFO

Keywords:
Microplastics
Food-grade salt
Brine
Quality
salinas

ABSTRACT

Microplastics are a pollutant of growing concern. Several studies have found microplastics in table salt worldwide in the last decade, although most have focused on already prepackaged salt. To the best of our knowledge, there is no previous research analysing the entire salt production process. In this study focused on solar evaporation salinas, brine and salt samples were obtained from each stage of production, starting with the entrance of seawater/brine until the final stage of ready-to-sell salt, in six sites in Spain. We extracted microplastics from each sample after 30 % H_2O_2 digestion and filtration through cellulose nitrate 5 μm pore filters. Microplastic fibres were optically analysed with an Olympus DSX1000. Results indicate that microplastics are present both in seawater and air, with atmospheric fallout identified as the primary source. Microplastic concentrations from the entrance to the salina till the inlet to the crystallizers ranges from 256 to 1500 items per liter and from 79 to 193 microplastics per kg for packaged salt were estimated. Artisanal salina F shows the highest content in microplastics. This study hopes to give insight into the origin and causes of microplastic pollution in solar evaporation salinas and contribute to preventing this form of pollution in food-grade salt.

1. Introduction

Plastic waste is one of the major pollutants of the natural environment. It is ubiquitous, abundant, and fast-growing. Depending on the size, a large part is present in the form of microplastics (between 1 μm and 5 mm in size), and nanoplastics, smaller than 1 μm [1]. With regard to their shape, microplastics can be irregular fragments, pellets, and threadlike fibres. This paper will focus on fibre-like microplastics.

The main sources of these pollutants are [2,3]: primary microplastics, those intentionally manufactured in small sizes that have not been through a significant transformation yet, and secondary microplastics, those that result from a degradation process of larger items and with a higher presence in the environment. Important sources of primary microplastics are wastewater [4], air-blasting cleaning media or any industrial process which use virgin microplastics as a raw or auxiliary material [5]. Secondary plastics can be formed by

* Corresponding author. ICAI School of Engineering. Universidad Pontificia Comillas, Alberto Aguilera, 25, 28015, Madrid, Spain.
E-mail address: mcledera@comillas.edu (M.M. Cledera-Castro).

https://doi.org/10.1016/j.heliyon.2024.e31609
Received 24 October 2023; Received in revised form 13 May 2024; Accepted 20 May 2024
Available online 21 May 2024
2405-8440/© 2024 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC license (http://creativecommons.org/licenses/by-nc/4.0/).

degradation of larger pieces of debris due to the exposure to sunlight, water, and/or sea salt [6]. They can also be found in the wastewater or air, resulting from the abrasion of tires on the road and fibres from clothes, sacks, and similar textiles [7].

Microplastics have been detected in many different locations, even in uninhabited areas [8,9] or human stools and placentas [10,11]. Water is the medium in which microplastics have most often been detected, including inland waters and groundwater, in ever-increasing abundance [3,12,13].

One of the main concerns of microplastics in water is how they will affect living organisms, because of the very slowly degradation, the potentially toxic chemical additives used in their manufacturing [3], and their role as vectors for pathogens and invasive species [12,14]. As plastic production and consumption are expected to grow in the years to come, the number of studies focused on marine and freshwater cleanup technologies is increasing, and costs and effectiveness of these technologies are being studied in greater detail [15].

Although the study of microplastics in marine and other aquatic environments has been ongoing since the late 1980s [16], the research in microplastic pollution in saline habitats actually started in the 2010s. Microplastics have been found in most samples, e.g., in 98.5 % of them in a review by Peixoto et al. [17], and in more than 100 commercial brands, according to Zhang et al. [18]. Most of the studies focus on commercial table salts that are already packaged, coming from a wide range of sources and locations [17,19–30], rather than in salinas themselves or the part of the salt-making process where microplastics enter the system. Salinas (i.e. salt making sites) [31] are wetlands in which salt is being obtained by solar evaporation of brine.

Fig. S1 in Supplementary Material shows a summary of natural sources of salt and salt-making processes and the most vulnerable steps to plastic pollution in red [32]. Rock salt is also included for the record. It can follow a dry process by using classical mining techniques, or can also be dissolved and then evaporated in a wet process Salt from lakes, wells, and seas is already dissolved in water, forming a concentrated brine, and needs to be recrystallized, usually by solar evaporation [33,34]. Other methods exist, such as seething (a form of forced evaporation), graduation (to concentrate brine but not crystallize the salt), and some alternative marginal methods not considered here [35].

Microplastic pollution in salinas may be due to different pathways. Lake, well, and sea soils and sediments are essential deposits of plastic pollution which can affect groundwater [36]. Solar evaporation processes, whether coastal or inland, are exposed to the wind and, therefore, airborne plastic pollution. Microplastics in commercial sea salts can be present in water during and after the crystallization process [37]. Storage and handling of salt at various stages of the salt making process can also pose a risk of plastic exposure, such as vehicles used during harvest which may cause wear and tear of tires, as well as the clothing and footwear worn by workers and visitors. How it is washed in the mechanical process exposes the salt to microplastics, and transportation usually involves conveyor belts and other mechanical devices made of rubber of with plastic coatings that the salt may come into contact with. Packaging, as the final step of the salt production, often requires the use of plastic parts and the longer the salt is stored, the higher the risk of microplastic contamination. Yang et al. [38] observed that rock/well salts were contaminated with microplastics during collecting, wind drying, and packaging production stages. Refined salt is, in theory, more prone to being polluted than artisanal salt due to the contact with mechanical processes. *Fleur de sel* –an unrefined type of salt that is typically produced on the surface of the crystallizers– is, on the other hand, more exposed to airborne plastic pollution, as well as capturing floating particles [27,39]. Also, due to its flat shape (as with other salt types such as scales), it is expected to trap more particles.

In this exploratory study, the presence of microplastic fibres in six solar evaporation salinas in Spain are analysed at different stages of the salt-making process. They exhibit different features, including their location (whether inland or coastal salinas), size of production (ranging from industrial to artisanal), and site abandonment due to rural exodus, a societal trend that began in the early 20th century in Spain [40]. This approach will provide insight into the pollutant's origin and the pathway with which it enters the process. This, in turn, will help prevent and mitigate microplastic contamination, an issue of growing concern to salt-making companies, public health officials, and the environment in general.

2. Material and methods

2.1. Sampling in the field

Both brine and salt samples were obtained from the different production stages of salt making at six solar evaporation salinas in Spain. Of these, two sites were located on the Mediterranean coast, one on the Atlantic coast and three, inland. Of the three inland locations, one had been abandoned in the 1940s. The six sample sets also represent different sizes and scales of operation, from mid-

Table 1
Solar evaporation salinas sampled.

Site [a]	Location	Scale	Size[b]	Natural protected area
A	Mediterranean	Industrial	Large	Yes
B	Mediterranean	Industrial	Medium	Yes
E	Inland	Abandoned	Small	No
F	Inland	Artisanal	Small	Yes
G	Inland	Industrial	Medium	No
H	Atlantic	Artisanal	Large	Yes

[a] Sample sets C and D are not considered here.
[b] Size in relative terms with respect to the range of sizes found in the scale category of solar evaporation salinas in Spain.

and large-size industrial to small artisanal sites. By sampling this choice of locations, all types of solar evaporation salinas in Spain are represented [41]. For confidentiality reasons, a map with the location of the sites cannot be provided and the sites were labelled as indicated in Table 1.

These samples cover the different stages of the salt-making process, from the collection of seawater (in coastal sites) or brine (in inland sites or wells), to the market-ready salt. In a solar evaporation salina, salt is obtained by letting the seawater or brine concentrate in a succession of ponds of escalating salt concentration. The seawater or brine is introduced into the facilities either by pumping or by gravity flow. The first pond series are the evaporators, in which water has a low concentration of salt (usually around 5–10 %) and sedimentation of small particles occurs. From there, the water is led to the concentrators, where the concentration is increased to 20–25 % and other salts, such as carbonates or sulphates, precipitate and the brine is thus purified. In the third stage, the brine is led into the crystallizers, where the sodium chloride precipitates and can be harvested. Often, inland salinas, which are fed from brine wells, only have the last two stages, as the brine comes with a higher concentration from the ground. In some types of salina, the remaining liquid after crystallization, the so-called mother liquor, is discharged into the environment, while in other sites this highly concentrated brine is used for salt production and/or washing [35,40]. Once harvested, the salt is piled up in heaps and let to drip and dry, typically in an outdoor setting, before being transported to storage and packaging. In the case of industrial saltworks, this process is mechanized and extra stages are added such as washing and grinding (pers. obs.).

All the stages are listed below (between brackets, the type of samples obtained) and shown in Fig. S2 (Supplementary Material). Stages 1–4 correspond to liquid samples and stages 5–9 to solid samples.

1. Entrance to the salina (seawater)/spring (brine).
2. Entrance to evaporators and/or concentrators (brine).
3. Inlet to crystallizers (brine).
4. Outflow from crystallizers (mother liquor).
5. Crystallized salt in crystallizers (coarse salt and *fleur de sel*).
6. Washed salt, when applicable (coarse salt).

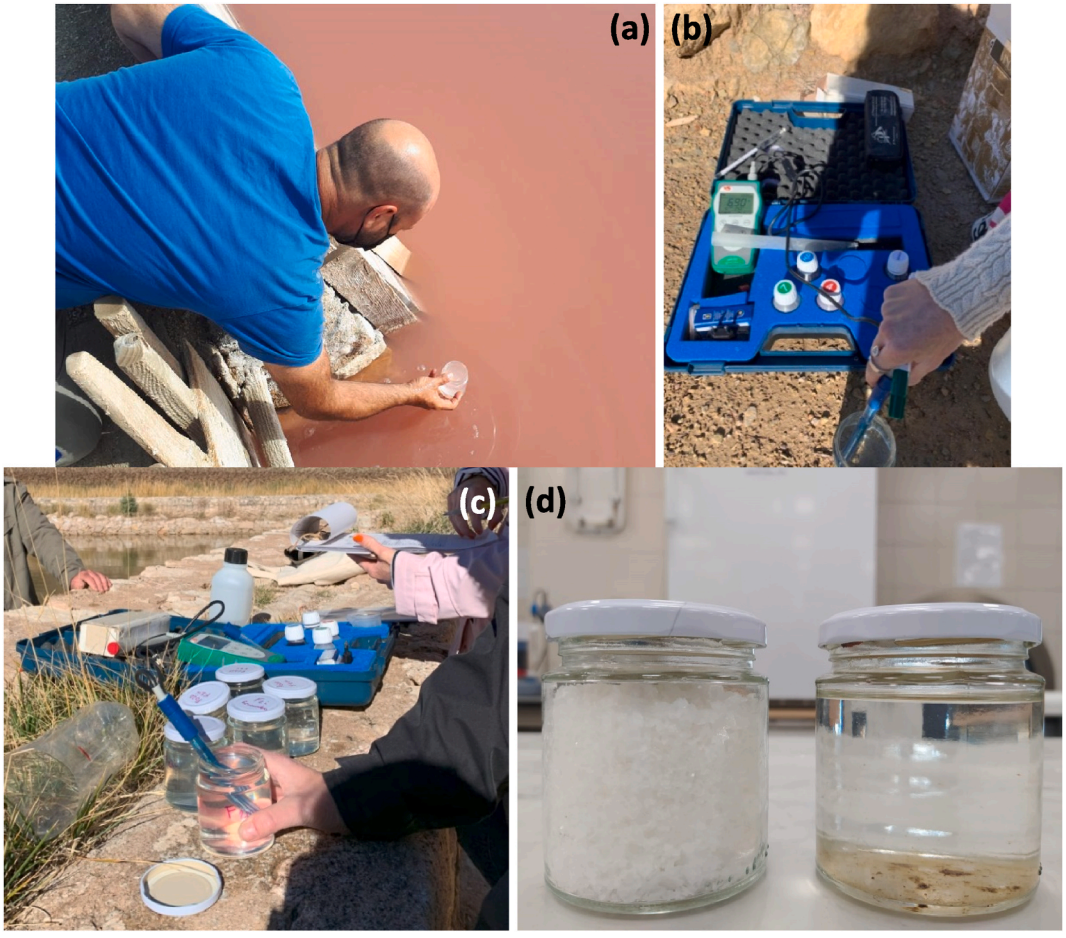


Fig. 1. Panel a, sampling in the field; Panel b and c, pH and salinity measurement; Panel d, samples.

7. Stacked salt for at least 6 months, when applicable (coarse salt).
8. Salt packaged during harvest season (coarse salt and *fleur de sel*).
9. Salt packaged for more than 6 months (coarse salt and *fleur de sel*).

The choice of samples indicated above should represent the critical points in the salt-making process, i.e., where brine or salt is transferred from one stage to another. Not all sample collection points could be considered due to the nature of salinas themselves, their production process (the scale and the types of salt produced) and the salina conservation state. Wet samples were obtained at 1 m from the shore, avoiding areas of natural accumulation of debris due to water and brine currents and/or wind. Sampling was done using a hard PET 500 mL bucket, filling three 250 mL glass jars with metal lids, to avoid cross contamination. Solid samples (coarse salt and *fleur de sel*) from the tops or sides of mounds were scooped with a metal spade or by hand, previously having been rinsed with freshwater. Three replicas were obtained from each stage and recorded the specific data on a form prepared for this purpose. This form (Fig. S3 of Supplementary Material) included: location (name of the salina, municipality, coordinates); date and time; person taking the sample; and weather conditions of the previous 3 days, with maximum and minimum temperatures recorded, precipitation, relative humidity of the air and wind, and any field observations deemed relevant to assist in interpreting any potential anomalies. Each jar was labelled with the corresponding code, as described above. The salinity and pH were measured for all samples. After the samples were taken, they were stored at room temperature without direct sunlight. Fig. 1(a–d) shows sampling in the field, the measurement of salinity and pH in field, and one sample in the glass jar.

In this study, the analysis of microplastic content involves more than just counting their numerical presence in each stage. The comparison also considers the proportion of micropastics within each stage relative to the total count across all stages. For this purpose, stages were categorized into two fundamental groups.

1. *Production group*: Stages 1–5. The comparison of this group was mainly carried out according to proportions.
2. *Consumption group*: Stages 6–9. As each salina provides different types of salt, or none (if abandoned), the comparison of the six salinas was carried out based on the number of microplastics for this group.

2.2. Sampling processing in the lab

Prior to starting the laboratory procedures, all surfaces were cleaned alcohol-moistened paper. Cotton laboratory coats and nitrile gloves were worn to prevent contamination. All glassware used in the laboratory was rinsed with pre-filtered water.

2.2.1. Materials and sample preparation

Each sample collected from field, consisting of 250 mL, whether brine or salt, was placed in 1L glass bottle. To place the coarse salt or *fleur de sel* in the bottle, a funnel and paper filter were necessary. All bottles were labelled with the names of the respective samples. 10 mL of 30 % hydrogen peroxide (H_2O_2) from Merck, Darmstadt, Germany, was added to each bottle and they were sealed with a lid. One bottle was used as a blank for the set of experiments, with just hydrogen peroxide, to verify that there was no microplastic contamination during the laboratory procedures. All bottles were adequately sealed and placed into an oven at 65 °C for 24 h to degrade the organic matter. Following this, 800 mL of ultrapure water were added to all the bottles using a scale cylinder. All samples were homogenized to dissolve the remaining salt and filtered using a Kitasato filter set. A filter of cellulose nitrate with 5 μm pore size and 47 mm diameter was introduced using tweezers, with precautions taken not to contaminate it during handling. A vacuum valve was used to filter all the samples, for excessively dirty samples, more than one filter per sample were used. Once the filtration was finished, the filter was carefully placed into a labelled Petri dish and stored in darkness.

2.2.2. Measurement and classification of microplastics

A DSX1000 microscope (OLYMPUS®, Tokyo, Japan) was used for the measurement and classification of microplastics by size and

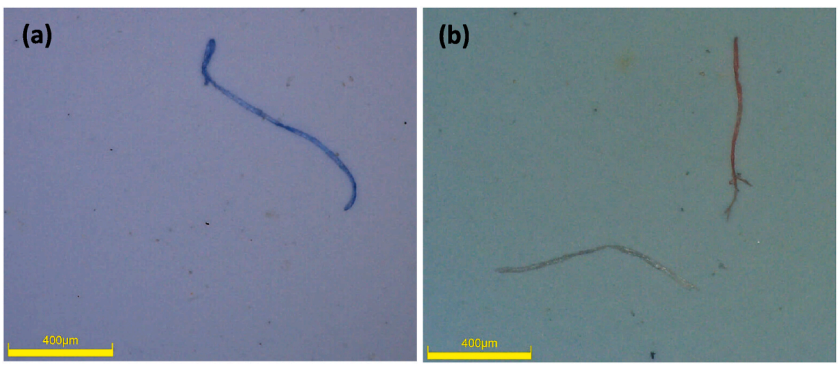


Fig. 2. Microphotographs obtained by the DSX1000 microscope OLYMPUS®, stage 2 of salina A; Panel a, blue fibre; Panel b, red and transparent fibres. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

colour. Two sample images of a fibre obtained by the microscope are shown in Fig. 2 (a,b). Microphotographs from the previously mentioned samples were analysed using the IC Measure® software. Microplastic fibres were categorized based on colour, including: Black, Blue, Brown, Green, Red, Transparent, White, and Yellow. Size ranges were specified as follows: 5–6.3 μm, 6.3–20 μm, 20–63 μm, 63–200 μm, 200–630 μm, and 630 μm −5 mm.

2.2.3. Data analysis

Tests were repeated in three runs, in each stage. ANOVA statistical analysis, with a 95 % confidence level was conducted to assess the mean differences between stages (both colour and size of microplastics) within the same salina, and the mean differences between salinas for each stage. ANOVA was then able to determine whether the observed differences were statistically significant. Furthermore, DMS and Tukey post-hoc contrasts can be used to help to identify significant differences between pairs of groups. If the p-value resulting from the analysis is below a pre-defined significance level (commonly 0.05), it suggests that at least one group differs significantly from the others. ANOVA analysis results are shown in Tables S1–S10in the Supplementary Material.

3. Results

3.1. Comparison by proportions

A comparative analysis has been conducted at different sites where the number stages differed, depending on various factors, such as the state of the facilities, the scale and size of operation, and the final product types.

Table 2 shows the number of microplastics measured at each stage for each salina, per liter for liquid samples (stages 1–4) and per kg for solid samples (5–9). As it was not possible to take a sample from every stage for each salina, the total number of microplastics for each salina cannot be compared. The comparison of the number of microplastics in stages 1–3 shows that the inland artisanal salina F exhibits the highest content. Conversely, the comparison of stages 8 and 9 indicates that microplastic content in salt packaged is similar in industrial and artisanal salinas. Dwiyitno et al. [42] reported higher microplastic contamination in traditionally produced sea salt compared to geomembrane and tunnel pond methods, in three different solar evaporation methods in Java, Indonesia. On the contrary, results published by Thiele et al. [43] concluded that traditionally harvested sea salts contain significantly fewer microplastics than industrially harvested ones. Regarding the rest of salinas, they present a similar content in microplastics, except for salina B which shows the lowest content in these contaminants. Microplastic fibre concentration across the six salinas ranges from 79 to 193 items per kg in stages 8 and 9. This concentration is lower than the mean microplastic concentration in sea salts studied by Thiele et al. [43], between 74-1155 microplastics per kg. They also reviewed 35 previous studies focused on food-grade salts. Three of them reported that microplastic content in Spanish salts ranges from 50 to 380 items per kg, indicating a consistent pattern with our results.

3.1.1. Production group

Stages 1, 2, and 3 are the only common stages to all the salinas studied and, therefore, the comparative analysis was carried out from them. Fig. 3 illustrates the microplastic proportion at each stage relative to the stages 1–3 for each salt. As can be seen, the abandoned salina E stands out from the others, since most microplastics appear at the first stage. A comparison of active salinas shows a different trend between inland salinas, F and G, and coastal salinas, A, B, and H. Regarding only coastal active ones, there is a decreasing trend in terms of the number of microplastics per stage. The higher concentration of microplastic in coastal salinas may be attributed to the higher concentration of these contaminants in sea water compared to inland water, and its deposition in evaporators and concentrators could decrease the number of microplastics observed in brine.

Fig. 4 shows the size proportion of microplastics for each stage in salina A (Panel a), salina B (Panel b), and salina H (Panel c). As can be seen, the 200–630 μm and 630–5000 μm ranges dominate in all salinas. Although it should be noted that in salina H the proportions for the smallest sizes are minimal (ranges 5–6.3 μm and 6.3–20 μm), these sizes in salinas A and B have representations of

Table 2
Number of microplastics (per liter stages 1–4 or per kg stages 5–9) measured at each stage for each salina.

Stages	Site					
	A	B	E	F	G	H
1	268	119	443	284	189	249
2	192	72	17	644	280	219
3	156	65	59	572	127	100
4	120	39	-[a]	-[a]	-[a]	-[a]
5	107	57	-[a]	168	-[a]	-[a]
6	44	85	-[a]	-[a]	101	-[a]
7	227	-[a]	-[a]	169	-[a]	-[a]
8	140	79	-[a]	-[a]	96	-[a]
9	193	-[a]	-[a]	173	-[a]	173
Microplastics in stages 1–3	**616**	**256**	**519**	**1500**	**596**	**568**
Total microplastics	1447	516	519	2010	793	741

[a] No sample was taken in this stage.

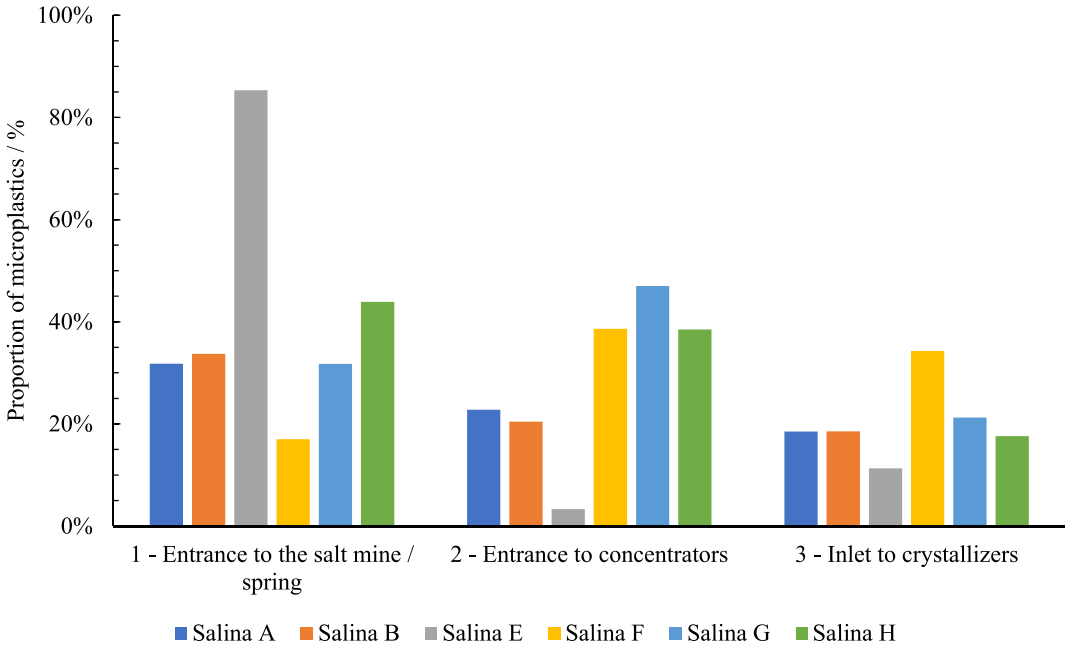


Fig. 3. Proportion of microplastics at each stage for the production group, stages 1, 2, and 3 (all salinas).

between 10 and 15 %.

Fig. 5 illustrates the distribution of colours in salinas A (Panel a), B (Panel b), and H (Panel c). As it can be observed, transparent microplastic fibres are the most prevalent, followed by blue, red, and black. This colour distribution aligns with patterns observed in studies on microplastics in food-grade sea salts [38,43].

3.1.2. Consumption group

The study of the salt consumption group was conducted in a different way. To begin with, the abandoned salina E was excluded since it was not possible to obtain product samples. As not all the stages of the process were available for each salina, the number of microplastics was considered rather than their proportions.

Results in Table 2 show that the number of microplastics is similar in these stages. Particularly, salt packaging processes seem not to have any influence on the number of microplastics present in it, since they do not vary significantly from one stage to another. This phenomenon can be seen when comparing stages 7 and 9 of salina F. Results from salina A show that salt after packaging contains a slightly higher quantity of microplastics than the salt before packaging. There is a clear difference between stages 6 and 8 in this salina, but the difference is not so significant between stages 7 and 9. This may be due to the treatment the salt undergoes after washing, and not because of its packaging. Finally, the wide range of the number of microplastics in the final products when comparing inland and coastal salinas does not show any pattern related to the location of the site where the salt is made.

Fig. 6 shows the proportion of a given range with respect to all sizes, for salinas A (Panel a), B (Panel b), F (Panel c), G (Panel d), and H (Panel e). As can be seen, the two largest ranges (200–630 μm and 630–5000 μm) stand out, while the others are present in very low proportions. Fig. 7 shows the proportion of colours in salinas A (Panel a), B (Panel b), F (Panel c), G (Panel d), and H (Panel e). It was observed that the predominant colour is transparent, followed by black, blue, and red. Black colour microplastics slightly increase in the final stages, which may be related to the tires of the vehicles being used during the harvest of the salt as well as the conveyor belts for transportation. To determine its origin, analysis using Fourier Transformed Infra-Red (FTIR) technology needs to be performed.

3.2. Differences between inland and coastal salinas

As observed prior, a comparison of the proportion of microplastics of the inland active salinas, F and G, at stages 1, 2, and 3 in Fig. 3 shows that microplastic content increases from stage 1–2 and decreases from 2 to 3. This is likely due to the fact that groundwater entering the concentrator ponds stays in contact with the air, increasing the microplastic pollution. After this, microplastic fibres could settle at the bottom of the evaporating and concentrating ponds. In coastal salinas, A, B, and H, the number of microplastics follows a decreasing trend from stage 1 to stage 3. However, the fundamental difference is that stage 1, in this case, is the one with the highest proportion of microplastics when compared to the others. This may be because the incoming sea water was already contaminated, but not the groundwater. Regarding salt as final product, we did not observe significant differences between inland and coastal salinas at stages 8 and 9. Microplastic content is similar in salt coming from non-marine and marine environments, which supports results published by Iñiguez et al. [20]. Unpacked and packed salt (stages 7 and 9) have similar content in microplastics in salinas A and F, which led to the conclusion that the final packing process does not have any influence, as had been previously observed by Peixoto

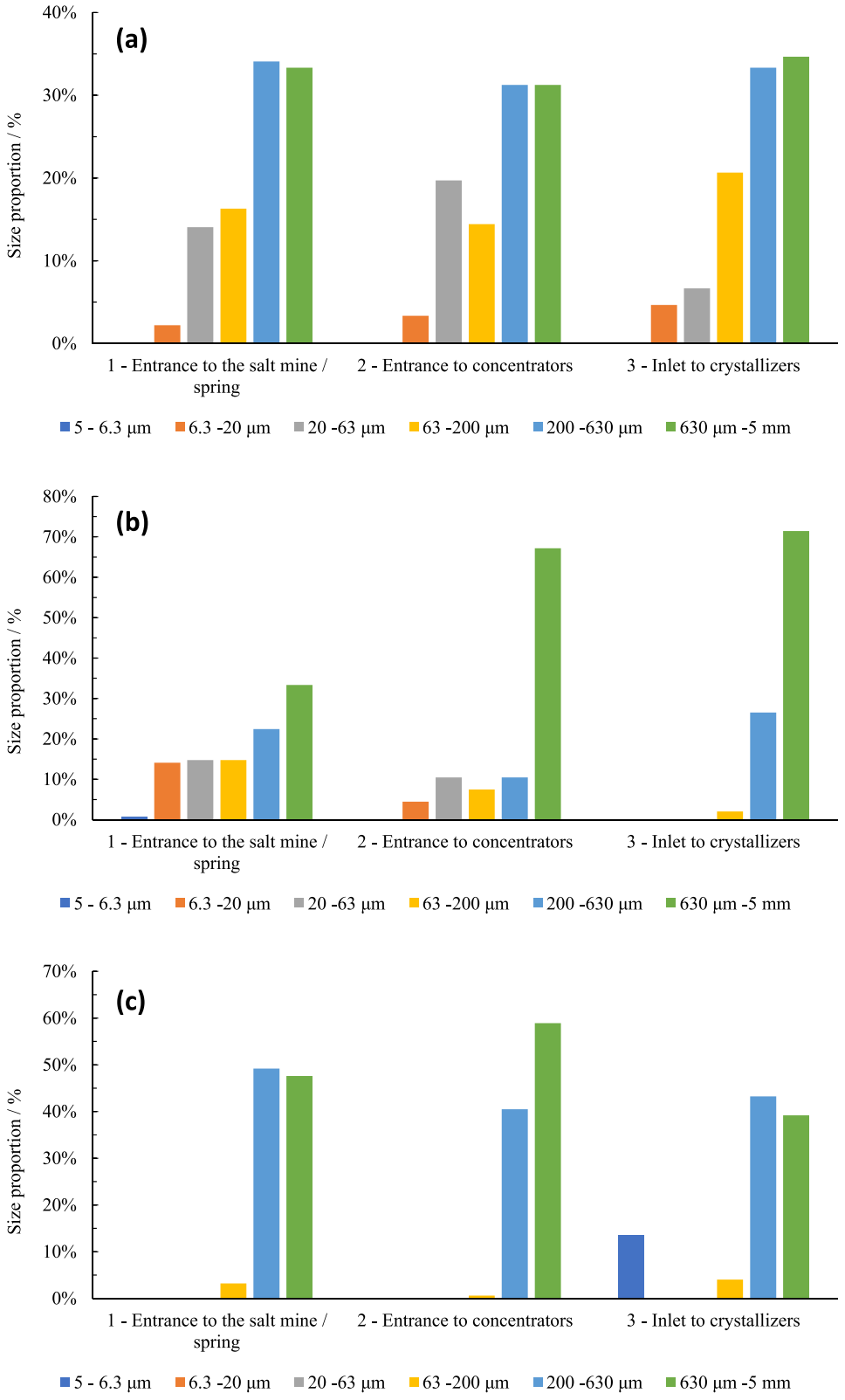


Fig. 4. Size ratio per stage: Panel a, salina A; Panel b, salina B; Panel c, salina H.

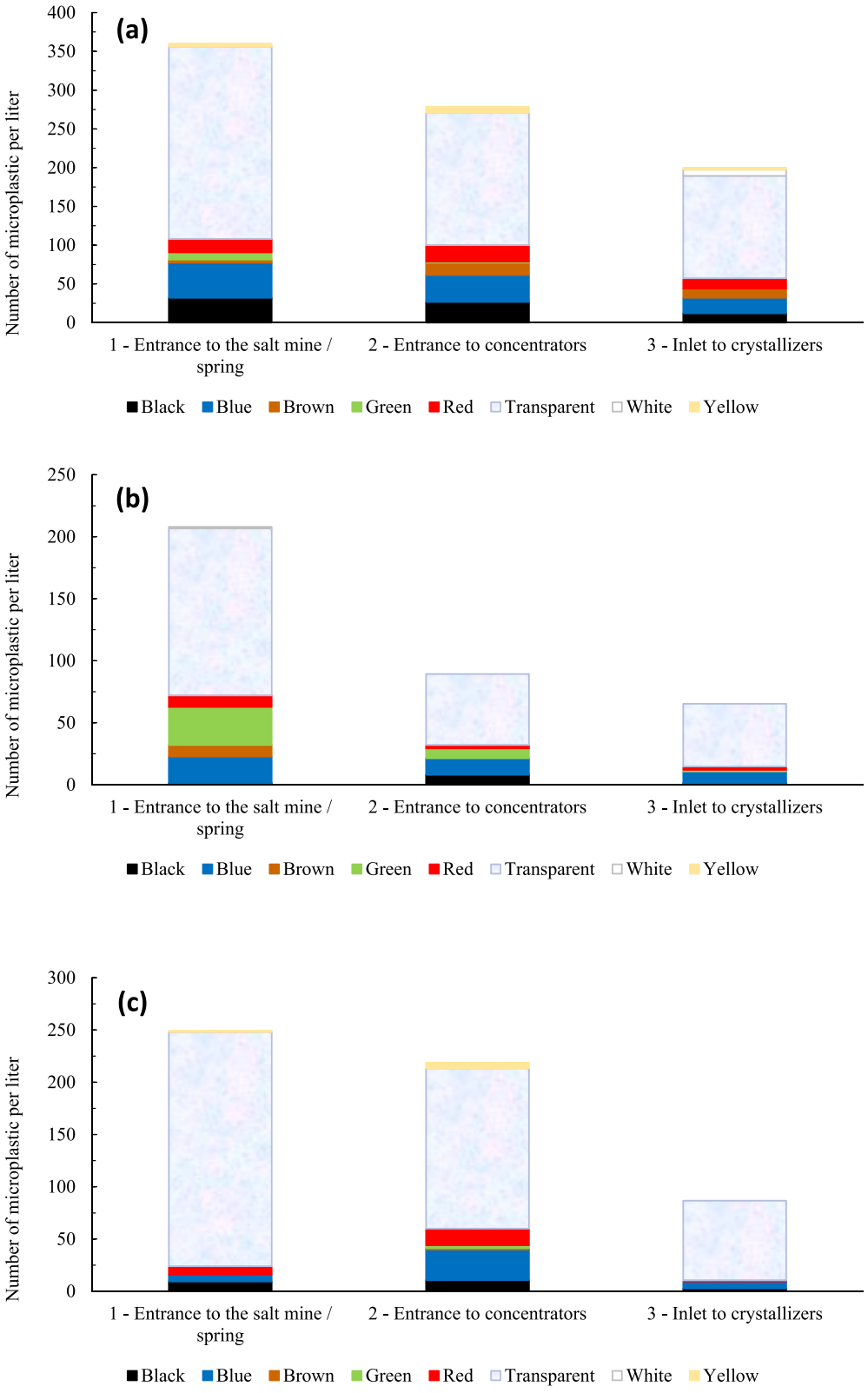


Fig. 5. Distribution of colours (number of microplastics per liter): Panel a, salina A; Panel b, salina B; Panel c, salina H. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

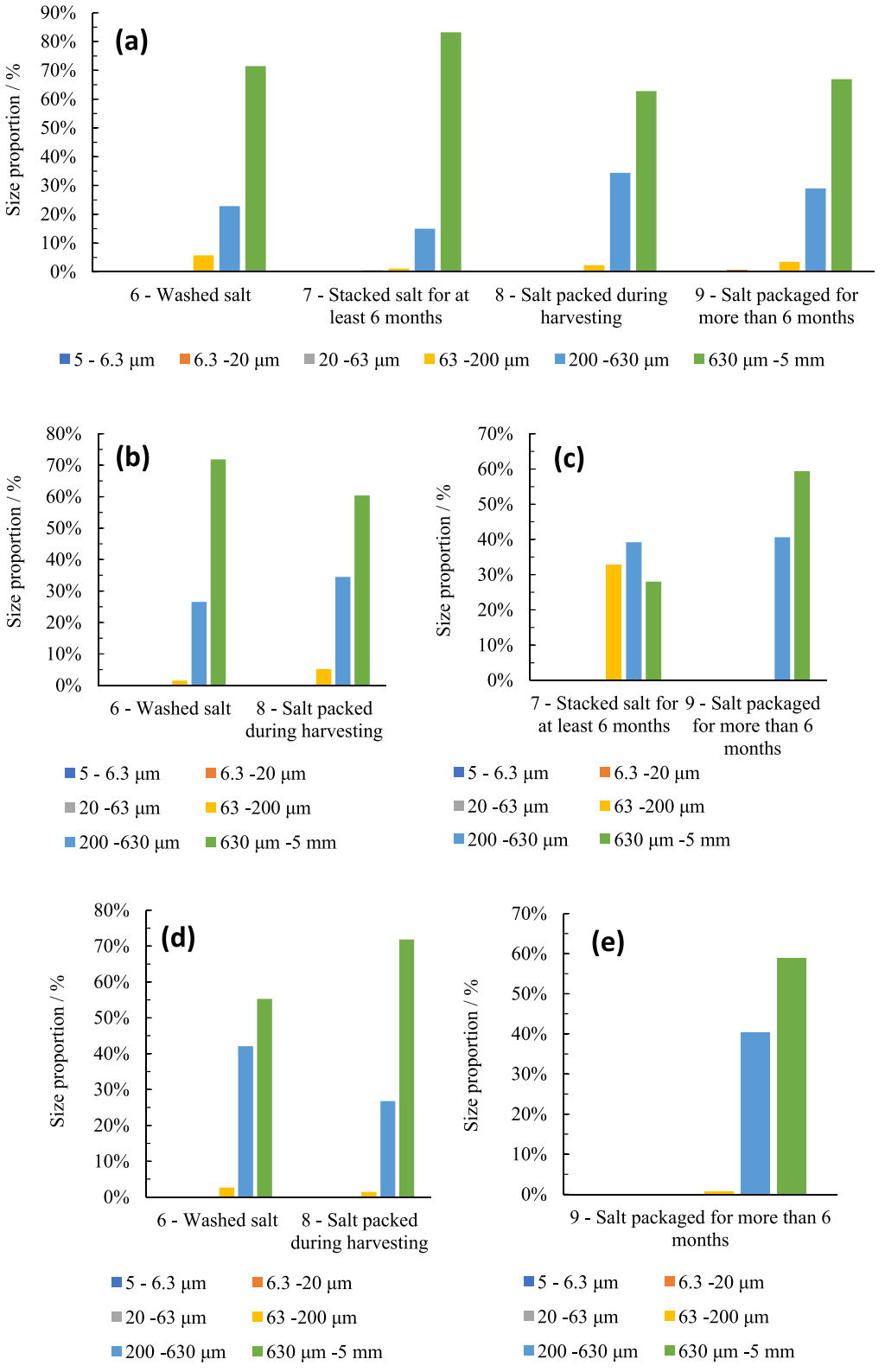


Fig. 6. Size ratio per stage: Panel a, salina A; Panel b, salina B; Panel c, salina F; Panel d, salina G; Panel e, salina H.

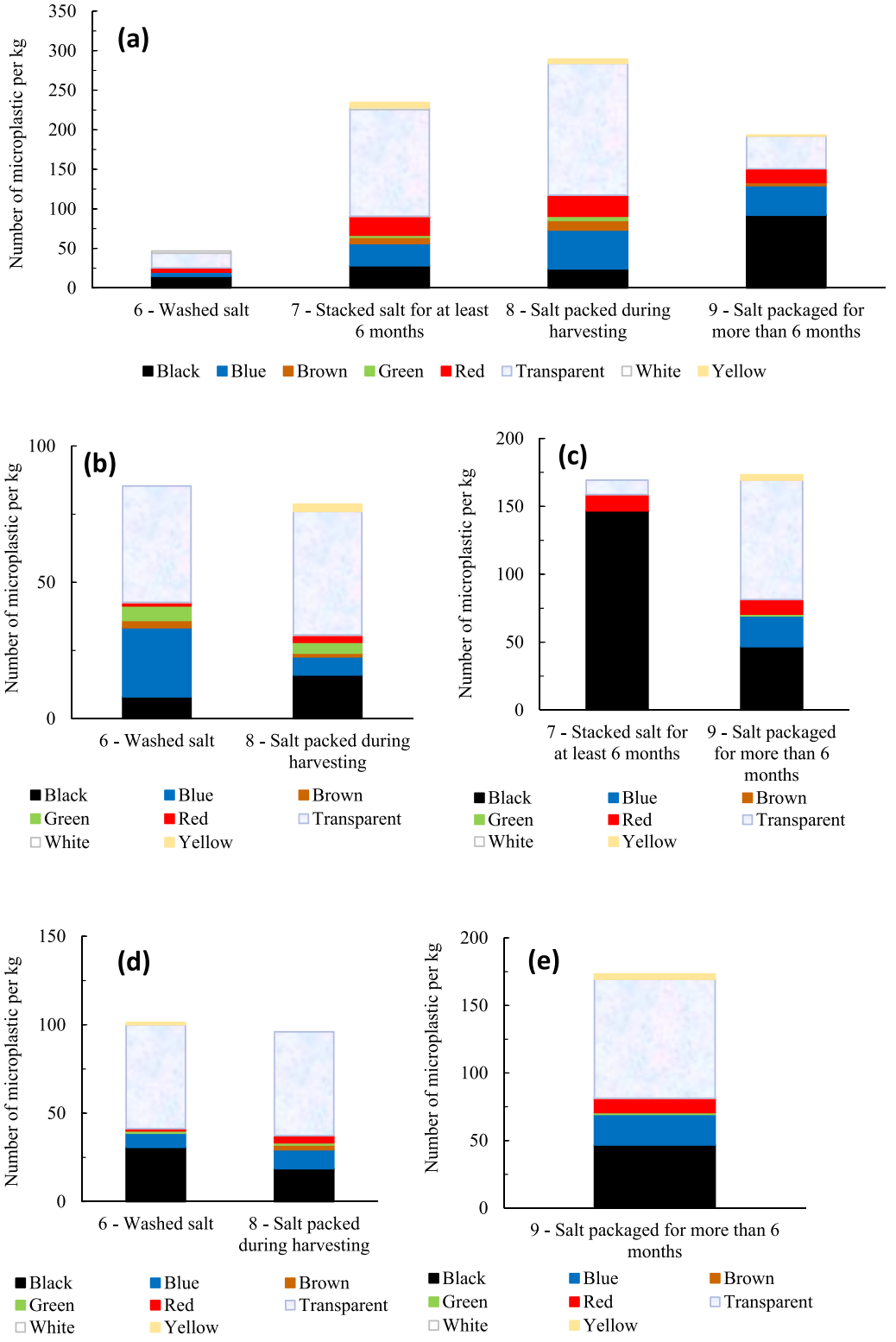


Fig. 7. Number of microplastics per kg by colour and stage: Panel a, salina A; Panel b, salina B; Panel c, salina F; Panel d, salina G; Panel e, salina H. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

et al. [17].

Fig. 8 shows the size ratio distribution of the inland salinas F and G for stages 1–3. It can be seen that the two largest ranges (200–630 μm and 630–5000 μm) are the most representative ones. Samples from stage 2 in salina G also show an important percentage of microplastics in the range 6.3–20 μm. Coastal salinas also presented large range size ratios, as seen in Fig. 4. The existence of any pattern cannot be determined by visual analysis. In addition, the fact that there are already large differences only between the inland salinas seems to indicate that the air pollution of each area was considerably different. As studied by Allen et al. [44], microplastics can be transported through air, not only from areas with high population but also from sea to land in the form of sea spray, which has a significant influence on the concentration of microplastics observed.

Related to the colour of the microplastic fibres, transparent is the most representative colour in both coastal and inland salinas. But it has also been observed that black fibres are more present in inland salinas than coastal salinas.

3.3. Abandoned salina

Fig. 9 Panel a shows the variability of the number of microplastics in the abandoned salina E in stages 1–3. It is observed that the amount of microplastics at the first stage is much higher than those obtained at the second and third stages. This first stage consists of a very exposed lagoon that used to serve as a brine reservoir. In addition, the sample was taken on the shore, risking windward accumulation of microplastics. Meanwhile, stages 2 and 3 are located in more sheltered locations and with less exposure to open air. These results constitute strong evidence of microplastic pollution by atmospheric fallout at this abandoned site, which is located 5 km away from the nearest town, on a slope sheltered by a forest with no human activity within its boundaries. Panel b in Fig. 9 represents the different size ranges of microplastics in stages 1–3, showing the predominance of large size ranges, 200–630 μm and 630–5000 μm. Finally, the variability of colours studied in Panel c shows great differences in comparison with other sites, in terms of the predominance of black colour and a low number of transparent microplastics.

4. Discussion

Inland salinas function differently from coastal ones. In the first case, the brine comes from groundwater that has been salinized due to the passage through salt deposits. Meanwhile, coastal salinas obtain the brine directly from seas and oceans. Thus, it is possible to think that, if there are microplastics present in the production stages of inland salinas, they may come either from groundwater through filtration, or by contact with the air when the water emanates to the surface.

It has been observed that microplastic pollution in inland salinas mainly occurs due to air pollution, since in both salinas F and G,

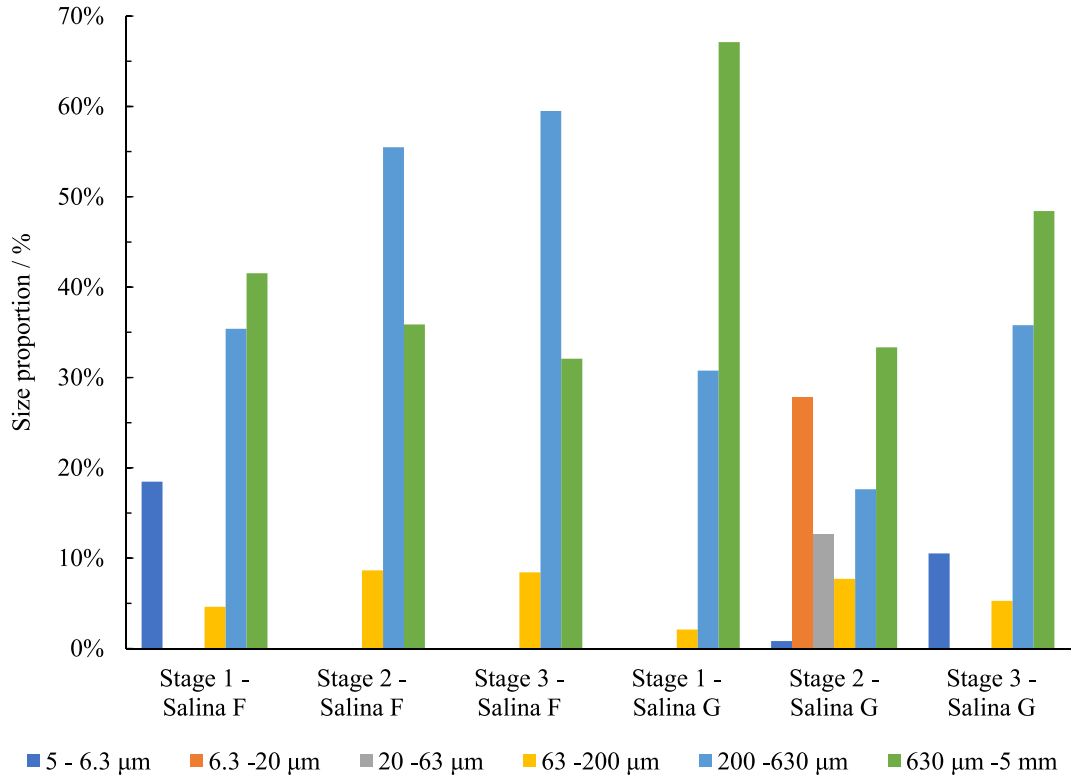


Fig. 8. Size ratio per stage in salinas F and G.

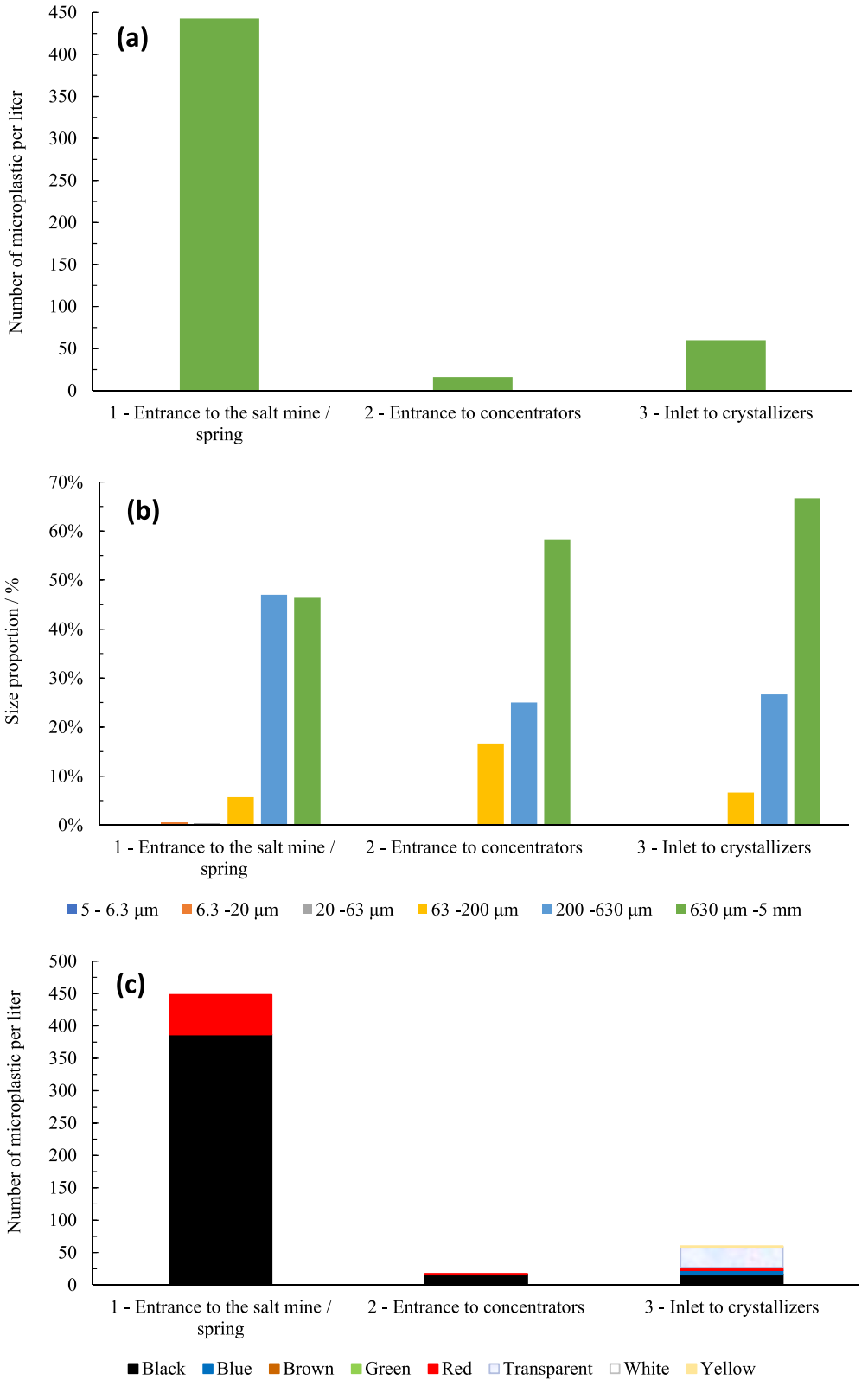


Fig. 9. Salina E: Panel a, number of microplastics per liter per stage; Panel b, proportion of sizes per stage; Panel c, number of microplastics per liter by colour per stage. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

stages 2 and 3 contain a higher number of microplastics in comparison to the first one (water inlet). This is not the case with coastal salinas, which seems to indicate that the sea is the most polluted source. However, no correlation is found between air pollution and the studied salinas, since the characteristics of microplastics are very diverse in each site, which seems to indicate that the amount and types of microplastics present in the air are very diverse and deeply depend on local environmental conditions and the proximity to populated areas. Knowledge of microplastic atmospheric pollution is still limited. Their small density and size facilitate their transportation by wind [45]. Fibres are the main shape of microplastics from atmospheric fallout; they persist for a long time in the atmosphere, and are transported over long distance. Beside this, about 50 % of the fibres observed in atmospheric fallout were longer than 1 mm [44,46]. In this study, larger size ranges dominate, suggesting atmospheric fallout from nearby environments as the likely source of pollution. These salinas are natural protected areas in Spain but they are not sufficiently separated from human activity to prevent atmospheric pollution downwind and most of them host a broad range of human activities within their surroundings. Therefore, the diverse geographical location of these salinas should be kept in mind in order to infer the influence of air pollutants from different origins. Studies on prevailing winds and the presence of nearby human settlements and activities would help clarify this.

Both coastal and inland salinas experience a decrease in microplastic pollution as water passes through different ponds until crystallizers, potentially due to particle sedimentation. Subsequent processes, such as stacking in mounds, increase microplastic numbers. However, microplastic fibres in the final packaged salt do not vary by origin, coastal or inland, and salt packaging does not significantly alter microplastic content in final products.

Transparent fibres are the most common colour, except for salina F (stage 7) and E, followed by blue and black, potentially linked to salt worker clothing and vehicle tires.

Finally, the abandoned inland salina E has been studied as an isolated case because of its particular characteristics. The water present in the ponds does not come from groundwater, but it is accumulated from precipitation instead and has been exposed to inclement weather. It has been seen that the predominant colour of the microplastic fibres found is black, instead of transparent as in the others salinas (except for stage 7 of salina F). In order to study this phenomenon, it would be convenient to analyse the density of the microplastics, and the likelihood to turn up within the samples. This would require an in-depth study with micro-FTIR technology, which will be performed in future studies, along with the characterization of microplastic fragments.

5. Conclusions

This study provides an analysis of microplastic fibres at different stages in six Spanish salinas, considering coastal or inland locations and artisanal or industrial production. Results for inland salinas indicate that microplastic contamination occurs once the water emerges from the spring and occupies the ponds in contact with open air. Coastal and inland salinas both experiment a decrease in microplastic pollution as water (brine) passes through different ponds until crystallizers, potentially due to sedimentation. Subsequent processes, such as stacking in mounds, significantly increase microplastic numbers. Microplastic fibres in final packaged salt do not vary by its origin, coastal or inland. Also, it has been observed that salt packaging does not significantly alter the number of microplastics contained in the final products.

Transparent microplastic fibres were the most common colour, except for salina F (stage 7) and E, followed by blue, which corresponds to the common clothes used by salt workers, and black, related to tires and rubber conveyors.

Larger size ranges dominate. This shows that microplastic pollution from atmosphere fallout would come from the nearby environment. If the radius of pollution transport was higher, the colours found would be similar. The abandoned inland salina E stands out due to its unique characteristics, indicating pollution from nearby human activities.

This study presents the originality of analysing the type of salt extraction process, and not just the final product, being able to discern both the nature of the salina (coastal or inland) and the number and type of stages within the salt obtaining. The results contribute to understanding the origin of contamination in salt production processes.

6. Data availability statement

All data are fully available on request.

CRediT authorship contribution statement

M.M. Cledera-Castro: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Conceptualization. **K. Hueso-Kortekaas:** Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Conceptualization. **C. Sanchez-Mata:** Investigation, Formal analysis. **C. Morales-Polo:** Writing – review & editing, Visualization, Validation, Methodology, Investigation, Formal analysis, Conceptualization. **J. Calzada-Funes:** Writing – review & editing, Visualization, Investigation. **N. Delgado-Mellado:** Writing – review & editing, Investigation. **R. Caro-Carretero:** Writing – review & editing, Visualization, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

We are greatly indebted to the hospitality and interest of managers and owners of the solar evaporation salinas studied, for granting access and facilitating the sampling process. The handling and analysis of samples in the laboratory and with the microscope has counted with the valuable contribution of María Balbás, Cristina Espinosa de los Monteros, Ana Fegies, Auon Haq, Óscar Travesí, Lucía Manzano, José María Subirá, and Philip Thomson. We are very grateful to the Comillas Pontifical University for the support and grants received through the *Proyecto Propio Microplasticos en Salinas (*PP2020_01*)*.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.heliyon.2024.e31609.

References

[1] J.P.G.L. Frias, R. Nash, Microplastics: finding a consensus on the definition, Mar. Pollut. Bull. 138 (2019) 145–147, https://doi.org/10.1016/j.marpolbul.2018.11.022.
[2] M. Cole, et al., Microplastics as contaminants in the marine environment: a review, Mar. Pollut. Bull. 62 (12) (2011) 2588–2597, https://doi.org/10.1016/j.marpolbul.2011.09.025.
[3] J.-Q. Jiang, Occurrence of microplastics and its pollution in the environment: a review, Sustain. Prod. Consum. 13 (2018) 16–23, https://doi.org/10.1016/j.spc.2017.11.003.
[4] L.S. Fendall, M.A. Sewell, Contributing to marine pollution by washing your face: microplastics in facial cleansers, Mar. Pollut. Bull. 58 (8) (2009) 1225–1228, https://doi.org/10.1016/j.marpolbul.2009.04.025.
[5] A. Lechner, et al., The Danube so colourful: a potpourri of plastic litter outnumbers fish larvae in Europe's second largest river, Environmental Pollution 188 (2014) 177–181, https://doi.org/10.1016/j.envpol.2014.02.006.
[6] M. Eriksen, et al., Plastic pollution in the world's oceans: more than 5 trillion plastic pieces weighing over 250,000 tons afloat at sea, in: H.G. Dam (Ed.), PLoS One 9 (12) (2014) e111913, https://doi.org/10.1371/journal.pone.0111913.
[7] D. Habib, D.C. Locke, L.J. Cannone, Synthetic fibers as indicators of municipal sewage sludge, sludge products and sewage treatment plant effluents, Water Air Soil Pollut. 103 (1/4) (1998) 1–8, https://doi.org/10.1023/A:1004908110793.
[8] N. Evangeliou, et al., Atmospheric transport is a major pathway of microplastics to remote regions, Nat. Commun. 11 (1) (2020) 3381, https://doi.org/10.1038/s41467-020-17201-9.
[9] M. González-Pleiter, et al., First detection of microplastics in the freshwater of an antarctic specially protected area, Mar. Pollut. Bull. 161 (2020) 111811, https://doi.org/10.1016/j.marpolbul.2020.111811.
[10] P. Schwabl, et al., Detection of various microplastics in human stool: a prospective case series, Ann. Intern. Med. 171 (7) (2019) 453–457, https://doi.org/10.7326/M19-0618.
[11] A. Ragusa, et al., Plasticenta: first evidence of microplastics in human placenta, Environ. Int. 146 (2021) 106274, https://doi.org/10.1016/j.envint.2020.106274.
[12] C. Díaz-Mendoza, et al., Plastics and microplastics, effects on marine coastal areas: a review, Environ. Sci. Pollut. Control Ser. 27 (32) (2020) 39913–39922, https://doi.org/10.1007/s11356-020-10394-y.
[13] G. D'Avignon, I. Gregory-Eaves, A. Ricciardi, Microplastics in lakes and rivers: an issue of emerging significance to limnology, Environ. Rev. 30 (2) (2022) 228–244, https://doi.org/10.1139/er-2021-0048.
[14] M. Junaid, et al., Enrichment and dissemination of bacterial pathogens by microplastics in the aquatic environment, Sci. Total Environ. 830 (2022) 154720, https://doi.org/10.1016/j.scitotenv.2022.154720.
[15] Brouwer, et al., Assessing the performance of marine plastics cleanup technologies in Europe and North America, Ocean Coast Manag. 238 (2023) 106555, https://doi.org/10.1016/j.ocecoaman.2023.106555.
[16] J.G.B. Derraik, The pollution of the marine environment by plastic debris: a review, Mar. Pollut. Bull. 44 (9) (2002) 842–852, https://doi.org/10.1016/S0025-326X(02)00220-5.
[17] D. Peixoto, et al., Microplastic pollution in commercial salt for human consumption: a review, Estuar. Coast Shelf Sci. 219 (2019) 161–168, https://doi.org/10.1016/j.ecss.2019.02.018.
[18] Q. Zhang, et al., A review of microplastics in table salt, drinking water, and air: direct human exposure, Environ. Sci. Technol. 54 (7) (2020) 3740–3751, https://doi.org/10.1021/acs.est.9b04535.
[19] L. Yang, et al., Challenges and strategies for solid-state anaerobic digestion of lignocellulosic biomass, Renew. Sustain. Energy Rev. 44 (2015) 824–834, https://doi.org/10.1016/j.rser.2015.01.002.
[20] M.E. Iñiguez, J.A. Conesa, A. Fullana, Microplastics in Spanish table salt, Sci. Rep. 7 (1) (2017) 8620, https://doi.org/10.1038/s41598-017-09128-x.
[21] A. Karami, et al., The presence of microplastics in commercial salts from different countries, Sci. Rep. 7 (1) (2017) 46173, https://doi.org/10.1038/srep46173.
[22] S. Gündoğdu, Contamination of table salts from Turkey with microplastics, Food Addit. Contam. 35 (5) (2018) 1006–1014, https://doi.org/10.1080/19440049.2018.1447694.
[23] J.-S. Kim, et al., Global pattern of microplastics (MPs) in commercial food-grade salts: sea Salt as an indicator of seawater MP pollution, Environ. Sci. Technol. 52 (21) (2018) 12819–12828, https://doi.org/10.1021/acs.est.8b04180.
[24] M. Kosuth, S.A. Mason, E.V. Wattenberg, Anthropogenic contamination of tap water, beer, and sea salt, in: Z. Zhou (Ed.), PLoS One 13 (4) (2018) e0194970, https://doi.org/10.1371/journal.pone.0194970.
[25] M. Renzi, A. Blašković, Litter & microplastics features in table salts from marine origin: Italian versus Croatian brands, Mar. Pollut. Bull. 135 (2018) 62–68, https://doi.org/10.1016/j.marpolbul.2018.06.065.
[26] C.K. Seth, A. Shriwastav, Contamination of Indian sea salts with microplastics and a potential prevention strategy, Environ. Sci. Pollut. Control Ser. 25 (30) (2018) 30122–30131, https://doi.org/10.1007/s11356-018-3028-5.
[27] M. Fischer, I. Goßmann, B.M. Scholz-Böttcher, Fleur de Sel—An interregional monitor for microplastics mass load and composition in European coastal waters? J. Anal. Appl. Pyrol. 144 (2019) 104711 https://doi.org/10.1016/j.jaap.2019.104711.
[28] M. Renzi, et al., Microparticles in table salt: levels and chemical composition of the smallest dimensional fraction, J. Mar. Sci. Eng. 7 (9) (2019) 310, https://doi.org/10.3390/jmse7090310.
[29] M.N. Sathish, I. Jeyasanta, J. Patterson, Microplastics in salt of tuticorin, southeast coast of India, Arch. Environ. Contam. Toxicol. 79 (1) (2020) 111–121, https://doi.org/10.1007/s00244-020-00731-0.
[30] M. Sivagami, et al., Extraction of microplastics from commonly used sea salts in India and their toxicological evaluation, Chemosphere 263 (2021) 128181, https://doi.org/10.1016/j.chemosphere.2020.128181.

[31] M. Smiechowska, Ecological threats for coastal systems in the context of acquiring and providing the quality of sea salt. A review, Journal of Research and Applications in Agricultural Engineering 63 (3) (2018) 118–122.
[32] K. Hueso Kortekaas, et al., Microplastics in food-grade salt: how bad is the problem? El Alfolí 28 (2021) 11–19. https://repositorio.comillas.edu/xmlui/bitstream/handle/11531/65476/Microplastics_in_food-grade_salt_how_bad_is_the_problem.pdf?sequence=1.
[33] J.F. Carrasco Vayá, K. Hueso Kortekaas, *Los paisajes ibéricos de la sal. 1, Las salinas de interior*. Guadalajara: Asociación de Amigos de las Salinas de Interior, 2008. Print.
[34] EUsalt, About salt [WWW Document], Website Eur. Salt Prod. Assoc. URL (2024). https://eusalt.com/. (Accessed 16 February 2024).
[35] K. Hueso Kortekaas, Salt in our veins. The Patrimonialization Processes of Artisanal Salt and Saltscapes in Europe and Their Contribution to Local Development, Universitat de Barcelona, 2017. Ph.D. Thesis, https://www.tdx.cat/handle/10803/418811. (Accessed 17 July 2023).
[36] Horton and Dixon, Microplastics: an introduction to environmental transport, WIREs Water 5 (2018) e1268, https://doi.org/10.1002/wat2.1268.
[37] R. Serrano, et al., Non-target screening of organic contaminants in marine salts by gas chromatography coupled to high-resolution time-of-flight mass spectrometry, Talanta 85 (2011) 877–884, https://doi.org/10.1016/j.talanta.2011.04.055.
[38] D. Yang, et al., Microplastic pollution in table salts from China, Environ. Sci. Technol. 49 (2015) 13622–13627, https://doi.org/10.1021/acs.est.5b03163.
[39] A. Sofia Soares, et al., Microplastic pollution in Portuguese saltworks, in: A. Devlin, J. Pan, M. Manjur Shah (Eds.), Inland Waters - Dynamics and Ecology, IntechOpen, 2021, https://doi.org/10.5772/intechopen.91476.
[40] K. Hueso Kortekaas, J.F. Carrasco Vayá, Biodiversity of inland saltscapes of the Iberian Peninsula, Nat. Resour. Environ. Issues 15 (2009) 30. Article, https://digitalcommons.usu.edu/nrei/vol15/iss1/30.
[41] J.-F. Carrasco-Vayá, K. Hueso-Kortekaas, Los paisajes ibéricos de la sal I: Las salinas de interior, Asociación de Amigos de las Salinas de Interior (2008).
[42] D. Dwiyitno, et al., Influence of various production methods on the microplastic contamination of sea salt produced in Java, Indonesia, Environ. Sci. Pollut. Control Ser. 28 (2021) 30409–30413, https://doi.org/10.1007/s11356-021-14411-6.
[43] C.J. Thiele, et al., Microplastics in European sea salts – an example of exposure through consumer choice and of interstudy methodological discrepancies, Ecotoxicol. Environ. Saf. 255 (2023) 114782, https://doi.org/10.1016/j.ecoenv.2023.114782.
[44] S. Allen, et al., Examination of the ocean as a source for atmospheric microplastics, PLoS One 15 (5) (2020) e0232746, https://doi.org/10.1371/journal.pone.0232746.
[45] C. Edo, et al., A nationwide monitoring of atmospheric microplastic deposition, Sci. Total Environ. 905 (2023) 166923, https://doi.org/10.1016/j.scitotenv.2023.166923.
[46] G. Chen, et al., Mini-review of microplastics in the atmosphere and their risks to humans, Sci. Total Environ. 703 (2020) 135504, https://doi.org/10.1016/j.scitotenv.2019.135504.