

1 From the ocean to jellies forth and back? Microplastics along the commercial  
2 life cycle of red algae

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14  
15 **Abstract**

16 Red algae are increasingly exploited for direct consumption and for production of gelling  
17 agents like agar and carrageenan, widely employed in food and personal care products.  
18 In this article we identify knowledge gaps about microplastics in the whole commercial  
19 life cycle of gelling red algae, from their marine production to the final wastewater  
20 treatment. Recommendations for new research include studies of microplastics  
21 deposition on red algae at sea, during the industrial process of production of gelling  
22 agents, and indeed about improvements of microplastics retention in wastewater  
23 treatment plants.

24  
25 *INTRODUCTION*

26 Marine litter studies have been on the rise for the last decades, being  
27 microplastics (MP) an emerging focus. That is because MPs act like vectors for toxic  
28 compounds such as Bisphenol A (Rochester, 2013), an endocrine disruptor, and heavy  
29 metals (Brennecke et al., 2016; Bradney et al., 2019; Pachana et al., 2010). These toxics  
30 can cause several diseases to marine fauna and finally to the human consumer  
31 (Andrady, 2011; Cole et al., 2011; Brennecke et al., 2016). MPs are the most abundant  
32 litter particles in the seas (Galgani et al., 2015; Barcelo and Pico, 2020). This is due to the  
33 continuous degradation of bigger plastic elements (like fishing nets and strains, see  
34 Figure 1) that form secondary MP ( Cole et al., 2011; Carbery et al., 2018; Setälä et al.,  
35 & Cole, 2018), and also to the release of primary MP in cosmetics and industrial products  
36 (Lei et al., 2017; Zhou et al., 2020). Primary release MP are incorporated in the product  
37 during the industrial process, as in personal care products, detergents, paints, abrasives,  
38 agriculture and others (European Commission (DG Environment) intentionally added

39 microplastics in products.  
40 <https://ec.europa.eu/environment/chemicals/reach/pdf/39168%20Intentionally%20a>  
41 [dded%20microplastics%20-%20Final%20report%2020171020.pdf](https://ec.europa.eu/environment/chemicals/reach/pdf/39168%20Intentionally%20added%20microplastics%20-%20Final%20report%2020171020.pdf)). MPs, whose largest  
42 length is 5 mm (Arthur et al., 2009), are found floating in the water column (Lavender &  
43 Thompson, 2014), deposited on the bottom (e.g. Pabortsava & Lampitt, 2020), and also  
44 associated to biota: inside animals (Cole et al., 2014) and attached to the external  
45 surfaces of marine plants (Gutow et al., 2016; Goss et al., 2018).

46 The presence of MP in water and biota leads to their introduction into marine  
47 food webs starting in lower levels (Setälä et al., 2014; Goss, Jaskiel, & Rotjan, 2018) and  
48 then arriving at higher levels by trophic transfer (Farrell & Nelson, 2013; Carbery,  
49 Connor, & Thavamani, 2018; Nelms et al., 2018; Zhang et al., 2019). Since MPs are not  
50 absorbed in the intestinal tract in significant quantities, they do not accumulate in  
51 animal organs and tissues (Akoueson et al., 2020); thus the trophic transfer depends on  
52 the MPs ingested by the prey at the time of predation (Priscilla, Sedayu, & Patria, 2019),  
53 and MP concentration in higher trophic levels may be diluted. For this reason, although  
54 significant MPs ingestion has been demonstrated from their presence in faeces (Schwabl  
55 et al., 2019), the risk of human consumption of MP via marine animals is debatable,  
56 especially from large fish where muscle is the main edible tissue. In contrast, ingestion  
57 of primary producers (algae) implies the ingestion of MP deposited on or adhered to  
58 their surface, which may pose a risk to the human consumer (Carbery, O'Connor, &  
59 Palanisami, 2018; Waring, Harris, & Mitchell, 2018). Many studies have been published  
60 related to MPs in the marine environment and trophic transfer in animals (e.g. Farrell &  
61 Nelson, 2013; Chagnon et al., 2018). However, some marine species that are highly  
62 consumed by humans have not yet received much attention regarding the risk of MP  
63 transfer to humans: red algae.



64  
65 *Figure 1: Macroplastic (Right) and Microplastic (Left) found in El Arañón and Zeluán beaches respectively*  
66 *(Ría de Avilés, Asturias, Northern Spain). Photographs taken by D. Menéndez.*

### 67 *Red algae and their gelling agents*

68 Red algae (Rhodophyta Wettstein, 1901) is an algal division mainly comprised by  
69 macroalgae (Díaz González et al., 2004). They are highly consumed worldwide due to  
70 their nutritional benefits (Kumar et al., 2008). According to Díaz González et al. (2004),  
71 the genus *Porphyra* is the most important because its species are widely cultivated for  
72 food production: Nori in Japan, Purple Laver in Wales. *Palmaria palmata* is also intensely

73 cultivated for its antioxidant benefits (Yuan, Bone, & Carrington, 2005; Yuan, Carrington  
74 & Walsh, 2005).

75 Other red algae of high interest for humans are gelling agents' producers:  
76 agarophytes and carrageenaphytes  
77 (<http://www.fao.org/3/y4765e/y4765e04.htm#bm04.3>). Agar and Carrageenan,  
78 respectively, are the gelling agents extracted from them (Usov, 1998). These compounds  
79 are sulphated polysaccharides present in the cell wall, where they represent 15%-30%  
80 of dry weight in Agar producers and 30%-80% in carrageenan producers (Rioux &  
81 Turgeon, 2015). Agar is principally extracted from the following species: *Gelidium*  
82 *sesquipedale*, *Gracilaria* (*G. gracilis*, *G. dura* and *G. bursa-pastoris*), *Gelidiella acerosa*,  
83 *Pterocladia capillacea* and *Anhfeldtia plicata* ( Salt, 1976; Prasad et al., 2007; Lee et al.,  
84 2017; Titlyanov et al., 2017; Lebbar et al., 2018; ) and carrageen from *Chondrus crispus*,  
85 *Mastocarpus stellatus*, *Kappaphycus alvarezii*, *Gigartina* (*G. decipiens*, *G. skottsbergii*  
86 and *G. stellate*), *Sarcothalia crispate* and *Euचेuma striatum* ( Craigie, 1978; Epifanio et  
87 al., 1981; Díaz González et al., 2004; Tasende et al., 2013; Hughes et al., 2018).

88 Gelling agents are commonly used in research (functional biology,  
89 pharmaceutics, molecular biology...) (Bhattacharya, Dey & Bhattacharyya, 1994; Shilpa  
90 et al., 2011) and in cosmetics and food industry (Aliste et al., 2000; Joshi et al., 2018).  
91 Moreover, algal gelling agents are a perfect substitute for animal gelatine. Both Agar (E-  
92 406) and Carrageenan (E-407) are commonly used as food additives due to their gelling,  
93 thickening and nutritional properties (Rioux & Turgeon, 2015; Cunha & Grenha, 2016;  
94 Lee et al., 2017). Seaweed species are harvested to elaborate vegan, vegetarian and  
95 religion-friendly products (e.g. halal, kosher), and to reduce animal exploitation. The  
96 industry of gelling red algae products is estimated to produce more than 370 million  
97 dollars in benefits according to FAO (<http://www.fao.org/3/y5600s/y5600s07.htm>).  
98 Due to their wide use, it is important to know their role in the transfer of hazardous  
99 elements like MP from the ocean to consumers.

#### 100 *MP and red algae, what do we know?*

101 Figure 2 represents the cycle of red algae where MP can be transported. The  
102 cycle starts with the adhesion of MP to the algal surface (step 1), which, although not  
103 much studied yet, has been demonstrated in *Fucus vesiculosus* (Gutow et al., 2016),  
104 *Pyropia* sp. (Li et al., 2020), *Endocladia muricata* (Saley et al., 2019) and others. Then the  
105 algae production (step 2) will introduce more MP because nets, boxes and other tools  
106 employed to cultivate or harvest red algae are made of different types of plastic (Li et  
107 al., 2016; Simeonova & Chururkova, 2020). Red algae can be directly consumed – sold  
108 fresh or dry for human consumption, so humans will directly ingest MP carried by them.  
109 Alternatively, they can be industrially processed (step 3) to extract the gelling  
110 compounds and use them in different fields (cosmetics, food). Those compounds will  
111 contain the MP carried by red algae, and others may be added purposely (primary MP),  
112 for example in cosmetics like tooth paste and scrubs (Napper et al., 2015; Duis & Coors,  
113 2016; Anagnosti et al., 2021). Finally, either consumed directly or as part of gelling  
114 industry, MP contained in red algae will be part of wastes/debris present in wastewater

115 to be treated in Wastewater Treatment Plants (WWTP). Hospido et al. (2004) call WWTP  
 116 “ecological treatment systems” where pollutants are removed from water before it is  
 117 returned to the environment. Although their efficiency of MP retention can be up to  
 118 90% or even higher, a part of them escapes their retention systems (Murphy et al., 2016;  
 119 Masiá et al., 2020), and will return to the sea (step 4).

120

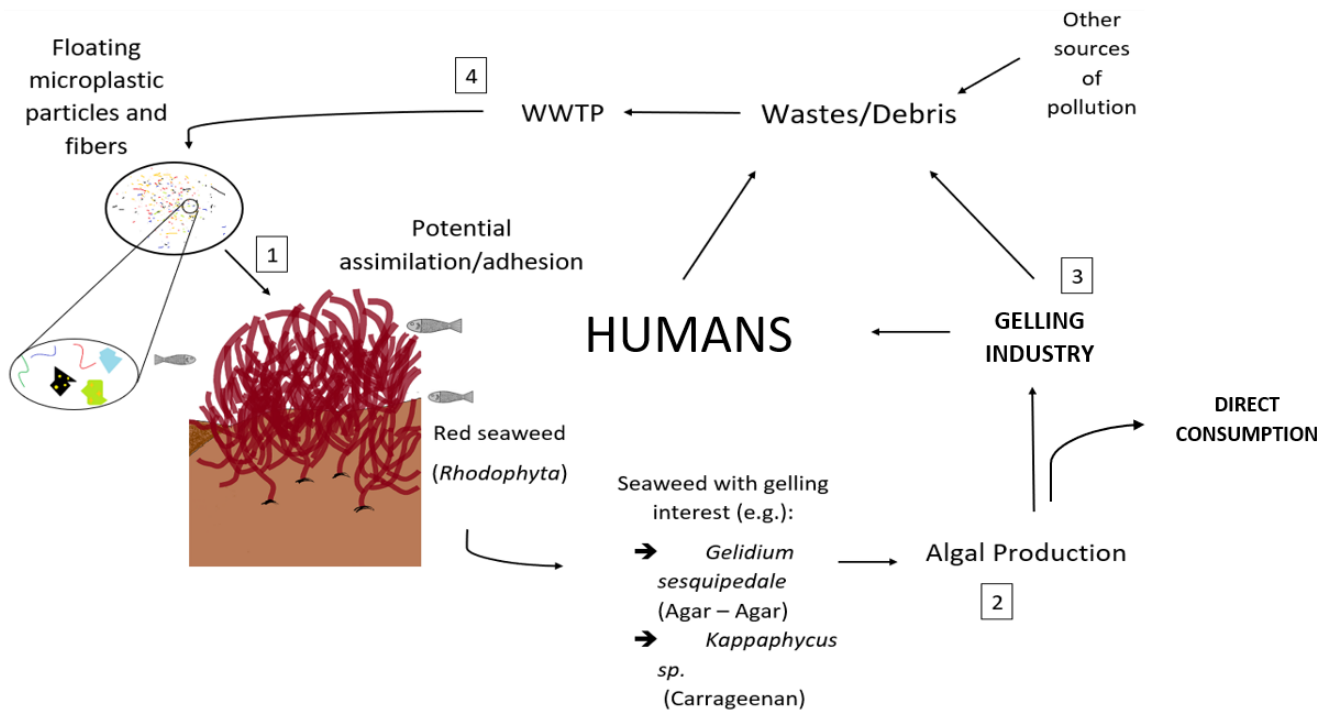


Figure 2: The MPs cycle through red algae used by humans. Steps 1, 2, 3 and 4 are critical for MP increase or retention.

121 Here we consider only the regular, legal cycle; in addition, we could add those  
 122 industrial and household wastes that are irregularly treated, directly dumped, or  
 123 accidentally deposited in rivers and seas. These have been estimated to be the majority  
 124 of wastewater volumes in some regions ([https://www.unwater.org/water-facts/quality-  
 125 and-wastewater-2/](https://www.unwater.org/water-facts/quality-and-wastewater-2/))

126 *MP and red algae, what do not we know?*

127 The first gap of knowledge in step 1 (Figure 2) is the lack of information of MP  
 128 content in many species of red algae exploited for direct human consumption and for  
 129 gelling compounds. Due to their use in food and cosmetic industry (Duis and Coors,  
 130 2016; Welden & Cowie, 2017) their study from the point of view of MP contents is  
 131 considered essential (Guerranti et al., 2019). However, according to Li et al. (2020), very  
 132 few studies have been published to evaluate how MPs affect macroalgae and how those  
 133 MPs are transferred to the food web. Studies reporting MP on red algae at sea are  
 134 limited to a few edible species such as *Porphyra* sp. (Li et al., 2020). Many more studies  
 135 are needed about MP content in red algae, especially in important genera such as  
 136 *Gelidium*, *Gracilaria*, *Kappaphycus*, *Eucheama* and others.

137 The second knowledge gap occurs in Step 2 (Figure 2). The MPs produced during  
 138 the extraction of red algae (and in general of seaweed) are still not well known. Plastic  
 139 nets and protection gloves (made of neoprene) are commonly used in fishing and  
 140 aquaculture industry to manipulate all products, and are considered sources of MP  
 141 (Montarsolo et al., 2018). Elements used in red algae harvesting and handling processes  
 142 could be considered MP pollution sources, but currently there is another lack of  
 143 information here (Duis & Coors, 2016).

144 In step 3 (Figure 2) there is another gap. Although gelling products are widely  
 145 used, MPs non-intentionally introduced during their industrial life cycle have not been  
 146 considered yet. For example, air floating MP are a pollution source in all environments  
 147 (Gasperi et al., 2015; Gasperi et al., 2018), and is logically expected to occur in the  
 148 industry of gelling products. It is difficult in the industrial environment to avoid this  
 149 pollution due to the current great dependence on plastic. Also, post-industrial processes  
 150 such as packaging in plastic bags can be considered as a MP source (Wagner, 2017;  
 151 Gerritse et al., 2020). Moreover, although products with primary MP have been  
 152 thoroughly studied (e.g. Leslie, 2014; Duis and Coors, 2016; Guerranti et al., 2019), Agar  
 153 and Carrageenan have yet to be examined as potential sources of MP in human diet to  
 154 date.

155 Knowledge gaps about MPs, summarized in Table 1, occur at the WWTP level too  
 156 (step 4 in Figure 2). Indeed, these gaps are not specific of MP derived from red algae but  
 157 general and can be applied to all MPs. As explained above and in other studies  
 158 (Ziajahromi, Neale, & Leusch, 2016; Edo et al., 2020), the improvement of WWTP  
 159 systems for MP retention is needed to ensure they do not enter into running waters and  
 160 arrive in the sea.

161

162 *Table 1: Summary of needs and gaps in the cycle of gelling agents' producers.*  
 163 *WWTPs: wastewater treatment plants.*

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	<b>STEP 1</b>	<b>STEP 2</b>	<b>STEP 3</b>	<b>STEP 4</b>
<b>What do we know</b>	Microplastics are commonly adhered to algal surfaces.	Plastic gear may add microplastics to the production chain in fishing and aquaculture.	Gelling agents can contain microplastics due to industrial handling and processing.	WWTPs are sources of microplastics because they are not specifically prepared to retain them.
<b>What we have to focus on</b>	Algae producing gelling agents are widely exploited but are not studied as microplastic vectors.	Microplastic input during algae harvesting and handling is not well known.	All steps in the industrial processing of algae, from microplastics in the air to the packaging of the final product.	The improvement of microplastics retention systems in WWTPs.

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### *Conclusions and recommendations*

169 Red algae production is in great demand because their derived products are  
170 widely employed in daily life. Here we have detected knowledge gaps in several steps of  
171 the life cycle of commercial red algae regarding their content of MP, from primary  
172 production to the treatment of final wastes. Recommendations for future studies begin  
173 with the analysis of MP deposition on or adherence to all exploited species of red algae.  
174 Subsequent studies could address their industrial handling during harvesting and  
175 aquaculture, as well as the sources of contamination in the process of obtaining agar  
176 and carrageenan. The amount of MP in agar and carrageenan should be analysed given  
177 their wide use in food products. Finally, WWTPs should continue to be studied in order  
178 to improve their efficiency of MP retention systems.

179

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