

1 **Abundance of plastic microbeads in Hong Kong coastal water**

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3 Wing Kwan So^{a,b}, Kayi Chan^{a,b}, Christelle Not^{a,b*}

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5 ^aDepartment of Earth Sciences, University of Hong Kong, Pokfulam, Hong Kong

6 ^bSwire Institute of Marine Science, University of Hong Kong, Cap d'Aguilar, Hong Kong

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8 *corresponding author: Christelle Not cnot@hku.hk

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12 **ABSTRACT**

13 To address the rising concern over the use of plastic microbeads in personal care and cosmetic
14 products, countries worldwide have started taking legislative actions to ban microbeads. Yet, the
15 degree of contamination of coastal waters by plastic microbeads is rarely reported. Surface manta
16 trawls were conducted to investigate the presence of microbeads in the southern coastal waters of
17 Hong Kong. Considering only the size fraction of 0.3 to 1 mm, 60% of samples were found to
18 contain microbeads. Microbeads accounted for 3.6% of the total microplastics collected and
19 microbead abundance ranged from 0 to 380,129 pcs/km². The shapes, sizes, colours, and
20 composition of microbeads found in our samples were similar to those from tested facial scrubs,
21 suggesting that pelagic microbeads collected in this study very likely originated from the cosmetic
22 products available locally. Microbeads represent a non-negligible part of the microplastics found
23 in surface coastal waters.

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25 **1. Introduction**

26 The increasing presence of plastic litter in the ocean is a growing concern of the public.
27 Approximately 5.25 trillion plastic particles weighing 268,940 tonnes are believed to have reached
28 the ocean between 2007 to 2013 (Eriksen et al., 2014). Plastic pieces bigger than 5 mm are
29 identified as macroplastic and pieces equal or smaller than 5 mm are identified as microplastic by
30 the scientific community (Arthur et al., 2009). Microplastic particles have been found in several
31 marine environments such as gyres, coastal surface waters, the deep sea, and beaches, and their
32 size restricts their recovery during cleanup efforts (Lee et al., 2015; Lusher et al., 2014; Moore et
33 al., 2001; Van Cauwenberghe et al., 2013). In the marine environment, microplastic originates
34 from either the degradation of macroplastic pieces, which then are referred to as secondary
35 microplastic, or direct release in their original size, which then are referred to as primary
36 microplastic (Barnes et al., 2009).

37 Primary microplastics are manufactured by the industry in differing sizes depending on their usage.
38 For example, microbeads are primary microplastics smaller than 1 mm, mainly used in personal
39 care and cosmetic products (Schneiderman, 2014). Several studies have investigated the variety of
40 physical characteristics (i.e. shape, colour) and composition of microbeads from personal care or
41 cosmetics products (Cheung and Fok, 2017; Fendall and Sewell, 2009; Napper et al., 2015;
42 Schneiderman, 2015). They observed that microbeads are principally blue, white or transparent
43 with spherical shape and mainly polyethylene (PE) but some have polypropylene (PP) or
44 polystyrene (PS) compositions (Zitko and Hanlon, 1991; Fendall and Sewell, 2009; Napper *et al.*,
45 2015). Microbeads from personal care products sold in Hong Kong are principally a combination
46 of colourless particles in granular shape and blue or colourless spherical pieces, mostly composed
47 of low density polyethylene (LDPE; Cheung and Fok, 2017). Effluent outfall is considered the
48 major source contributing to microbead pollution (Leslie, 2015; Magnusson and Norén, 2014;

49 Murphy et al., 2016). As the substitutes of natural scrubbers in personal care and cosmetics
50 products, plastic microbeads are designed to be ditched down the drain (Rochman et al., 2015;
51 Schneiderman, 2014). Murphy et al. (2016) estimated that microbeads are effectively but not
52 totally captured in the grease of the wastewater treatment, leading to a non-negligible release into
53 the aquatic environment. Between 80 to 98% of microbeads retention by wastewater treatment
54 have been suggested, depending on the technology of the treatment plant (Duis and Coors, 2016;
55 Murphy et al., 2016) still it was estimated that over 94,000 microbeads could wash down the drain
56 in a single use and in each cubic meter of treated sewage discharge up to 7,000 microbeads could
57 pass through wastewater treatment screens and directly enter the sea (Napper et al., 2015; Rochman
58 et al., 2015). Only a few studies have investigated the presence of microbeads in the ocean as both
59 their size and their identification are difficult in the natural environment (Cheung and Fok, 2016;
60 Isobe, 2016). Microbeads, which have a diameter around 100-200 μm (Fendall and Sewell, 2009;
61 Napper et al., 2015), are not collected by regular surface trawling as the mesh net is typically 335
62 μm . In addition, the differentiation between non-spherical microbeads and secondary microplastics
63 in samples is problematic. With these limitations, the identification of microbeads in previous
64 studies are clearly underestimations; yet they are the best estimates available to evaluate the role
65 of cosmetics and personal care products on microplastic pollution in coastal waters (Eriksen et al.,
66 2013; Isobe, 2016).

67 Even though recent experiments have demonstrated the role of microbeads as a transport vector
68 for chemicals (Napper et al., 2015), their threat to the marine environment is still debated
69 (Koelmans et al., 2017; Rist et al., 2018). However, the source of microbeads, in comparison to
70 secondary microplastics, is unique and therefore, any changes in personal care and cosmetics
71 regulations will directly impact the presence of microbeads in the marine environment. Over the
72 last five years, several countries such as the UK, Canada, and New Zealand, several states in the

73 USA, and private sector industries have started to ban or stop the usage of microbeads in personal
74 care products, limiting their input into the ocean (Rochman et al., 2015). Bans can be perceived as
75 extreme but several natural replacement solutions for microbeads are available to cosmetics
76 companies, allowing them to easily stop the contribution of microbeads to the plastic pollution
77 observed in the ocean.

78 Here, we aim to quantify the microbeads present in Hong Kong coastal waters and evaluate their
79 relative importance within the microplastic fraction. Using 147 samples from the southern region
80 of Hong Kong, we investigated the spatial and temporal variability of the presence of microbeads.

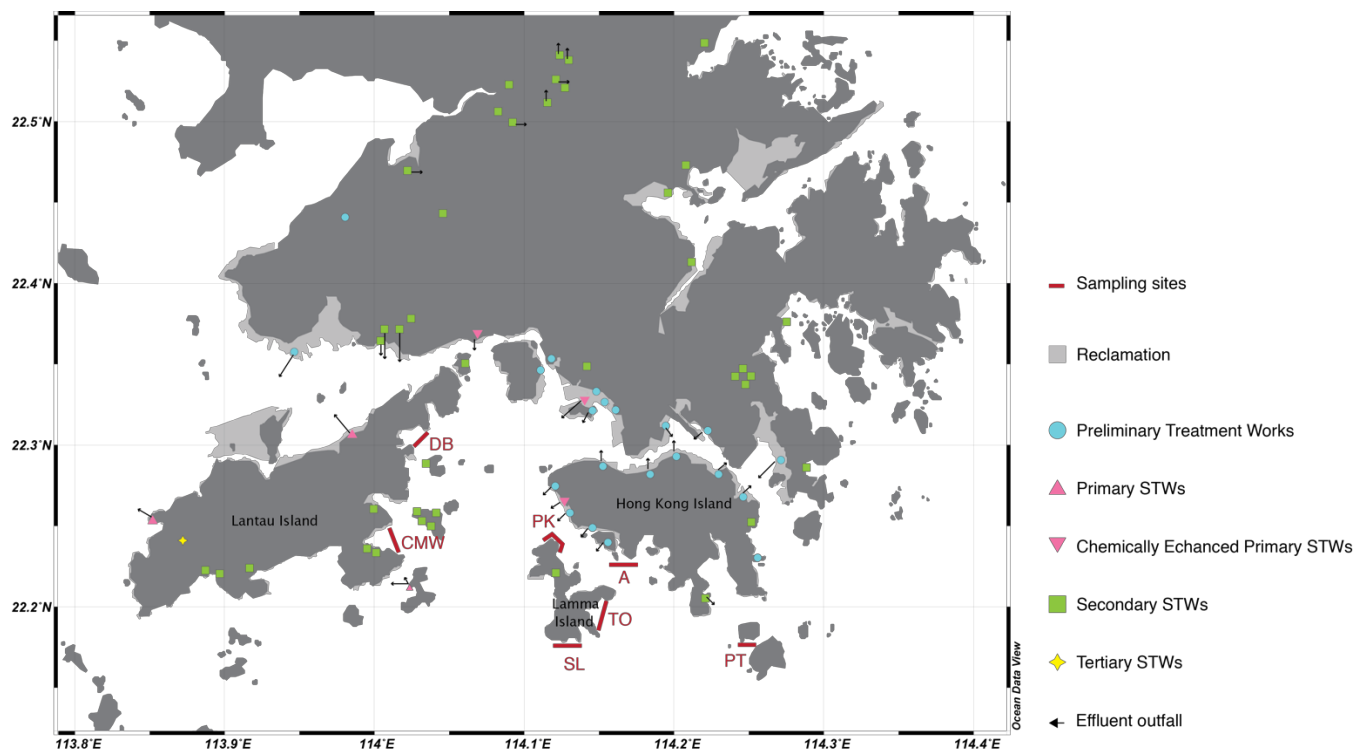
81 **2. Materials & Methods**

82 *2.1. Sample collection*

83 Hong Kong is surrounded by the South China Sea and situated at the estuary of the Pearl River.
84 Seven locations around the Hong Kong territories were chosen as sampling stations, namely
85 Aberdeen (n = 37), Chi Ma Wan (n = 27), Discovery Bay (n = 1), Pak Kok (n = 60), Po Toi (n =
86 13), South Lamma (n = 1), and Tung O (n = 8) (Fig. 1). From February 2016 to April 2017,
87 opportunistic surface water trawls were conducted at each site and a total of 147 samples were
88 gathered throughout the year. A 15-minute surface manta trawl, using a sampling net with opening
89 of 0.5 x 1m and a mesh size of 335 μ m, was performed at a constant speed of 2 knots to collect
90 samples. Samples were first stored in a glass bottle, transported to the laboratory at the University
91 of Hong Kong, and kept inside the refrigerator until they were processed. Abundance of
92 microbeads is presented by surface area estimated as follow:

93 $\text{Area (km}^2\text{)} = \text{speed of vessel (ms}^{-1}\text{)} \times \text{duration of the trawl (s)} \times \text{width of the net (m)} / 1000000$

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95

96 **Figure 1.** Map of Hong Kong with the seven sampling locations: Aberdeen (A), Chi Ma Wan
 97 (CMW), Discovery Bay (DB), Pak Kok (PK), Po Toi (PT), South Lamma (SL), and Tung O (TO).
 98 Sewage treatment works are located with information about their effluent outfall direction and the
 99 type of treatment (Drainage Services Department, 2015). Dark grey represents the natural
 100 coastline whereas the light grey represents coastline after land reclamation.

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102 2.2. Sample processing

103 Each sample was separated into two size fractions by sieving through a 5 mm and 0.3 mm stainless-
 104 steel mesh sieve. All materials remaining on the sieves were transferred to clean beakers and dried
 105 completely in an oven at 50°C for 24 hours. The smaller size fraction samples (0.3-5 mm)
 106 underwent wet peroxidation using 30% hydrogen peroxide and 0.05M aqueous iron (II) sulphate
 107 solution at 70°C to oxidize organic matters (Masura et al., 2015). If organic matter was still visible,
 108 samples were further subjected to alkaline digestion with 10% potassium hydroxide solution
 109 (Foekema et al., 2013). Sodium chloride was later added to the samples to increase the aqueous
 110 density and samples were settled in filter funnels overnight for density separation. Only the floating
 111 plastic debris were collected and screened through a 0.3 mm mesh sieve (Masura et al., 2015).

112 Samples were again thoroughly dried in an oven at 50°C for 24 hours and separated into two size
113 fractions of 1 to 5 mm and 0.3 to 1 mm by sieving through stainless-steel sieves with mesh sizes
114 of 1 mm and 0.3 mm. Plastic samples smaller than 1 mm were preserved in glass petri dishes for
115 visual sorting of microbeads under a stereomicroscope (40x; stemi 305; Carl Zeiss Microscopy
116 GmbH). Effort to limit sample contamination included ensuring that all materials used in sample
117 processing were thoroughly cleaned before usage and covered after each step. Microplastic
118 particles with spherical shapes within the 0.3-1 mm size fraction were considered microbeads and
119 as such were sorted and counted. Due to the impossibility to distinguish granular microbeads with
120 secondary microplastic produced by the degradation of bigger plastic pieces combined with the
121 limitation of the mesh size of the net used to collect the sample, microbeads abundance presented
122 below is underestimated as in the only two previous studies, yet it represents a primary estimate.

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124 *2.3. Fourier Transform Infrared (FTIR) Spectroscopy Verification*

125 Suspected microbeads (hereafter regarded as “microbeads”) were selected for FTIR (Spectrum
126 Two, Perkin Elmer) verification. A random selection of 95 microbeads, with at least one from each
127 sample where microbeads were present, were scanned with a spectral resolution of 4 cm⁻³ at a
128 range of 4000 cm⁻¹ to 400 cm⁻¹. Samples were identified with the aid of Perkin Elmer standard
129 polymer spectrums and compared to the composition of microbeads from the locally sold facial
130 scrubs tested in Cheung and Fok (2017).

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132 *2.4. Data analysis*

133 All statistics were performed in R Studio using R version 3.4.0 (R Core Team, 2017). In order to
134 test the potential spatial and temporal variability of microbead abundance in coastal surface waters,
135 a one-way ANOVA comparing the difference in microbead abundance between different sites,

136 months and seasons was performed. In addition, to evaluate the impact of weather parameters such
 137 as rainfall and wind magnitude, and sewage volume outflow and total suspended solids on the
 138 abundance of microbeads, we performed correlation analyses using the ‘ggpubr’ package
 139 (Kassambara, 2017). Spearman correlations were chosen due to a lack of normality in the data.
 140 Rain and wind data were obtained from the Hong Kong Observatory and sewage data was obtained
 141 from the Drainage Services Department of Hong Kong.

142

143 3. Results

144 More than half of the samples (60%) collected from seven sampling sites contained microbeads.
 145 A total of 1698 spherical microbeads, accounting for 3.59% of all microplastics, were found in the
 146 147 samples examined (Table 1). The abundance of microbeads found per sample ranged from 0
 147 to 380,129 pcs/km² with an average of 12,429 ± 3,929 pcs/km².

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149 Table 1: Mean abundance and percentage of microbeads in Hong Kong.

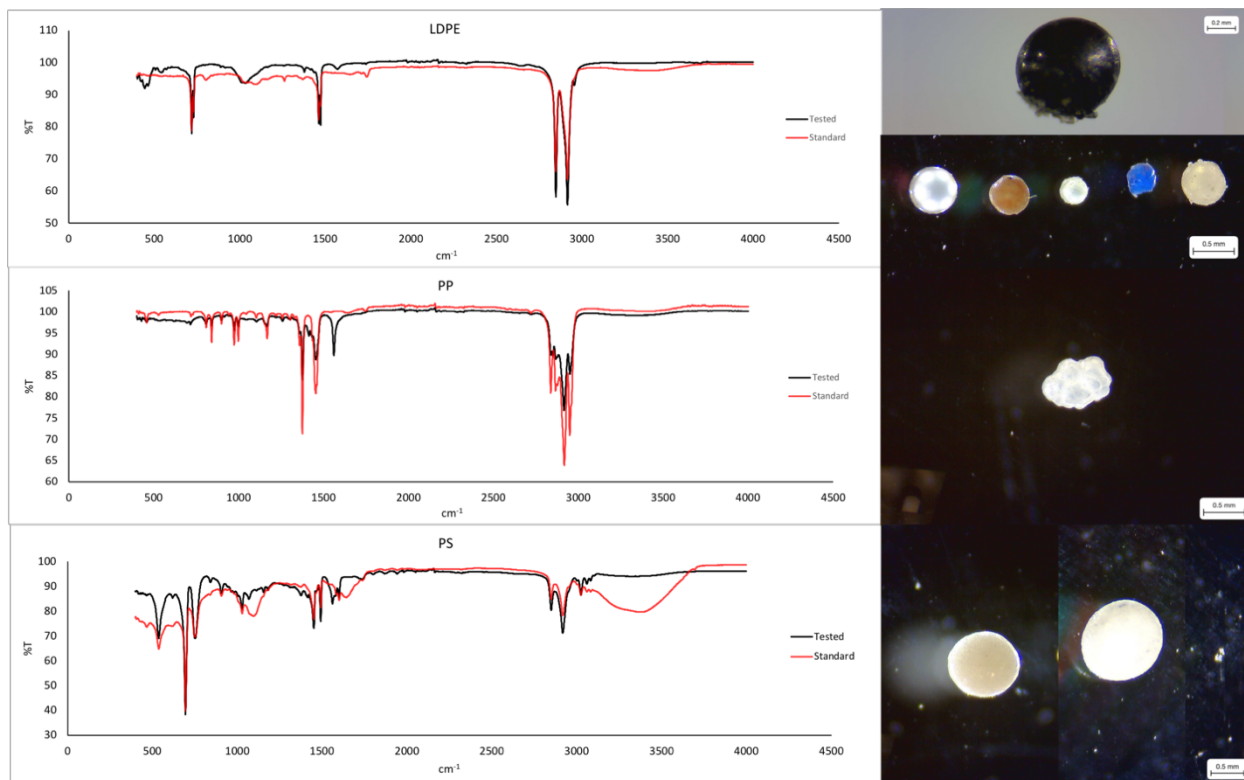
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Site		Size (mm)	Mean abundance (nps/km ²)	Standard deviation (nps/km ²)	Range (pcs/km ²)	no. of MBs	no. of MPs	%
Southern Hong Kong	(n = 147)	0.3-1	12429	47634	0 - 380129	1698	47241	3.59
Aberdeen	(n = 37)	0.3-1	3495	6310	0 - 29158	127	3518	3.61
Chi Ma Wan	(n = 27)	0.3-1	8759	19666	0 - 87473	219	3768	5.81
Discovery Bay	(n = 1)	0.3-1	15119	n.a.	15119	14	83	16.87
Pak Kok	(n = 60)	0.3-1	17298	56660	0 - 380129	960	30132	3.19
Po Toi	(n = 13)	0.3-1	2907	6821	0 - 24838	35	972	3.60
South Lamma	(n = 1)	0.3-1	0	n.a.	0	0	27	0.00
Tung O	(n = 8)	0.3-1	46301	127925	0 - 362851	343	8741	3.92

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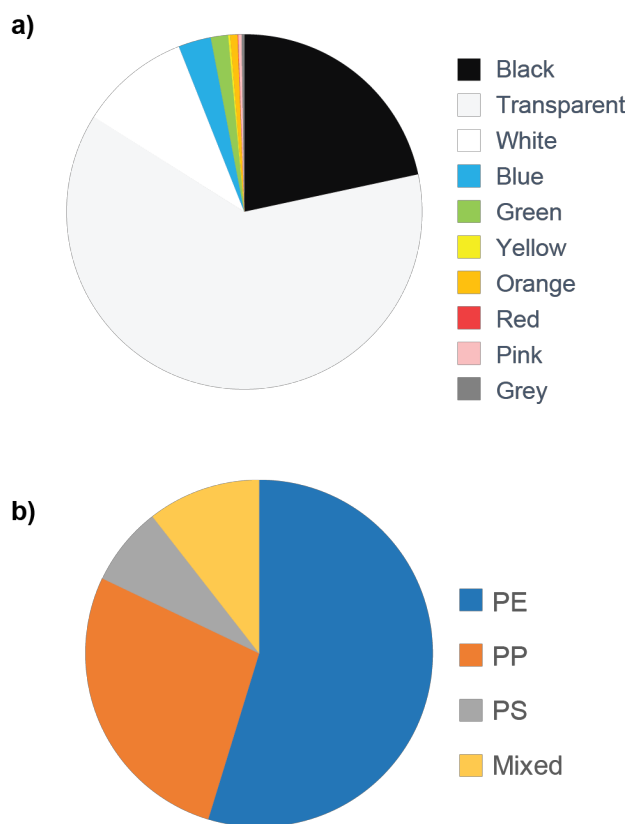
153 Microbeads found in the samples were mainly transparent (62%), black (22%), white (10%), blue
 154 (3%), and green (2%) (Figure 3). Most of them were spherical in shape (97%) and some were
 155 grape-like (3%). Out of the 95 microbeads analysed by FT-IR, 55% were identified as polyethylene
 156 (PE), 27% as polypropylene (PP), 7% as polystyrene (PS) and 11% as mixed plastic composition
 157 (Figure 2 and Figure 3), indicating a 100% rate of identification of plastic microbeads.



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160 **Fig. 2.** Photo of microbeads present in Hong Kong water with their composition spectrum obtained
 161 by FT-IR.

162 Samples from Tung O showed the highest mean concentration (46301 ± 45228 , $n = 8$), followed
 163 by Pak Kok (17298 ± 7315 , $n = 60$), Discovery Bay (15119 , $n = 1$), Chi Ma Wan (8759 ± 3785 , n
 164 $= 27$), Aberdeen (3495 ± 1037 , $n = 37$), Po Toi (2907 ± 1892 , $n = 13$), and finally South Lamma
 165 which was free of microbeads ($n=1$; Table 1). However no statistically significant difference was
 166 observed between the locations ($p = 0.16$). Interestingly, the percentage of microbeads per site had
 167 a different spatial variability. Samples from Discovery Bay showed the highest percentage (16.9%),
 168 followed Chi Ma Wan (5.8%), Tung O (3.9%), Aberdeen (3.6%), Po Toi (3.6%), Pak Kok (3.2%),
 169 and lastly South Lamma (0%; Figure 3) but this observation is driven by Discovery Bay where
 170 only one sample was collected and without this sample, the proportion of microbeads is quite
 171 similar between the sites.



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173 Fig. 3. Proportion colour composition from the 1698 microbeads collected within Hong Kong
 174 water (a) Proportion of the chemical composition of the 95 microbeads measured by FT-IR (b).

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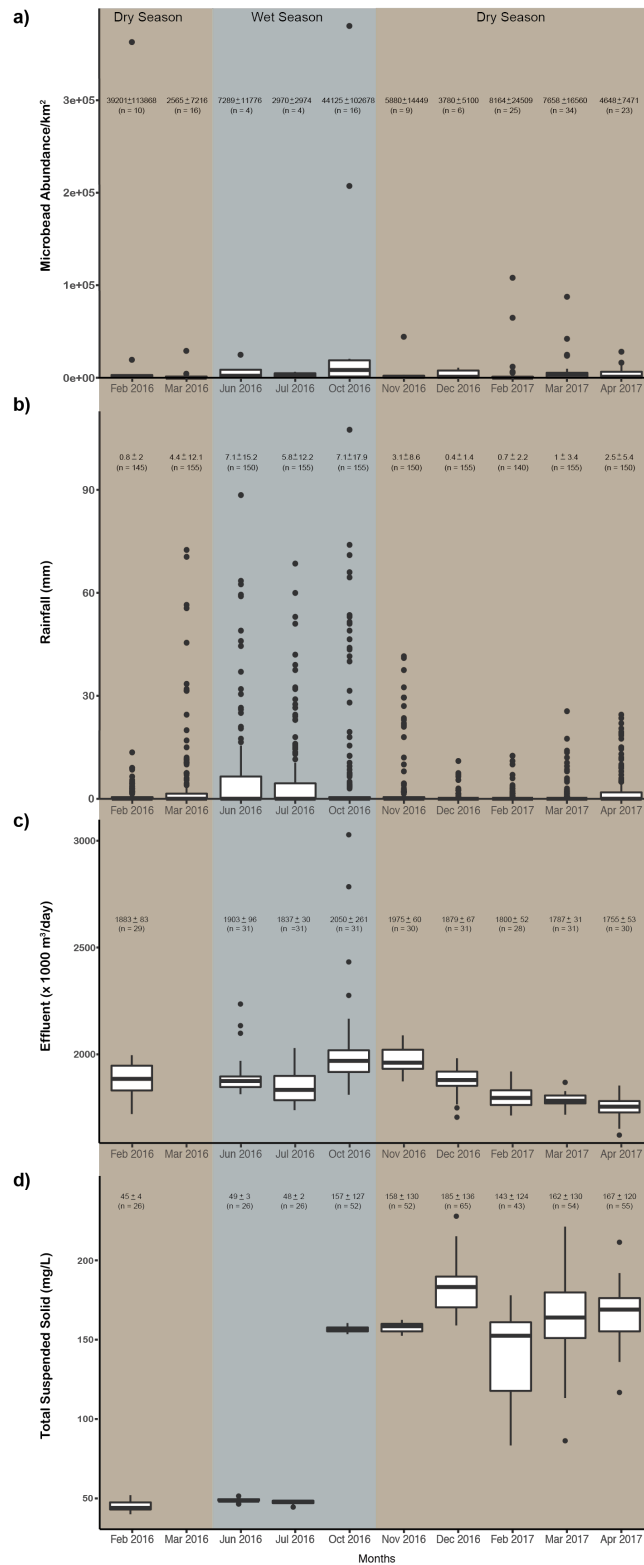
176 The average concentration of microbeads found in the sampling sites remained around $12429 \pm$
 177 47634 pieces/ km^2 throughout the year and no significant difference was observed between months.

178 However, three samples with very high abundance were considered as outliers. One sample was

179 taken on February 4th, 2016 and two were taken on October 20th, 2016. Both in absence and

180 presence of the outliers, a significant difference is observed between seasons (wet: November to

181 April vs dry: May to October), with the wet season having a higher abundance of microbeads.



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183 Fig. 4. Temporal variability in the abundance of microbeads in surface water from Hong Kong (a),
 184 in the rainfall (b), volume outflow of waste water treatment plant (c) and quantity of total
 185 suspended solid (d) between February 2016 to April 2017.

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4. Discussion

Hong Kong, with its high population density, is believed to be at high risk of pollution by plastic microbeads. We found a total of 1698 microbeads within the 75 days of sampling. Colours and composition of the microbeads found in our samples matched with the study of microbeads from local personal care and cosmetic products done by Cheung and Fok (2017) where transparent microbeads and LDPE/PE composition were dominant (Figure 3). Therefore, we are confident that all microbeads gathered originated from the personal care and cosmetic products sold in Hong Kong or in the Pearl River area. Considering various factors such as the percentage of the population using facial scrubs, density of microbeads in facial scrubs, escape rate of microbeads, etc., it was estimated that 342.2 billion microbeads were emitted into Hong Kong waters (Cheung and Fok, 2016). Here we collected a total of 1698 microbeads, suggesting that only a small quantity of microbeads is found in coastal waters. However, direct comparison between microbeads discharge and abundance in surface water have to be done carefully since several factors will affect the quantification of microbeads in the water. As mentioned in the introduction, microbeads correspond to synthetic exfoliators which are usually spherical, but irregularly shaped microbeads are also present with general size ranges between 0.1 mm and 1 mm (Gouin et al., 2015; Napper et al., 2015; Rochman et al., 2015). The size of microbeads present in locally sold cosmetic products was typically between 0.024 mm and 1 mm (Cheung and Fok, 2017). Therefore, counting only spherical microplastic from the 0.3 to 1 mm size fraction underestimates the microbeads present in the surface water; thus, our estimate should be taken as the minimum abundance of microbeads in marine waters. More precise estimations are technically limited by the complexity involved in differentiating between irregularly shaped microbeads and secondary microplastics,

210 and the common usage of 335 μ m mesh net. Consequently, 60% of the surface water samples, with
211 between 1 and 380129 pcs/km² microbeads, constitute a conservative evaluation of the microbead
212 contamination of Hong Kong waters. In addition, sewage treatment works have been shown to
213 effectively but not totally captured microbeads in the grease of the wastewater treatment, leading
214 to non-negligible release in the aquatic environment (Murphy et al., 2016). In Hong Kong, 69
215 sewage treatment works with different levels of technological advances are operated to handle the
216 domestic sewage from different districts (Figure 1; Drainage Services Department, 2017) but for
217 all of them the grease removal is undertaken, which results in a significant quantity of microbeads
218 retained. Yet, about 7% of the population (510,000 persons) in Hong Kong reside in rural areas
219 that are not connected to the public sewage facilities but rely on some rather simple wastewater
220 treatment systems such as septic-tank-and-soakaway and dry-weather-flow interceptors to handle
221 the untreated sewage (Audit Commission, 2016; Cheung and Fok, 2016). Here, despite the fact
222 that most of the waste waters treated will limit the input of microbeads in coastal water, the
223 technical limitation of identification of irregular microbeads, and the absence of collection of the
224 microbeads smaller than 0.3 mm, we show that microbeads form a non-negligible proportion of
225 the microplastics present in Hong Kong surface coastal waters. The close match between both the
226 colour and the composition of the microbeads found in coastal waters with those from cosmetics
227 and personal care products sold in Hong Kong confirm the sources of the microbeads present in
228 the surface coastal waters.

229 No significant spatial difference is observed in the abundance of microbeads in surface waters or
230 the number of samples containing microbeads between the different locations sampled, suggesting
231 that the distance or the type of the wastewater treatment plant (primary vs secondary) does not
232 influence the quantity of microbeads present. For example microbead abundance at Discovery Bay
233 or Chi Ma Wan, which are close to several secondary sewage treatment plants, is not lower than

234 in Aberdeen or Pak Kok, which are close to primary sewage treatment plants. Overall, our data
235 suggests the absence of a link between the proximity or type of sewage treatment plant and the
236 abundance of microbeads in the surrounding waters. Thus, it supports the idea that grease removal,
237 which is performed at all sewage water treatment plants in Hong Kong, is efficient at retaining
238 microbeads (Murphy et al., 2016).

239 No significant difference in the microbead abundance is observed between the different months
240 (Figure 4). We also observed no correlation between the abundance of microbeads and rainfall
241 ($p=1.00$), wind intensity ($p=0.18$), or quantity of total suspended solids ($p=0.23$), suggesting that
242 none of these parameters influence the quantity of microbeads present in surface coastal water.
243 However, a significant difference is observed between seasons, where the wet season (June to
244 October) has a significant higher abundance of microbeads when rainfalls are higher, more
245 variable and driven by extreme events. Also, a positive correlation between rainfall and sewage
246 outflow is observed, validating the link between sewage effluent and microbeads release in coastal
247 water. In addition, we observed three outliers with abundance at least an order of magnitude higher
248 than the average abundance observed during the sampling period. One of these sample was collect
249 on February 4th, 2016 and two were collected on October 20th, 2016. These two days are
250 characterized by rainfall and sewage outflow higher than the average for the sampling period and
251 higher than the average of their respective month, suggesting that these three parameters influence
252 the abundance of microbeads, especially during “extreme” events. For example, on October 20th,
253 the average rainfall of the previous two days was 71 mm and the average sewage outflow was
254 2905 ($\times 1000 \text{ m}^3/\text{day}$) in comparison to the October 2016 month average of 7 mm of rainfall and
255 2050 ($\times 1000 \text{ m}^3/\text{day}$). Previous studies have also highlighted the role of major rain events and
256 storm water increase the abundance of microplastic collected (Lattin et al., 2004; Moore et al.,
257 2002; Yonkos et al., 2014). A positive correlation is observed between the abundance of

258 microbeads and the volume of sewage outflow when outliers are included, validating the impact
259 of extreme event in the abundance of microbeads in surface waters. In comparison, wind has been
260 recognized to enhance microplastic transport to the deeper part of the ocean and therefore decrease
261 the abundance of microplastic in surface waters (Kukulka et al., 2012; Reisser et al., 2015). Here,
262 wind intensity was not different between these specific days and the average of the sampling period
263 or the average of the month. In addition, we observed a much higher variability in the data from
264 the dry months (November 2016 to April 2017), when sampling efforts were concentrated. This
265 suggests that the opportunistic sampling conducted had an influence and did not allow us to cover
266 the full range of weather conditions, especially rainfall. When looking at the rainfall data over the
267 sampling period we noticed that several days have more than 100 mm of rain, but none of these
268 days were sampled. Our data suggests a link between rainfall, and therefore sewage outflow, with
269 microbeads abundance in surface water. More samples are needed to confirm the direct link or the
270 existence of a threshold.

271

272 **5. Conclusion**

273 Microbeads, spherical plastic pieces from 0.3 to 1 mm in size, has been identified in 88 surface
274 water samples from Hong Kong coastal waters. Based on comparable shapes, colours and
275 compositions, the microbeads collected most likely originated from personal care and cosmetics
276 products sold in Hong Kong. The average abundance of microbeads in the surface water was
277 estimated at 12429 ± 47634 pieces/km². However, this estimate represents a minimum value of
278 the microbeads present since it does not take into account microbeads smaller than 0.3 mm and
279 microbeads with irregular shape. Irrespective of this limitation, our study confirms that microbeads
280 represent a non-negligible part of the microplastic pollution of Hong Kong coastal water.

281

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