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Litter behaviour on Mediterranean cobble beaches, SE Spain

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ABSTRACT

Despite the large research effort on reporting quantities of coastal litter, the dynamics of this litter is not yet sufficiently understood. Litter inputs in five cobble beaches located in the Mediterranean (Spain) were studied over three months during winter by biweekly litter tagging. Plastic represented the dominant material that reached the beaches (77%). In remote and narrow beaches, storms constituted the main driver in litter dynamics, favouring the accumulation of floating items such as plastic bottles and wood fragments as well as the largest but contrasting effects, increasing litter inputs and outputs from the beach, respectively. In rural beaches, beach users, mainly fisher people, but also tourists, contributed to a notable input of litter to the beach. Burial and exhumation of litter were reported as common occurring processes. Better management actions are required to improve beach environmental quality.

1. Introduction

Litter is a major and constantly growing global environmental issue (Tudor and Williams, 2004; Rech et al., 2014). Several million tonnes (Mt) of litter currently enter the ocean every year from different sources (Jambeck et al., 2015) and is a cause of great concern to beach managers (Tudor and Williams, 2008). Marine litter or debris is been defined as any persistent, manufactured or processed solid material discarded, disposed of or abandoned in the marine and coastal environment (UNEP, 2009, 13). The vast majority of litter derives from land-based sources such as villages, towns, landfills, private households, etc. and, because of improper disposal, passes through rivers and streams into the ocean (Golik and Gertner, 1992; Sheavly and Register, 2007; Rech et al., 2014). Litter can also be directly discarded in coastal environments by beach users (Nachite et al., 2019), as well as having a marine-based origin, from recreational and fishing boats, aquaculture and shipping in general (Earll et al., 1999; Coe and Rogers, 2012). Litter constitutes a serious hazard (Williams et al., 2013; Campbell et al., 2019) as has been shown by several previous studies during decades (Wallace, 1985; Azzarello and Van Vleet, 1987; Pemberton et al., 1992; Laist, 1997; Duncan et al., 2017). Birds, fish, invertebrates, mammals, reptiles and amphibians have been found entangled in marine debris (Ocean Conservancy,

2010). Lost or abandoned fishing gear and plastic bags are the greatest impact items to marine wildlife (Ocean Conservancy, 2016). Other dangerous items to wildlife include consumer products, such as food wrappers, straws, caps/lids, and smoking-related items (Ocean Conservancy, 2016).

The shoreline is widely used in litter monitoring because of its connection to land-based sources and accessibility, therefore litter accumulates there (GESAMP, 2019). Beach litter along the world's coasts is not only an important topic in marine pollution (Bergmann et al., 2015; Williams and Rangel-Buitrago, 2019), but also affects economies such as tourism (Botero et al., 2013a; Krelling et al., 2017; Houston, 2018). The presence of beach litter is one of the "Big Five" reasons to determine beach choice (Williams and Micallef, 2009; McKenna et al., 2011; Botero et al., 2013b) and beaches can be classified according to their anthropogenic environment, into different beach typologies such as resort, urban, village, rural and remote (Williams and Micallef, 2009; Williams, 2011). Litter categories and abundance can depend on the number of visitors (Nachite et al., 2019; Asensio-Montesinos et al., 2019b). In remote beaches located on islands, marine-based sources usually are the most important, e.g. fishing (Kaviarasan et al., 2020) and often observed is beach litter from foreign countries (Williams and Simmons, 1997a; Kei, 2005; Topçu et al., 2013).

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Plastic is the most common litter material found on the coast (Kusui and Noda, 2003; Bergmann et al., 2015; Simeonova and Chuturkova, 2019). It is estimated that 70% of marine litter remains on the ocean bottoms, while the other 30% is retained equally between the water column and beaches (UNEP, 2005). Global plastic production reached 368 Mt. in 2019 (Plastics Europe, 2020) and considering the amount of litter that has been yearly discarded into the environment over decades, it is expected that large quantities of litter will end up on the coast in the next few years. Furthermore, the coast acts as a great "barrier" between terrestrial and aquatic ecosystems. On urbanised coasts, beaches are generally cleaned during the bathing season by the various administrations (Williams, 2011), but in rural areas, cleaning activities in beaches are not generally performed and litter may remain for several months (Anfuso et al., 2020; Ecoplaya Alicante, 2020; Maiklem, 2020).

These isolated areas are suitable for accurate estimates of beach litter abundance (Lavers and Bond, 2017; Dunlop et al., 2020; Ryan, 2020), however, a limited number of researchers have attempted to quantify short-term dynamics of beach litter (Williams and Tudor, 2001). Different methodologies have been applied, e.g. daily collections of marine litter (Eriksson et al., 2013; Chitaka and von Blottnitz, 2019), beach litter monitoring using webcams (Kako et al., 2010), periodical litter assessments (Prevenios et al., 2018) and litter mark-recapture/tagging (Williams and Tudor, 2001). Among these methods, litter tagging provides accurate information on the litter dynamics by periodical quantification of the inputs (Garrity and Levings, 1993; Bowman et al., 1998; Williams and Tudor, 2001; Kataoka et al., 2013).

The aim of this paper was to study the dynamics of marine litter in rural and remote cobble beaches located on the southeastern coast of Spain. A periodical litter mark-recapture/tagging following Williams and Tudor (2001), was used to quantify litter inputs and correlate them with environmental parameters.

2. Materials and methods

2.1. Physical background

The study area is located in the southeast of the Iberian Peninsula, in the municipalities of "Campello", "La Vila Joiosa" and "Calp", in the province of Alicante. Five beaches were selected for this study, all located between the External Zone of the Betic *Cordillera* and the coastal waters of the Western Mediterranean Sea (Fig. 1). The northern part of the Alicante coast is characterized by several cliffed sectors (Yébenes et al., 2002), as a result of intensive faulting and folding. The proximity of mountain ranges to the sea has completely conditioned the coastal profile and gives the beaches great scenic beauty (Alfaro et al., 2008; Asensio-Montesinos et al., 2019a).

The involved beaches are mainly composed of cobble and boulder sediments, with sizes ranging from -6 to -9 Phi (64–512 mm, according to Blott and Pye, 2012) and show reflective morphodynamic states (Masselink and Short, 1993). They are high energy beaches with little sign of anthropogenic activities/structures (Fig. 2) that could affect the natural processes and morphology, which are only influenced by waves and tide (Wright et al., 1987). Tides in the area show a micro-tidal range (<2 m, according to Davies, 1964). In microtidal beaches, litter accumulations are usually concentrated at the high tide level and backshore area (Kei, 2005; Asensio-Montesinos et al., 2020a), which has been considered an efficient trap for litter (Bowman et al., 1998). All beaches are characterized by accumulations of Posidonia oceanica "banquettes" that reach >1 m in height and cover several square metres of beach surface. When the leaves of this Mediterranean plant accumulate on the beach, they protect against erosion processes because they contribute to stabilization of coastal sediments (Belmonte et al., 2001). These accumulations come from submerged seagrass meadows that in the past occupied large areas (Sánchez-Lizaso et al., 1994).

According to the beach typology classification of Williams and Micallef (2009), beaches no. 1 and 2 are essentially rural with few users daily observed (0-10) during the study period. Rural beaches are located outside the urban/village environment and are not accessible by

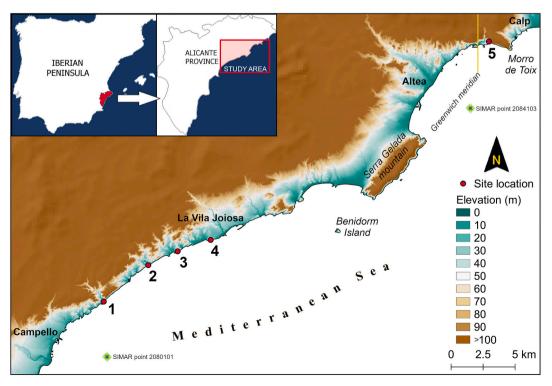


Fig. 1. Location of study area with the position of the beaches sampled.

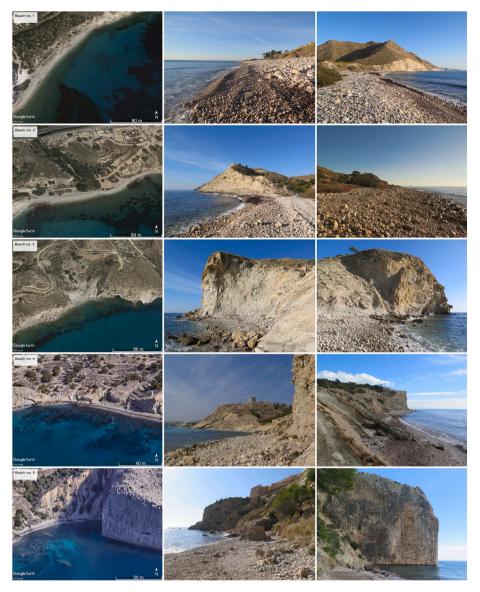


Fig. 2. In rows, the five beaches investigated. The first column shows the beaches in 3D with a bird's eye view. The second and third columns respectively show a southward and eastward view of each beach.

public transport (Williams and Micallef, 2009). While beaches no. 3, 4 and 5 are remote (i.e. remote Mediterranean beaches are defined by difficulty of access, mainly by foot after a walk of 300 m or more) with very few users observed (0–5, see Fig. 2), i.e. all studied beaches are characterized by a low abundance of users. Users observed on the beaches were usually fishermen that spend several hours on the beach and in some cases, people hiking and, at the end of the study period, by tourists. Rural and remote areas investigated in this paper are valued by beach users for their natural qualities. Additional data for each beach are presented in Table 1.

2.2. Field surveys

Data were collected in seven field campaigns carried out from December 2020 to March 2021, at five beaches ranging in length between 50 and 250 m, all of them consisting of cobbles backed by cliffs (Figs. 1 and 2). The distribution of surveyed coastal sites was selected according to their geomorphological characteristics and other logistical reasons, such as proximity. Satellite images (from Google Earth), bibliography (DPA, 2010; Iváñez-Rugero, 2020) and previous field studies (Asensio-Montesinos et al., 2019a, 2019b) were used to select the least

visited and most remote beaches in the province of Alicante. All are unpopular and/or have difficult access compared to common Mediterranean beaches. In line with previous litter studies in remote cobble beaches (Anfuso et al., 2020), the entire beach surface was covered and assessed by two researchers that moved along cross-shore parallel transects spaced 2 m apart. The beach surface was examined from the strandline to the backshore area because the pattern of litter distribution on cobble beaches is often irregular (Williams and Tudor, 2001). The backshore included a cliff base and ancient beach ridges, composed of cobbles, pebbles and P. oceanica "banquettes" (Fig. 2). The backshore is the most diversified zone of a cobble beach because different sediment grain sizes coexist and a great variety of litter items was observed there. The five beaches were visited between 8 a.m. and 3 p.m. Litter was classified according to a Masterlist of beach litter with 183 litter categories that take into consideration the type of material and size following "The Environment Agency and The National Aquatic Litter Group" (UK) (EA/NALG, 2000), "The United Nations Environment Programme" (UNEP, 2009), "OSPAR Commission" (OSPAR Commission, 2010), and finally, "The National Oceanic and Atmospheric Administration" (Opfer et al., 2012). A few new categories were also added such as sanitary masks, bracelets, bins, fishing rod pieces, hair

Table 1Location and main characteristics of investigated beaches.

Beach no. and name	1. Lloma de Reixes	2. Carritxar	3. Xarco–Caleta ^a	4. Malladeta	5. Racó del Corb	
Municipality	Campello	La Vila Joiosa	La Vila Joiosa	La Vila Joiosa	Calp	
Geographical coordinates	38°27′27.40″N	38°28′56.53″N	38°29′29.42″N	38°29′55.88″N	38°37′56.77″N	
	0°20′31.92″O	0°18′4.94″O	0°16′28.92″O	0°14′41.35″O	0° 0′36.96″E	
Length assessed (m)	336	258	50	200	113	
Width max. assessed (m)	20	12	12	7	18	
Width min. assessed (m)	11	8	7	0	8	
Area assessed (m ²)	4621	2451	672	1280	1469	
Orientation	NNE-SSW	ENE-WSW	NE-SW	ESE-WNW	NE-SW	
Facing to	ESE	SSE	SE	SSW	SE	
Geomorphology	Open beach	Open beach	Pocket beach	Open beach	Pocket beach	
Surrounding geological	Marls, calcarenites and	Marls, calcarenites and	Marls, calcarenites and	Conglomerates, sandstones,	Marls, sandstones and	
materials	limestones	limestones	limestones	gravels, sands, silts and clays	conglomerates	
Cobbles origin	Alluvial deposits and	Alluvial deposits and	Cliff slides	Cliff slides	Cliff slides	
	cliff slides	cliff slides				
Beach typology	Rural	Rural	Remote	Remote	Remote	
Type of access and difficulty	By foot: normal	By foot: difficult	By foot: very difficult	By foot: difficult	By foot: very difficult	
	By car: difficult	By car: very difficult	By car: not possible	By car: not possible	By car: not possible	
Distance to paved road	300 m	154 m	1117 m	310 m	305 m	
Slope of access to the beach	Low (<5%)	Low (<5%)	Medium (5-25%)	Medium (5-25%)	High (>25%)	
Parking and no. of spaces	Yes (<10)	Yes (<10)	No	No	No	
Level of occupation	Low	Low	Very low	Very Low	Low	
Recreational activities	Angling, camping and	Angling, camping and	Enjoy the coastal	Enjoy the scenery and picnicking	Enjoy the scenery and	
observed on the beach	picnicking	picnicking	scenery		picnicking	
Services	Bins	Bins	No	No	No	

^a This beach has no official name and it is located between the beaches of 'El Xarco' and 'La Caleta'.

bands, hats and caps, etc. "Pottery & ceramics" category was not counted/marked because very little debris was observed, and such items were part of the environment and usually remain there. In addition, "Pottery and ceramics" on beaches could be of historical interest (Rouillard et al., 2014; Maiklem, 2020). Data were represented by litter abundance and average litter accumulation rates on the surveyed beaches for a 15 day time-span (i.e. the average time interval between following surveys) per 100 m beach to make data obtained easily comparable with other studies. The accumulation process is not usually a linear one because it records great temporal variability, therefore daily accumulation rates presented within this study are only indicative.

By cleaning a specific area and using marking methods, it is possible to record litter input rates, accumulation times and movement patterns (Williams and Simmons, 1997b). The methodology used in this paper was applied 20 years ago on a UK cobble beach by Williams and Tudor (2001). This was probably the first study to highlight the importance of burial and exhumation of litter on cobble beaches and demonstrated the rapid recolonization of litter on an unfrequented pocket beach (Williams and Tudor, 2001). Within the framework of this research, on 1st December 2020, the five investigated beaches were cleaned of all visible surface litter. Then, every two weeks during three months (13th December 2020-17th March 2021) a total of seven surveys were carried out. During each survey, all beach litter was counted and marked in situ with a specific waterproof permanent colour on each survey to differentiate newly arrived items ("fresh" litter) from the ones observed in previous surveys (Williams and Tudor, 2001). The colours used were: red (13th December); blue (28th December); green (12th January); orange (28th January); yellow (12th February); brown (27th February); and purple (17th March). Only items of litter observed on the beach surface were counted and those items that were floating, or outside the study area were not included in these surveys. Photographic evidence of many tagged items was obtained to verify their reappearance and condition in subsequent surveys. From the results obtained in this type of study, the probability of double counting as a consequence of litter disintegration is minimal (Williams and Tudor, 2001). Waterproof permanent paint may rub off on some specific items over the weeks, e.g. on rusty metal items, but this issue was solved by re-marking these items with their corresponding colour during each survey.

For relating litter inputs to environmental factors, oceanographic data series during the study period were obtained from the "Puertos del

Estado" website (www.puertos.es, accessed on 19th April 2021). Datasets of wind (average wind speed and direction) and wave parameters (significant wave height, peak period and prevailing wave direction) are obtained from the closest SIMAR ("SImulación MARina") points to the studied beaches, i.e. the beaches nos. 1, 2, 3 and 4 are closer to the SIMAR point 2080101 and beach no. 5 to the SIMAR point 2084103 (Figs. 1 and 3). The SIMAR virtual database is obtained through numerical wave modelling from wind time series by means of an energy balance equation that has been validated by numerous studies and used in practical applications along the Spanish coast (Tomás et al., 2004). In addition, the maximum and minimum daily ambient temperature data were obtained from "Agencia Estatal de Meteorología" (datosclima.es, accessed on 20th April 2021).

2.3. Statistical analysis and data processing

To compare litter composition among beaches and surveys, a nonparametric multi-dimensional scaling (nMDS) was performed. Objects are represented as points, such that the distance between points matches the observed dissimilarities (Groenen and van de Velden, 2005). Additionally, the cluster technique was carried out as a complementary classification technique to the nMDS. For this purpose, the dataset was normalised, i.e. the number of items per beach was converted into items/m². Then, according to the characteristics of the dataset and the differences between the minimum and maximum values, it was decided to perform a fourth root transformation. Analyses were made based on the Bray-Curtis dissimilarity index (Bray and Curtis, 1957) as the abundance of the litter items is zero-inflated. Two independent analyses were performed; one for the fresh litter data and another one for the total litter data. In the nMDS plots, to identify litter categories that showed the highest weight in the ordination of the beaches during the different surveys, the litter categories with a Pearson correlation above 0.5 were represented as vectors from a central point in a twodimensional space. The orientation of the vectors with respect to the distribution of the beaches is related to the composition of the most important items present, which in turn is related to the use made of each beach. The analyses were conducted using PRIMER V.6 + PERMANOVA (Plymouth Marine Laboratory, UK). All statistical tests were conducted with a significance level of $\alpha=\text{0.05}\text{.}$

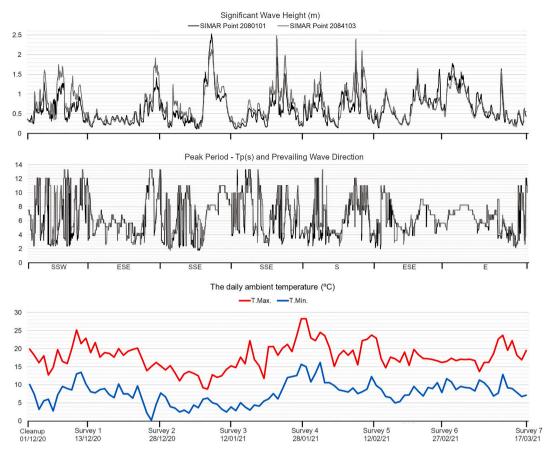


Fig. 3. Environmental parameters: Significant wave height (m), peak period or Tp (s), prevailing wave direction and daily ambient temperature. Sources: "Puertos del Estado" (www.puertos.es) and "Estaciones meteorológicas de AEMET" (datosclima.es).

3. Results

3.1. Beach litter dynamics

During the clean-ups at the beginning of this study (1st December 2020), from one to five large bags of litter were removed per beach. Due to uncertainty in previous accumulation times, the collected beach litter was not counted. Two weeks later, 150 fresh items were recorded at the first survey (13th December 2020), rising to 722 by the final survey (17th March 2021, Table S1). A total of 130 litter categories were recorded that principally included plastic, metal, cloth, paper, rubber, wood, glass, and other materials.

An amount of 3418 litter items were counted on the five beaches investigated during the entire study period, 2410 of them were constituted by fresh items (Table S1). The rest (1008) were remaining items that were counted more than once (Table S1). Fig. 4 shows all beach litter accumulation over the study period. Results showed that inputs during the studied period were quite different within surveys, but after some time the accumulated litter tended to become similar among beaches (Fig. 4). Litter counted at a specific survey, tended to disappear over time, but despite this a few items generally remained. Beaches no. 1 and 2 recorded the highest amount of litter (Table S1, Fig. 4); however, they are also the largest beaches (Table 1, Fig. 2). Considering the seven surveys carried out at each beach, the average number of litter items per 100 m beach ranged from 66, 42, 56, 29 to 63 items from beach no.1 to beach no.5, respectively. The number of litter items/m² was very low and similar for all beaches (Fig. S1), all falling in the category "Very clean" according to the Clean Coastal Index (CCI; Alkalay et al., 2007). In the study area there is little connectivity between beaches, but it is important to give an idea about this process, as it can be of great interest for researchers and local administrators.

Accumulation rates ranged from 0.4 to 6.9 items·day⁻¹·100 m⁻¹ (Table S2). There was a notable increase in the number and accumulation rates of items in the third survey (Fig. 4 and Tables S1 and S2) due to a strong marine storm that occurred during the days preceding the survey (Fig. 3). On beach no. 4 this trend was not observed because of the reduced width of the dry beach (in some cases less than 1 m) so waves during the storm were expected to reach the cliff base resulting in the formation of reflected waves that favoured erosive processes that transported litter items offshore (Fig. 2). Generally, from the first to the last survey, was observed an increasing trend in the number of new items appearing on beaches, particularly in the last two field visits (surveys 6 and 7, Figs. 4 and S1; from more detail of these items see Fig. 5). Concerning beach users' abundance, a clear increase of paper fragments related to users was observed during the study period: only one fresh paper fragment was recorded at the first survey and 30 in the final (Table 2). Different specific examples of beach litter dynamics are displayed in Fig. S3.

Multivariate analysis showed that beaches no. 1 and 2 presented similar litter composition, while the rest presented different trends (Figs. 6 and 7). In beach no. 3, the remains of *P. oceanica*, as well as litter items, have also accumulated in less quantity than in nearby beaches (Table S1). Results of surveys carried out in the beach no. 4 also presented a different trend from the one observed at other beaches. Beach no. 5 shares geomorphological characteristics with beach no. 3 but it receives as many users as beaches 1 and 2 (Table 1). With regard to the seven surveys, they tend to become similar over time (Fig. 6 and Table S4).

3.2. Beach litter composition and typology

Fresh beach litter was composed of plastic (77.1%), followed by

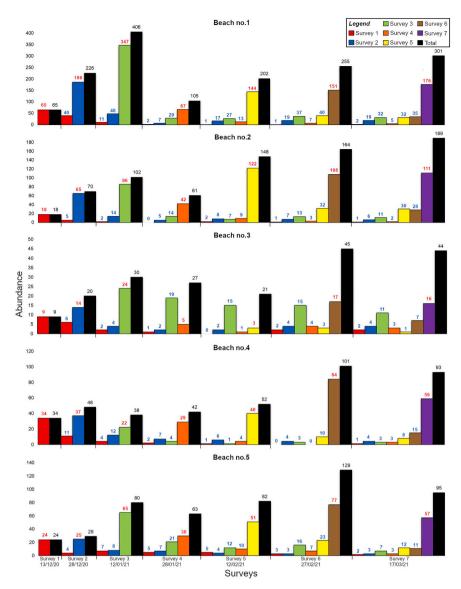


Fig. 4. Litter abundance in the five studied beaches (no. items per beach). Numbers in red represent the accumulated fresh litter content among consecutive surveys. Numbers in blue represent the remaining litter. The colours of each bar belongs to the litter that was counted in the same survey (see the legend and X-axis): in survey no. 1, new litter items were marked in red, in survey no. 2 new litter items were marked in blue (and litter marked in red was also counted and considered as remaining litter); in survey no. 3, new litter items were marked in green and litter marked in red and blue, i.e. the remaining litter, was also counted, and so on. The black bars represent the total amount of litter, i.e. remaining + fresh litter, at each survey. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

metal (7.3%), cloth (5%), paper & cardboard (3.9%), rubber (2.3%), wood (1.5%), other materials (1.3%), glass (1.2%), and organic (0.4%). The most abundant litter items (>2%) in the study area have been identified for the entire period (Table 2) and included hard plastic pieces (2.5–50 cm, 25.1%), foamed plastic pieces (2.5–50 cm, 12%), film plastic pieces (2.5–50 cm, 7.1%), cigarettes, butts & filters (5.4%), caps/lids (3.4%), Drinks (bottles, containers and drums) < 2 L (3%), paper fragments (2.5–50 cm, 2.8%), cloth pieces (2.5–50 cm, 2.6%), fishing line (angling, 2.6%), and metal fragments (2.5–50 cm, 2.4%).

The bulk of the litter observed were plastic pieces, due to the high energy hydrodynamic conditions associated with cobble beaches and the high buoyancy of plastics. Table 2 shows the number of the most abundant fresh litter items during the study period, e.g. plastic pieces (Fig. 5d). Most litter categories recorded few items. For example, some rare or uncommon residues observed in rural and remote beaches were cosmetics, sandals, toys (Fig. 5e), tyres, sponge scourers, sunglasses, rags, nail clippers, among others. Cigarette butts, caps/lids (Fig. 5f), drinks, paper fragments and others items (Fig. 5g–k), which are abundant in the Mediterranean (Nachite et al., 2019; Vlachogianni, 2019) and in other places (Ocean Conservancy, 2016), were present in this study in only small proportions (<6%). This is substantiated by some litter categories, being positively correlated to beach users, e.g. cloth pieces (*CL15*), paper fragments (*PP11*) and cigarette butts (*PL24*, Fig. 6).

Other categories such as hard and film plastic pieces (PL63, PL66) were also positively correlated. Rural beaches no. 1 and 2 are similar in terms of litter composition related to beach use, accessibility, management, etc. (Table 1, Figs. 6 and 7). Several rubber fragments (RB10) and octopus elastic straps (RB13) were found during the first surveys on beach no. 3 (Fig. 6). The latest surveys of remote beaches no. 4 and 5 also resemble the surveys carried out on rural beaches no. 1 and 2 (Fig. 6). The similarity of values obtained between consecutive surveys increased in the latest surveys (Table S4). Cluster analysis shows that, regarding fresh litter, the most comparable surveys take place in beach no. 1, e.g. surveys 2, 3, 5 and 6, Fig. 7 the most accessible and visited beach of this study (Table 1). Other groupings can be seen in the surveys of beaches no. 4 and 5 (Fig. 7). However, it is again observed that beach no. 3 is the most different from the rest because it is the least visited, the most remote and the smallest beach (Table 1 and Fig. 7). The survey 3 of beach no. 4 is very different from the rest, as mentioned above, due to the erosive processes produced by the storm that removed a large part of beach litter (Fig. 7). Regarding total litter, the results of Cluster analysis show obvious groupings where the composition of litter from the same beach is similar throughout the study (see groupings by colour in Fig. 7).

Some of the litter items observed during the study were potentially dangerous to users and animals that live or frequent beach. For example, fishing-related debris (Fig. 5l) such as lures, hooks (Fig. 5l, m) and

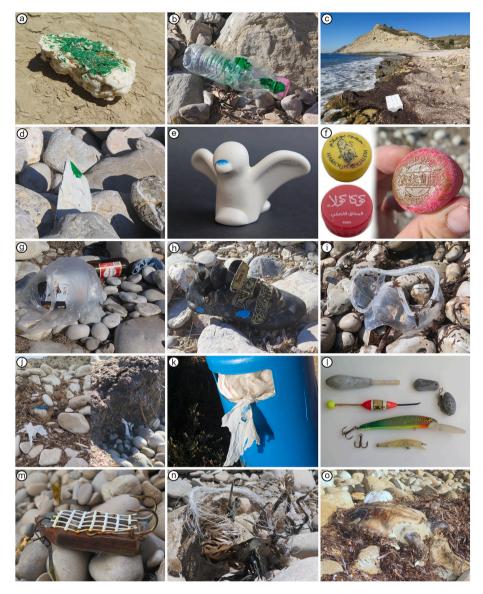


Fig. 5. Examples of litter items: a) piece of expanded polystyrene foam; b) plastic bottle; c) foamed plastic box; d) hard plastic piece; e) Playmobil Bird White, Part Number: 6102170; f) caps/lids; g) litter accumulation; h) climbing shoe; i) dive mask; j) buried litter; k) litter bin; l) fishing related items; m) fishing lure with hooks; n) fishing line; o) marine turtle.

Table 2Number of most abundant fresh litter (i.e. top ten litter items) during the study period recorded at each survey.

Beach litter categories	S1	S2	S3	S4	S5	S6	S7
Hard plastic pieces 2.5-50 cm	41	67	136	49	119	120	72
Foamed plastic pieces 2.5-50	13	86	101	13	20	23	34
cm							
Film plastic pieces 2.5-50 cm	13	17	22	18	30	39	33
Cigarettes, butts & filters	0	15	39	1	14	26	34
Caps/lids	3	13	25	6	7	17	12
Drinks (bottles, containers and	2	3	41	2	11	8	6
drums) < 2 L							
Paper fragments 2.5-50 cm	1	4	4	4	10	15	30
Cloth pieces 2.5-50 cm	2	7	10	7	14	17	6
Fishing line (angling)	6	11	9	1	6	15	15
Metal fragments 2.5-50 cm	7	6	8	8	8	9	11
Total	88	229	395	109	239	289	253

fishing lines (Fig. 5n) can cause lacerations, entanglements and other serious problems to humans and animals. There was little fishing-related debris, mostly linked to shore angling. Fishing-related debris were found on all studied beaches, being more abundant in beach no. 1 and 2, specially surveys 6 and 7 (Table S5). Other hazardous items are broken glass, nails, screws, razors, knives, wires, some metal and plastic fragments (Fig. 5d), and other cutting items (all of them observed in this study), which can cause foot lacerations and other potential dangers (Whiting, 1998; Williams et al., 2013). Other harmful litter items detected during the study period were fiberglass fragments, sanitary masks and manufactured/processed wood fragments. Biohazard items were also recorded and related to the presence of sewage-related debris that accounted for 1.2% of all fresh litter observed, some examples were sanitary towels, tampon applicators, cotton bud sticks, eye drops and condoms. Further, a few dead animals were observed, sometimes so degraded that identification was difficult: a sea turtle (probably Eretmochelys imbricata, Fig. 50), a fish (Trachinus draco), two seagulls (probably Larus michahellis), a cuttlefish (Sepia officinalis), worms for fishing (Perinereis aibuhitensis) and a small goat, the head of which was found in another part of the beach.

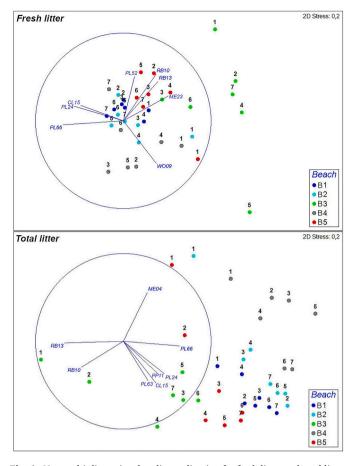


Fig. 6. Non multi-dimensional scaling ordination for fresh litter and total litter based on the litter categories abundance. The colours (blue, cyan, green, grey and red) correspond to each of the five beaches, while the labels 1 to 7 correspond to the surveys. Vector labels refer to litter categories that showed a Pearson correlation above 0.5: *CL15* (cloth pieces), *MEO4* (drink cans), *ME23* (metal fragments), *PP11* (paper fragments), *PL24* (cigarette butts), *PL52* (fiberglass fragments), *PL63* (hard plastic pieces), *PL66* (film plastic pieces), *RB10* (rubber fragments), *RB13* (octopus elastic straps), *WO09* (matches & fireworks). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

4. Discussion

4.1. Litter composition and sources

Beach litter composition helps to identify the litter source (EA/NALG, 2000; Bergmann et al., 2015; Ryan, 2020) as well as the typology and use of the beach (Ocean Conservancy, 2010; Asensio-Montesinos et al., 2019b). Results obtained in this study concerning beach litter composition differed from other studies that have reported higher percentages of plastic (82.6–92.2%) and, generally, lower percentages of other materials in different seasons (Kusui and Noda, 2003; Topçu et al., 2013; Asensio-Montesinos et al., 2019b, 2020a, 2020b; Nachite et al., 2019; Vlachogianni, 2019). In some of these studies, "Paper & cardboard" category showed higher proportions than this study because they were counted on beaches in months where users were more abundant. Reliable observations have also been reported in other countries, such as Bulgaria, Cuba and Morocco (Botero et al., 2017; Nachite et al., 2019; Simeonova and Chuturkova, 2019).

The bulk of the litter observed in this study consisted of plastic pieces that have different composition and their origin is often unknown (Bergmann et al., 2015). A very similar result accounted for the highest percentage of plastic pieces (2.5–50 cm, i.e. 26%) were recently reported by Vlachogianni (2019) on 23 Mediterranean beaches of coastal

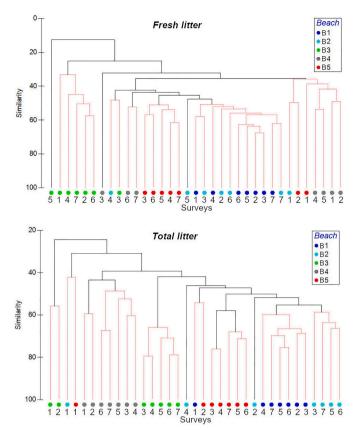


Fig. 7. Cluster analysis for fresh and total litter based on the litter categories abundance.

and marine protected areas, including beaches of different typologies (e. g. resort, remote, rural, village and urban) defined by Williams and Micallef (2009). Asensio-Montesinos et al. (2020b) estimated that most of the hard plastic pieces on Cádiz province beaches (Andalusia, Spain) are composed of polyethylene (PE, 35%), polypropylene (PP, 31%), polystyrene (PS, 13%), polyethylene terephthalate (PET, 4%), polyvinyl chloride (PVC, 3%) and other types of plastics (14%). In Europe, these types of plastics generally coincide with the highest plastics demand by resin type in 2019 (Plastics Europe, 2020). In this study, many of these hard plastic pieces (approx. 35%) comprised transparent fragments of beverage bottles, usually made of PET. This proportion was estimated in some of the surveys that recorded the greatest litter abundance, and bottle fragments accounted for between 23% and 72% of all hard plastic pieces. Foamed plastic pieces come from marine and terrestrial inputs. These fragment types observed in remote beaches are mainly composed of expanded polystyrene foam (PS) and polyurethane foam (PU, Anfuso et al., 2020), which have high buoyancy due to their composition (Mark, 1998). This fragmentation of foamed plastic occurs due to their exposure to sunlight and abrasion processes (Biber et al., 2019; Turner, 2020). In some beaches located in SW Spain, film plastic pieces are composed of PP (59%), PE (27%), PET (3%), PVC (3%) and other materials (8%, Asensio-Montesinos et al., 2020b).

Concerning beach litter origin, litter comes from both local and foreign sources (Wade et al., 1991; Munari et al., 2016; Ryan, 2020) and allochthonous litter can severely endanger marine ecosystems through the biological invasion of non-native attached biota (Rech et al., 2016). In this study a large amount of litter categories had a local origin and reflect a contamination linked to beach users and near-residents that could have deposited litter directly on the beach surface. Approximately, 30% of the sampled litter was directly related to beach users (fishermen, beachgoers, sportsmen, etc.). However, other items with positive buoyancy had an unknown origin. Examples of litter directly

related to users found in this investigation were hair bands and clips, bracelets, sun glasses, sandals, pieces of a beach umbrella, cigarette packets, cutlery, glass fragments, some fishing related items, foil wrappers, food wrappers, etc. as well as recent accumulations of food-related litter (Fig. 5g). Litter items related to sports have a local origin and were probably abandoned or lost in the vicinity of the beaches. Some examples are the tennis balls observed on beach no. 4, probably coming from one of the numerous tennis courts near this beach. Other items related to climbing were found on beach no. 5; e.g. a steel carabiner and a climbing shoe (Fig. 5h), where climbing is regularly practised on the beach cliff (Fig. 2). Snorkel tubes and diving masks used to appear arbitrarily on all beaches (except beach no. 3). They were probably lost by beachgoers during the summer and have been washed ashore by the sea during the winter period (Fig. 5i). Other items related to sport were observed only once, for example a swim cap and a digital wristwatch. Further items may have been left for many years at sea or buried in marine sediment until they washed up on beaches. Some of these particular examples are a model of rubber sandal very common in the 1980s and 1990s (Hobeky) and a small plastic toy from 1999 (Fig. 5e) which had bryozoans attached.

Local users were also responsible for the 6.2% of fresh litter consisting of fishing-related debris, ranging from 4.6 to 10.2% among beaches (Table S5). Although these percentages are low, they are higher than those reported in other beach assessments in the Mediterranean and Atlantic littoral of Spain (1.2 to 3.1%; Asensio-Montesinos et al., 2019b, 2020a), which could be because those investigated in previous works included village and urban areas where fishing does not usually occur. The present study agrees with previous ones that beach typology is a key factor that determines beach litter composition, an observation also reported by other studies (Rangel-Buitrago et al., 2018; Nachite et al., 2019). However, the percentages found in this study coincide within the range of other reported works for the Mediterranean Sea where fishing-related items overall comprise less than 10% of total litter items found (Vlachogianni et al., 2018, 2020). In contrast, in three Galician beaches (NW Spain), litter related to fishing and aquaculture represented an average value of 14, 23 and 38% (Gago et al., 2014) because Galicia province has the largest fleet and the highest aquaculture production in Spain. During the surveys it was observed that fishermen generated not only fishing-related debris, but also general litter, i. e. beer cans, cigarette butts, cable ties, adhesive tape roll and fragments, lanterns, foil wrappers, tools, clothes, etc. Wildlife can be injured or killed by discarded fishing lines, hooks and nets (Ocean Conservancy, 2010, 2016; Hardesty et al., 2015) while fishing weights (made of lead, Fig. 5l) are highly toxic if ingested (Haig et al., 2014). Humans and their litter may have been responsible for the death of animals, as has already been demonstrated on numerous occasions in the case of turtles and seabirds (Ocean Conservancy, 2010; Duncan et al., 2017). Dead animals on the beach can also be a biological hazard to other species because they can be a source of food, with lethal consequences (National Geographic, 2019). Microbiological hazards are related to the presence of dead animals and other litter items that can present high amounts of Escherichia coli (Philipp, 1993). The bacterium E. coli may be associated with dog faeces, observed at the end of the study (surveys 6 and 7; beaches no. 2, 4 and 5) when an increase of beach users occurred. A litter increase at the end of the study was probably due to the increase of users (often observed in small groups) and beach recreational activities linked to the improvement in weather conditions (Fig. 3). Additionally, it could also be because an increase of items probably due to fragmentation of plastic pieces mainly made of foam (Fig. 5a).

Nearby coastal settlements are responsible for the sewage-related debris, i.e. sanitary towels (essentially), tampon applicators, cotton bud sticks, eye drops and condoms that accounted for 1.2% of all fresh litter observed. They represent most offensive litter contamination type (Tudor and Williams, 2008) and their presence has been documented on many coasts around the world (Tudor et al., 2002; Munari et al., 2016; Botero et al., 2017; Ocean Conservancy, 2010). Recently, on the Alicante

coast, sewage-related debris reached average values of 6.5% (spring) and 2.5% (summer) for all beach litter (Asensio-Montesinos et al., 2019b). Differences compared to this study are mainly due to seasonality, which is related to the number of resident people near the coast, and the number and type of sites studied (e.g. the five beaches investigated were far from rivers or streams and villages or cities).

Finally, in some cases, identification of labels on the litter (Kei, 2005; Smith et al., 2018; Asensio-Montesinos et al., 2020b; Ryan, 2020) showed a foreign origin. Several plastic cap/lids were found with different inscriptions in Arabic and Chinese languages (Fig. 5f). In the first case, Arabic bottle caps may come from ships (Ryan, 2020) or could also be transported by currents from their country of origin, e.g. Morocco, Algeria or Tunisia. In the second case, Chinese bottle caps may come from Chinese ships (Ryan, 2020), which cross the Mediterranean Sea through important routes such as the Suez Canal and the Strait of Gibraltar. The same model of Chinese bottle cap (Fig. 5f) was found on the beaches of New South Wales, Australia (Smith et al., 2018) and recently on the west coast of Svalbard (Falk-Andersson et al., 2021).

4.2. Distribution patterns of beach litter

Accumulation rates recorded within this study were lower than reported in Corfu Island (Ionian Sea, Central Mediterranean), Cape Town (South Africa), and Cousine Island (Seychelles, Prevenios et al., 2018; Chitaka and von Blottnitz, 2019; Dunlop et al., 2020) and were, together with litter patterns, clearly influenced by both anthropogenic and environmental factors.

On the one hand, the type of users determines litter categories and the number of users determines litter abundance, but management (through clean-up operations) can help to reduce the amount of litter. On the other hand, environmental factors have a major impact in litter distribution. The capacity of litter to recolonize natural beaches within a short time period was also reported by Williams and Tudor (2001). For example, the high buoyancy of plastics and sea-storms, e.g. before survey 3, favoured their transport across the sea surface to the coast (Table 2). Wind is another relevant factor in litter distribution. Eriksson et al. (2013) also demonstrated that the combined effects of environmental factors such as maximum tide height and wind speed and direction are related to variation in daily litter accumulation rates. During the study period, the highest wind speed values were recorded in the first month and mostly blew from inland (max. wind speed: 19.3 m/s from WNW direction), resulting in a considerable number of foamed plastic fragments observed in survey 2 (Table 2). According to the Beaufort Scale, wind at these velocities "breaks twigs off trees, generally impedes progress and the wave crests begin to break into spindrift".

Beaches no. 1 and 2 were managed during the study period by the municipal services. Cleaning actions were carried out and consisted of emptying of litter bins and occasionally in removal by hand of the largest visible items. This would explain the disappearance of gross litter and other items of a considerable size, such as bottles or fish boxes (Fig. 5b, c). For example, at survey 3 on beach no. 1, of the 36 plastic bottles counted, only 4 of them were observed again at the following survey. In addition, as observed in one case, isolated beach users performed individual removal of beach litter of some of the bulkiest items, such as large bottles of water. The abundance of some litter categories changed over the study period due to storm occurrence and the abundance of beach users. As an example, concerning storms, 41 new bottles appeared after the storm that took place in early January (survey 3, Table 2).

When a certain amount of litter appeared on the beach, over time this same amount tends to decrease and generally small items from previous surveys tend to remain (Fig. 4, Table S3). Different dynamics of the recorded litter was observed:

- (i) there were litter items that once observed remained on the beach during the whole study period;
- (ii) few litter items were observed on the beach only once;

- (iii) others items appeared and remained for several weeks and then disappeared;
- (iv) some litter observed on the beach on a certain day disappeared weeks later and was seen again some time later (some litter presented this trend more than once, i.e. October 2021 some marked litter such as small plastic fragments and processed wood still remained on the studied beaches);
- (v) few floating litter observed from the beach, disappeared and were seen again weeks later on another beach.

In the first case/example, items that remained on the beach were small, heavy and had negative buoyancy (e.g. metal and glass fragments). In the second and third cases, beach litter disappeared because it was removed by people or buried into the sediment (Fig. 5j), transported away by wind or waves, or simply was not seen by observers [e.g. during survey 4 (28th January 2021) on beach no. 1, a film plastic piece marked in blue colour was observed inside a litter bin (that came from survey 2 carried out on 28th December 2020), Fig. 5k]. The fourth case can be related to the last three circumstances mentioned above. The fifth case demonstrates that litter may be exchanged between nearby or adjacent beaches. In survey 3 of beaches no. 1 and 2, two wood fragments were marked (in green) on each beach, two weeks later (survey 4), no wood fragments were observed on beach no. 1 and the three wood fragments appeared on beach no. 2 (Table S3). This is supported by coastal orientation, NE-SW aligned, and oceanographic data. The prevailing approaching wave direction before the third and fourth surveys was from the SSE, confirming the possible movement of items from beach no. 1 to beach no. 2 (Fig. 3).

Litter buried on a beach has often been underestimated. Some studies have shown that a large proportion of beach litter remains buried in the sediment before being exhumed (Williams and Tudor, 2001; Lavers and Bond, 2017). Litter fragments frequently become buried and are later exhumed on beaches, with smaller fragments tending to remain in the sediments for longer periods (Kusui and Noda, 2003). A lot of beach surveys have ignored the litter buried in the sediments because appropriate survey methods have been lacking, and very few studies have sampled buried litter (Kusui and Noda, 2003). Some researchers have applied methods for surveying litter buried in the sand (Ogi and Fukumoto, 2000; Kusui and Noda, 2003; Ryan et al., 2009; Lavers and Bond, 2017) but on cobble beaches it is more difficult to carry out this type of sampling. A substantial proportion of beach litter may be buried: e.g. in Henderson Island, 68% of beach debris (<10 cm), was buried in the sediment (Lavers and Bond, 2017), while in the beaches of Senegal the density of buried litter was 25 times higher than at the beach surface (Tavares et al., 2020). A notable decrease in the number of litter items observed between successive surveys, e.g. from S6 to S7, Fig. 4 could be largely due to the burial of many of these items within the cobbles, but other factors such as personal recollections by beach visitors or environmental factors may also play an important role. The present study has demonstrated the burial and exhumation of some specific litter items such as hard plastic pieces, foamed plastic pieces, plastic and metal caps/lids, film plastic pieces, metal fragments, rubber fragments, etc. However, during surveys, it is difficult to know exactly which items first seen were buried. Some labelled litter were buried and then were exhumed, these form part of the remaining litter, as can be seen in Fig. 4 and Table S1.

Changes in litter quantity between surveys five to six, illustrate the process of exhumation.

- Beach 1, green coated litter increases. Mainly hard plastic pieces, film plastic pieces, and metal/plastic bottle caps.
- Beach 2, green coated litter increases. E.g. foamed plastic pieces, caps/lids, foil wrappers, string and cord.
- Beach 3, red, blue and orange coated litter increases. E.g. fragments (foamed plastic, rubber, film plastic), tubes, cables and sanitary towels.

- Beach 4 orange coated litter disappears in survey 6 but reappears in survey 7. E.g. fragments (metal, foamed plastic and rubber).
- Beach 5, green coated litter increases. E.g. fragments (metal, paper, film and foamed plastic) and shoe insoles.

Beach accumulation surveys can be used to estimate litter flows into the marine environment, but litter inputs from the sea can be influenced by numerous factors, including weather conditions, ocean currents and coastal geomorphology (Anfuso et al., 2011; Chitaka and von Blottnitz, 2019). The complex coastal morphology of the study area (e.g. the presence of headlands and extended rock shore platforms) determines the sheltering of some beaches to wind, waves action and litter inputs not associated with longshore currents. For example, beaches no. 3 and 5 are pocket beaches (Fig. 2) only directly affected by wind/wave approaching from a limited range of directions because they are protected by headlands and submerged rock shore platforms. Such structures divide the coast into sectors or basic units also named morphological "cells" (Anfuso et al., 2011). Cells are usually limited by natural features (e.g., a promontory) or human structures (e.g., a port) and may include coastal stretches belonging to different municipalities, provinces or countries. The interaction between wave propagation patterns and limits of a cell (typology, dimensions, etc.) determines the distribution of erosion/accretion areas within a cell (Anfuso et al., 2011, 2014). Similarly, longshore distribution patterns of beach litter are also controlled by coastal compartmentalisation. Two of the main morphological cells observed in this study (Alicante Cape-South of Serra Gelada and Northeast of Serra Gelada-Toix Cape, Fig. 1) are separated by a natural limit (i.e. Serra Gelada mountain, Benidorm) that gives rise to cliffs more than 400 m high covering 6 km of coast between the bays of Benidorm and Altea (Yébenes et al., 2002). Several sub-cells are created within these two cells due to the emplacement of anthropogenic structures (such as harbours and breakwaters), which form artificial fixed limits that allow only a one-way transport of floating litter (Fig. 1). In the first cell (from Alicante Cape to Serra Gelada mountain), litter transport between beach no. 1 and no. 2 occurs because there are no natural or anthropogenic structures that prevent it (Fig. 1). Among beaches no. 2, 3 and 4 it is more difficult for litter items to move because there are small headlands that act as retaining barriers (Figs. 1 and 2). In the second cell (from Serra Gelada mountain to Toix Cape, Fig. 1), beach no. 5 is very far away from the rest. It is very difficult for litter to be transported from one of the first four beaches to the latter (or vice versa) because beach no. 5 is separated from the rest by the Serra Gelada sea cliffs and, secondarily, by anthropogenic structures such as harbours and breakwaters located in "La Vila Joiosa", "Benidorm" and "Altea" municipalities.

4.3. Litter influences: human and environmental factors

Previous observations regarding the abundance and composition of litter and relationships with human and environmental factors were supported by statistical analyses using nMDS and cluster, which are suitable to compare litter composition in different areas (Tudor et al., 2002; Rech et al., 2014; Rangel-Buitrago et al., 2018; Asensio-Montesinos et al., 2019b).

Beach litter composition was similar for beaches no. 1 and 2 due to the similarities between these two beaches. Both are rural beaches where the same beach activities are carried out (angling, picnicking, etc.). Beach no. 3 is different from the rest since it receives few visitors because it is not very popular and presents a difficult access. Moreover, being a small pocket beach bordered by headlands, it receives little inputs from longshore transport. Beach no. 4 has very distinctive geomorphological and usage characteristics that differ from the rest. Beach no. 5 is the most similar, from a morphological point of view, to beach no. 3, but records more visitors, so the nMDS analysis groups beach no. 5 close to beaches no. 1 and 2. Regarding surveys, initially they are less similar as there is less litter on the beach, but as new inputs

linked to storm events or beach use take place, the similarities increase between samples. Generally, the last surveys (6 and 7) tend to be similar in all beaches, but in beach no. 3, due to the increase of specific litter related to beach users, e.g. cigarette butts due to the improvement of weather conditions. This observation can be explained by the fact that beach no. 3 is by far the most remote of all sampled beaches and thus, no or little increment of beach users was to be expected.

A strong storm had important implications for litter composition in some beaches. This is observed in survey 3 of beach no. 4, where erosional processes at the cliff base linked to the storm could have removed a large part of the beach litter, resulting in the lowest accumulation rate reported in this beach during the study. Also, survey 3 of beach no. 3 recorded the arrival of fresh litter, which was quite different from other surveys and constituted the largest accumulation rate reported in this beach during the study. A storm might have the capacity to increase the input of litter to the beach simulating the cases of other beaches that have important inputs from other sources, such as beach users. The effect of beach storms seems to be more important in narrow beaches and ones that are more remote.

The analysis of the structure of the total litter showed connections by beach and survey. Trends were similar to those observed in the analyses for fresh litter but in this case, groupings by beach are clearer and there is less overlap between points (Fig. 6). Survey 1 is the most different due to the short-period accumulation time. As litter accumulates through time the composition of beaches no. 1, 2, 4 and 5 tend to become similar and related to beach users' presence and high litter abundance. Generally, remote beaches had more unique characteristics that differentiate them from the rest except when there is an increase in the number of beach users. Surveys that most differ from the rest were usually the first ones (surveys 1 and 2, Fig. 7) because the beaches were cleaned at the beginning of the study and they had little accumulated litter. Both analyses (nMDS and Cluster) coincide in the grouping patterns according to litter composition and abundance. Therefore, both methods have been demonstrated to be useful in perceiving differences and similarities between surveys and beaches. This is very useful to relate litter presence with factors such as users, beach use, occurrence of storms, etc.

5. Conclusions

This study shows the behaviour of litter on five cobble beaches and its relationship with human and environmental parameters. The understanding of beach litter inputs and dynamics on this type of natural beaches, which are increasingly valued and visited by users, will allow better coastal management measures to improve their current environmental status. This research has shown that on the beaches investigated, when beach users are not very abundant, litter accumulates in small amounts (2 items day $^{-1}$ ·100 m $^{-1}$). Observations carried out in this paper show that much of the recorded litter consisted of buried items that later were exhumed. Litter burial and exhumation processes on cobble beaches are recurrent, so litter should be removed as soon as possible to avoid burial and subsequent exhumation. Energetic storms bring fresh litter from the sea generally contributing to the increase of specific litter categories, such as plastic bottles. People are also responsible for the appearance of other specific litter items on remote and rural beaches. While some people such as fishermen and other marine users discard litter of different materials and sizes (fishing line, cigarette butts, small and large plastic bags, single-use plastics, hairbands, pens, lanterns, drinks, food containers, etc.), other people such as cleaners and volunteers, collect it. However, during the period of this research, an imbalance was observed in the area investigated and litter amounts increased because i) clean-up operations are not frequently carried out; ii) usually litter items removed included only those observed on the beach surface and iii) recollected litter during the clean-up operations includes only the largest and most visible items (e.g., bottles, large bags, etc.). Therefore, smaller items are expected to remain buried or embedded within cobbles for long periods while larger items are

expected to have a limited life because they are likely to be removed by waves and currents and during beach clean-up operations. The type of litter material is also an influencing factor, as heavier and more durable items such as glass or metal will tend to stay on the beach longer than lighter and less durable items such as plastic. Even when appropriate clean-up activities are carried out, some litter items composed of different sizes and materials remain for several weeks, even months, especially small items.

The best way to achieve a clean cobble beach is to prevent litter from reaching it. Litter will continue to appear on the beach surface because of exhumation and supplies from the sea (or land) to the shore, so cleanup actions are essential. Indeed, more specific measures can be taken to prevent direct dumping of litter by beach users, and at the same time, to avoid subsequent burial of beach litter. Regarding management actions, it is possible to make some recommendations to reduce beach pollution, according to results obtained in this work. Occasionally, more precise clean-up actions could be carried out, in which smaller litter is collected and not ignored. Education is the key and the promotion of a responsible behaviour can help to reduce the amount of litter ending up in the coastal environment. Sound environmental management of recreational activities should be mandatory and in remote and rural beaches: angling, picnicking and camping should be controlled. In many cases, these activities are practised by beach users in the same day. It essentially requires better enforcement of existing laws to prevent pollution and inform and punish polluters. Beach fines could be the answer to change beach users' erroneous behaviours. More restrictive laws and environmental campaigns focused on fishermen and other kinds of users are also required.

Lastly, in future similar studies, the sampling period between successive surveys (two weeks) could be reduced since the wave climate changes very rapidly and beach users' abundance varies greatly, strongly conditioning the quantity and typology of daily accumulated litter.

CRediT authorship contribution statement

Conceptualization, A.-T.W.; Data curation and Investigation, F.A.-M.; Funding acquisition and Resources, C.S.-L.; Methodology and Software, F.A.-M. and C.S.-L.; Supervision and Validation, G.A. and A.-T.W.; Visualization, G.A. Writing - original draft, F.A.-M. and G.A.; Writing - review & editing, A.-T.W and C.S.-L.

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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2021.113106.

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