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# On mechanical fragmentation of single-use plastics in the sea swash zone with different types of bottom sediments: Insights from laboratory experiments



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#### ABSTRACT

Mechanical fragmentation of four commonly used plastics, from 2-cm squares or cubes to microplastics (MPs, < 5 mm), is experimentally investigated using a rotating laboratory mixer mimicking the sea swash zone with natural beach sediments (large and small pebbles, granules, sand). Macro-samples were prepared from brittle not-buoyant PS (disposable plates), flexible thin film of LDPE (garbage bags), highly buoyant foamed PS (building insulation sheets), and hard buoyant PP (single-use beverage cups). With a great variety of behaviors of plastics while mixing, coarser sediments (pebbles) have higher fragmentation efficiency than sands (measured as the mass of generated MPs), disregarding sinking/floating or mechanical properties of the samples. It is confirmed that, under swash-like mixing with coarse sediments, the MPs tend to burry below the sediment surface. The obtained relationship between the mass of MPs and the number of items is similar to that for MPs floating at the ocean surface.

#### 1. Introduction

Longevity of plastics under environmental conditions makes these synthetic materials both irreplaceable in almost all human activities and also potentially dangerous as persistent environmental contaminants. Relatively small plastic particles (microplastics (MPs), < 5 mm (GESAMP, 2015)) can be misidentified as food by marine biota and climb up the food chains (e.g., Farrell and Nelson, 2013; Bouwmeester et al., 2015; van Franeker and Law, 2015; Chagnon et al., 2018; Guo and Wang, 2019b). Uncertainty in possible effects of MPs on marine life, fast increase in plastic contamination of aquatic systems, impossibility to remove MPs from the environment - all these aspects raise serious concern among researchers from many environmental disciplines. Marine MPs attract special attention since they can be transported by currents over huge distances, and are found nowadays in very remote areas from pole to pole (e.g., Lusher et al., 2015; Munari et al., 2017; Waller et al., 2017). Application of ocean circulation models to problems of transport and fate of plastic litter and MPs is still limited (van Sebille et al., 2019) since these pollutants change their physical properties with time spent in the marine environment (Chubarenko et al., 2018). Besides, proper parameterizations of relevant processes for numerical models are still missing.

MP particles found in the environment are conventionally divided into those of primary and secondary origin (e.g., Bergmann et al., 2015). Primary MPs are directly produced as micro-particles, e.g., preproduction pellets or plastic spherules in cosmetic scrubs and tooth-paste. Since 1970th, when pollution of oceanic beaches of New Zealand by pre-production pellets was reported for the first time (Gregory, 1977), their leakage in the environment has become much stricter controlled, and this has already shown the effect (van Franeker and Law, 2015). The same is true for the use of primary MPs in cosmetics: in many countries it is completely banned today (The Guardian, 2018). Thus, both sources of primary MPs are more or less known, and its release into the environment can be controlled and (potentially) reduced.

Secondary MPs originate from fragmentation of larger plastic objects. It is commonly accepted that MP fragments make up the majority of MPs in the ocean (Cózar et al., 2014; GESAMP, 2015). Fragmentation, as a process of disintegration of an object, can be facilitated by degradation of plastic material. Plastics can undergo a number of different types of material degradation in the environment, such as bio- or UV-degradation, thermal, *mechanical*, thermo-oxidative destruction,

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and hydrolysis (Browne et al., 2007; Andrady, 2011; Anderson et al., 2016). Among them, oxygen and sunlight (UV-radiation) are considered the most important factors of abiotic material degradation in natural environments (Andrady, 1998; Gewert et al., 2015). In particular, in the terrestrial environment, light-induced oxidation is shown to be orders of magnitude faster compared to other types of degradation processes (Wang et al., 2016). Fragmentation due to material degradation typically produces small particles, e.g., from 30 nm to  $60\,\mu m$  in a UV-degradation study by Lambert and Wagner (2016). Fragmentation to pieces of mm-size range, often observed in the ocean (e.g., Cózar et al., 2014), requires certain external mechanical forcing (exerted by waves, wind-induced mixing, etc.). This mechanical fragmentation is accelerated by prior material degradation. In water, plastics are effectively protected from solar UV radiation, winds, high temperatures and frost, etc., so material degradation becomes much slower (Gregory and Andrady, 2003; Barnes and Milner, 2005; Arthur et al., 2008; Corcoran et al., 2009; Ryan et al., 2009; Andrady, 2011; Anderson et al., 2016). Thus, understanding of mechanical fragmentation processes becomes important. In particular, mechanical fragmentation in the swash zone along the shores, under the breaking waves and moving sediments, seems to be of primary importance for the generation of secondary MPs.

Abrasive wear and fragmentation of materials is commonly investigated on the base of laboratory experiments and tests, because the results depend on too many factors, from the very abrasive and abrasion materials to external factors like temperature, pressure, mechanical energy, etc. (e.g., see (Lancaster, 1969) for polymers). Wear of market plastics under the conditions they were developed for is investigated during both pre- and post-production stages. For the case of wear and fragmentation under "unexpected" sea-swash conditions, it is crucial to reproduce the real process (contacting materials, temperature, etc.) as close as possible. Following this idea, a set of experiments was performed, covering 4 common plastics and 4 natural sediment types, at room temperature, atmospheric pressure, and very moderate "wave energy", so that only some pebbles or upper 1–1.5 cm of sand were able to move.

The process of mechanical fragmentation of plastic objects in the marine environment has only been addressed in a few studies up to now, and mainly in the context of facilitation of degradation due to exposure to UV radiation. Song et al. (2017) conducted experiments under accelerated weathering conditions in the laboratory, with UVexposure for up to 12 months followed by mechanical abrasion with sand in rotating glass containers for 2 months. The authors used samples of low-density polyethylene, polypropylene, and expanded polystyrene and showed that the UV-exposure followed by mechanical abrasion is an efficient way of MPs generation (µm size range). Similarly, Kalogerakis et al. (2017) examined the fragmentation of highdensity polyethylene films from single-use supermarket plastic bags under laboratory-simulated onshore and nearshore conditions. During the 6 months of the experiment, they monitored the tensile strength and molecular weight of the samples, analyzed FTIR-spectra, measured weight loss, and applied image processing of photographs of the plastic strips before and after mild mechanical stress. Samples placed in seawater proved to be resistant to fragmentation compared to those on sand over the 6-month period of the weathering experiment. The results confirm that weathering of plastics in the terrestrial environment proceeds significantly faster onshore compared to weathering of plastics floating in seawater (Kalogerakis et al., 2017). Complementary information comes from observations by (Corcoran et al., 2009): examination of weathering of plastics from Kauai beaches (Hawaii) showed that granular oxidation textures were concentrated along mechanically weakened fractures and along the margins of the plastic particles, i.e., that mechanical degradation, in its turn, facilitates oxidation processes. Overall, very little published information can be found concerning mechanical degradation and fragmentation of plastics in the coastal zone (Cooper, 2012). Importantly, the disintegrating effect of UV-exposure, oxidation, microbial degradation (e.g., see Tsiota et al., 2018) becomes significant at the time scales of more than several months, while direct mechanical action of breaking waves and abrasion by sediments is able to fragment plastic objects within days or even hours (Jungnickel et al., 2016; Efimova et al., 2018).

The goal of this paper is to disclose most general and characteristic features of the process and the results of mechanical fragmentation of macroplastics, made of commonly used new polymers, subject to swash-zone-like mixing with different kinds of natural beach sediments.

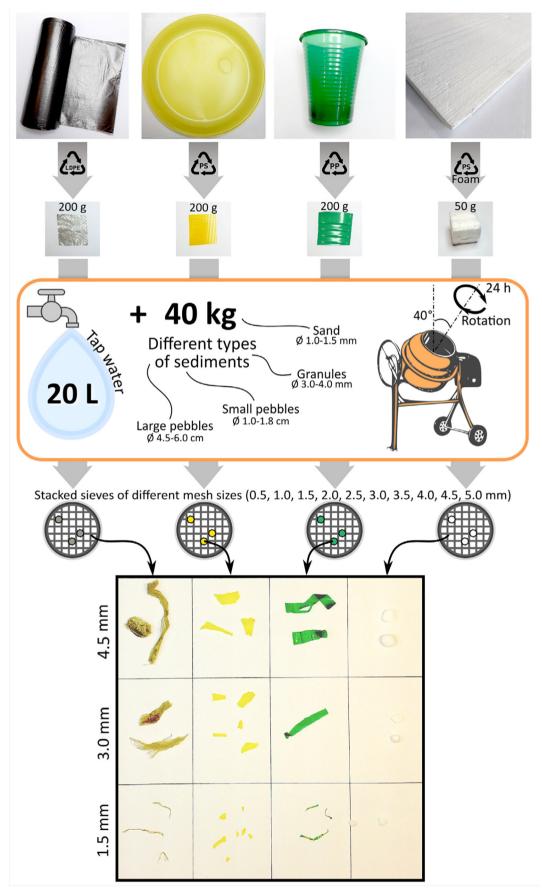
#### 2. Materials and methods

A set of laboratory experiments (16 runs: 4 types of plastic x 4 types of sediments) was designed to address the following questions: (i) how do the samples, made of different types (floating, sinking, flexible, foamed) of commonly used plastics, behave in the swash zone with different kinds of beach sediments (sand, granules, small and large pebbles), (ii) how do plastics change over time, (iii) comparatively, how effective are different sediments in mechanical fragmentation of macrosamples into MPs.

## 2.1. Laboratory set-up and procedure

A laboratory mixer with an inclined axis of rotation (40 degrees; frequency of rotation 30 rev/min) was used to mimic the wave breaking on the shore (see also (Efimova et al., 2018)). A standard 100-litre concrete mixer (ALTRAD POLAND S.A., 0.55 kW) with slight modifications was used: internal metallic blades were removed and the axis of rotation was fixed at the pre-selected (not standard) position. The mixer was loaded with 40 kg of natural sediments and 201 of tap water (Fig. 1). The inclination of the axis was carefully adjusted to the given amount of the mixing material in order to reproduce the wave up-rush/ back-wash strong enough to cause rolling/sliding of the upper layer of sediments at one side of the mixer. The maximum run-up height in this construction was about 20 cm. This way, sediments and plastic samples put inside the mixer experienced submersion and emersion at every mixing cycle, with rolling/sliding motion of the upper sediment layer, as it takes place in the sea swash. The characteristic bore of the "wave breaking" at the plunging side of the sediments' surface, and rolling pebbles/sliding sand layers at their rising side reproduced wave swash of quite moderate intensity. The inclination of the rotation axis, rotation frequency, volume of water and mass of sediment were the same for all the experiments, with the aim to keep the same mixing energy.

For every one of 16 runs, the given mass of the plastic samples of the certain plastic type (50 g for foamed plastic, 200 g for other plastic types) was placed in the mixer together with water and natural manually calibrated marine sediments. During the run, the dynamical behavior of macro- and micro-plastics in the swash-like mixing, their interaction with sediments, changes in material properties were visually monitored and fixed in protocols. For inspection of plastics distribution within sediments, the mixer was stopped occasionally. After cumulative 24 h of rotation, plastics were manually separated from the sediments using running water, then washed through a set of standard metallic sieves (mesh sizes from 0.5 to 5 mm, with the step of 0.5 mm), and dried at room temperature until constant weight. Dry mass of plastics from every sieve was obtained with the precision of 0.0001 g (analytical scales Sartogosm-MB-210A, working range 1 mg-210 g). Mass of plastics retained by the 5 mm sieve was considered as remaining in the category of "macro-plastics"; cumulative mass of plastics on the sieves from 4.5 mm down to 0.5 mm was regarded as "large microplastics" (l-MPs). With two types of plastics (solid and foamed forms of polystyrene), a significant deficit in the total dry mass left on all the sieves (0.5-5 mm) was found, with many tiny particles observed by naked eye in the out-flowing water (i.e., of the size < 0.5 mm). This mass deficit gave an estimate for the amount of "small microplastics" (s-MPs, < 0.5 mm). The number of particles in each size fraction of l-MPs was calculated either directly during visual inspection of the sieves, or



(caption on next page)

Fig. 1. A scheme of laboratory experiment on mechanical fragmentation of plastic samples in the sea swash zone with different beach sediments. Plastic samples  $(2 \text{ cm} \times 2 \text{ cm})$ , made of common market products were placed for 24 h in the rotating mixer with an inclined axis of rotation, filled with natural calibrated marine sediments and tap water. After 24 h of mixing, plastic material was washed through the set of sieves (typical examples of particles at particular sieves are shown in the bottom part of the figure).

recalculated from the mass using the number of particles in sub-samples, weighed on analytical scales.

The described above labor and time-consuming procedure of manual plastic separation/washing through the nest of sieves/weighing of fractions was designed, tested, and applied in order to (i) reveal in detail the way of fragmentation and specific features of the MP fragments of different plastics; possible changes in material properties and properties of MP fragments with decreasing particle size, and (ii) minimize possible errors, especially in counting of the number of MP particles. At the stage of separation of plastics from sediments, control measures (repeated washing of sediments) indicated that the mass of plastics remaining in the sediments in all the cases was < 1 g, i.e., < 0.5% of the initial mass of plastic. The l-MPs, collected from each of the 9 sieves (from 4.5 to 0.5 mm), were weighed separately with the precision of 0.0001 g, that leads to the error of about 0.001 g for the total mass of l-MPs, and is negligible in comparison to the error at the separation stage. For calculation of the number of l-MPs, maximum bias was contributed by the precision of analytical scales in the cases when calculation of particles in sub-samples was required. Final accuracy of estimation of the number of particles in such cases was about 3% of the total number of l-MPs particles. Thus, the estimated error of measurement of MPs mass is < 1%, while for the number of l-MPs particles it is < 3%.

As a general rule throughout the experiments, we considered the sieve mesh rather than real length as the characteristic scale of the sediment grains/fragments/particles. This is important especially for MPs particles since many of them have elongated shapes, which makes sorting by sieving complicated (Allen, 2003). In the field observations, often the maximum particle length is reported as the length scale, however the net/sieve mesh size is also a common parameter.

## 2.2. Choosing types of plastics

Types of plastics typically found on the beaches and in the sea/ocean coastal zone tend to reflect common industry production trends. For example, a study from Italy (Vianello et al., 2013) found the most predominant MPs to be PE (48%) and PP (34%); Frias with co-authors (Frias et al., 2014) found PE, PP and polyacrylates (PA) dominating along the Portuguese coast; on the beaches of Hawaii, PE (85%) and PP (14%) are dominating (Carson et al., 2011); in (Claessens et al., 2011), the analyzed MPs in coastal and offshore sediments consisted of PP, PS, and PP; (Esiukova, 2017; Esiukova and Chubarenko, 2018) found that foamed PS is the most common type of MPs on the beaches of the southeastern Baltic Sea. Based on these observations, we chose PE, PP, and PS (in its solid and foamed modifications) for our fragmentation experiments.

Since about a half of the produced plastic is used today in low-value products designed for disposable single-use (PlasticsEurope, 2015), and exactly these single-use products are shown to be the major part of the beach and marine litter (Carson et al., 2011; Claessens et al., 2011; Vianello et al., 2013; Frias et al., 2014; Esiukova, 2017; Zobkov and Esiukova, 2017; Balčiūnas, 2018; Haseler et al., 2018; Schernewski et al., 2018, among many others), we produced our squared  $2 \text{ cm} \times 2 \text{ cm}$  macro-plastic samples for the experiments from the following new products bought on the market (see photos in Fig. 1): single-use polypropylene beverage glasses (PP, measured in the lab material density is  $0.86 \text{ g cm}^{-3}$ ), disposable polystyrene plates (PS,  $1.05 \text{ g cm}^{-3}$ ), and the thinnest found ( $5 \mu \text{m}$ ) garbage bags from low-density polyethylene (LDPE,  $0.92 \text{ g cm}^{-3}$ ). As an example of foamed material, building heat insulator Styrofoam was chosen (PS-foam,

0.01 g cm<sup>-3</sup>): its spherules are mentioned in many studies as abundant pollutant on marine beaches (e.g., Claessens et al., 2011; Duis and Coors, 2016; Esiukova, 2017). Styrofoam plates of the standard thickness of 2 cm were manually cut into cubes with a 2-cm side using electrically heated thin wire. While selecting the plastics for the experiments, we made a preference to those that have significantly different mechanical and hydrodynamic properties (Chubarenko et al., 2016; Bagaev et al., 2017; Bagaev et al., 2018): (i) floating rigid flat PP squares with very smooth surface, (ii) floating thin flexible LDPE films, (iii) sinking rigid flat PS squares with productionally-corrugated surface, and (iv) very light foamed PS cubes composed of small spherules. As it was expected, the behavior of the samples in the "mixer swash", celerity and way of their fragmentation was self-specific in all the cases.

#### 2.3. Sediments

Four different types of natural marine sediments (according to the Udden-Wentworth grain-size scale (Wentworth, 1922)) were used in experiments: very coarse sand (1–1.5 mm), granules (3–4 mm), medium-size pebbles (1–1.8 cm), and very coarse pebbles (4–6.4 cm). The sediments (40 kg each) were collected on the open shores of the south-eastern part of the Baltic Sea using pairs of sieves with the corresponding mesh sizes. Some of the exceptionally long pebbles, which passed through the larger sieve but were retained by the smaller one, were manually removed, but in general we kept natural shape distribution, not discriminating between rounded, elongated, or irregular stones.

## 3. Results and discussion

Laboratory experimental studies on mechanical fragmentation of polymers are the most effective way towards understanding and quantifying their potential threat to the environment and ecosystems. This is because the process of fragmentation and its results depend on various factors and external environmental conditions (e.g., Lancaster, 1969), while laboratory conditions allow for their selective examination. The driving question of this study is the comparison of fragmentation efficiency of different types of beach sediments, in order to support the investigations of plastic contamination in physical oceanography and numerical modeling. For the sake of comparability of the results, only new plastics were chosen for the experiments. Influence of plastic weathering, degree of its crystallinity, etc., was not examined in the experiments, but is discussed later. However, we find it important to report the most typical qualitative features, since the knowledge of the qualitative behavior and changes of physical properties of different plastics with time during the fragmentation process helps a lot in explanation of in-situ observations.

## 3.1. Different plastics versus different sediments: qualitative features

Polypropylene (PP) samples appeared to be the most resistant to mechanical degradation. As follows from visual inspection during the experiments, not before 8 h from the beginning of mixing, some slight signs of degradation became visible: being initially smooth and transparent, the PP samples first got dull and rough to the touch, and then started to show longitudinal cracks. These longitudinal cracks seem to manifest not the PP material feature, but the production technology used for all the plastic cups: photo of both PP and PS cups found on the beach displays the same manner of cracking (Fig. 2). After 12 h of mixing with the coarsest sediment – large pebbles – the rectangular 2-



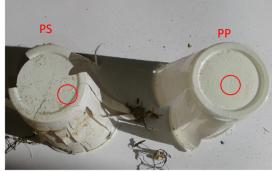


Fig. 2. Fragmentation of plastic cups in marine environment. The longitudinal cracks seem to manifest the production technology used for all the plastic cups: PP and PS cups found on the beach display the same manner of cracking.

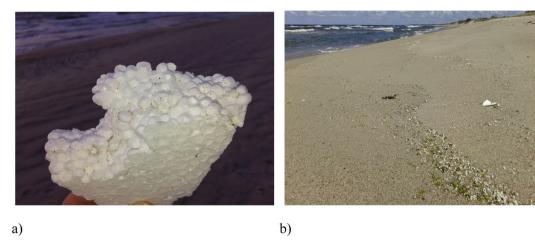


Fig. 3. (a) Initial stage of fragmentation of foamed PS in the sea swash zone: breaking to individual spherules. (b) Stripes of groat-like PSf particles on the beach along the wrack line are much typical for this kind of plastics.

cm-long fragments (still having sharp edges) began detaching from the main square sample plates (see photo of green PP stripes on Fig. 1). After 24 h, no > 0.5% of the total PP mass has passed into the "microplastics" category (see Section 3.3 below for further details). The samples were floating and actively "caught" into the swash-like mixing, but immediately rose to the surface when the rotation was stopped. Changes in their surface morphology after 24-h-long mixing with the coarsest sediments are illustrated by images in the Supplement (Table S1). The images were obtained using a fluorescence microscope Olympus of M319/12-OG BX43F (Konstantinova et al., 2016).

Low-density polyethylene films (LDPE) tended to become distributed first in water, then also within the sediment, despite the fact that their density is (slightly but) less than that of water. Plunging into the sediment was the weakest with the sand. Like PP samples, the LDPE samples are also very resistant to fragmentation: the macro-samples first get 2-4-8 times fold (in rectangles, smaller squares, tubes), and only some of them became deformed (stretched). With coarser sediments, the "folding" begins right after 1 h of mixing. At the end of the experiment, the folded samples squeeze through the 4.5-4-mm sieves, so that even on the 3.5 mm sieve the majority of "microplastics" are still not fragments. Below this 3.5-mm-sized sieve, the stretched "threads" become the major MPs type. At the 2.5 mm-sieve and below, thin LDPE thread-like filaments are sometimes found felted in lumps, in some cases - tangled with tiny fibers (external contamination from sediments or air in laboratory). After about 3 h of mixing with large pebbles (and about 10 h of mixing with sand), the samples lost their buoyancy: they did no longer float, rather, they all were submerged under the water surface or buried in sediments, being evenly distributed among stones and sand. Being folded, many of the LDPE samples caught sand grains or chips of stones, small pieces of paint from the mixer walls, tiny fibers (external contamination), etc. After 24 h of the experiment, quite many samples in macro-plastic fraction became longer/stretched; all the samples became very soft, just like an old rag.

Rigid polystyrene (PS) samples appeared to be the most prone to mechanical cracking and fragmentation. With their density slightly larger than that of water, the samples were very effectively mixing with sediments. Initially (at production) ribbed surface became smooth very soon (in 1–2 h); the square samples got rounded shapes, smoothed edges, and cracked sides. Towards the end of the experiment (24 h), a lot of material was transferred into the "s-MPs" category (< 0.5 mm). In contrast to LDPE films, the PS samples did not catch any other particles to their surface; they were relatively easy to separate from the sediment. Still, at finer sieves (1–0.5 mm), small aggregates/clews were found, indicating the same tendency of the smallest MPs to get tangled with any other possible materials.

Foamed polystyrene (**PSf**) samples are extremely light and hydrophobic. They tended to float as an "island" in the central part of the mixer and could not be much involved into mixing with the sediments. Since the sample cubes are composed of small individual spherules, the initial step of their fragmentation is detaching of these spherules from the cubes' edges and sides (**Fig. 3**). In the experiment with large pebbles, this process exhibited already in 1 h after the beginning. Towards the end of the experiment (24 h), all the cubes were much rounded, but still kept their macro-size, while many of the detached spherules were destroyed into smaller fragments. Thus, when detached from the highly-buoyant "mother-cube", the micro-particles are no longer resistant to further fragmentation. They become deformed, however still floating (i.e., still containing some air inside the bubbles), easily-drying and not-

sticking other particles to their surface. Overall, the photo of plenty of individual groat-like PSf particles along the wrack line on the beach (Fig. 3) looks very typical for this kind of plastic.

Remarkable is quite a specific behavior of plastic samples versus particular sediments. The most exciting result of the "swash mixing" of all the types of plastics with coarse sediments is a clear tendency of plastics to clog below the stones. The larger the stones – the clearer is this tendency, with certain variations for different plastics: with large (4–6.4 cm) pebbles, the (initially floating) LDPE films became buried under the stones (all over the bottom area) already in 3 h of mixing; with small pebbles – after about 9 h, with granules – the clogging beneath sediment was observed only for the sinking PS and the LDPE samples. In our experiments with sand, the LDPE films were rarely found buried, however lots of wrecks of PS samples were found deep in the sand. Light PP samples were captured only by large pebbles while they were moving (rolling), but when rotation was stopped the majority of them floated up to the surface.

The identification of the mechanisms of dynamic failure of the used materials requires further targeted efforts. However the observed qualitative behavior of plastics may provide some suggestions. In general, the impact behavior of plastic materials during dynamic fragmentation varies between ductile and brittle (e.g., Shockey, 1985). In our case, under repetitive shock load with abrasion, the LDPE films showed mainly ductile response, while PP and PS (solid) were obviously more brittle. Crystallinity of polymers is shown to be important for mechanical properties (Shockey, 1985; Andrady, 2017): crystalline polymers show generally higher strength & lesser ductility compared to amorphous polymers. The degree of crystallinity for the chosen plastics varies (Guo and Wang, 2019a) from amorphous (PS) to semi-crystalline (PE and PP), however PS fragmented mainly as a brittle material, while LDPE - as a ductile one. This shows that the degree of crystallinity of a polymer is not the leading factor for fragmentation under such a specific load as it is in the sea swash zone.

## 3.2. Mass of the generated MPs

The results of all 16 runs are presented in Table 1 and Fig. 4(a). The MPs masses are expressed in percent from the initial mass of macrosamples of the given plastic. This way, values in Table 1 and Fig. 4(a) show the fraction of initial mass of macro-samples of the given plastic, converted into the category of MPs by mechanical fragmentation due to interaction with the particular sediment. Table 1 shows the measured reduction of initial mass of plastics (with indication of particular contributions of l-MPs (0.5–5 mm) and s-MPs ( $<0.5\,\mathrm{mm}$ ), where possible), while Fig. 4(a) presents only the mass of l-MPs, directly collected from the sieves. The particularly small amounts of l-MPs (e.g., for PP) are also reliable and have an error of <0.5% from their value, since they represent the particles directly collected from the sieves and weighed on analytical scales.

As it follows from Table 1, the easiest for fragmentation among the selected plastics is **solid PS**: only 0.2% of its initial mass (0.4 g)

remained in macro-size class (on the 5 mm sieve). Averaged over all the sediment types, the fraction of MPs mass for PS is about 28% of its initial mass. Even though such an averaging has quite questionable physical meaning, it is still suitable for comparison of different plastics within this study, since they underwent similar sets of experiments. Solid PS showed a significant deficit of mass remaining on all the sieves (from 5 mm to 0.5 mm) after the experiment, which means that a considerable part of it was transferred to "s-MPs" category (below 0.5 mm here). This deficit of mass amounted to 17.9/3.7/0.3/0.7% of the initial mass (200 g) for the experiments with large pebbles/small pebbles/granules/sand, respectively. This way, the total loss of mass of macro-plastic category for PS amounted to 99.8/9.7/0.3/0.7%, i.e. the PS samples were almost completely fragmented during 24-h-mixing with large pebbles, and practically not fragmented by granules and sands.

The LDPE films are the second type in terms of effectively generating mass of MPs, with the averaged over all the sediment types percentage of MPs mass of 13%. In this case, however, the situation is qualitatively different: along with really fragmented pieces, many macro-samples were folded (up to 8 times), and passed into the MPs category. Some of the folded samples captured sand grains/granules/ small fragments of stones, so that the final integral mass of plastics on all the sieves (from 0.5 mm to 5 mm) was 9.1/5.8/2.5/8.9% (for large pebbles/small pebbles/granules/sand, respectively) larger than the initial mass (200 g). To make the "fragmentation efficiency" data comparable to other plastics, which did not show such a growth of mass, the percentages of mass in Table 1 and Fig. 4(a) are taken for the LDPE samples not from the initial mass, but from the final integral mass (which is 2.5-9.1% larger). This is equivalent to the assumption, that all size fractions of LDPE particles equally capture the sediments, which is quite reasonable for thin flexible films. As an overall result (see Table 1 and Fig. 4(a)), about 1/3 to 1/5 of the mass of LDPE macro-samples was transferred into the MPs category by mixing with pebbles, while granules converted only about 2%, and sand again was the least effective in fragmentation to quantify it reliably by the used methodology (fragmented  $\ll 1\%$ ).

For the **foamed PS**, the average fraction of mass converted by the four sediment types into mass of MPs amounts to about 12%. That is mainly due to the generation of s-MPs ( $< 0.5\,\mathrm{mm}$ ): its amount 2–10 times exceeded the amount of l-MPs ( $0.5-5\,\mathrm{mm}$ ). Foamed PS eventually had a large portion of macro-samples (about 73/90/94/94% for large pebbles/small pebbles/granules/sand, correspondingly) plus 19/9/5/5% of "s-MPs"-particles, with only about 8.4/0.8/0.9/1% of particles in the l-MPs size range (shown in Fig. 4(a)). This shows that when the individual spherules are detached from the highly buoyant macrosample, they are very soon fragmented into small-sized particles.

The most resistant to mechanical fragmentation and abrasion among the selected types of plastics was **PP**. The generated l-MPs particles were all manually counted and weighed at analytical scales, indicating the largest fragmentation "efficiency" for large pebbles, which was much < 1% of initial mass during 24 h of the experiment.

Table 1

Efficiency of particular sediment types (large pebbles, small pebbles, granules, sand) in conversion of the mass of PS, LDPE, PSf, PP macro-samples into the mass of micro-plastics (0.5–5 mm) during 24 h of mixing (expressed in percent of the initial mass of macro-samples in the given run; except for LDPE, see the text). Where possible, particular contributions of l-MPs (0.5–5 mm) and s-MPs (< 0.5 mm) are provided in parentheses. The maximum estimated error is within 0.5% from the presented values.

29.0	27.4 (8.4 + 19)	0.1 (0.1 + 0)	1000
20.5	9.8 (0.8 + 9)	0.003(0.003+0)	10,000
1.9	5.9 (0.9 + 5)	0.001 (0.001 + 0)	6000
0.2	6 (1 + 5)	0.004 (0.004 + 0)	1500
145	5	25	
	20.5 1.9 0.2	20.5 9.8 (0.8 + 9) 1.9 5.9 (0.9 + 5) 0.2 6 (1 + 5)	20.5     9.8 (0.8 + 9)     0.003 (0.003 + 0)       1.9     5.9 (0.9 + 5)     0.001 (0.001 + 0)       0.2     6 (1 + 5)     0.004 (0.004 + 0)

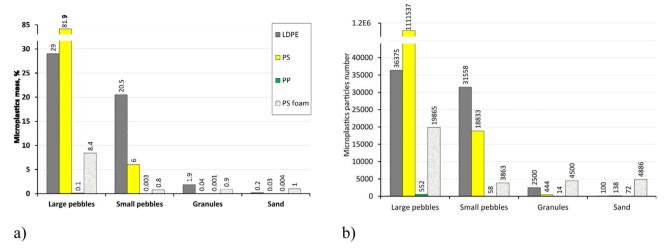


Fig. 4. The results of laboratory experiments on mechanical fragmentation of LDPE, PS, PP, and PS-foamed during 24-h mixing with different types of sediments: (a) fraction of mass (in % from the initial mass) converted into l-MPs (0.5–5 mm); (b) number of l-MPs particles generated. Note the break at vertical axes.

Summing up, different plastic types in our study can be ranked in order from the highest to the lowest MPs particles generation efficiency as follows: (1) solid PS (the mean mass fragmented by all four sediment types is 27% of initial mass), (2) LDPE (13%), (3) foamed PS (12%), PP (< 1%). Interestingly, Lambert and Wagner (2016) found quite a similar ranking of plastics by the number of produced MPs particles in their UV-degradation study: PS/PP sheet/PE pellet/PP pellet/PP film, with the number of particles generated after 112 days of exposure related as 3.8/1.9/1.6/1.08/1. The particles, however, were of much smaller size range – from 30 nm to 60  $\mu$ m.

The laboratory set-up reproduces only very moderate mechanical "wave action", able to roll only some pebbles or sheet-flow motion of the upper sand layer. This way, the reported experiments provide the time scales of plastic fragmentation under a low-energy forcing, while under more energetic impact of stronger waves the fragmentation will proceed faster. Quantitative evaluations of the fragmentation efficiency under higher-energy forcing require using of the corresponding correction coefficients, which should, presumably, scale with the significant wave height squared (as the measure of the wave energy). Still, based on laboratory data only, the comparison of fragmentation efficiency of different sediments under the same mechanical forcing is correct. Inter-comparison of general fragmentation efficiency of particular sediments in the experiments indicate that the coarser is the sediment, the larger is the fraction of the mass of the plastics, converted by mechanical fragmentation into the category of MPs (see Table 1 and Fig. 4(a)). This general rule is valid disregarding the particular sinking/ floating behavior of different plastic types (Khatmullina and Isachenko, 2016): the ratio of plastic masses, fragmented during 24 h into MPs by large pebbles and sand, is 143 for sinking solid PS, while for floating plastics it makes up 145 (LDPE), 5 (PSf), and 25 (PP), see Table 1 (last row). These values indicate that beaches covered by large pebbles are 5–145 times more effective than sandy beaches in converting the mass of available plastics into the mass of MPs. Sand and granules showed quite similar fragmentation efficiency. The last column of Table 1 shows, however, that the fragmentation efficiency of the considered sediment types is 1000-10,000 times different for different polymers. This way, the behavior at dynamic fragmentation, along with shape, size, and density, is one more highly diverse feature of plastics in marine environment (Chubarenko et al., 2018).

Weathering of plastics in the environment makes them more brittle (e.g., Andrady, 2017), favoring even faster mechanical fragmentation. Still, for the same degree of plastic weathering, higher fragmentation efficiency of coarser sediment should hold, because it is shown in the experiments to be valid for brittle plastics as well. One more environmental factor influencing mechanical properties of plastics is

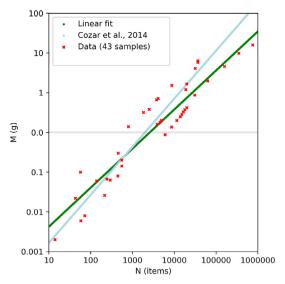
temperature: at a lower temperature all the materials become harder and more brittle. Again, the comparison of the fragmentation efficiency of different kinds of sediments in the given region holds, as long as the environmental temperature regime is similar.

In the environmental context, these results highlight an extreme importance of clastic shores and headlands in the generation of marine MPs. A number of investigations (Debrot et al., 1999; Convey et al., 2002; Eriksson and Burton, 2003; Thiel et al., 2013) already pointed out the role of rocky shores exposed to surface waves: they capture larger debris items and serve as a "grinding mill", then exporting fragments back to the sea. Rocky shores and cobble/pebble beaches are also reported to capture MPs not at the beach surface, but at some depth under the cobbles and small rocks (McWilliams et al., 2018), which is in full agreement with the laboratory observations.

## 3.3. MPs mass versus number of items

Field observations most often report MPs pollution in number of particles - per kilogram of sediment, or liter of water, or squared meter of the beach area or ocean surface (Claessens et al., 2011; Cózar et al., 2014; Bergmann et al., 2015; Bagaev et al., 2018; Khatmullina and Chubarenko, 2019). At the same time, the only data available for practical evaluation of the net emission of the MPs into the marine environment is the statistical information on the net plastic production and the average amount of mismanaged plastic waste; both are typically expressed in mass units. Numerical models also require conservative units, while the number of items is not conservative. Moreover, numerical models need calibration, which is possible only via comparison with monitoring data, expressed in non-conservative number of items of MPs particles. Taking advantage of laboratory experimentation, we paid especial attention to this practically important question: how the mass of the generated MPs is linked to the number of MPs particles. Calculations were performed for 1-MPs only (0.5–5 mm).

The total number of the generated l-MP particles of different polymers, collected from the sieves (mesh size from 0.5 to 4.5 mm) after 24 h of mixing of macro-samples with different kinds of bottom sediments, is presented versus sediment types in Fig. 4(b). In general, the same tendency remains: the coarser is the sediment, the larger is the number of generated l-MPs items. The distribution, however, differs from that of the mass (shown in Fig. 4(a)). This difference arises from the different distribution of the number of particles by size for the used polymers. For example, PS (in both its solid and foamed modifications) produces more l-MPs of smaller fractions in comparison to LDPE, so the masses of l-MPs from these polymer types differ more than their numbers. A deeper consideration of distribution of l-MPs by size faces



**Fig. 5.** Relation between the mass of MPs and the number of items. The comparison of linear fits (in log-log presentation) for the data on MPs floating on the ocean surface from (Cózar et al., 2014, shown in blue) and for our experimental data on mechanical fragmentation (in green). Note logarithmic scales on both axes. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

principal methodical difficulties because the MP particles (in contrast to natural sediments) have very different shapes, which influences the results of their sieving (Efimova et al., 2018).

Linear dependency between the mass and the number of l-MPs in log-log presentation was found in (Efimova et al., 2018) for the case of fragmentation by large pebbles. Here, we cover four most typical plastics and four most typical beach sediments, and compare the obtained result with observations in the ocean (Fig. 5).

Cózar with co-authors (Cózar et al., 2014) combined data sets from four studies on floating plastic debris in the open ocean, and on the base of 570 data points in total found the relationship between the mass and the number of plastic items in the size range from 0.16 mm to 158 mm. Cózar et al. (2014) mention that the vast majority of the plastic items in their study consisted of fragments of larger objects, while industrial resin pellets represented only a small fraction (< 2%) of all encountered items, and the textile fibers were found only occasionally and were excluded from the analysis. This makes our data well comparable: our set also includes only fragments of larger plastic objects, no primary pellets, no fibers. Fig. 5 displays (in logarithmic axes) the Cózar's dependency along with the combined results of our experiments on mechanical fragmentation: 16 points from the present study and 27 points from (Efimova et al., 2018). An analytical linear dependency log<sub>10</sub> M (g) =  $a \cdot \log_{10} N(items) - b$  was obtained by the least squares method, and its statistical characteristics are shown in Table 2, along with those reported for the data by (Cózar et al., 2014). Explicit expressions of the relations read:

 $M = 4.4 \cdot 10^{-4} N^{0.98}$  this study,

 $M = 1.0 \cdot 10^{-4} N^{1.21}$  field data (Cózar et al. 2014),

where M is expressed in g, and N – in items. The dependencies

summarize data for MPs particles (excluding fibers) in the size range of  $[0.16\,\mathrm{mm};\ 158\,\mathrm{mm}]$  and  $[0.5\,\mathrm{mm};\ 5\,\mathrm{mm}]$ , correspondingly, valid for the number of particles N>10.

Fig. 5 and Table 2 show quite an amazing similarity of linear fits for the laboratory and field data. This speaks in favor of the hypothesis (Chubarenko et al., 2018) that mechanical fragmentation on the coastlines is probably the main mechanism of generation of marine MPs. Since the slope of the field-data regression (Cózar et al., 2014) is some 20% larger than that of the laboratory data, this might indicate that there is some mechanism that makes oceanic particles heavier through their lifetime: the more oceanic MPs particles are collected, the larger their cumulative mass excess is compared to just mechanical-fragmentation dependence. This effect might be attributed, for example, to the bio-fouling or aggregation of MPs with organic or sediment particles (Bergmann et al., 2015).

The contribution of the competitive to the mechanical fragmentation mechanism of UV-induced fragmentation of plastics seems to be not manifested in the Cózar data. The UV-degradation produces not the large fragments but many smaller particles (100–200  $\mu$ m in (Andrady, 2017), 30 nm–60  $\mu$ m in (Lambert and Wagner, 2016), < 50  $\mu$ m for foamed PS in (Eo et al., 2018)). Such particles have smaller cumulative mass but a large number of items, which shall contribute data points at the lower right part of the M(N) field on Fig. 5, i.e., out of the Cózar scatter.

## 4. Conclusions

Laboratory experiments on mechanical fragmentation of single-use plastic products made of most common plastics (LDPE, PP, and PS in its solid and foamed modifications) were carried out in a rotating mixer filled with water and different kinds of natural marine beach sediments (pebbles, granules, sand). The set-up was adjusted to mimic moderate-energy mixing in sea swash zone. Experimental studies of similar purpose are common to disclose abrasive wear of materials under certain external conditions of interest, however they have never been conducted before for plastics under sea-swash like mixing, which is a combination of repetitive shock load and abrasion.

Experiments show that the coarser is the sediment, the larger is the fraction of mass of the plastics transferred by mechanical fragmentation into the category of MPs (< 5 mm) during the same period of time. This general rule is valid for all four types of plastics used in our experiments, disregarding their particular sinking/floating behavior or mechanical properties, e.g., large pebbles showed about 140 times higher fragmentation efficiency than sand for both sinking solid PS and floating LDPE film. An extreme importance of clastic shores in generation of marine MPs is highlighted once again, in accord with previous field investigations (Debrot et al., 1999; Convey et al., 2002; Eriksson and Burton, 2003; Thiel et al., 2013). It is confirmed that, under swash-like mixing with coarse sediments, the MPs tend to burry below the sediment surface. This is in accord with field observations, too (McWilliams et al., 2018).

Time scales of mechanical fragmentation in the swash-like mixing are much shorter than those for other degradation/fragmentation mechanisms: even new materials, used in this study, could be 20, 30, up to 99.8% fragmented down to MPs during 24 h of very moderate forcing. In natural environment, the UV-, oxidative, or other material degradation processes should even facilitate the mechanical

Table 2
Statistical characteristics of a linear model for relationship between the number of l-MPs particles and the mass of l-MPs in the experiments and for the field data compilation by (Cózar et al., 2014): SE is the standard error of the estimated slope *a*, *R* is the correlation coefficient, *N* is the number of samples in the dataset.

MODEL	p-Level	a (slope)	b (intercept)	SE	$R^2$	N
Linear regression ( $y = a x + b$ ) Cózar et al., 2014	$ 2.5E - 22 \\ p < 0.0001 $	0.9833 1.21	-3.3567 -3.99	0.0505 -	0.90238 R = 0.95 $R = 0.85$	43 570

fragmentation.

The relationship between the mass of MPs and the number of items, obtained via mechanical fragmentation of common plastics by four most typical types of beach sediments, is similar to the dependence deduced from MPs collected at the ocean surface (Cózar et al., 2014). This indicates that mechanical fragmentation at coasts can be a significant source of MPs in the ocean.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.marpolbul.2019.110726.

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#### Declaration of competing interest

The authors declare no competing interests.

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