Transport of marine microplastic particles: why is it so difficult to predict?[1]

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Abstract: Marine microplastic particles (MPs, <5 mm) exhibit wide ranges of densities, sizes, and shapes, so that the entire MPs "ensemble" at every time instant can be characterized by continuous distributions of these parameters. Accordingly, this community of particles demonstrates distributions of dynamical properties, such as sinking or rising velocity, critical shear stress, and the re-suspension threshold. Moreover, all the MPs' properties vary significantly with the time spent in marine environment and with particular conditions experienced by the particle on its journey. A brief review of the present-day numerical efforts towards prediction of MPs transport shows the prevalence of the Lagrangian particle tracking approach, especially for floating litter. In a broader context, the present practice of MPs transport modelling follows the "selective" strategy (e.g., only a certain sub-class of MPs, or specific processes, are considered, sometimes in only one- or two-dimensional setting). The heterogeneous nature of MPs, their enormous longevity and movability in marine environment, and the wide spectrum of the involved environmental processes suggest further integration (or coupling) of different models in future, as well as application of other types of models (ensemble modeling, chaos theory approaches, machine learning, etc.) to the problems of MPs transport and fate in the marine environment.

Key words: microplastics, transport, modelling.

1. Introduction

Microplastic particles (MPs, <5 mm) are found nowadays in all marine environments, from pole to pole, from the water surface to the deep bottom sediments (Ivar do Sul and Costa 2014). As a part of plastic marine debris, MPs pollution is of great concern due to MPs' ability to be transported over large distances causing potential harm to ecosystems (Cole et al. 2011; Cózar et al. 2017; Wright et al. 2013). MPs have various densities, shapes, and sizes, and all of these properties change with the time spent in the marine environment due to biofouling, weathering, mechanical degradation, and other external factors (Jahnke et al. 2017; Kooi et al. 2017; GESAMP 2015). As a result, physical transport characteristics of MPs are complicated and very specific (Chubarenko et al. 2016; Zhang 2017). Thus, MPs are a principally new pollutant, whose transport in marine environments deviates from that traditionally deduced from modeling of conservative tracers, Lagrangian floats,

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or oil spill patches. By the same token, MPs are only to some extent similar to natural particles, like sediment grains, phytoplankton, fecal pellets, or marine snow.

Although a lot of effort has been put into in situ observations on MPs in different compartments of marine and freshwater environments, global understanding of mass balances between sources and sinks, exact amounts of litter present in the world ocean, and its pathways and accumulation zones is still lacking (Hardesty et al. 2017). Numerical modelling is a promising tool to elaborate the required holistic view of MPs (as well as marine plastic litter in general) in the world ocean. In its turn, relevant modeling depends on thorough conception of MPs dynamics in the real environment; however, only initial steps have been made in this regard (Ballent et al. 2012; Reisser et al. 2015; Chubarenko et al. 2016; Kaiser et al. 2017; Khatmullina and Isachenko 2017; Kooi et al. 2017). In contrast, behavior of other particles in the water column (e.g., natural sediment grains, fecal pellets, phytoplankton) have been quite extensively studied in sedimentology and hydrodynamics (e.g., Soulsby 1997; Turner 2002). Careful application of existing knowledge concerning other particles may provide the necessary theoretical background for studying MPs transport (Filella 2015; Chubarenko et al. 2016).

In this review, we highlight some aspects of the problem of adequate prediction of MPs transport in the marine environment. We briefly review the present-day knowledge on MPs characteristics, which are specific and different from those of other (either anthropogenic or natural) marine particles. Over time, both the physical properties, such as size, shape, and integral density, and variable fundamental dynamical properties, such as the settling or rising velocity and the re-suspension threshold, have been changing. We make a short (and obviously not exhaustive) overview of successful modern numerical modeling studies of MPs transport, and suggest some directions for further development.

2. Variability of physical properties of MPs

The behavior of any particle in a fluid is determined by its physical properties, of which the density, size, and shape make the largest (both quantitative and qualitative) contribution to its dynamics (Dietrich 1982; Filella 2015; Zhang 2017). Physical properties of a particle determine not only the forces acting on it and the resulting physical processes (settling, re-suspension, beaching, etc.), but also the processes related to the MP itself, including adhesion of molecules to the surface (Teuten et al. 2009), aggregation with other particles (Long et al. 2015; Besseling et al. 2017), biofouling (Kooi et al. 2017), and degradation (Gewert et al. 2015; Bandow et al. 2017; Song et al. 2017).

2.1. Material density versus integral particle density

For particles found in the marine environment, the initial density of plastics (at production) varies from about 0.05 g cm^{-3} (for polystyrene foam) to 2.1–2.3 g cm^{-3} (for polytetrafluoroethylene) (Chubarenko et al. 2016). Use of fillers, stabilizers, and other additives during the production causes deviation of the plastic material density from the table value (Andrady 2017). By their material density, MPs could have either positive or negative buoyancy in sea water. This defines two important external conditions: (*i*) whether the MP will stay at or near the surface or sink, and (*ii*) which marine organisms may be encountered in its surroundings (Andrady 2017). In the worldwide production of 311 million tons of plastic in 2014, obviously positively buoyant plastics (polypropylene, foamed polystyrene, polyethylene) comprise at least about 50%, whilst at least about 20% (solid polystyrene, PVC, and PET) are negatively buoyant (PlasticsEurope 2015). Because MPs are prone to bio-fouling and sinking with time, 50% is a zero-order approximation of the maximum fraction of floating MPs in oceans.

2.2. Size and shape

Size of a plastic particle is the only parameter by which it is defined as a MP. However, the exact size limits are still not universally accepted: the upper limit is taken as 5 mm (Arthur et al. 2009), but the lower limit is often confined to the mesh size used in the particular field survey, and in some cases is taken at a few micrometres (Hidalgo-Ruz et al. 2012; Filella 2015; Andrady 2017). The distribution of MPs by size was attained for MPs in the upper part of the water column (Cózar et al. 2014, 2015; Isobe et al. 2014; Enders et al. 2015; Reisser et al. 2015; Lebreton et al. 2018). On the beach, size distribution data were reported in some case studies (e.g., Chubarenko et al. 2018b). Overall, data on sizes of MPs found in the environment are still scarce (Hardesty et al. 2017). Such distribution surveys are typical for sedimentology (called grain-size analysis) and should be strongly recommended to MPs monitoring reports (Filella 2015).

MPs demonstrate a variety of shapes, which are typically categorized during visual inspection of samples (Rodríguez-Seijo and Pereira 2017). One way of generalization could be the dividing of MPs into three groups: quasi-one-dimensional (fibers, lines), quasi-two-dimensional (films, flakes, flat thin particles), and three-dimensional (3D) (irregular fragments, pellets, ovoids, cylinders, spherules) (Chubarenko et al. 2016) particles.

Ranges of MPs' density, size, and shape overlap with those of other natural marine particles (Table 1). Analysis of transport mechanisms and rates obtained for other particles could be applied to MPs (Ballent et al. 2012; Hardesty et al. 2017; Khatmullina and Isachenko 2017).

2.3. Evolution of MPs properties in the marine environment

Physical properties of MPs are permanently changing in natural environments, altering their dynamics and transport pathways (Galloway et al. 2017; Hardesty et al. 2017; Jahnke et al. 2017). Under environmental conditions plastic particles exhibit different types of weathering processes: photooxidation, thermooxidation, hydrolysis, biodegradation, and mechanical fragmentation due to wave action in the swash zone (Andrady 2011; Gewert et al. 2015). As a result, molecular structure of the polymer changes, cracks are formed at the surface, particles become brittle and break down into smaller pieces, implying change of the density, size, and shape of the particles (Gewert et al. 2015; Galloway et al. 2017; Halle et al. 2017). When combined, the weathering mechanisms could lead to more rapid breakdown of MPs than in the case of each of the mechanisms separately (Song et al. 2017). Rates of MPs weathering and fragmentation depend on plastic type and morphology (Shah et al. 2008; Gewert et al. 2015; Bandow et al. 2017; Song et al. 2017; Efimova et al. 2018); however, the exact rates of MPs weathering taking place in the environment and not in laboratory conditions are still poorly investigated (Gewert et al. 2015).

Almost any solid surface introduced to the natural environments offers a new substrate for colonizing and fouling organisms (Abarzua and Jakubowski 1995), and plastics are no exception (Galloway et al. 2017). The presence of biofilm at the surface of plastic pellets was reported as long ago as 1972 by Carpenter and Smith (1972), and recent works demonstrate the presence of diverse microbial communities on the surface of weathered microplastic debris (Zettler et al. 2013; Reisser et al. 2014). Colonization by organisms is supposed to increase particle density and sinking capacity, explaining why light plastics are found in marine sediments (Morét-Ferguson et al. 2010; Woodall et al. 2014); however, not many works report the quantitative effect of biofouling using in situ exposure experiments (Ye and Andrady 1991; Fazey and Ryan 2016; Kaiser et al. 2017). Biofouling is supposed to be especially important for initially positively buoyant particles that remain at or near the water surface — in the photic zone — where biofilm formation is possible. Initially negatively buoyant particles would most probably leave the photic zone prior to

Table 1. Size, shape, and density of marine particles.

Particle	Density (g/cm^3)	Size (mm)	Shape
MPs	0.05–2.1 (Chubarenko et al. 2016)	<5 (Arthur et al. 2009)	Quasi-1D (fibers, lines), quasi 2D (films, flakes, flat thin particles), 3D (irregular fragments, pellets, ovoids, cylinders, spherules)
Amber	1.05–1.1, up to 2.00 (Chubarenko and Stepanova 2017)	—	Rather irregular but definitely volumetric (3D) shapes; many of them break in parts in stormy waves and have sharp fresh cleavages (Chubarenko and Stepanova 2017)
Quartz sand	2.65	0.062–2 (very fine to very coarse sand) (Wentworth 1922)	Irregular, 3D, rounded, angular (Blott and Pye 2008)
Calcerous sand (coccoliths, corals, pieces of carbonate materials)	2.6–2.7	0.062–2 (very fine to very coarse sand) (Wentworth 1922)	Irregular, flat, variety of shapes associated with their biological origins, rod, blade, disc, and equant, rough surface textures and jagged edges (Wang et al. 2018)
Fecal pellets	1.1–1.2 (Yoon et al. 2001), density is reported to vary according to the quality and quantity of food ingested by the zooplankton (Turner 2002)	from 0.4–4 to tens of μm, minipellets (Turner 2002)	Ellipsoid, cylindrical with rounded or tapered ends, conical, spiral (Yoon et al. 2001), spherical (Turner 2002)
Marine snow	1.03–1.3 (Maggi 2013); ~1.03 (Iversen and Ploug 2010)	1–4 (Iversen and Ploug 2010)	Irregular, 3D aggregates of multiple particles; aggregates may originate from abandoned larvacean houses, diatom and dinoflagellate flocs, fecal aggregates, and aggregates of miscellaneous detritus (Turner 2002)
Oil droplets	0.72–1.04 (Fingas 2017)	0.001–0.070 (Lunel 1993)	Spherical

the formation of substantial biofilm. Biofouling processes depend not only on the fouling species, water temperature and transparency, and availability of nutrients and sunlight in water, but also on the particle surface-to-volume ratio, and thus strongly depend on the particle's shape and size (Chubarenko et al. 2016; Fazey and Ryan 2016; Kooi et al. 2017).

The effective (integral) density of a particle could also be changed due to interaction with other organic or inorganic particles, natural colloids, and suspended soils. Besseling et al. (2017) experimentally showed that MPs undergo hetero-aggregation with clay particles, and modeled that the resulting excess of mass on the surface of MPs leads to particle sedimentation and further retention at the bottom of the estuary. Other mechanisms that potentially enhance vertical flux of MPs to the ocean floor are incorporation into fecal pellets or marine aggregates consisting of phytoplankton cells and detritus (Wright et al. 2013; Long et al. 2015; Michels et al. 2018).

It is important to mention here that while the rates of MPs evolution presumably vary for different types of plastics, in general, changing of properties in the natural environment is known and reported also for other particles. Oil spills are quite similar to the MPs in this vein. Oil discharged to the sea surface is carried by currents and is subjected to various processes, evaporation, emulsification, dissolution, photolysis, dispersion, biodegradation, etc., which depend on the initial properties of the oil (Fingas 2017). In the marine environment, under the action of waves and turbulence, oil patches can be completely or partially broken into droplets of various sizes (Lunel 1993). Oil droplets mixed in the upper layer of the water column can interact with each other, merge or break into smaller droplets,

aggregate with other mineral particles and sink, be available for interaction with marine organisms — some of these processes are also typical for MPs. At the same time, physical, chemical, and biological processes that erode or modify oil as it drifts and spreads occur on different time scales from several hours to months or even years (Korotenko et al. 2004). The similarity of these two types of marine pollution indicates the possibility to apply numerical models developed for oil spills to the MPs investigation.

3. Dynamical properties of MPs

In environmental hydrodynamic modeling, motion of the particle in the flow is classically described by its fundamental parameters, such as terminal sinking velocity and critical shear stress of re-suspension. The former characterizes the free fall of the particle through a still water column, while the latter indicates the threshold flow velocity below which the particle cannot be re-suspended from the bottom.

3.1. Sinking and rising velocity of MPs

Bottom sediments are supposed to be the end-point for all the MPs present in the water column (Andrady 2011; Woodall et al. 2014; Kooi et al. 2017; Bagaev et al. 2018). Terminal sinking (or settling) velocity is defined as a maximum velocity of the particle falling in a still fluid without acceleration (e.g., Hallermeier 1981; Dietrich 1982). This is a crucial hydrodynamic parameter of a non-buoyant particle that has been extensively studied for more than 100 years in sedimentology, hydrodynamics, and also in more application-focused disciplines like hydraulic engineering, pipeline construction, etc. (Hallermeier 1981; Dietrich 1982; Ayazi Shamlou 1987). For MPs, this parameter determines residence time of a particle spent in the water column, and the possible distance to which it could be transported by currents in a real environment.

Rising velocity (upward terminal velocity in Isobe et al. (2014) and Hinata et al. (2017)) is the corresponding parameter introduced for particles with positive buoyancy; it reflects the balance between the same forces (gravitational, buoyancy, and drag forces). In the field of MPs research, it is required for analysis of MPs migrations due to biofouling and defouling, transport and beaching of MPs under wave-induced motions in sea swash zone, capturing of buoyant MPs into the water column by wind- and wave-induced mixing, etc. Although the existing works on rising velocity did not accomplish extensive comparison of experimental values and observations with the semi-empirical and theoretical predictions developed in hydrodynamics, discovered tendencies repeat those that have already been known for behavior of particles in a fluid. Both settling and rising velocities depend on the size of the particle, on the density difference between particle and fluid, and on the shape, which determines the manner of settling or rising and the presence of secondary movements (Kukulka et al. 2012; Ballent et al. 2013; Reisser et al. 2015; Kooi et al. 2016; Kowalski et al. 2016; Khatmullina and Isachenko 2017). Experimental values of settling velocity of different MPs range between 1 and 127 mm/s, equivalent to about 86 m – 11 km per day (Kowalski et al. 2016; Bagaev et al. 2017; Khatmullina and Isachenko 2017). Some of the existing semi-empirical formulations of terminal settling velocity developed for sand grains (Dietrich 1982; Ahrens 2000) showed good correspondence with experimental data on settling of MPs spheres and cylinders (Khatmullina and Isachenko 2017). Rising velocities of MPs sampled in the North Atlantic subtropical gyre span between 1 and 43 mm/s, or, between 86 m and 3.7 km per day (Kukulka et al. 2012; Reisser et al. 2015; Kooi et al. 2016).

In a real environment, a particle's settling and rising rate deviates from the terminal settling or rising velocity. Motion of relatively small and light MPs in natural (generally stratified) waters is significantly influenced by turbulence and currents (see, e.g., Nielsen 1993). Increased turbulence near the surface is responsible for establishing an exponential

decrease of particle concentration with depth in the subsurface mixed layer: because of the wind mixing, floating MPs become submerged and start to float to the surface with different rates according to their properties, and the extent of the decrease is inversely proportional to wind speed (Kukulka et al. 2012; Reisser et al. 2015; Kooi et al. 2016). Many plastics have densities only slightly lower or higher than that of seawater, and the level of turbulence required for retaining such MPs in suspension (submerged below surface or above sea bottom) is relatively small (Filella 2015). Ballent et al. (2012) have experimentally shown that susceptibility to turbulence depends on MPs' size and shape: large, irregularly shaped pieces were easily submerged by surface turbulence, whereas spherical pellets were the most resistant. Another factor influencing the particle's vertical velocity while falling though the water column is highlighted in Cheng (1997) and Baldock et al. (2004): the particle is not isolated from other grains, and their presence modifies the settling velocity of an individual particle due to mutual interference among them. Moreover, collision with organic and inorganic particles and biofouling lead to a change of settling velocity of the resulting aggregate (Wright et al. 2013; Long et al. 2015; Besseling et al. 2017; Kooi et al. 2017).

Ocean density discontinuities also interfere with settling process predicted for a particle in a still unified fluid. Non-buoyant MPs will slow down as the density difference between particle and water decreases, which may lead to accumulation of MPs at mid-water depths with high density gradients (Ye and Andrady 1991; Kooi et al. 2017).

Most of the aforementioned aspects concerning vertical transport of MPs were also investigated for other marine particles. Settling velocity of fecal pellets varies according to their size, density, and shape (Yoon et al. 2001; Turner 2002). Density discontinuities in a water column are known as the "accumulation zones" of marine snow (MacIntyre et al. 1995). Turbulent mixing is attributed to the accumulation of fecal pellets in the upper layers of the water column (Turner 2002). Therefore for deeper understanding of MPs motion in the water column it may be beneficial to consider existing studies on the transport of other particles in the ocean (see Table 1).

3.2. Re-suspension threshold

Magnitude of critical shear velocity (or shear stress) characterizes the threshold below which water currents are unable to re-suspend particles from the bottom. This parameter is vital for modeling of transport of particles settled to the bottom in deep areas, or saltated over a coastal slope under the influence of surface waves, or beached–recaptured by swash, etc. This key topic has received very little attention up to now. The only investigation was reported by Ballent et al. (2013), where standard plastic pellets were re-suspended in a rotating chamber. Chubarenko et al. (2018a) mentioned an attempt to apply to MPs the classical Shields methodology (Shields 1936), commonly used to determine critical shear stress for natural sediments. The bottom of a 10 m long laboratory channel with unidirectional step-wise changeable flow was covered either by natural marine sand (1–1.5 mm), or by granules (3–4 mm), or by pebbles (1–2 cm). MPs of various shapes (irregular 3D fragments, spherules; flakes; fishing line cuts, flexible threads), size, and density were used. These tests provided an insight into the new MPs-related challenge: in contrast to natural sediment grains, which are to be re-suspended in the sea from the bottom covered by particles of similar properties, the MPs are to be re-suspended from natural sediments, (i.e., from the bed covered by particles differing from MPs by the size, shape, and density). Two parameters seem to be important for MPs re-suspension: (*i*) the ratio of the characteristic size of MP to the bed roughness (i.e., to the grain size of the bed load sediment), and (*ii*) the shape of the MP. The latter defines the manner in which the particle responds to the flow: 3D particles tend to roll over the bed surface; flat particles suddenly saltate from the

bottom and then fly long distances over the bed until next settling; one-dimensional lines and threads roll over the sediments until they turn along the direction of the current, and then begin saltatating. Overall, initiation of motion of the same very particle at the same kind of bed load significantly depends on the initial particle orientation, and is especially variable at coarser sediments. After beginning their motion over pebble-covered rough bottom, all the particles tend to be caught in-between stones, and it is only sudden or stochastic turbulent bursts that can bring them back into the flow (Chubarenko et al. 2018a). Straightforward practical conclusion for further field and modeling applications is the fact that the key factor for re-suspension of MPs is the bed roughness: the larger the grain size of the bed load, the larger *the range of variability* of the magnitude of the critical shear stress.

4. Modelling of MPs transport — current progress and open questions

Numerical modeling is one of the most effective tools in investigation of dynamics and transport of contaminants in both marine and freshwater environments.

Several studies used one-dimensional models to obtain physically relevant conclusions about vertical transport and distribution of MPs. The effects of different types of turbulence in the ocean surface boundary layer on the vertical distribution of buoyant tracers, including influence of the Langmuir circulation and turbulent kinetic energy input due to breaking waves, were modeled using the large eddy simulations approach (Kukulka et al. 2012; Brunner et al. 2015; Kukulka and Brunner 2015). Enders et al. (2015) analyzed transition between buoyancy-dominated and turbulence-dominated vertical distributions of polyethylene MPs of three size classes in the upper 100 m layer (Enders et al. 2015). Theoretical model based on settling, biofilm growth, and ocean depth profiles for light, water density, temperature, salinity, and viscosity in addition to simple sinking and rising of particles predicts oscillating vertical movement of MPs related to biofouling and dependent on the particle size and density (Kooi et al. 2017). The influence of MPs physical properties (size, shape, density) on their respective rates of windage, settling, and effects of biofouling were described using simple analytical calculations (Chubarenko et al. 2016).

The Lagrangian approach (or numerical particle-tracking) based on introduction of virtual particles that are allowed to move freely through the hydrodynamic field simulated by ocean circulation models is most frequently used to investigate transport of marine debris (Lebreton et al. 2012; Maximenko et al. 2012; van Sebille et al. 2012) including MPs (van Sebille et al. 2015; Hardesty et al. 2017; Lebreton et al. 2018) on both global and regional scales (e.g., Kubota 1994; Isobe et al. 2014, 2015; Neumann et al. 2014; Critchell et al. 2015; Mansui et al. 2015; Critchell and Lambrechts 2016; Liubartseva et al. 2016; Bagaev et al. 2017; Carlson et al. 2017; Li et al. 2018). Because the majority of field data on MPs in the ocean, available for models' verification, consists of manta–neuston net surface samples, MPs Lagrangian modelling is predominantly focused on the transport of floating tracers at the ocean surface (van Sebille et al. 2015; Hardesty et al. 2017). Advection mechanisms implemented in the modeling frameworks include sea surface currents in all cases, and sometimes other factors, (e.g., Stokes drift (Kubota 1994) and windage (Kako et al. 2014; Bagaev et al. 2017; Lebreton et al. 2018)). Global hydrodynamic models coupled with Lagrangian models to simulate transport of floating litter and MPs were able to predict general patterns of distribution (e.g., accumulation of particles in the "garbage patches"), to some extent seasonal variation of the patch location and its spatial dimensions (Lebreton et al. 2018), and to give estimations on the temporal increase of the MPs' abundances in the patch based on current estimations of mismanaged plastic waste and future plastic waste scenarios (Isobe et al. 2019). However, predicting more or less realistic (absolute) values of MPs pollution is out of scope of such models, yet, also due to the lack

of relevant amount and quality of field data. Known discrepancy between the estimations of annual plastic litter discharge to the oceans (4.8–12.7 million tonnes for 2010 (Jambeck et al. 2015)) and modelled estimates of total weight of the floating plastic litter at the ocean surface (0.27 million tonnes (Eriksen et al. 2014)) is explained by the presence of effective removal processes (Koelmans et al. 2017), such as settling due to biofouling, beaching, incorporation to the ice, etc., which are not specifically considered in the global models.

MPs transport "below the surface" was also studied using the Lagrangian approach in more detailed regional or process-oriented studies. A two-dimensional XZ model accounting for buoyancy, friction, and Stokes drift allowed reproduction of nearshore dynamics of buoyant MPs in the Seto Inland Sea, Japan (Isobe et al. 2014). Terminal rising velocity was parameterized using three formulas for spherical particles of different diameters with the density of polyethylene as of the most common type of MPs according to the field data from the study region. Simulated 3D transport of non-buoyant MPs in the Nazaré canyon, Portugal, revealed up–down-canyon motions of particles under tidal forcing, seasonal variation of MPs transport, and allowed estimation of MPs residence times and average transport rates along the canyon (Ballent et al. 2012, 2013). Settling rates, bedload motion, critical and resuspension shear stress were parameterized using experimental data acquired for one type of the high density preproduction pellets. Effect of buoyancy on the residence time of MPs in Lake Erie, USA and Canada, was evaluated by comparing numerical results of a 3D model with passive Lagrangian tracers with vertical diffusion ("neutral buoyancy particles") and with surface currents only ("buoyant particles") (Cable et al. 2017). Such comparison showed that "neutrally buoyant plastics" were flushed to the coastline several times slower (and remained then in the nearshore zone according to the model setting) than the "floating" ones, thus having greater residence times.

Critchel and Lambrechts (2016) suggested a depth-averaged two-dimensional model coupled with Lagrangian particle tracking, which implements various aspects of MPs transport in the coastal zone. They highlighted greater sensitivity of numerical solutions to the source location, turbulent diffusivity, re-suspension of beached plastics, and the rate of degradation of macroplastics to MPs, than to other processes (Critchel and Lambrechts 2016). In a recent paper by Jalón-Rojas et al. (2019) the most complex Lagrangian-type model mentioned here was tested. The 3D TrackMPD model accounts for many physical processes (advection, dispersion, windage, sinking, settling, beaching, and re-floating of particles) and evolution of MPs properties due to biofouling and degradation. Sensitivity tests revealed that sinking has a dramatic impact on MPs trajectory and fate, followed by turbulent dispersion and beaching. Although such sophisticated models seem to be quite promising, they still require realistic parameterizations and validation based on experimental or field data.

With all the advantages of the Lagrangian-type as a representative of deterministic models in simulating of the behavior of a certain MPs particle or MPs-related process in circulation models, important general questions remain unanswered, like variation in time of general (observed) concentrations of MPs in a region or in the ocean, distribution of MPs between geographical regions or environments (water, sediment, and beach), general removal rate from the surface, etc. These — real-life — aspects require a holistic representation of MPs as an ensemble of particles, despite of the fact that every individual particle has its own basic initial properties (density, size, shape) and its own time rate of change of those processes under the particular environmental conditions. In this regard, probabilistic models and modeling approaches could be considered, accounting for distributions of parameters that are inherent for MPs. As an example, Granado et al. (2019) use Bayesian networks successfully for beach litter forecasting. Various probabilistic methods

are applied in marine sciences for similar (transport and fate of MPs) problems (e.g., an entropy theory is used for suspended sediment transport (Khorram and Egril 2018), ensemble model with uncertainty analysis is applied for forecasting of chlorophyll-*a* concentrations (Shamshirband et al. 2019), recurrent neural network and improved evidence theory predicts water quality (Li et al. 2019), machine learning approach is used to predict the settling velocity of non-cohesive particles (Goldstein and Coco 2014)). Such models may help in answering the most general and most practically needed questions related to the contamination of environments by MPs, considered as an *ensemble of particles*, with a wide range of properties permanently changing with time. This will also require adjustment of field data collection, with an emphasis on acquiring a normalized distribution of parameters rather than absolute concentrations or characteristics.

5. Conclusions and outlook

Global plastic debris models, based on simulation of the ocean circulation, reproduce well the observed accumulation of floating macro- and microplastics in the subtropical gyres and convergence zones. Other MPs-related transport issues still require further field verification. With all the success of the used approaches, some of the key specific features of marine MPs still remain practically not addressed. These features include (*i*) inherently heterogeneous composition of MPs "community" by size, density, and shape (this necessitates also the range or spectrum of dynamical properties of MPs); and (*ii*) intrinsic evolution of all the MPs properties, depending on the particular environmental conditions experienced by a particle, and the time spent in the marine environment. For such a specific contaminant, the deterministic physical models are able to clarify only some questions. This indicates the need to apply not only MPs-specific parameterizations in the circulation models (e.g., the random walk term, or the differential particle loss from the surface), but also alternative approaches, able to deal with ensembles of particles characterized by a spectrum of properties. Initial steps may include the incorporation of probabilistic dependencies into deterministic models (e.g., for sinking or fragmentation processes). Promising alternative approaches may be employment of, for example, ensemble forecasting (where the real solution should fall within the predicted ensemble spread), application of the chaos theory (e.g., use of the entropy theory for suspended sediment transport), ecosystem-scale modeling (e.g., for bioaccumulation), statistical (probabilistic) models, machine learning algorithms.

In summary, numerical modeling of MPs transport and accumulation pattern seems to be one more challenge of the emerging "MPs science", along with methodical issues, monitoring problems, and shortage of field data. A practical way forward today could be a continuation of the "selective" strategy in modelling of MPs behaviour in natural waters, with the main modelled "classes" of MPs still to be determined.

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