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Abstract

Plastic is a widespread marine pollutant, with most studies focusing on the distribution of floating plastic debris at the sea surface. Recent evidence, however, indicates a significant presence of such low density plastic in the water column and at the seafloor, but information on its origin and dispersion is lacking. Here, we studied the pathways and fate of sinking plastic debris in the Mediterranean Sea, one of the most polluted world seas. We used a recent Lagrangian plastic-tracking model, forced with realistic parameters, including a maximum estimated sinking speed of 7.8 m/d. Our simulations showed that the locations where particles left the surface differed significantly from those where they reached the seafloor, with lateral transport distances between 119–282 km. Furthermore, 60% of particles deposited on the bottom coastal strip (20 km wide) were released from vessels, 20% from the facing country, and 20% from other countries. Theoretical considerations furthermore suggested that biological activities potentially responsible for the sinking of low density plastic occur throughout the water column. Our findings indicate that the responsibility for seafloor plastic pollution is shared among Mediterranean countries, with potential impact on pelagic and benthic biota.

Keywords: low-density plastic, marine pollution, water column, seafloor, transport, sinking speed

Synopsis: Minimal information is available on the fate of plastics sinking from sea surface. Here we show that they potentially travel hundreds of km and that accountability for seafloor pollution is shared among Mediterranean countries.

1 Introduction

Plastic pollution represents a major threat to the oceans, causing socio-economic damage and impacting tourism, fishing, and marine ecosystems (1, 2). More than 914 marine species have been reported to accidentally ingest or be entangled by plastic, a number expected to increase in the near future (3). In addition, plastic debris is a vector for invasive species and persistent organic pollutants (4, 5, 6). Around 300,000 metric tons of plastic have been estimated to float at the sea surface (7). However, they only represent a tiny portion of the plastic expected to enter the marine environment each year (8, 9, 10), suggesting that plastic may be even more present in other ocean compartments. Even if the amount of plastic entering the oceans (e.g., via rivers) and the plastic budget are still open questions (10, 11, 12), a growing body of evidence suggests that plastic debris is present not only at the sea surface, but in the whole water column and sea bottom (e.g. refs. 13, 14, 15, 16, 17, 18). Surprisingly, observations reported plastic debris composed of polymers lighter than seawater in the water column, down to at least 1000 m depth (14).

Several processes are responsible for the removal of plastic debris from the sea surface, the most studied being biofouling (19, 20) - the colonisation of plastic debris by bacteria, algae or invertebrates - which increases its relative density. This process continues until, eventually, the debris leaves the surface, and has been documented by laboratory and, recently, by in situ studies (21, 22, 23, 24). Other processes that could lead to the removal of surface plastic debris are aggregation in marine snow and faecal pellet formation after ingestion (25, 26). However, information about the pathways and fate of settling plastic debris is lacking. A common assumption is that plastic debris reaches the seafloor where it has left the surface (27). This assumption has also been used to estimate the seafloor plastic concentration (16).

The aim of this study is to assess the validity of this assumption and to improve the understanding of plastic transport from the sea surface to the sea bottom. This information is essential to solve the plastic budget problem, to understand plastic fate, and for mitigation strategies. For that purpose, we use a Lagrangian plastic-tracking model to analyse (*i*) the potential pathways of plastic debris from the surface to the seafloor, (*ii*) its distribution once it reaches the seafloor, and (*iii*) its potential sources. We focus on plastic whose absolute density is lighter than seawater (referred to as low-density plastic, LDP), usually smaller than 5 mm in size (28). This includes low- and high-density polyethylene and polypropylene, which represent 88% of the LDP debris floating at the surface of the Mediterranean Sea (29), and about half of the produced plastic globally (30, 6). We only focus on the sinking phase of LDP debris, as multiple works already studied its cycle from the moment it is released at sea until it starts sinking (e.g. (31, 27, 32)). We do not consider high density plastic debris, which is expected to sink directly to the seafloor (17, 33) and about which there is little information, nor extremely light items which mainly float at the air-sea interface. Lagrangian methods are widely used to describe the transport of particles in the ocean and are suited to describe the transport of LDP debris. These models can cover areas wider than observations and can describe forward and backward trajectories useful to identify sources and pathways.

This study is a first modeling effort to evaluate the transport dynamics and fate of LDP in the Mediterranean Sea, which is an ideal case study for two main reasons: (*i*) globally, it is one of the most plastic polluted seas (28, 34); (*ii*) its plastic pollution at the sea surface has been intensively studied, allowing us to estimate key physical parameters such as plastic debris sinking speed and to set initial conditions. In particular, we combine recent observations of plastic concentration in the water column, the largest Mediterranean Sea database of floating LDP debris to date, one of the best performing drag models, and estimates of the location and amount of LDP leaving the sea surface. The latter metric was obtained from the first Lagrangian model quantitatively validated in the Mediterranean Sea (31), by assuming that the probability of a LDP leaving the surface increases with the time it spent in water according to a prescribed function. With this information, we are also able to calibrate the model and perform a detailed sensitivity test. We then provide an assessment of the paths and fate of LDP sinking in the Mediterranean sea as well as an estimation of the contribution of the different countries to the coastal seafloor pollution. Finally, we consider the implications of the calculated density differences between generic sinking LDP debris and seawater to assess the role of biological transport throughout the water column, consistent with recent studies.

2 Materials and Methods

2.1 The numerical Lagrangian model: velocity field and trajectory computation

Here, we simulate the path of LDP debris once they start sinking from the surface (due to biofouling or other processes) down to 1000 m. We do not analyse what occurs to debris prior to that (i.e., from the moment it was released at sea to when it left the sea surface), as several studies already investigated this question (e.g. (31, 27, 35)). Hence, this study can be seen as a continuation of those works. To this aim, we used the TrackMPD model (36), an advanced 3D Lagrangian model recently developed to simulate the fate of plastic debris at sea. The TrackMPD model reads the velocity field offline, and computes the trajectories with a Runge-Kutta scheme of order 4-5 both in time and space. Time steps were set to 3 hours. Model initialisation and simulated scenarios are reported in Subsec. 2.3.

The velocity field used to simulate the virtual particle trajectories was a high resolution configuration of the NEMO model (NemoMed36; $1/36° \times 1/36°$) provided at daily intervals, with 50 stretched vertical levels (sigma coordinate system), developed by Arsouze et al. (37). This circulation model was initialised with sea surface temperature and salinity fields and took into account riverine freshwater runoff. It included the vertical component of the velocity field w as well. This product has already been used to simulate 3D virtual plastic trajectories in the Mediterranean Sea (38).

A Stokes component was added to the NemoMed36 velocity field. The Stokes product (MEDSEA_HINDCAST_WAV_006_012), with a spatial resolution of 1/24°, was spatially interpolated over the grid of NemoMed36, and was summed to its upper layer. This allowed us to take into account Stokes drift due to waves at the surface, which affects particles in the first meters of their sinking. Stokes effect indirectly includes windage and is known to affect plastic fate (39).

2.2 Constraining the model settings

A key parameter to study the fate of (biofouled) sinking LDP debris in the water column is its settling speed. In situ observations of LDP settling speed are not available to date. A few studies have measured the settling velocity of biofouled plastic particles under laboratory conditions and found that a large

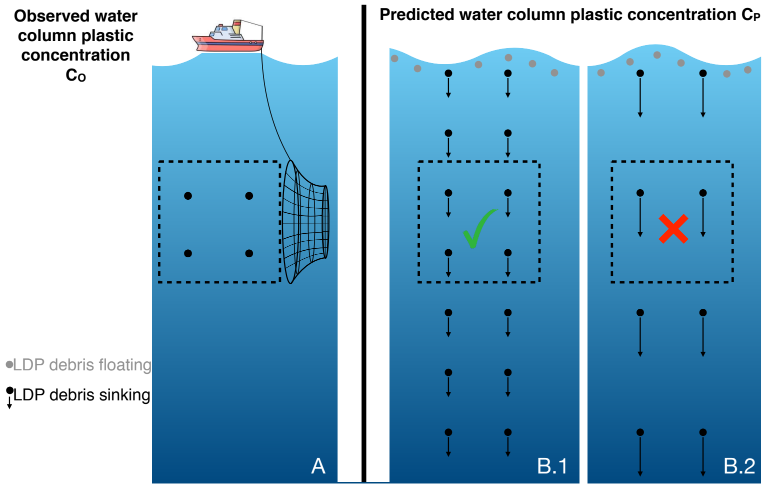

Figure 1: Illustrative scheme of the paradigm used to calculate the sinking speed of LDP debris. The predicted water column LDP concentration C_P decreases when LDP sinking speed increases (panels B.1 and B.2). In order to have C_P equal or greater to a given value observed C_O (panel A), the LDP sinking speed must not be larger than a given value. In this case panel B.1 represents the maximum possible sinking speed of LDP debris. Panel B.2 shows that, if a larger sinking speed is considered, then C_P is lower than C_O.

number of parameters, both physical (e.g. particle polymer, shape and size; water temperature and salinity) and biological (e.g. biofilm growth and density) can impact the sinking speed (33, 24, 19). The large spectrum of sinking LDP and environmental conditions in the ocean make difficult the definition of settling speed in numerical simulations. For these reasons, we calculated a (maximum) representative sinking speed value to calibrate our model, ensuring that mean simulated and observed LDP concentrations in the water column have the same order of magnitude. We assumed that the water column LDP concentration is the result of a linear fall of LDP debris from the sea surface. The larger the LDP sinking speed, the lower the water column LDP concentration for a given rate of plastic submergence from surface (Fig. 1). By taking the lowest water column LDP concentration measured to date as the lower boundary, we can derive a maximum sinking speed, which will be the reference value in the simulations. Details of the calculation are provided in the following subsections. The choice of a linear fall of LDP debris is evaluated by integrating the largest LDP database in the Mediterranean Sea to date ($\sim$ 75,000 debris) and with a drag model (Subsec. 2.5).

2.2.1 Observed water column plastic concentrations C_O

Here, we report the current literature on observed plastic concentrations in the ocean water column (hereafter C_O) in Table 1 (adapted from Liu et al., (40)) which is used to calibrate the simulated plastic concentration in the model. We did not consider the measurements sampled at a depth shallower than 50 m to exclude mixing layer processes, except for the study of Lattin et al., (41), as it is among the only 4 studies which measured the LDP mass concentration (μg/m^3) to date. The highest concentrations were measured by Pabortsava et al., (42) (940 μg/m^3) and Lattin et al., (41) (150 μg/m^3), the lowest by Egger et al., (14) (1.6 μg/m^3, averaged on the 5–1000 m depth layer).

Four studies measured C_O in coastal areas of the Mediterranean Sea (43, 15, 44, 45). Baini et al., (43) and Lefebvre et al., (15) found similar concentrations (0.23$\pm$0.20 particles/m^3; 0.26$\pm$0.33 particles/m^3, respectively). Vasilopoulou et al., (44) and Rios-Fuster et al., (45), sampling in shallower waters ($<$50 m), and close to the coast, found water column plastic concentration $\sim$200 times larger (41$\pm$22 and 67$\pm$52 particles/m^3, respectively). Interestingly, Rios-Fuster et al., (45) found LDP debris down to 50 m depth.

As the studies on the Mediterranean Sea did not measure the mass of plastic debris, we compared them with the concentrations of Egger et al., (14) which reported the lowest concentrations in μg/m^3 and particles/m^3 and can serve as a reference value of minimum plastic concentration in the water column to obtain in our simulations. Egger et al., (14) collected LDP debris larger than 500 μm, 76% of plastic debris collected by Baini et al., (43) was larger than 500 μm, while the mean size of plastic debris found by Lefebvre et al., (15) was (1.81$\pm$1.42) mm, indicating that most of debris was larger than 500μm. Therefore, Egger et al., (14), Baini et al., (43), and Lefebvre et al., (15) studies collected plastic debris of similar size range, but the concentration measured by Egger et al., (14) was $\sim$200 times lower (0.001 particles/m^3). Hence, Mediterranean water column plastic concentrations are probably larger than those measured by Egger et al., (14) also when considering the mass (μg/m^3). This is also corroborated by the fact that the Mediterranean is one of the seas most affected by plastic pollution, with surface LDP concentrations comparable to those found in the North Pacific Garbage Patch (NPGP) (28, 34, 43). Biofouling was predicted to be larger in several regions of the Mediterranean Sea than in the oligotrophic NPGP area (32), suggesting that more LDP particles might leave the surface in the Mediterranean. However Mediterranean Sea studies (43, 15, 44) sampled lower water volumes, inducing possible biases towards higher concentrations (46, 40, 47). Therefore, we considered a minimum water column LDP concentration $C_{O_{MIN}} = 1.60$ μg/m^3

Sampling regions	Methods	Samples pretreatment and polymer analysis	Volume/sample (L)	Depth (m)	Mesh size of nets [a] or filters (sieves) [b] (µm)	MPs size (mm)	Plastic concentration (items/m^3)	Plastic concentration (µg/m^3)
Arctic Central Basin (Kanhai et al., 2018, ref. 48)	CTD sampler	Direct filtration; µ-FT-IR spectrometer	7-48	8-4,370	250 [b];1.20 [b]	0.25-5.00	0–100	/
Mariana Trench (Peng et al., 2018, ref. 49)	CTD sampler	Direct filtration; Raman spectroscopy	35-180	2,673-10,904	0.22 [b]; 0.30 [b]	1.00-3.00 (mostly) [c]	$(2–14)\cdot 10^6$	/
Baltic Sea proper (Bagaev et al., 2017, ref. 50)	CTD sampler	Direct filtration; Combination of visual identification and UV lamp	7.79-30	0.5-217.5	174 [b]	N/A [d]	80 (fibers)	/
Sumba coastal waters, Indonesia (Cordova et al., 2018, ref. 51)	CTD sampler	Direct filtration; FT-IR spectrometer	10	5-300	0.45 [b]	0.30-1.00 (mostly) [c]	44±25	/
Baltic Sea, Sweden (Gorokhova, 2015, ref. 52)	Plankton net	Direct observation; visual identification	6,200-9,200	0-100	90 [a]	0.05-0.30 (mostly) [c]	2000–3000	/
Tuscany coastal waters, Mediterranean Sea (Baini et al., 2018, ref. 43)	Plankton net	Direct observation; FT-IR spectrometer	500-30,000	2-120	200 [a]	1-2.5 (mostly) [c]	0.26	/
Gulf of Lions (Lefebvre et al., 2019, ref. 15)	Plankton net	Direct observation; FT-IR spectrometer	N/A [d]	0-100	200 [a]	0.24-4.93	0.23±0.20	/
Cyprus coasts (Vasilopoulou et al., 2021, ref. 44)	Plankton net	Direct observation; DMIL microscope and SLX-3 stereoscope	N/A [d]	0-50	200 [a]	0.2-5	41.31±22.41	/
Spanish coast (Rios-Fuster et al., 2022, ref. 45)	Niskin bottles	Direct filtration; FT-IR spectrometer on 10% of items	5	5-50	1.2 [b]	0.1-8.4	1860±1430 (67 without fibers)	/
Northeast Pacific (Goldstein et al., 2013, ref. 53)	Plankton net	Direct observation; FT-IR spectrometer	N/A [d]	0-210	202 [a]	N/A [d]	0.05	/
Santa Monica Bay (Lattin et al., 2004, ref. 41)	Multi-net trawls	Freshwater floatation; No polymeric identification	N/A [d]	0-14.80	333 [a]	2.80-4.75 (mostly) [c]	/	150
North Pacific Garbage Patch (Egger et al., 2020, ref. 14)	Multiple Opening and Closing Net	Direct observation; Raman spectroscopy	136,000-5,039,000	0-2,003	333 [a]; 500-15,000 [b]	N/A [d]	0.001	1.60
Baltic Sea, Russia (Zobkov et al., 2019, ref. 54)	Submersible pump	Organics removal prior to the refiltration (174µm); µ-Raman spectroscopy	2,500-3,500	0.5-91	174 [b]	0.17-1.00 (mostly) [c]	32	/
Korean coastal waters (Song et al., 2018, ref. 55)	Submersible pump	NaCl solution floatation prior to the refiltration; µ-FT-IR spectrometer	100	3-58	20 [b]; 5 [b]	0.02-5.00	423 (50 if only particles < 5 mm)	/
Monterey Bay (Choy et al., 2019, ref. 13)	In-situ filtration device	Direct observation; Raman spectroscopy	1,007-2,378	5-1,001	100 [b]	N/A [d]	1–10	/
HAUSGARTEN observatory (Tekman et al., 2020, ref. 56)	In-situ filtration device	Organics removal prior to the filtration; µ-FT-IR spectrometer	218-561	1-5,351	32 [b]	0.01-0.15	95±85	/
West Pacific and East Indian Ocean (Li et al., 2020, ref. 47)	In-situ filtration device	Wet peroxide oxidation; µ-FT-IR spectrometer	10,000	2–4001	60 [b]; 1.60 [b]	0.03–6.33 (mean 0.67)	0.2–3.5	/
Atlantic Ocean (Pabortsava et al., 2020, ref. 42)	In-situ filtration device	Organics removal prior to the filtration; µ-FT-IR spectrometer	500–1500	0–201	55; 25	0.032–0.651	2300	940
Atlantic Ocean (Zhao et al., 2022, ref. 57)	Multiple Opening and Closing Net; WTS-LV pump	Direct observation; visual identification, µ-FT-IR spectrometer	440-1765	0-5200	200 [a]	SMP: <100µm; LMP: > 300µm	SMP: 0-244.3; LMP: 0-0.011	SMP:0-20.83; LMP: 0-15.3

a: mesh size of net used in the reference.
b: mesh size of filter or sieve used in filtering process.
c: only size range of MPs was presented in the literature.
d: N/A represented no available information.

Table 1: Literature on Mediterranean water column plastic concentration (adapted with permission from Liu et al., (40), Copyright 2020 Elsevier).

2.2.2 Annual estimates of LDP plastic entering the Mediterranean Sea.

We defined N_{TBY} as the number of tons of LDP leaving the Mediterranean surface every year due to biofouling or other processes. N_{TBY} was obtained assuming that ~11.5% of the amount of floating plastic debris entering the Mediterranean Sea every year from coastal sources or vessel discards (N_{LDP_Y}) left the sea surface during that period due to biofouling or other processes. This percentage was based on the study by Baudena et al., (31), and was similar to the value obtained in a previous study (9.2 %, 27). Thus:

$$N_{TBY} = 0.115 \cdot N_{LDP_Y} \ . \tag{1}$$

N_{LDP_Y} was estimated using the findings of ref. (8). The authors calculated the number of tons of plastic debris entering the global ocean in 2010. Considering that ~50% of plastic is LDP (mainly low- and high-density polyethylene and polypropylene, 30, 6), about 100,000 tons of LDP debris were estimated to enter the Mediterranean Sea in 2010. This quantity includes also extremely light and large LDP not considered here. However, these constitute less than 1% of items collected in the Mediterranean Sea (28). Overall, the assumed quantity represents a compromise between recent estimates, both lower (58, 59) and higher (11, 10). Furthermore, we stress that this value was adopted in recent studies modelling plastic-debris dispersion in this basin (27, 38, 31). The sensitivity of the results with respect to this parameter is studied by simulating a further scenario (Subsec. 2.3).

2.2.3 Estimation of the maximum sinking speed of LDP debris SS_{MAX}

N_{TBY} can be converted into a flux F of LDP mass leaving each m^2 of sea surface every day by considering the Mediterranean surface ($\simeq 2.5\times10^{12}$ m^2):

$$F = \frac{N_{TBY} \cdot 10^6}{365 \cdot 2.5 \cdot 10^{12}} \ \frac{g}{d\, m^2} \ = 1.10 \cdot 10^{-9} \, N_{TBY} \ \frac{g}{d\, m^2} \tag{2}$$

The predicted concentration of the falling particles in the water column C_P (expressed as g/m^3) depends on their vertical sinking velocity SS. We stress that the flux of particles sinking from the sea surface is a release of individual particles (the LDP debris) at discrete intervals (Fig. 1), similar to a *rain effect*. Thus, the concentration C_P was considered as the ratio between F (Expr. 2) and SS (expressed in m/d, Fig. 1):

$$C_P = \frac{F}{SS} = \frac{N_{TBY}}{SS} 1.10 \cdot 10^{-9} \ \frac{g}{m^3} \tag{3}$$

Expr. (3) implies that the more slowly LDP debris sinks, the greater the resulting water column plastic concentration is, and vice versa (Fig. 1, panels B.1 and B.2). If N_{TBY} increases (and we assume the same sinking speed), so does the water column plastic concentration. Here, C_P was derived considering the

same sinking speed and mass for all the particles. We also derived an expression for the predicted concentration by considering individual sinking speed and mass, and showed that these did not affect C_P, nor the conclusions of our paper (Supporting Information S.1).
By inverting Expr. (3), we could obtain an expression for the sinking speed of the LDP debris, SS:

$$SS = \frac{N_{TBY}}{C_P} 1.10 \cdot 10^{-9} \ , \tag{4}$$

from which it was possible to obtain a maximum estimate of the settling speed SS_{MAX} in the Mediterranean Sea. To do so, we considered the estimate of N_{TBY} for the Mediterranean Sea (11,500 tons/year, Subsec. 2.2.2) and imposed C_P=$C_{O_{MIN}}$ (1.60 μg/m^3, Egger et al., (14), Subsec. 2.2.1):

$$SS_{MAX} = \frac{N_{TBY}}{C_{O_{MIN}}} 1.10 \cdot 10^{-9} \ , \tag{5}$$

By substituting these values in Expr. (5), we obtained SS_{MAX}=7.8 m/d.
In summary, in order to have a minimum water column plastic concentration of 1.60 μg/m^3, considering a maximum load of 11,500 tons of biofouled LDP leaving the surface each year, LDP debris should sink with a maximum settling speed of 7.8 m/d. Considering a greater water column plastic concentration, and/or a lower load of biofouled LDP per year would lead to a lower sinking speed.
We stress that using the C_O values of Baini et al., Lefebvre et al., Vasilopoulou et al., (43, 15, 44) (converted in plastic mass) as $C_{O_{MIN}}$ rather than those of Egger et al., (14), SS_{MAX} would have been lower. Similarly, using a lower N_{LDP_Y} (and thus a lower N_{TBY}) would have lead to a lower SS_{MAX} value. Nevertheless, we considered a two fold value of N_{TBY}, by carrying out a simulation with a sinking speed set to twice SS_{MAX} (15.6 m/d, Subsec. 2.3).

2.3 Model initialisation and simulated scenarios

The simulated particles were considered representative of LDP debris of all sizes, with the exception of extremely light foamed plastics (such as polystyrene foam) or air filled objects which tend to stay suspended at the air-sea interface. The latter represent less than 1% of plastic debris collected in the Mediterranean Sea (28). In general, 95% of LDP debris collected in the Mediterranean Sea are less than 5 mm in size (28). LDP debris were considered to sink, assuming that biofouling, weathering or other processes decrease the buoyancy of these particles. Virtual LDP particles were released at the surface at daily intervals between January 1, and December 31, 2010. This time interval is consistent with the residence time of plastic debris at the Mediterranean surface (31, 27). To initialise the particle starting positions, we used the results of Baudena et al., (31), which simulated the path of LDP debris from their release at sea (by coastal cities, river mouths, and vessels) to the moment they started sinking. In that work, the authors assumed that the probability of a simulated LDP particle leaving the sea surface (due to e.g. biofouling, etc.) increases with the time spent in water. This probability peaked in correspondence with the biofouling time (the period of time necessary to induce sinking, see Supplementary Fig. 2 in Baudena et al., 31). In this way, the authors calculated the Mediterranean surface sinking rate (the amount of LDP debris that disappears from the surface each day in a square kilometer of sea surface). To strengthen that metric Baudena et al., (31) considered different biofouling times (between 50–200 days, based on literature values) and obtained similar estimates when considering 16 different parameterisations and an ensemble average. Hence, in our study, particles were released proportionally to the Mediterranean surface sinking rate, i.e. the larger the surface sinking rate in a region, the larger the number of particles released there. The choice of the dataset of Baudena et al.(31) is further motivated by the fact that it was the first Lagrangian model quantitatively validated in the Mediterranean Sea.

7,652,197 virtual particles were released in total, using four sinking speed values (7.8 m/d, Scenario 1; 4 m/d, Scenario 2; 2 m/d, Scenario 3; 1 m/d, Scenario 4; Table 2). The choice of the maximum value used for the sinking speed (SS_{MAX}=7.8 m/d) is motivated in Subsec. 2.2. All virtual plastic particles of a given scenario were advected for a time period (provided in Table 2) long enough to reach at least 1000 m depth. For scenario 4 (sinking speed of 1 m/d), particles were advected for 1000 days. At the end of the simulation, 90% of particles reached 1000 m depth or were deposited. The 10 % left were at an average depth of 900±100 m, thus very close to reaching 1000 m depth. Thus, we use their final position to calculate the seafloor concentration (Subsec. 2.4). The particles were considered as non-inertial passive tracers with a constant sinking velocity, which were transported by currents and by isotropic horizontal and vertical diffusion (diffusivity coefficient K_h and K_v, respectively).

We used K_h=10 m^2/s and K_v=5$\cdot$10^{-5}m^2/s, in line with the values used in previous plastic studies (27, 36, 58, 31). In order to test the sensitivity of the results to the choice of the diffusivity coefficients, different K_h and K_v values were used (Scenarios 5–8, Table 2). In order to evaluate the role of the vertical component of the current field w on the simulated LDP concentration, w was set to zero in scenario 9 (Table 2). Scenarios 5–9 were run with the same sinking speed of Scenario 1, namely 7.8 m/d. In order to evaluate the sensitivity of the results with respect to a potentially larger SS_{MAX} (which could be due to a larger flux of LDP leaving the surface, Subsec. 2.2.3), in Scenario 10 we used a sinking speed of 15.6 m/d. Finally, in Scenario 11 we evaluated the sensitivity of the results with respect to the release conditions: to do so, the surface sinking rate of Baudena et al. (31) was varied at each location of ±10% randomly. This new sinking rate was used to initialise the particle release locations of that scenario.

2.4 Model output analyses

One of the limitations in simulating particle trajectories from the surface to the seafloor in a deep basin such as the Mediterranean Sea is the elevated computational cost. To overcome this issue, we calculated the concentration of the simulated particles on a virtual layer at 1000 m depth. In the regions with a seafloor shallower than 1000 m, we kept the original depth (provided by the velocity field domain (37)). 1000 m was chosen as the reference depth because it is usually considered as the upper boundary of the deep sea. Thus, LDP reaching this layer are therefore considered as sequestered in the deep sea. Furthermore,

Nº Scenario	Current velocity field	Vertical component of currents included (yes/no)	Sinking speed particles (m/d)	Release period	Advective period (days)	K_h (m^2/s)	K_v (m^2/s)	Notes
S1	NemoMed36	yes	7,8	1 Jan-31 Dec 2010	450	10	$5 \cdot 10^{-5}$	
S2	NemoMed36	yes	4,0		550			
S3	NemoMed36	yes	2,0		1050			
S4	NemoMed36	yes	1,0		1050			
S5	NemoMed36	yes	7,8		450	5	$5 \cdot 10^{-5}$	
S6	NemoMed36	yes	7,8		450	15		
S7	NemoMed36	yes	7,8		450	10	$1 \cdot 10^{-5}$	
S8	NemoMed36	yes	7,8		450		$10 \cdot 10^{-5}$	
S9	NemoMed36	no	7,8		450	10	$5 \cdot 10^{-5}$	
S10	NemoMed36	yes	15,6		225	10	$5 \cdot 10^{-5}$	
S11	NemoMed36	yes	7,8		450	10	$5 \cdot 10^{-5}$	Surface sinking rate varied of ±10%

Table 2: Parameters used for each of the ten simulated scenarios.

this assumption allowed us to simulate sinking speeds down to 1.0 m/d, which would not have been possible for further depths due to computational costs. The concentration on a deeper virtual seafloor, at 2000 m depth, was calculated for scenarios 1–3 only, and is reported in Supporting Information S.2.
Further, we calculated the concentration of particles deposited less than 20 km from the coast (hereafter the coastal strip). We chose this distance because it is associated with the inner average continental shelf, exploited by industrial and recreational fishery and essential for tourism activity (60, 61). This concentration was calculated for each Mediterranean country. For each deposited particle we identified if it was released by a land (coastal city or river mouth) or a sea (vessel discard) source. To this aim, we tracked each particle backward in time from the seafloor to the surface (using our trajectories) and from the surface to its release source (using the trajectories calculated by Baudena et al., 31). If the original release source was land based, we determined the corresponding source country as well. The plastic sources considered were those used in Baudena et al., (31), coastal cities (62), river mouths (63), and vessel discards (64).
We analysed the connectivity between Mediterranean surface and seafloor by considering the starting and final position of each particle and by calculating a connectivity matrix (65) at two different resolutions. Further details are reported in Supporting Information S.3.
To understand the pathways of the LDP particles reaching a zone of high LDP concentration, located north-east of the Balearic archipelago (Fig. 3A), we calculated their crossroadness (66, 31). This metric provides, for a given point, the percent of the trajectories that passed in its neighborhood (defined as a circle of radius 0.1°). Further details are reported in Supporting Information S.4.

2.5 Density of sinking LDP debris from a drag model

In Subsec. 2.2.3, we estimated a maximum sinking speed SS_{MAX} of LDP debris equal to 7.8 m/d. Here we calculate the difference of LDP and seawater densities necessary to obtain such value. To this aim we use (i) the largest LDP debris dataset collected in the Mediterranean Sea to date and (ii) the drag model of Dioguardi et al., (67).

Mediterranean Sea LDP debris database

We exploited the largest LDP field dataset in the Mediterranean Sea to date, collected during the Tara Expedition (122 stations, ref. 28, https://zenodo.org/record/5538238). This dataset provided the ferret (considered representative of the particle size d_p), sphericity Φ, and circularity χ of each LDP debris collected at the sea surface (75,030 items in total). We used these debris physical properties to estimate the velocity at which they are expected to sink once biofouled.

Drag model to calculate the sinking velocity

Van Melkebeke et al., (68) evaluated eleven drag models estimating the vertical sinking velocities of plastic particles with different characteristics, such as size, shape, and density. The best performing drag model was the one reported in (67, average error of 13.20%), which calculated the vertical sinking speed SS_{DM} as follows:

$$SS_{DM} = \sqrt{\frac{4gd_p\Delta\rho}{3C_d\rho_{SW}}}\ , \qquad (6)$$

where g is the gravity acceleration (9.81 m/s^2), d_p is the particle size, $\Delta\rho$ is the difference between the particle density and that of seawater, ρ_{SW}. C_d is the particle drag coefficient, which the authors expressed as follows:

$$C_d = \frac{24}{Re_p}\left(\frac{1-\Psi}{Re_p}+1\right)^{0.25} + \frac{24}{Re_p}\left(0.181\,Re_p^{0.65}\right)\Psi^{-Re_p^{0.08}} + \frac{0.4251}{1+\frac{6881}{\Re_p}\Psi^{5.05}}\ ,$$

where Re_p is the particle Reynolds number.

$$Re_p = \frac{\rho_{SW}SS_{DM}\,d_p}{\mu_f}\ ,$$

with μ_f being the water dynamic viscosity. The range of validity of Eq. (6) is 0.03< Re_p <10^4, which was respected with the parameters used. Ψ is the shape factor, defined as the ratio between the particle sphericity Φ and circularity χ. The vertical sinking velocity SS_{DM} depends therefore on the six parameters, d_p, $\Delta\rho$, ρ_{SW}, μ_f, Φ, and χ. We considered a seawater density ρ_{SW}=1027 kg/m^3 and a viscosity μ_f=0.00109 Pa·s; the latter is representative of a seawater temperature of 20° (https://www.engineeringtoolbox.com/sea-water-properties-d_840.html), which is the mean Mediterranean surface temperature. As d_p, Φ, and χ we used the d_p, Φ, and χ of each of the 75,030 debris collected during the Tara expedition (Subsec. 2.5). Therefore, $\Delta\rho$ represents the only unknown parameter in Expr. 6.

Different $\Delta\rho$ values were tested in Expr. (6), ranging from 0.001 to 0.1 kg/m^3. The Matlab function provided in Dioguardi et al., (67) was used to

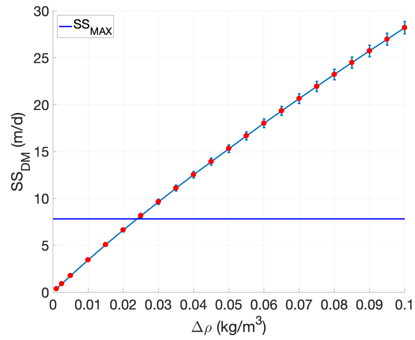

Figure 2: Mean theoretical sinking speed SS_{DM} of 75,030 LDP debris (y-axis, subsec. 2.5) as a function of the difference between the density of the biofouled LDP debris and the seawater $\Delta\rho$ (x-axis). SS_{DM} are reported as mean values (red dots) with standard deviation (errorbar). The blue horizontal line identifies the SS_{MAX} value calculated in Subsec. 2.2.3 (7.8 m/d).

calculate 75,030 SS values, which were then averaged together. We found that SS_{DM} matched SS_{MAX} for $\Delta\rho \simeq 0.025$ kg/m^3 (Fig. 2). The $\Delta\rho$ obtained did not change considerably when the predicted water column plastic concentration C_P was calculated considering also the individual mass of each of the 75,030 LDP debris (Supporting Information S.1). We also performed a sensitivity test to analyse the robustness of the mean SS_{DM} while varying the three other parameters d_p, ρ_{SW}, and μ_f. These calculations indicated that they did not affect the mean SS_{DM} value (Supporting Information S.5) significantly.
Overall, above considerations show that only a minor excess density of 0.025 kg/m^3 compared to seawater is required to reach a maximum sinking speed of 7.8 m/d. Smaller $\Delta\rho$ are related to lower sinking speeds.

3 Results

3.1 Concentration of deposited LDP debris and distance travelled during its sinking

The spatial distribution of particles deposited on the 1000 m depth virtual layer (Fig. 3B, C, D, and E;) was consistently different from the distribution of particles leaving the surface (Fig. 3A) for all the sinking speeds considered. When reducing the sinking speed, the difference increased (Fig. 3B to Fig. 3E). This result was particularly visible in the western Mediterranean: while large amounts of virtual particles were predicted to leave the surface north of the Balearic archipelago, the concentration at 1000 m depth in this same region was relatively low for all the scenarios considered. Conversely, the 1000 m concentration was greater north-east of the Balearic archipelago, in a region where the surface sinking rate was lower (cyan rectangle in Fig. 3A). This region, covering 1.63% of the Mediterranean surface, contained 1.36% of the virtual LDP particles leaving the Mediterranean surface. Notably, at 1000 m depth, it accumulated the 3.07 %, 4.68%, 6.66%, and 7.36% of the virtual LDP particles for scenarios 1–4, respectively. The LDP particles deposited there were mostly

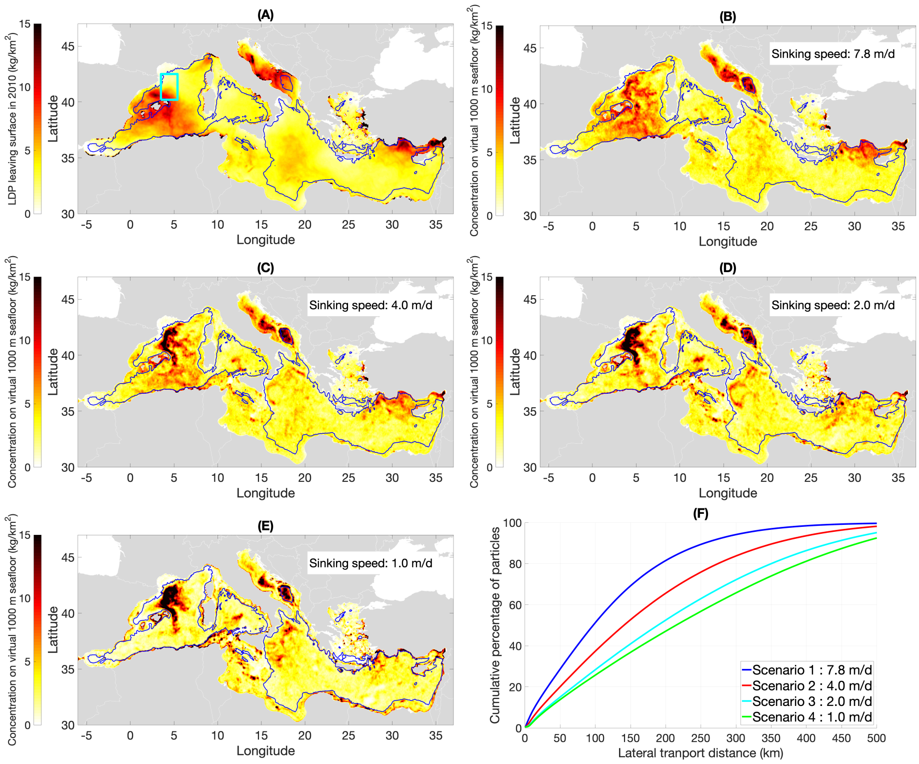

Figure 3: **(A)**: amount of virtual LDP particles that left the Mediterranean surface in 2010 (the surface sinking rate integrated between January 1, and December 31, 2010; kg/km^2; adapted with permission from Baudena et al., (31), Copyright 2022 Nature). **(B–E)**: concentration of virtual LDP particles released in 2010 deposited on a virtual seafloor at 1000 m depth, for scenario 1, 2, 3, and 4, respectively. The virtual 1000 m seafloor was built by considering all the seafloor locations deeper than 1000 m as 1000 m deep. The blue lines in panel A–E indicate the 1000 m isobath. **(F)**: cumulative pdf of the lateral transport distance (the distance between the location in which the particle left the surface and the location in which it reached the virtual 1000 m seafloor) of the 7,652,197 virtual particles released in 2010, for scenario 1 (blue line), 2 (red line), 3 (cyan line), and 4 (green line). The cyan rectangle in panel (A) shows the region considered for the statistical analyses.

from the south and the east of this region, as supported by supplementary analyses (Supporting Information S.4)

The connectivity analysis between the locations where particles left the surface and where they reached the seafloor indicated that between 90–95% of the particles (scenarios 1–4) deposited at a certain location (defined by a ∼111 km size square) started to sink in other regions (Supporting Information S.3). In the Southern Adriatic Sea, where a hotspot of enhanced LDP accumulation was identified, between 32–50% of the particles were from adjacent regions. Other LDP accumulation regions were detected across the basin, such as in the Tyrrhenian and Ionian Seas, in the Strait of Sicily, or in the Eastern Mediterranean.

The spatial variability between the concentration of virtual LDP particles leaving the surface and reaching the 1000 m virtual seafloor was confirmed also by the cumulative pdf of the lateral transport distance (Fig. 3F). This was defined as the distance between the locations at which particles left the surface and those where they reached 1000 m depth. The mean lateral transport distance for scenario 1 was 119±99 km; for scenario 2: 172±136 km; for scenario 3: 232±177 km; and for scenario 4: 282±218 km. The percentage of virtual LDP particles with a lateral transport distance larger than 100 km was 30% for scenario 1, increasing to more than 60% for scenario 4. This conclusion was robust to changes in the horizontal and vertical diffusivity, to the exclusion of the vertical component of the velocity field, to the increase of the sinking speed of the LDP particles up to the double of SS_{MAX}, and to changes of the surface sinking rate used to initialise the particle starting locations (Supporting Information S.6).

In situ observations of seafloor plastic concentration in the Mediterranean are sparse in space and time and differ in methodology. In addition, these collected all types of plastic, while here we focus on LDP only. Hence, a quantitative comparison with our model results is currently not possible. Despite these constraints, our model predictions qualitatively agree with in situ measurements (databases and their references in Supporting Information S.7). Largest seafloor concentrations are reported in the Central Adriatic Sea, close to the Turkiye shore, and in the Balearic archipelago, where our model predicts the largest LDP concentrations. Conversely, the lowest concentrations were measured on the Eastern Sardinia shelf, in the Northern Tyrrhenian Sea, and close to Alicante (Spain), in agreement with our model predictions (further details in Supporting Information S.7).

3.2 LDP concentration on the coastal strip of Mediterranean countries

The concentration of particles deposited on the coastal strip (i.e. in this study within 20 km from the coast, Subsec. 2.4) ranged between 5.5–8.9 kg/km^2 (scenarios 1–4), which was between 11–69% larger than the concentration of particles sinking from the sea surface above (i.e., less than 20 km from the coast; 5.2 kg/km^2). The bottom coastal strips of Algeria and Turkiye showed the largest particle concentrations (15.6 and 13.9 kg/km^2, respectively, obtained from an ensemble average of scenarios 1–4; Fig. 4).

We tracked the particles deposited on the bottom coastal strip backward in time (based on Baudena et al., (31), Methods), and we found that ∼60% were from vessel discards, while ∼40% were released from land sources. On average, ∼20% of the particles deposited on the coastal strip of a given country were from the land sources of the same country, while ∼20% were from other countries, with some variability. For example, 53% of particles deposited on the coastal strip of Cyprus were from neighbouring countries. The corresponding values for Croatia and Syria were 47 and 46%, respectively. Conversely, 73% of particles deposited on the coastal strip of Turkiye were from Turkish coastal sources. Finally, 83% of the particles deposited on the coastal strip of Egypt were from vessel discards. This proportion was similar for particles deposited on the coastal strip of Malta and Libya (84 and 83%, respectively). Results obtained for scenarios 1–4 separately (Supporting Information S.8) were consistent with this pattern.

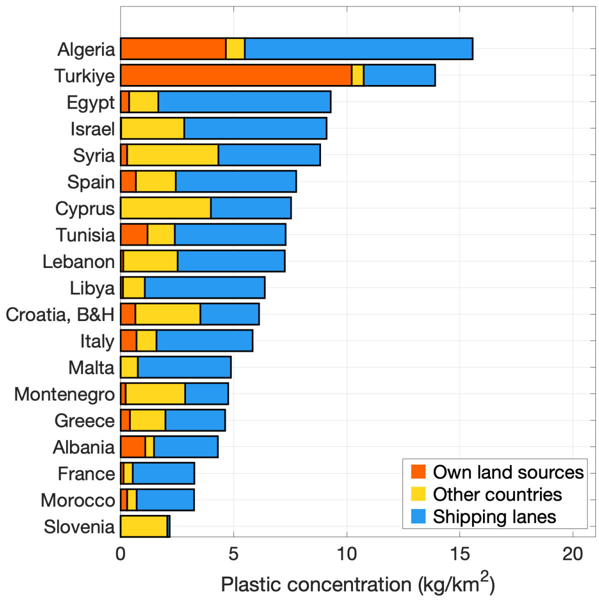

Figure 4: Concentration of particles released in 2010 which deposited on the coastal strip (defined as the region less than 20 km from the coast) of the different Mediterranean countries, obtained from the ensemble average of scenarios 1–4. For each country, the red rectangle represents the amount of particles deposited on its coastal strip which were released from its own land sources, the yellow rectangle represents those released from other countries, and the blue rectangle those directly released at sea.

4 Discussion

4.1 Pathways and fate of sinking LDP debris

The study of the pathways of sinking LDP debris has highlighted that the locations where LDP debris left the surface did not coincide with the locations where it was found at depth, as assumed by recent studies (27, 16). This stressed the importance of a three dimensional approach to study plastic dispersion (69). When neglecting the vertical component of the currents, the accumulation of virtual LDP particles in specific regions slightly decreased, e.g. for the area north-east of the Balearic archipelago (last panel in Fig. S.6). When considering a virtual seafloor down to 2000 m (Supporting Information S.2), the accumulation slightly decreased as well. This indicated an important role played by vertical current shear and by horizontal stirring on the accumulation of LDP particles, in coherence with recent studies (70, 38). For instance, we observed a region of LDP particle accumulation at 1000 m depth in the Adriatic Sea. This matched remarkably well with a recently discovered persistent bottleneck structure in the circulation of this sea (71), which may be responsible for this LDP accumulation. In a specific region north-east of the Balearic archipelago, the number of LDP debris deposited increased for slower sinking speeds. Crossroadness analyses indicated a possible mechanism of particle accumulation at depth (Supporting Information S.4): particles are transported toward this area due to the regional converging circulation. At the same time, particles sinking more slowly spend more time suspended in the water column, traveling larger distances. Hence, the probability they end up in that region increases.
Our simulations also pointed to the fact that a large fraction of LDP particles could potentially reach the deep sea: 48–63 % of the virtual LDP particles leaving the surface were transported to 1000 m depth, while 38–46 % reached 2000 m in Scenarios 1–3.
Even if further information is needed to quantitatively validate our model outputs, our results were in general agreement with in situ observations of seafloor plastic concentration (Supporting Information S.7), corroborating our findings. In addition, our results were robust with respect to horizontal and vertical diffusivity changes, removal of the vertical velocity current component, variation of the LDP debris sinking speed, and changes of starting sinking locations (Supporting Information S.6).

4.2 Bottom coastal LDP pollution.

The estimated LDP concentration on the bottom coastal strip increased for decreasing sinking speeds (from 5.6 kg/km^2 for 7.8 m/d sinking speed to 8.9 kg/km^2 for 1 m/d). This concentration was 11–69% higher than the concentration of LDP particles leaving the sea surface in the same region (5.2 kg/km^2). This indicated that currents at depth tend to propel debris towards coastal regions. This agrees with the pattern of surface currents, which are expected to retain LDP debris in the majority of Mediterranean coastal regions (27, 31). Notably, $\sim$ 20% of the particles deposited in the coastal region of a given country were from neighbouring countries, while$\sim$ 60% were from maritime sources. The high percentage of deposited LDP debris originating from maritime sources (60%) is due to the fact that these particles spend more time at the surface than particles released from land sources, which tend to strand quickly and are therefore less biofouled. In general, while previous studies suggested that each country is the primary responsible for the plastic pollution of its own beaches (27, 31), LDP debris on the bottom coastal strip seems to be from multiple Mediterranean countries and especially from shipping lanes. For instance, Egypt has been reported to have large rates of beaching plastic debris, mostly released from its own land sources (27, 31): however, we suggest that its coastal strip pollution is mainly due to LDP particles released at sea or from other countries. Overall, LDP pollution emerges as a shared problem in the Mediterranean basin, as particles polluting the bottom coastal strip were mostly released at sea or from distant countries.

4.3 Implications from comparing the maximum sinking speed SS_{MAX} with the sinking speed from a drag model SS_{DM}

Several biological processes are suspected to affect LDP debris throughout the water column. Kooi et al., (20) theorised a progressive colonisation of LDP, which is expected to decrease and eventually cease below the euphotic layer (due to mineralization or scraping of plastics by copepods; 72). Also, fragmentation fosters the slowdown of settling LDP debris, as the vertical sinking velocity decreases with decreasing debris size (73). Hence, LDP debris may be resuspended and colonised again (20), as modelled by Lobelle et al., (32), Fischer et al., (70), and Tsiaras et al., (74). Other biological activities can occur below the euphotic zone, such as the biofilm formation via heterotrophic organisms that do not necessarily require light to grow (70). Aggregation in marine snow and consumption by zooplankton may cause plastic debris to sink, whereas remineralization at depth would remove organic mass from plastic debris, making it rise again (75, 76). Zooplankton faecal pellets usually do not reach the seafloor due to coprophagy (see a review in 77), potentially releasing buoyant plastic debris. In addition, faecal pellets containing plastic debris are more subject to fragmentation (25), potentially enhancing resuspension. Chemical processes can affect the buoyancy of LDP debris as well, especially in regions where biological activities are limited, such as in the NPGP. For example, weathering of debris causes hydrogen abstraction with oxygen substitution penetrating deeper into the polymeric matrix, altering its absolute density (78). Crystallinity also increases over time during degradative attack of the amorphous regions leading to an increase in density (79).

However, these processes, potentially responsible for the sinking of LDP debris, have been observed only in laboratory studies or, in situ, uniquely at the sea surface (21, 22, 23, 24), with the exception of the recent observation of LDP in marine snow (80). The presence of LDP at depth is hence poorly understood.

To investigate this question, we used a drag model and the largest collection of Mediterranean LDP debris to date. We calculated the difference of density between sinking LDP debris and seawater necessary to obtain a theoretical sinking speed SS_{DM} equal to SS_{MAX}=7.8 m/d. We obtained $\Delta\rho$=0.025 kg/m^3. As seawater density increases with depth (using a conservative estimate, about 1 kg/m^3 every 100 m; de la Fuente et al., (81)) LDP debris should stop sinking after 2.5 m if its density does not increase meanwhile. Therefore, our results suggest that the biological processes proposed by the aforementioned studies (e.g. 20, 75, 76) occur also below the surface.

This conclusion is corroborated by the fact that currents, in the Mediterranean Sea, seems unable to transport LDP debris at great depths. Indeed, Soto-Navarro et al., (38) have shown that vertical currents redistribute plastic debris mainly in the first 100 meters of the water column. Onink et al., (82) predicted a transport to greater depths primarily due to internal tides, but only for a small proportion of LDP debris (<1%). Tsiaras et al., (74) studied the water column plastic concentration in some regions of the Mediterranean Sea, and found that model predictions were orders of magnitude lower than observations. In addition, de la Fuente et al., (81) argued that neither the vertical nor horizontal currents affect the water column debris concentration in the Mediterranean Sea significantly (see in particular their Figure 5). Fischer et al., (70) suggested a larger impact of vertical transport, but only when associated with an intense biological activity.

All in all, our results point to the fact that LDP debris may persist in the water column for time windows larger than previously suspected (e.g. 27, 16) and travels for hundreds of kilometers. This can increase their bioavailability and their potential negative impact for marine biota (3, 83). This study provides an upper limit for the LDP sinking speed that can be used to constrain future plastic-tracking studies. Further information on concentration of LDP in the water column and on the seafloor, as well as observations of in situ sinking speeds are urgently needed, given the potential damage of plastic debris on pelagic and benthic ecosystems.

4.4 Limits and perspectives

The Lagrangian simulations were subject to approximations. We used a constant sinking velocity of LDP particles from the surface to 1000 m depth, while this can vary, due to seawater density variation or biochemical processes. The sinking speed was considered equal for all the particles, while this may not be the case (Subsec. 2.5). We focused on the 1000 m (and 2000 m, Supporting Information S.2) depth layer, while several Mediterranean areas are deeper than 3000 m. Particles were released for one year only, and land based and maritime plastic sources used to calculate the surface sinking rate (our initial condition) were subject to high uncertainties (12, 27, 31). The surface sinking rate needs further refinement, including processes such as fragmentation, seasonality and spatial variability (e.g. 82). The horizontal and vertical diffusivity were considered as homogeneous through the basin and constant in time, while they can have both spatial and temporal variability. These choices were due to the fact that information about concrete ways of parameterising these dynamics (e.g. the change in time of the sinking speed) were not available or not validated by observations to date.

Therefore, while the concentration of LDPs on the seafloor is affected by high uncertainties, our results represent a first step forward in the modelisation of sinking LDP debris, as evidenced also by the agreement with in situ seafloor observations. The hotspots of plastic debris accumulation on the seafloor as well as its transport pathways may be used to design optimal sampling or removal strategies (e.g. 31). These could be focused both at large or regional scales, and may benefit from future improvements of TrackMPD simulated processes.

The previous considerations advocate for further research efforts, as additional information is essential to deepen the knowledge on the biological processes affecting the vertical path of plastic debris (e.g. refs. (32, 70, 74, 76)), and to implement the characterisation of the hydrodynamical field transporting it (for instance, by increasing its spatio-temporal resolution). Also, resuspension from the seafloor or funnelling effects (for instance due to canyons) should be investigated (16, 17, 84). This information is needed to improve plastic-tracking models and, more generally, to mitigate plastic pollution.

Supporting Information

Use of individual sinking velocities and particle mass; LDP seafloor concentration at 2000 m; surface-seafloor connectivity; crossroadness analyses; sensitivity test of SS_{DM}; sensitivity test of LDP seafloor concentration; in situ observations of plastic seafloor concentration; sensitivity test of coastal strip LDP concentration.

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Data availability

All the data necessary to produce all the figures of the main text, including the LDP seafloor concentration, the coastal strip pollution, and the SS_{DM} vs $\Delta\rho$ relationship, are publicly available at https://doi.org/10.5281/zenodo.7350455. The surface sinking rate used to initialise the particle release is available at https://doi.org/10.5281/zenodo.5931213. The in situ plastic concentrations are available at https://doi.org/10.5281/zenodo.5538237. The TrackMPD code is available at https://github.com/IJalonRojas/TrackMPD.

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