Science of the Total Environment 777 (2021) 145944

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

The dynamics of microplastics and associated contaminants: Data-driven Lagrangian and Eulerian modelling approaches in the Mediterranean Sea

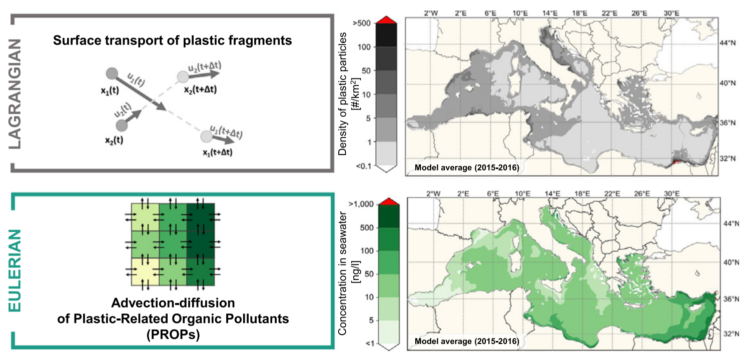
Federica Guerrini *, Lorenzo Mari, Renato Casagrandi *

Dipartimento di Elettronica, Informazione e Bioingegneria, Politecnico di Milano Via Ponzio 34/5, 20133 Milano, Italy

HIGHLIGHTS

- Dynamics at sea of Plastic-Related Organic Pollutants (PROPs) modelled for the first time.
- Data-informed particle sources affect modelled patterns of marine microplastics.
- Distributions of microplastics and PROPs are different when modelled separately.
- Future models should couple the dynamics of microplastics and PROPs at sea.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 26 November 2020
Received in revised form 13 February 2021
Accepted 14 February 2021
Available online 20 February 2021

Editor: Ouyang Wei

Keywords:
Microplastics
Marine pollution
Advection-diffusion
Oceanographic reanalyses

ABSTRACT

Plastic pollution is widespread in the global oceans, but at the same time several other types of hydrophobic pollutants contaminate the marine environment. As more and more evidence highlights, microplastics and polluting chemicals are intertwined via adsorption/desorption processes. A thorough assessment of their total impact on marine ecosystems thus requires that these two kinds of pollution are not considered separately. Here we compare the outcomes of two complementary, data-driven modelling approaches for microplastic dispersal and for Plastic-Related Organic Pollutants (PROPs) in the marine environment. Focusing on the Mediterranean Sea, we simulate two years of Lagrangian particle tracking to map microplastic dispersion from the most impacting sources of pollution (i.e. coastal areas, the watersheds of major rivers, and fishing activities). Our particle sources are data-informed by national census data, hydrological regimes, and vessel tracking data to account for spatial and temporal variability of mismanaged plastic waste generation. These particle-based simulations are complemented with a simulation of the dynamics of primary pollutants in the sea, obtained via an advection-diffusion Eulerian model. While providing further understanding of the spatiotemporal distribution of microplastics and the dynamics of PROPs at a Mediterranean-wide scale, our results call for the development of novel integrated modelling approaches aimed at coupling the dynamics of microplastics with the chemical exchanges occurring through them, thus promoting a holistic description of marine plastic pollution.

1. Introduction

Marine plastic pollution has been first detected just a couple of decades after the beginning of the large-scale production of plastic materials in the 1950s (Carpenter and Smith, 1972). Since then, the magnitude of the phenomenon of marine plastic pollution has considerably grown, driven by the continuous increase in the production of plastic materials

* Corresponding authors.
E-mail addresses: federica.guerrini@polimi.it (F. Guerrini), renato.casagrandi@polimi.it (R. Casagrandi).

https://doi.org/10.1016/j.scitotenv.2021.145944

(PlasticsEurope, 2020), with just about 20% of them being reprocessed at their end-of-life (Geyer et al., 2017). Likewise, the scientific interest in the issue of marine plastic pollution has also remarkably increased, starting from its most macroscopic evidence, i.e. the detection of oceanic garbage patches (Moore, 2008), down to the subtler pollution caused by micro- and nanoplastics (Syberg et al., 2015). During transport by currents, microplastics in the marine environment are subject to other concurrent processes of different nature: *physical*, such as degradation and fragmentation (Kalogerakis et al., 2017); *biological*, like colonization by microorganisms (Fazey and Ryan, 2016) or ingestion by an array of marine species, from invertebrates and fish (Cole et al., 2011) to fin whales (Fossi et al., 2012); and *chemical* (Caruso, 2019). Along this latter direction, microplastics have been found to adsorb hydrophobic chemicals from the surrounding environment, thereby potentially concentrating chemical pollutants and a whole variety of compounds (such as in the case of plastic cigarette butts, Marinello et al., 2020), and carrying around the additives used in the production of plastics (Teuten et al., 2009; Rochman, 2015; Tourinho et al., 2019; Schmidt et al., 2020). This double role of microplastics poses an additional toxicological threat to marine species when they ingest them (Alimba and Faggio, 2019; Compa et al., 2019; Guerrini et al., 2019).

The design of data-informed, targeted policies to contrast marine plastic pollution requires a thorough understanding of the sources of plastic waste and its consequent fate at sea. On-field sampling, although crucial for monitoring the phenomenon, can be rather challenging because campaigns can cover only limited areas, and are often constrained by time and weather. An additional and important complication for monitoring is that microplastics are not easily observable due to their size. For these reasons, simulations with numerical models using ocean currents have been regarded as an effective tool to estimate the role played by different sources, sinks, and pathways to study pollution from microplastics in the marine environment (Hardesty et al., 2017). Simulation models identified the Mediterranean Sea as one of the areas most polluted by microplastics globally (Lebreton et al., 2012), predicting concentrations comparable with those found in oceanic gyres (Eriksen et al., 2014; Cózar et al., 2015; van Sebille et al., 2015), as later confirmed by sampling campaigns (Suaria et al., 2016). This convergence of modelling predictions and field observations is especially worrisome for the potential impacts of microplastic pollution on the rich biodiversity of the Mediterranean marine ecosystem (Coll et al., 2010), which is already heavily impacted by human activities and the consequent pollution (Micheli et al., 2013). With regards to organic pollutants, modelling approaches have mostly been used to study local events (e.g. oil spills) rather than investigating their basin-scale discharge and dispersal. Indeed, further work is needed in this direction to complement the physical description of pollution from microplastics with its chemical-related side.

Despite being closely related, microplastic particles and Plastic-Related Organic Pollutants (PROPs) behave differently in the marine environment. While small-sized plastics can be approximated as point-like tracers passively transported by currents, PROPs are not just advected by surface currents, as particles are. In fact, they also diffuse in response to chemical gradients and thermal agitation, and may in some cases react with the surrounding environment. The Lagrangian and the Eulerian frameworks are the two most frequently used modelling approaches in fluid dynamics (Batchelor, 2000), and are also convenient to model the advection-diffusion-reaction processes of pollutants in the atmosphere and the oceans (see for example Zannetti, 1990; Dippner, 2004; van Sebille et al., 2018). Simply put, while Lagrangian schemes track punctual elements that follow an instantaneous flow, as in the case of microplastic particles in many models of marine plastic pollution, Eulerian models are typically used to describe flows of substances at fixed, specific locations, like the faces or the vertices of a grid.

In this work, we used those two frameworks to describe pollution due to either microplastic particles or the chemicals related to them. This choice allowed us to better account for the differences in the chemical and physical properties of microplastics and hydrophobic pollutants that consequently affect their dynamics at sea. Focusing on the Mediterranean Sea, here we use oceanographic reanalyses to simulate two years (2015–2016) of Lagrangian tracking for floating microplastic particles, following a widely adopted framework for the study of plastic pollution at both global (Lebreton et al., 2012; Eriksen et al., 2014; van Sebille et al., 2015) and Mediterranean scales (Liubartseva et al., 2018; Macias et al., 2019; Kaandorp et al., 2020; Mansui et al., 2020; Soto-Navarro et al., 2020), with the particular care of accounting for data-informed inputs from coastlines, rivers, and fishing activities. To deepen our understanding and monitoring of the source-to-sink pathways of microplastic particles in the Mediterranean Sea, we divide the basin into five geographically relevant regions and track the origin of plastic particles released within each of them by either input type (coastal, riverine, fishing-related) or source region. We also describe the advection-diffusion processes of PROPs that are known for accumulating on plastics with a simple Eulerian model applied to the surface layer of the Mediterranean Sea and run over the same time span as the Lagrangian simulations. To the best of our knowledge, this work is the first two-sided modelling attempt directly comparing microplastic pollution to its chemical counterpart; therefore, we kept our Lagrangian and Eulerian models realistic yet relatively simple in terms of forcings, surface dynamics, and other processes occurring at our relevant oceanographic scale to ensure a similar level of detail in both modules. Comparability between the two modules is key to providing here preliminary evidence of the need to integrate microplastic particle tracking with PROPs advection-diffusion in comprehensive numerical simulations of marine plastic pollution. These are two known and important facets of this complex issue, yet very rarely studied together from a quantitative perspective. Our modelling attempt thus represents a first step forward towards an integrated description of the intertwined dynamics of microplastics and PROPs at sea.

2. Methods

To better refine the description of the physical and chemical sides of plastic pollution, i.e. particles and PROPs, the model presented here is structured in two modules: a Lagrangian and an Eulerian one, both coded in Python 3 programming language.

2.1. Lagrangian simulations of microplastic particles

Following a Lagrangian framework, microplastics were assimilated to point-like particles whose passive movement is completely consistent with that of surface water masses. The zonal and meridional components of surface currents, as well as the geographical extent and gridding of our simulation domain, were obtained from the oceanographic reanalysis made available by the Copernicus Marine Environment Monitoring Service. More precisely, we used the Mediterranean Sea Physics Reanalysis (Simoncelli et al., 2014), which is based on the NEMO-OPA Ocean General Circulation Model (OGCM) and the OceanVAR data assimilation scheme. The reanalysis has a $1/16° \times 1/16°$ horizontal resolution, 72 layers of depth (in our model we used the data for the surface layer only), and daily temporal resolution. For each particle, the motion equations were solved using a Runge-Kutta scheme with an adaptive step size (Dormand and Prince, 1986). At every step of the simulation, the zonal and meridional components of current velocity were obtained using a tri-linear interpolation scheme.

For the sake of simplicity, we opted for a 2D description of particle motion in the surface layer of the Mediterranean Sea, neglecting vertical velocities and the vertical motion potentially induced by positive/negative particle buoyancy. This is equivalent to assume that our point-like microplastic particles have the same density as water ($\rho_{\text{particles}} = \rho_{\text{seawater}}$), and to disregard any mixing of particles to the water column, such as those possibily caused by wave motion and vertical turbulent diffusion (see e.g. van Sebille et al., 2020). Consequently, each particle was transported at a fixed depth assigned randomly in a 0–1 m range upon release.

Subgrid variability is here accounted for through two complementary mechanisms. The first is particle release, since the initial particle location is randomly assigned in a 100 m radius around the source point. The second is particle transport, as the interpolation of the gridded velocity fields at each particle's location is inherently sensitive to small changes in the input coordinates. These two mechanisms, applied on a large number of particles (see below) transported by velocity fields from an eddy-permitting ocean model, allow to capture subgrid variability in particle displacement (Koszalka et al., 2009; van Sebille et al., 2018).

Processes that remove microplastics from the sea surface, such as the sinking of particles due to growth of biofilm on microplastic or beaching, need also to be accounted for, because surface removal may act over shorter timescales (Fazey and Ryan, 2016) than our simulation period. We thus described the sinking process in our model by assigning to each particle a random duration of transport, after which the simulation of its motion was stopped and the particle was considered as removed from the system. Specifically, the advection time was extracted from an exponential distribution with an average of 50 days, and a maximum duration of transport truncated at 250 days (so that <1% of particles would persist for more than eight months on the sea surface). Following this methodology, about 50% of our particles were removed from the surface within the first 34 days of transport, a value that is coherent with the median sinking time provided by Fazey and Ryan (2016), who indicated a range from 17 to 66 days depending on the fragment size. Furthermore, microplastic particles were considered as beached, and therefore removed from the model, if at a certain time step they: i) were located within a coastal cell of the oceanographic grid, and ii) had null zonal and meridional velocities. We did not describe here the possible recapturing of beached particles caused by local weather conditions (e.g. high waves) and tidal-induced sea level rise. No other microplastic removal mechanisms, like fragmentation or ingestion by marine biota, were included in our simulations. Fragmentation of particles should not in fact be simulated as an actual removal process from the surface because it rather entails a reduction in size. Ingestion, which is sometimes considered to be negligible in terms of particle dynamics when compared with sinking and beaching (Koelmans et al., 2017), would require the simultaneous modelling of marine biota dynamics, which goes beyond the limits of the present study.

We simulated Lagrangian particle tracking over the period 2015–2016. This recent and relatively short time horizon allowed us to better refine our simulation by both informing particle input with realistic proxy data while releasing a large number (100 million) of particles per year. To account for plastic particles with long residence time in water (on the order of 8 months, as described above), it was necessary to warm-up the system by starting simulations 250 days before January 1st, 2015. In total, more than of 250 million particles were tracked over the whole Lagrangian experiment.

2.2. Characterization of the sources of plastic pollution

Mismanaged waste from coastal cities (Jambeck et al., 2015), rivers collecting surface water runoff (Lebreton et al., 2017), and accidental release during fishing activities (Macfadyen et al., 2009) are commonly regarded as the main sources of marine plastic pollution. In our simulations, we accounted for plastic pollution coming from each of these three sources by spatially distributing particle input locations i) along the Mediterranean coastline, ii) at the mouths of 100 selected rivers (with choice criteria detailed below), and iii) in several locations within the most active fishing grounds, as shown in Fig. 1. Each released particle was attributed to one of the three sources (coastal, riverine, or fishing) and one of the 21 countries facing the Mediterranean Sea. We allocated the amount of particles to be released from each source type

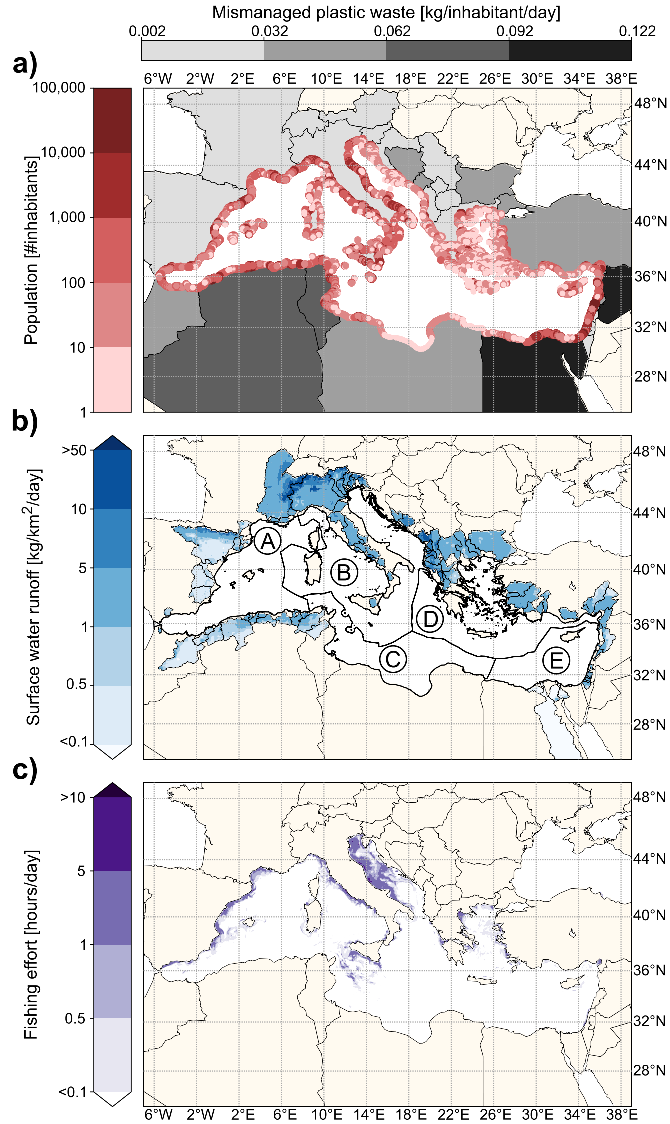


Fig. 1. Proxy data used to inform the sources of microplastic particles in the Lagrangian module of our model and Mediterranean regions used for particle sourcing. Displayed data is averaged over the time horizon of the simulation (2015–2016). a) Coastal population within the first 5 km from the shore, superimposed to the country-specific rate of mismanaged plastic waste generation, from Jambeck et al. (2015); b) average surface water runoff in the 100 river basins selected and region boundaries (see Section 2.1 for the complete definition of this proxy indicator and Figs. S1, S2, S3 for complete mapping); c) average daily fishing effort at each location.

by applying the coasts-to-rivers-to-ships ratio of 50:30:20 proposed by Liubartseva et al. (2018). At present, observations on plastic inputs from coasts, rivers, and fishing activities in the Mediterranean Sea have insufficient spatial and temporal coverage to inform patterns of particle release. For this reason, we identified for each source type a suitable proxy indicator of the amount of plastic waste entering the Mediterranean Sea that could account for the geographical and temporal apportionment of the number of particles released daily.

Coastal particle inputs (Fig. 1a) were set proportionally to the estimated quantity of mismanaged plastic waste produced by the populations living close to the coasts of the Mediterranean Sea. We chose to consider as coastal only the mismanaged plastic waste produced in the first 5 km adjacent to the coast; the contribution to plastic pollution by more inland locations was accounted for via riverine particle input (detailed below). For each coastal source, plastic input was estimated as the product between the population living near the source and the per capita production of mismanaged plastic waste for the country where the source is located. The former was elaborated from the Gridded Population of the World – Population Count 2015 (CIESIN, 2018), which has a spatial resolution of 2.5 min (approximately 5 km × 5 km); the latter was taken from Jambeck et al. (2015). From a technical viewpoint, coastal sources were positioned in the middle of the corresponding sea-facing cell border. Overall, a total of around 50 million particles was released from all coastal sources for each year of the simulation.

As for riverine sources (Fig. 1b), we selected the top-100 contributing rivers among all the Mediterranean coastal basins, i.e. those that discharge directly into the Mediterranean Sea, as retrieved from the HydroBASINS – HydroSHEDS products for Europe and Africa (Lehner and Grill, 2013). Rivers were ranked with an indicator accounting for both hydrological factors and waste mismanagement. This indicator was obtained as the product between the total mismanaged plastic in each watershed (computed with the same methodology used for the coasts, after Jambeck et al. (2015), extended to whole river basins) and the monthly-averaged surface runoff in the basin in 2015–2016, as given by the gridded FLDAS dataset (McNally and NASA/GSFC/HSL, 2018). For each cell at the finer scale of the population dataset, we multiplied surface water runoff by the amount of mismanaged plastic that can potentially reach the sea. For graphical readability of the map, in Fig. 1b we do not show the contribution given to the Nile river by Sub-Saharian countries, but a complete mapping of the components of the indicator is available in the Supplementary data (see Figs. S1, S2, and S3). To account for seasonal variations in the hydrological regime of the selected rivers, the indicator was made time-varying by calculating its value for each month of the simulation, so that the runoff in a cell in a certain month temporally modulates plastic release to the sea via rivers (Schmidt et al., 2017; van Emmerik et al., 2018). The monthly values of the indicator just defined were then summed up over each cell of the population dataset comprised within the selected basins to obtain weights for the apportionment of the number of particles to be released daily by each river. Around 30 million particles were released from all rivers for each year of simulation.

To account for plastic pollution due to accidental release during maritime activities, especially fishing (Fig. 1c), we released Lagrangian particles proportionally to the daily fishing effort (expressed in fishing hours/day) at each location, as registered by the Automatic Identification Systems (AIS) on the Global Fishing Watch dataset (https://globalfishingwatch.org/; Kroodsma et al., 2018). This global dataset spans over the years 2012–2016; however, a temporal gradient exists whereby the data becomes progressively more abundant in more recent years: for this reason, we used the daily data for the years 2015–2016 only. The data for these two years was processed to select all fishing vessels in the Mediterranean area not using fixed gears (like longlines, pots, and traps), under the assumption that gear loss happens mostly during handling. In fact, fishing activities typically release macroplastics, such as fishing nets and styrofoam boxes (Macfadyen et al., 2009). However, the movement of these larger plastic fragments on the sea surface cannot be properly described within our simple Lagrangian framework because of the presence of surface-related mechanisms, such as wave motion, wind entrainment, and drag. Other sea-based sources of primary microplastics indeed exist (for example, paint chips from weathered hulls; CCB Technical Report, 2017). Nonetheless, we chose not to differentiate particles released in fishing grounds from those generated by other sources. In total, about 20 million particles were released from sea-based sources for each year of the simulation.

No influx of particles was considered at the Strait of Gibraltar. In fact, to be coherent with our in-basin sources, any depiction of such input would necessarily rely on an arbitrary identification of the spatial scales to be accounted for, and would thus require to be informed by Atlantic circulation, which is beyond the already wide geographical scope of our study.

2.3. Eulerian model

The information available in the literature to quantitatively describe the dynamics of hydrophobic pollutants in the marine environment is somewhat sparse (Astiaso Garcia et al., 2019; see also Figs. S4 and S5 in our Supplementary data); thus, it could not be used to inform model inputs. For this reason, the Eulerian module of our model is intended as a coarse, yet reliable tool to produce reasonable projections of the spatio-temporal distribution pattern of PROPs in the surface waters of the Mediterranean Sea, rather than reconstructing or forecasting their actual concentrations. The Eulerian approach we followed to simulate PROPs is less computationally demanding than Lagrangian tracking, as the former is based on partial differential equations describing the dynamics of pollutant concentrations, while the latter accounts for the individual motion of every single microplastic particle. The Eulerian advection-diffusion equations were solved using FiPy (https://www.ctcms.nist.gov/fipy/; Guyer et al., 2009), a Python library based on a finite volume approach. Pollutants were advected by the same surface velocities as Lagrangian particles were (Simoncelli et al., 2014), and assumed to have a constant diffusivity over the whole oceanographic domain and the entire simulation period. Diffusivity of pollutants in water was calculated with the equation proposed by Schwarzenbach and Bouwer (1997), i.e.:

$$D_{iw}[cm^2s^{-1}] = \frac{13.26 \cdot 10^{-5}}{\eta^{1.14}\overline{V}_i^{0.589}} \tag{1}$$

where η is the pollutant viscosity (in centipoise, 10^{-2}g cm^{-1} s^{-1}) at the temperature of interest, and $\overline{V}_i$ is the molar volume of the chemical (cm^3 mol^{-1}).

Although diffusivity of a chemical depends on thermal agitation and, ultimately, on temperature, which may considerably vary over space and time, diffusivity values calculated with Eq. (1) for several temperature values in the range registered for the Mediterranean Sea were of the same order of magnitude as those obtained with a constant temperature. After an initial testing phase, we chose the diffusivity value obtained with 20°C, which is consistent with the average surface annual temperature of the whole Mediterranean Sea (Shaltout and Omstedt, 2014). In addition to describing advection-diffusion processes occurring because of marine currents and chemical gradients in the sea, our Eulerian module can also account for external inputs at selected locations. Since no reliable estimates of PROPs inputs exist at a Mediterranean-wide geographical scale, we estimated a total influx of these pollutants and allocated it among the sources of plastic pollution (coasts, rivers, and maritime sources) according to the same apportionment methodology described above for the distribution of Lagrangian particles. As prototypical PROP of reference, we selected phenanthrene, a typical Polycyclic Aromatic Hydrocarbon (PAH) detected in the marine environment (Berrojalbiz et al., 2011), for which we calculated the diffusivity as explained above. We chose not to implement any decay of pollutant concentration over time, thus indirectly assuming that the target pollutant may persist in the marine environment for a period of time much longer than our simulation time span. To spin up the Eulerian module of our model, we initialized the simulation by assuming a clean sea surface in 2005 and modelled the discharge, advection, and diffusion of the pollutant on daily time steps for 12 years (2005–2016), allowing the model to warm up before the beginning of Lagrangian simulations (mid-2014 to 2016).

2.4. Region apportionment

To monitor the spatial and temporal patterns of the simulated variables (number and location of particles, pollutant concentration), we divided the Mediterranean Sea into five large, geographically-coherent regions (Fig. 1b) that group the seascapes of pertinence of each of the Mediterranean-facing countries, i.e. the nations' maritime boundaries as in Marine Boundaries v11 (Flanders Marine Institute, 2019), namely: Ⓐ Western Mediterranean, Ⓑ Central Mediterranean, Ⓒ Southern Mediterranean, Ⓓ North-Eastern Mediterranean, and Ⓔ South-Eastern Mediterranean. For each region, we calculated the average particle count per square kilometer and pollutant concentration on each day of simulation. Particle counts could be further detailed by apportioning particles to their source type (coasts, rivers, fishing activities) and to their source region. In particular, while particles released from coastal and riverine sources were naturally attributed to their country of origin, those from fishing sources were assigned according to the flag flown by their vessel, as reported by the Global Fishing Watch records.

3. Results

Fig. 2 shows the geographical distribution of plastic particles, as projected by our Lagrangian module, in the five regions of the Mediterranean Sea identified in Fig. 1b. Fig. 2a reveals that higher particle densities are expected in the western and central Mediterranean (namely in the Iberian and Algerian Seas in Ⓐ, in the Adriatic Sea and the Strait of Sicily in Ⓑ, and along the Tunisian coasts in Ⓒ) than in the southern and eastern parts of the basin during 2015–2016. The Aegean Sea and the Turkish coasts in Ⓓshow higher particle counts than the high sea within the region, but still lower than those mentioned above. The coastal area in region Ⓔ represents a relevant exception within the eastern part of the Mediterranean, especially close to the Nile Delta, where modelled particle concentrations are the highest in the whole basin.

The temporal variations of average particle densities in the same regions are represented in Fig. 2b. Region Ⓒshows the lowest particle densities, which remain around 2 particles/km^2 during the whole simulation period. Regions Ⓑ and Ⓓ, both with particle densities of about 4 particles/km^2, present oscillating out-of-phase temporal patterns in time: in fact, in both 2015 and 2016, particle density is maximum around August in Ⓑ, while around the same weeks it is minimum in Ⓓ. Region Ⓐ presents the largest particle densities during February–April 2015. Starting from mid-April 2015, though, particle densities in region Ⓔ exceed those projected in all other regions and remain consistently the highest thereafter. As detailed below, particle densities in Ⓔ are strongly affected by the Nile inflow, which causes a peak density of 14 particles/km^2 during September 2016.

Further details on the contribution of each source type to the average density of plastic particles in the five Mediterranean regions are provided in Fig. 3. Region Ⓐ appears to be mostly affected by coastal particles, with a relatively constant contribution from fishing activities (Fig. 3a). Riverine particles cause the density peak observed in early

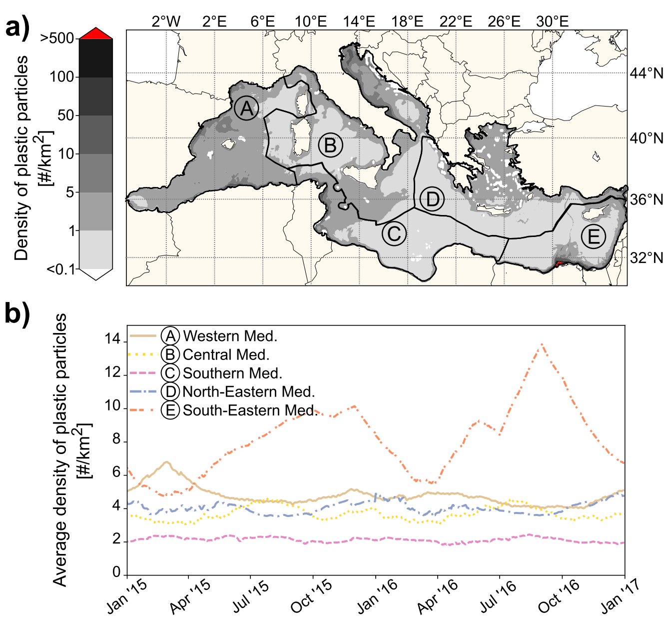


Fig. 2. Particle densities resulting from the Lagrangian simulations. a) Average particle density per square kilometer in 2015–2016; b) spatially-averaged regional particle density as a function of time.

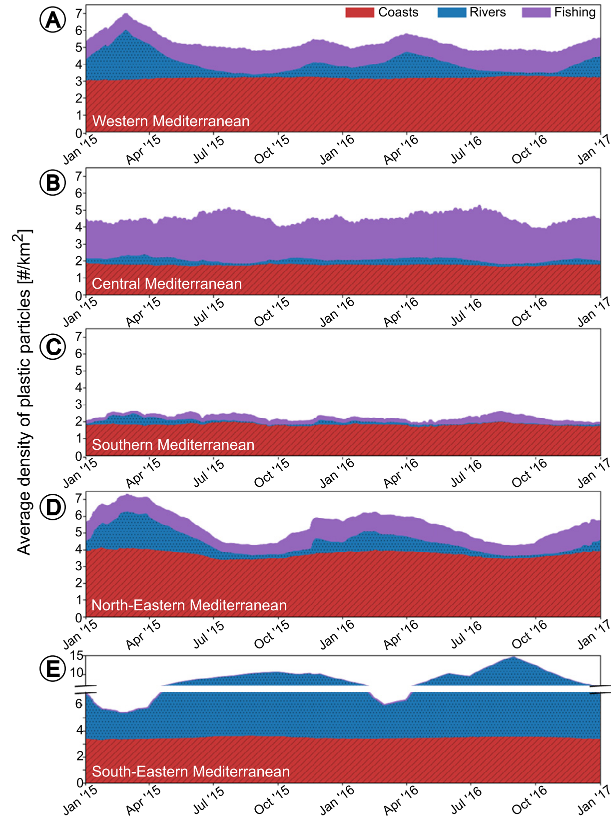


Fig. 3. Source attribution of particles per region, spatially-averaged. Top to bottom: Ⓐ Western Mediterranean; Ⓑ Central Mediterranean; Ⓒ Southern Mediterranean; Ⓓ North-Eastern Mediterranean; E South-Eastern Mediterranean.

2015, while reduced surface water runoff causes their quota to decrease during the summer months of both years of simulation. Despite the abundance of watersheds flowing into region Ⓑ (see Fig. 1b), the main contribution to particle density is here represented by fishing vessels (Fig. 3b), which also cause the summer maxima observed in this region. Coastal particles prevail in region Ⓒ, with smaller contributions from fishing activities and river sources (Fig. 3c). The contribution of riverine particles, in particular, shows some temporal variability that can be linked to increased surface water runoff during winter and spring months. Coastal particles represent the most important contribution to the density of plastic particles also in region Ⓓ (Fig. 3d). Here, their presence follows a seasonal pattern with a period of about one year, which is not as clearly visible in the other regions (except for Ⓐ). Riverine and maritime particles also follow a seasonal pattern in this region. In region Ⓔ, rivers disproportionately contribute to particle density, which attains here its highest values (7–13 particles/km^2, Fig. 3e). On the other hand, in this region the share of particles from fishing activities is the lowest among the five selected areas, according to the dataset we used.

In terms of apportionment, it is interesting to disentangle the contribution not only of each source type, but also of the geographical source region of the particles. Fig. 4 shows, for each region, the percentages of particles coming from any other region as a function of time. In general, most particles do not leave the region where they were released, thus contributing to local particle density. This result is not trivial, as in some cases the fraction of particles crossing the borders of their source region during their transport at sea are not negligible. For example, we calculated (details not shown) that about 27% of particles released in region Ⓒ from fishing-related sources leaves the region. Nonetheless, particles coming from other regions rarely contribute to plastic densities for more than 10%. This pattern is especially clear in region Ⓔ, where particles from Ⓑ, Ⓒ and Ⓓcontribute for less than 2% on average. By

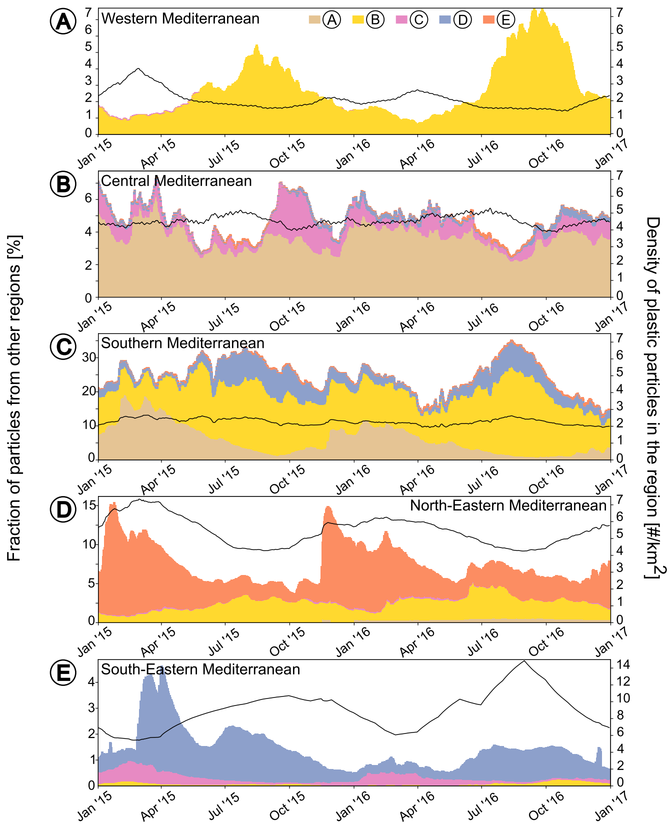


Fig. 4. Region-specific percentages of particles coming from another region in 2015–2016 (stacked, colored bar plots; left y-axis) as a fraction of regional particle density (black line; right y-axis). Top to bottom panels: Ⓐ Western Mediterranean; Ⓑ Central Mediterranean; Ⓒ Southern Mediterranean; Ⓓ North-Eastern Mediterranean; Ⓔ South-Eastern Mediterranean.

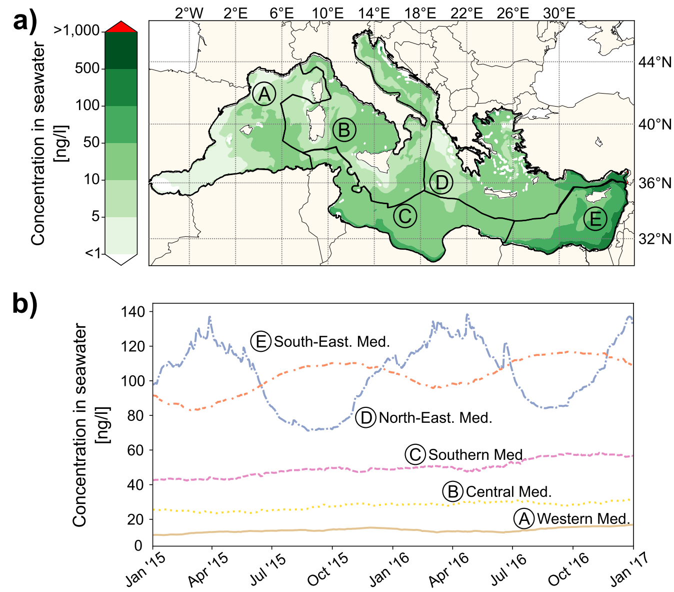


Fig. 5. Concentrations of the target pollutant (phenanthrene) as resulting from Eulerian simulations. a) Temporally averaged phenanthrene concentration in 2015–2016; b) spatially-averaged regional phenanthrene concentration as a function of time.

contrast, particles originating outside the region make up to 35% of the particles in Ⓒ, and up to 15% in the case of Ⓓ. Region Ⓒ, in particular, is the most diverse in terms of the origins of particles found within it, as it includes contributions from each of the five regions, despite being the region with the lowest average particle density. This diversity of geographic sources is probably due to marine circulation processes in the central-southern region of Mediterranean Sea. Temporal patterns can be identified also in this region, with an increment of particles from region Ⓐ during the first months of both years of simulation (up to 15%), while particles from Ⓓ increase their share during the summer and early fall (around 10%). Region Ⓓ too is significantly affected by particles coming from other regions: in 2016 there is even a minor contribution to region Ⓓ of long-travelling particles originating from Ⓐ. However, most of the particles from out-of-region found in Ⓓ come from Ⓔ, with a relevant seasonal increase during the first months of both years of simulation (Fig. 4d), just a few months earlier than the annual maximum in particle density for the region, occurring in late March (see also Fig. 3d). Indeed, the exchange of particles from Ⓔ to Ⓓ is also suggested by Fig. 2a, where a flow of trajectories connecting the Nile Delta (region Ⓔ) to the Turkish Gulf of Antalya in Ⓓ, across the Levantine Sea, is clearly visible. Particles coming from all regions are also found in Ⓑ; however, out-of-region particles contribute up to 7% to plastic density in the region, much less than in regions Ⓒ and Ⓓ. The least diverse picture in terms of particle sources emerges in Ⓐ (Fig. 4a), where particles come mostly from region B for the longest part of the simulated period.

The modelled distribution of the target PROP in the Mediterranean Sea, as obtained from the Eulerian module averaged over the period 2015–2016, is shown in Fig. 5. In particular, the map in Fig. 5a shows that pollutant concentration increases along a west-east longitudinal axis. The highest concentrations of PROPs can be found in the Gulf of Libya and along the coasts of the Levantine basin, with the exception of southern Cyprus. This zonal gradient is clearly visible also in Fig. 5b. In general, all five regions show a temporal increase of PROP concentrations over the simulation period (about 30% in each region, with the exception of region Ⓓ, where the overall increase amounts to less than 5%), which is consistent with the absence of removal mechanisms for the PROPs in our Eulerian module. Here, it can also be noticed that pollutant concentrations in regions Ⓓ and Ⓔ vary seasonally with out-of-phase signals. In fact, region Ⓓ is more polluted than Ⓔin winter and spring, an interesting temporal feature that cannot be captured by averaging the signals over time, as done in Fig. 5a.

4. Discussion

Although Mediterranean-wide data on microplastic pollution is still scarce, the average results obtained with our Lagrangian model (Fig. 2a) match reasonably well with the average marine litter concentrations modelled by Soto-Navarro et al. (2020) for floating particles (see their Fig. 2c) and with the results by Liubartseva et al. (2018) (see Fig. 3 therein). All three models predict high particle densities in zones like the coasts of the Gulfs of Valencia and Lion, both in region Ⓐ of Fig. 1b, the Adriatic Sea in Ⓑ, and along the Tunisian and Levantine coasts (Ⓒ and Ⓔ, respectively). In some areas, the predictions of our model differ from those drawn by previous modelling studies, as somehow expected because of the different time horizons used. As an example, in the central and southern parts of the Adriatic Sea, our simulations better agree with those by Liubartseva et al. (2018) in showing denser particle presence along the Italian coast than in the middle of the sea as found in the model by Soto-Navarro et al. (2020). Furthermore, the Aegean Sea is characterized by much lower values in surface plastic concentrations in both Liubartseva et al. and Soto-Navarro et al. than in our model (see again Fig. 2a). Liubartseva et al. identified as potential plastic density hotspots the southern part of the Ionian Sea (an area at the crossroad of regions Ⓑ, Ⓒ, and Ⓓ, in our subdivision of the Mediterranean Sea) and, to a lesser extent, the central part of the Levantine Sea, in the middle of region Ⓔ; however, both our model and the one by Soto-Navarro et al. agree on low particle densities in those areas. Finally, other Lagrangian modelling studies (Lebreton et al., 2012; Mansui et al., 2020) highlight the seasonality of plastic particle densities in the southern part of the Mediterranean basin (our region Ⓒ), while ours does not (see Fig. 2b for seasonal trends in Ⓒ), in agreement with the aforementioned work by Soto-Navarro et al.. Once again, these discrepancies may come also from differences in the time horizon of the simulations, the processes included, and especially the forcing terms applied. Despite being run over a shorter time span and having implemented a somehow simpler sinking mechanism than others, our model in fact relies on a finer description of particle sources in both space and time. Other than using high-scale demographic data, as well as time-dependent riverine flows from hydrological data, we also informed the release of maritime particle sources with high-precision data from fishing activities, where gear loss or intentional abandon may occur (Macfadyen et al., 2009). This choice is innovative with respect to the assumptions of both Liubartseva et al. (2018) and Soto-Navarro et al. (2020), who chose maritime transportation, which indeed may represent another relevant, sea-based source of plastic litter despite the adoption in 1973 of the International Convention for the Prevention of Pollution from Ships (MARPOL 73/78), which prohibited the discharge of all nonfood solid waste.

As shown in Fig. 3, coastlines are the most important sources of particles in regions Ⓐ, Ⓒ, and Ⓓ, a result that is similar to what Soto-Navarro et al. found in those areas (see their Fig. 3a). In our model, a relevant contribution from coastlines is indeed expected, as we (i) set coastal input to account for 50% of the total modelled particles, and (ii) assumed to have no changes in the coasts-to-rivers-to-fishing ratio in our coastal forcing due to population or mismanaged plastic waste generation rates in 2015–2016. Similarly, Soto-Navarro et al. adopted spatially-distributed, time-constant particle inputs. Interestingly, despite half of the particles released in the present study came from coasts, our results show that in some regions other source types contribute more significantly than coasts to average plastic density during either specific periods (like the peak of riverine particles in region Ⓐ in early 2015, Fig. 3a) or the whole simulation. Examples of this last category include region Ⓑ, where more than 50% of particles consistently come from fishing grounds (see Fig. 3b) or region Ⓔ, where the riverine contribution always outclasses the others (see Fig. 3e).

For all regions but Ⓔ, the presence of riverine particles is higher in winter and spring, as this period corresponds to the wet season in the northern hemisphere, thus to an increase in surface water runoff. It can be noticed that the number of particles discharged from river mouths in the wet season of 2015 is higher than in the wet season of 2016. This can be explained by the differences in the precipitation regime in the Mediterranean area in 2015–2016: in fact, although 2015 was an unusually dry year, the first months of 2015 were rainier than early 2016 (Kennedy et al., 2016; McNally and NASA/GSFC/HSL, 2018), and this caused an abundant surface water runoff and, ultimately, an increase of riverine particles released in our model. In region Ⓔ, the share of riverine particles seems to go against this seasonal trend, with more simulated particles in the dry season. As somehow expected, the Nile is the most significant river source there, and its diversified seasonal regime depends upon its watershed, which extends over climate zones that are quite different from those of the European Mediterranean countries. Moreover, the contribution of rivers to plastic particle pollution in region Ⓑ is noticeably lower than that of both coasts and fishing-related activities. The rivers flowing into region Ⓑ release a lower total number of particles than their counterparts in the other regions, although they are numerically prevalent (about one third of the rivers considered) and their basins are characterized on average, by the highest surface water runoff in the Mediterranean area (Fig. 1b). This seeming discrepancy is due to the proxy variable we selected for modulating particle release from rivers, which also accounts for plastic waste generation in their watershed. In fact, despite being densely populated (Italy in particular, see Fig. 1a), central Mediterranean countries have lower rates of mismanaged plastic waste generation compared to other countries, and this in turn affects the amount of particles released by their rivers in our simulations. Accounting for this effect explains the difference between our results and those presented by Soto-Navarro et al. (their Fig. 3b), who considered the Adriatic Sea to be significantly affected by the particles released from the mouth of the Po river. In fact, Soto-Navarro et al. geographically modulated particle release from rivers proportionally to their mean discharge, and the Po river has one of the highest values for the whole Mediterranean basin. River discharge is indeed crucial in modelling plastic release, as these variables are positively correlated (Schmidt et al., 2017). However, combining discharge with country-specific data about plastic waste mismanagement may be key to providing a more realistic proxy of actual particle release.

According to our results, particles from fishing grounds appear to have a low impact in regions Ⓒ and, especially, Ⓔ. In fact, looking at the distribution of fishing effort in the Mediterranean as measured by the data used here (Fig. 1c), it can be noticed that fishing effort is almost negligible in those two areas. This does not mean that no fishing activities occurred there in 2015–2016 (which is unlikely, as Egypt and Tunisia hold respectively 13.7% and 11.3% of the total Mediterranean fishing capacity; Conides and Papaconstantinou, 2020). Much probably, the lack of data can be explained by the limited use of AIS by fishing fleets operating in those areas (see Fig. 37.4 in Merino et al., 2019). Our modelling results thus structurally underestimate the impact of fishing activities on plastic pollution in those regions, which instead could be very a relevant source of microplastics there (see Fig. 3c in Soto-Navarro et al., 2020).

In general, particles released in a certain region are the most prominent responsible for plastic densities within that same region, as found also in a similar study (see Fig. 3 in Mansui et al., 2020). Particles sourced from out-of-region rarely contribute for more than 10% to average plastic densities in each area of our simulations (Fig. 4), with the exception of region Ⓒ, where inputs from all other regions account for up to 35% of particle presence. This specificity is due both to the location of region Ⓒ, which is the only one adjacent to all others, and to the Mediterranean surface circulation regime. In fact, entering the Mediterranean Sea from the Atlantic Ocean at the Strait of Gibraltar, surface currents prevalently flow along the African coasts towards the eastern part of the basin, first with the Algerian current and then with the Mid-Mediterranean jet current. In the Levantine basin, surface currents flow counter-clockwise along the coasts and around Cyprus. Then, with several eddies centered around the island of Crete and the coasts of mainland Greece, the prevailing currents flow towards the Ionian Sea, where they are redirected southwards along the eastern Sicilian coasts (Pinardi and Masetti, 2000). In our Lagrangian simulations, particles from the western and central parts of the Mediterranean Sea are carried by surface currents towards the southern part of the basin, so that regions Ⓐ and Ⓑ are the most frequent sources for the plastic particles found in Ⓒ. Furthermore, particles from regions Ⓓ and Ⓔ could respectively enter either Ⓒ or Ⓓ with the Ionian branch of the surface current. Due to this opposite current, particles from region Ⓔ are expected to reach Ⓒ from Ⓓafter being carried along the Levantine coasts, rather than by crossing directly the boundary between Ⓔ and Ⓒ. Notably, our results for region Ⓒ are coherent with the source attribution by Mansui et al. (2020) for particles within that same region. However, in the aforementioned work, the Levantine basin seems not to be reached by particles released west of 9°E, while in our model region Ⓓ is reached also by particles sourced in Ⓐ, consistently with the surface circulation regime summarized above. The opposite is not true: in fact, only particles from region Ⓑ (and rare particles from Ⓒ) can reach the westernmost region of the basin, coherently with the direction and the intensity of the westward (Ligurian-Provençal) current flowing from the central Mediterranean to the western part of the basin.

Finally, as far as between-region particle exchanges are concerned, it has to be considered that some out-of-region particles originated by fishing activities could actually be geographically sourced within the region of interest, depending on the nationality of the shipowner or in case of vessels fishing outside their country's maritime area of pertinence, which is not infrequent in the Global Fishing Watch dataset used here.

To the authors' knowledge, despite being known, studied, and policy-regulated for a longer time than marine plastic pollution, never has chemical pollution by hydrophobic components been modelled at a whole-Mediterranean scale. This lack of quantitative approaches is due to the complexity of this task, which requires three difficult steps. First, the identification of a target pollutant of relevance for the whole basin, which per se is not a trivial choice. Human activities, in fact, have been introducing a variety of chemical pollutants in the Mediterranean marine environment, including nutrients (Malagó et al., 2019), heavy metals (Migon et al., 2020), organohalogenated compounds (Fossi et al., 2004), hydrocarbons (Berrojalbiz et al., 2011), and plastic additives (Astiaso Garcia et al., 2019) and often gathered from sediments only (e.g. Merhaby et al., 2019). Third is the capacity of fully describing the dynamics of pollutants at sea, which involves transport by currents and chemical diffusion, but also their partitioning among environmental media (air, water, sediments; Dachs and Méjanelle, 2010), marine biota, and, not least, plastic particles (Teuten et al., 2009).

Indeed, in the Eulerian module of our model we had to tackle some of these complexities and uncertainties. As for the target pollutant, we focused on PAHs, phenanthrene in particular, as they have been frequently sampled in Mediterranean seawater (Berrojalbiz et al., 2011) and, more recently, found to be sorbed onto plastic particles (León et al., 2019), thus corresponding to our definition of PROPs. With regards to the sources of chemical pollutants, we chose to use the same spatial and temporal distribution already applied for particle release. This modelling assumption is in line with Gómez-Gutiérrez et al. (2007) who found that coastal cities and river inputs do play an important role of chemical pollution sources for the Mediterranean basin. To address the modelling complexities, here we assumed that phenanthrene in the marine environment is subject to advection-diffusion processes only, without accounting for interactions with the surrounding environment or removal mechanisms (which we deem a reasonable hypothesis, as it is categorized as a Persistent Organic Pollutant). This assumption turns the surface of our modelled Mediterranean Sea into a permanent sink for the target PROP, where its concentration can only increase through time. This explains two features of our Eulerian simulation results (Fig. 5). First, the time-series of average distribution of the model pollutant is smoother than its Lagrangian counterpart (Fig. 2a). Second, phenanthrene inflow and accumulation in the basin cause a positive trend in pollutant concentration for all regions.

Although available evidence is insufficient to inform the Eulerian module of our model, we can compare the few existing phenanthrene samplings with our simulation outputs to assess the overall performance of our PROP modelling, albeit with caution due to the qualitative nature of the comparison presented here. Nonetheless, the existing data makes it evident that measured pollutant concentrations are, in general, higher in the eastern part of the basin (see Fig. S4), and our results show a similar geographical distribution. In fact, a horizontal gradient in pollutant concentration throughout the Mediterranean basin is clearly visible in both panels of Fig. 5: the lowest values of our results are found in Ⓐ, close to the Atlantic inflow, and the highest are in the Levantine Sea, with Ⓓ and Ⓔ alternating as the most polluted areas, following fluctuating signals. This west-to-east gradient in surface PROP concentration is coherent with the Mediterranean surface circulation at basin scale, as summarized above. The identified pattern is also similar to that found by Macias et al. (2019), who used an advection-diffusion scheme to study the spread of a target tracer released from the main Mediterranean rivers. Although referring to plastic and not to chemical pollutants, their model is the closest match to our Eulerian scheme we could find in literature. This west-to-east gradient in the geographical distribution of the target pollutant projected by our Eulerian model differs from the results obtained with the Lagrangian approach (Fig. 2). In fact, in Fig. 2b, Ⓒ and Ⓔ appear as the regions with the lowest and highest particle density, respectively, while the time series of those quantities in Ⓐ, Ⓑ, and Ⓓ oscillate and occasionally intersect with each other. In contrast, in Fig. 5b the five regions stack up from west to east. Perhaps the most noticeable difference between Figs. 2b and 5b are the periodic oscillations of pollutant concentrations in Ⓓ and Ⓔ. Seasonal variations are more marked in D, dimming the relative increase of mean PROP concentration in the region when compared with the other ones. With a closer look, though, it can be noticed that the pattern followed by the curves of both D and E is coherent (albeit much amplified) with their Lagrangian counterpart in Fig. 2b.

5. Conclusions

Surface distribution of microplastics in the Mediterranean Sea has been simulated for two years (2015–2016), tracking more than 250 million particles using a Lagrangian approach. We distributed this large amount of particles among source points of different nature (coastal, riverine, fishery-related), time of release, and location by using relevant, data-informed proxy indicators. Once at sea, particle dynamics in our model accounts for removal mechanisms such as beaching and sinking, the latter being defined by a random transport duration, coherent with experimental sinking times. In addition to bringing further knowledge about the potential surface distribution of microplastics and PROPs in the Mediterranean Sea, our results advocate for the use of data-informed cues in the definition of sources of plastic debris and chemical pollution in future numerical experiments, as we verified that modelling results are noticeably affected by how forcing terms are defined. Furthermore, the apportionment of particles by their source region provides information about how connectivity between different regions of the Mediterranean basin may influence the exchange of microplastic particles. Yet again, we find that the marine environment has no borders. This observation is even more evident in a semi-enclosed basin like the Mediterranean Sea: the ever-increasing issue of marine plastic pollution urges for the accountability of coastal countries. Particle tracking is a powerful methodology that can help policy makers to inform targeted policies spanning multiple geographical scales, from local interventions to basin-wide regulations.

The physical problems caused to the marine environment by plastic waste are just one of the two sides of plastic-related pollution. Many pollutants are in fact discharged into the sea by human activities, and their interactions with plastic are being increasingly recognized as research progresses (Crawford and Quinn, 2017; Syberg et al., 2017; León et al., 2019). Therefore, a first, necessary step towards a thorough description of marine plastic pollution and its chemical interactions with the seascape requires to comprehensively model the dynamics of conventional pollutants at sea. Here we used phenanthrene as target PROP known for adsorbing on microplastic particles, and we modelled its advection-diffusion with a novel Eulerian model for the surface layer of the Mediterranean Sea. Opposite to the case of our Lagrangian simulations, in this case we could not modulate forcings (i.e., pollutant inputs) on the basis of relevant data. We thus distributed a properly tuned influx of pollution using the same source points and spatio-temporal release patterns already applied for the particle tracking model. However simple, our newly proposed advection-diffusion approach provides pollution patterns coherent with existing samplings in the Mediterranean area, and can thus contribute to an improvement of our knowledge about basin-wide surface dispersal dynamics of conventional pollutants.

The comparison between the results of our Lagrangian particle tracking module with relevant modelling simulations by other research groups, and of the advection-diffusion Eulerian module with sampling campaigns, suggests that this two-sided modelling framework can be effective in describing plastic-related marine pollution. Further improvements to the simulations performed here include prolonging the time horizons of the simulations. Longer simulation time spans, e.g. of the order of decades rather than years, would require a time-varying description of the drivers of pollution, i.e. population dynamics and changes in mismanaged plastic waste generation rates. Projecting geographical trends in the fraction of mismanaged plastics is far from trivial, because they may result from the balance between opposite mechanisms. While mismanaged plastic waste can be significantly reduced as a result of targeted policies and increasing awareness (Abbott and Sumaila, 2019), it may increase because of other factors, such as the, hopefully temporary, unprecedented rise in the global use of face masks and other personal protective equipment (Patrício Silva et al., 2021). Particularly challenging are the projections concerning the effects of climate change on surface water runoff (Blöschl et al., 2019) and fisheries (Allison et al., 2009). An improved modeling framework would also entail the identification of other sources of relevance for pollutant release, like industrial activities (Tobiszewski and Namieśnik, 2012). Also, the dynamics at sea of both plastics and PROPs would benefit from a more detailed description of processes disregarded here, such as plastic fragmentation (Kalogerakis et al., 2017), and pollutant removal mechanisms, as induced by partitioning of hydrophobic chemicals on sediments (Dachs and Méjanelle, 2010) that are then subject to settling. In fact, modelling dynamics occurring at the sea surface only may not exaustively describe marine pollution: sinking may convey significant amounts of both plastics (Kaandorp et al., 2020) and PROPs to the deep Mediterranean Sea.

On the top of this, considering microplastics themselves as vectors of hydrophobic pollutants would arguably represent a major improvement over our two-sided modelling approach. Here we described plastic particles and a target pollutant as two independent types of plastic-related pollution in the Mediterranean Sea (via Lagrangian or Eulerian modelling, respectively), but these are actually two sides of the same coin, whose dynamics are intertwined via adsorption/desorption processes. For this reason, a truly comprehensive assessment of the risks caused by microplastics and the pollutants associated with them on marine ecosystems worldwide needs to be informed by their complex interactions, thereby calling for the development of models suitable for this pivotal task in marine conservation.

CRediT authorship contribution statement

Federica Guerrini: Conceptualization, Methodology, Software, Investigation, Visualization, Writing – original draft. **Lorenzo Mari:** Conceptualization, Methodology, Investigation, Writing – review & editing. **Renato Casagrandi:** Conceptualization, Methodology, Investigation, Writing – review & editing, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

This study received funding from the H2020 project “ECOPOTENTIAL: Improving future ecosystem benefits through Earth observations” (GA 641762, http://www.ecopotential-project.). We are thankful to Tomas Lovato for discussions, and to Daniele Loiacono and Gaia Pani for suggestions on the graphical visualization of our results. We also thank two anonymous reviewers for critically reading the manuscript and suggesting improvements.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2021.145944.

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