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Modeling the transport of microplastics along river networks

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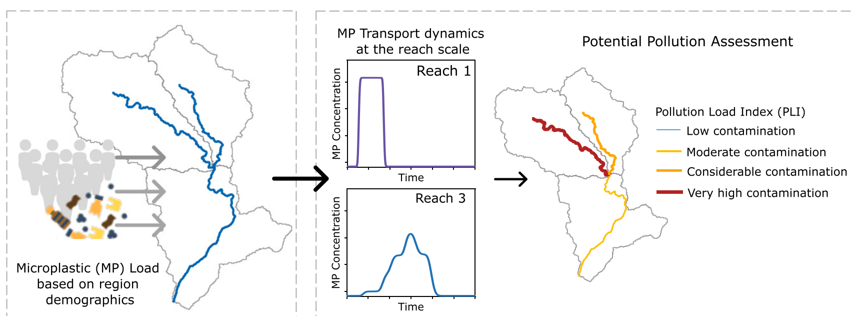
HIGHLIGTHS

- The proposed model allows to characterize microplastic (MP) fate along river networks.
- Microplastics load is assumed to be generated from anthropogenic activities.
- Model validity was assessed using literature data.
- Predicted microplastics allow to perform an assessment of the potential pollution.

GRAPHICAL ABSTRACT



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ABSTRACT

The excessive use of plastics in modern life has led to a significant increase in production and a corresponding rise in plastic waste generation. The slow degradation of plastics results in the introduction and accumulation of microplastics (MP) in the environment, posing environmental and health risks. River networks, acting as conduits between terrestrial and marine environments, play a crucial role in controlling the transport of MP. Predicting the complex processes of MP pathways in these environments is an ongoing challenge. To address this issue, we propose a model that integrates the advection-dispersion equation with anthropogenic MP loads and hydraulic river network characteristics. The validity of the model was assessed using literature data from three river networks worldwide. Model results show a good agreement between predictions and field observations ($R^2 = 0.72$). Consequently, predicted MP data was used to perform a potential pollution assessment through the pollution load index, revealing in most cases higher MP contamination in headwaters stream and a dilution effect along the river network. The structure of the proposed model allows its further implementation to account for other transport mechanisms, interactions with other emerging contaminants (i.e., pharmaceuticals), and connections with other riverine environments, making it a valuable tool for understanding and mitigating MP pollution.

1. Introduction

Research on the environmental impacts of microplastics (hereafter MP) has gained attention in the past few years, as underlined by the elevated numbers of publications tackling this important topic (Guerranti et al., 2020; Jenkins et al., 2022; J and Palmquist, 2021). MP are tiny plastic particles with a diameter ranging from 0.001 mm to 5 mm (Hartmann et al., 2019). They are present in everyday household

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products like personal care items, laundry detergents, and pharmaceuticals (usually referred to as primary MP) or generates by degradation of macroplastics (usually referred to as secondary MP). Although occurring at slow rates, the degradation of large plastic items through physical, chemical, and biological processes is considered an important source of MP (Machado et al., 2018; Zhang et al., 2021). Therefore, the significant increase in plastic production over the past few decades has led to a substantial rise of MP in environmental matrices (i.e. soil, water and air) worldwide (Geyer et al., 2017; Shams et al., 2021). Regarding the water matrix, only recently research focus shifted from marine to terrestrial and freshwater environments (Kallenbach et al., 2022) with streams and rivers considered the main export pathways for the MP observed in the world's oceans (van Wijnen et al., 2019; Strokal et al., 2023). MP find their way into rivers by means of precipitation, runoff, stormwater drainage networks and sewage systems after being treated by Waste Water Treatment Plants (WWTPs) (Shams et al., 2021; Boucher and Friot, 2017; Kay et al., 2018; Kiran et al., 2022; Müller et al., 2020a; Peng et al., 2021). By entering aquatic ecosystems, they threaten the human food chain by contaminating aquatic species (i.e. freshwater biota, fishes and their food chain) and water sources (Domenech and Marcos, 2021; Yuan et al., 2022). Additionally, MP can enter the food chain through the soil-plant system in agriculture, as well as through other non-aquatic pathways such as salt, sugar, and various food processing and packaging materials (Mamun et al., 2023). Research indicates that these particles also have negative impacts on human health, affecting the endocrine, digestive, reproductive, respiratory, and immune systems (Yang et al., 2022). In order to understand the potential pathways and impacts of MP on ecosystems and human health, different interdisciplinary efforts involving data collection, data analysis and interpretation, experimental and modeling activities are needed (see Szymańska and Obolewski (2020) and references therein).

In this context, mathematical models able to characterize the fate and transport of MP in riverine environments can be extremely helpful to detect the paths through which MP moves and interacts with and within different environments (i.e. water column and sediments). MP transport in fluvial settings is controlled by advection, dispersion, sedimentation, erosion and re-suspension as well as transformation processes such as: aggregation, dissolution, degradation, etc. The importance of these different mechanisms rely on a combination of environmental conditions, riverine hydro-morphological parameters and MP properties (i.e. size, density, shape, etc.) (see Kooi et al. (2018) and references therein). According to both the spatial (i.e. local reach scale, watershed scale up to the global scale) and temporal (i.e. flooding events, drought events, average yearly flow conditions) scales at which these transport-transformation processes are analyzed, methods and assumptions are subject to change (Krause et al., 2021; Mennekes and Nowack, 2023). Equally, data availability represent an important discriminating factor in the development and parameterization of these models (Mennekes and Nowack, 2023; Conkle et al., 2018). Models can be classified in a different way (see Uzun et al. (2022) and references therein); in the following, more attention is given on MP models developed and applied at river-networks or larger scales.

Some of the first mathematical models to characterize the fate of MP are based on existing tools developed to analyze contaminants (INCA - Contaminants (Nizzetto et al., 2016a)) or nutrients (Global-NEWS (Seitzinger et al., 2010)) transport (Nizzetto et al., 2016b; Siegfried et al., 2017). These models, nevertheless rigorous and based on realistic scenarios, are purely theoretical. The model of Nizzetto et al. (2016b) focuses on estimating MP fate at the catchment scale accounting for the retention processes occurring in soils and river sediments. The model of Siegfried et al. (2017) focuses on estimating MP fluxes emitted to sea from European rivers (i.e. at the continental scale) by considering point-sources inputs and accounting for riverine retention processes. A step forward has been made by models that starting from a similar theoretical background and using models developed and tested to analyze the fate of engineered nanoparticles (i.e. NanoDUFLOW (de Klein et al., 2016)), have been applied on real river systems (Besseling et al., 2017). In particular, Besseling et al. (2017) through NanoDUFLOW (de Klein et al., 2016) model simulated and interpreted the results of their theoretical model for detecting the transport-transformation processes controlling the fate of spherical nano and micro-plastics along a 40 km stretch of the river Dommel (Netherlands). With a similar idea, Domereq et al. (2022) propose a compartmental framework (the Full Multi), that consider different MP transport-transformation processes along and among 4 different aquatic environments (i.e. surface, flowing and stagnant water and sediment). According to their parameterization, the Full Multi can be used to *"describe rivers, lakes or ocean areas at different spatial resolutions and temporal scales"*. These models consider that all the possible transport and transformation pathways of MP depends both on their type, size, etc. and from the hydrodynamics of the peculiar: i) riverine environment (i.e. water column, sediment, water-sediment interface, etc.) or ii) other aquatic systems (i.e. lake) in which they move. Respect to previous models, these mathematical frameworks take into consideration also biological film formation, particle aggregation, sedimentation, re-suspension, polymer degradation, and particle burial into sediments.

Mennekes and Nowack (2023) use a similar approach representing, along the entire river-network of Switzerland, a compartmental model able to simulate the fate of MP along and among water column and sediments in both rivers and lakes. Respect to Domercq et al. (2022), Mennekes and Nowack (2023) reduces both the number of processes and compartments in favor of a reduction in the uncertainty associated to the large amount of input data necessary in the Full Multi.

All these models represent an important step towards assessing the behavior of MP in river networks. However, we need to take into account that increasing the number of transport-transformation processes as well as including the interaction among and along riverine compartments involves the increase in the data required by the model (i.e. data regarding particle characteristics, hydrological conditions, etc.) that are often missing (Conkle et al., 2018; Atugoda et al., 2022). To address this challenge, in order to reduce the model uncertainty, its complexity must be balanced by increasing the availability of data (both in input and for the parameterization of the processes). Unfortunately, the lack of standardized procedures for gathering, identifying, and quantifying MP, as well as methods for monitoring their presence in surface and subsurface riverine environments further complicate these challenges (Zhao et al., 2018).

Therefore, a possible alternative is to reduce the model complexity and testing its capability to capture the MP measured in different river networks. Following this strategy, our study has three primary objectives: 1) to develop a simplified yet robust modeling framework based on the advection-dispersion equation for characterizing the distribution and transport of MP in river systems; 2) to make use of open-source databases to set key parameters like MP loads and hydraulic characteristics, thereby fostering more accessible and replicable research (Klugman et al., 2011; Mai et al., 2019; CIESIN, 2018; Yamazaki et al., 2019); and 3) to establish a framework for a possible pollution assessment, enabling targeted mitigation strategies in river segments with high concentrations of MPs. To achieve the latter, and in the absence of a standardized method for evaluating the environmental risks associated with MP exposure, we employ the Pollution Load Index (PLI). This index serves as a quantitative tool to measure the overall pollution level of a particular location, by relating the concentration of MP to a reference condition (Tomlinson et al., 1980). It has been effectively utilized in prior research by several authors (Liu et al., 2022a; Xu et al., 2018; He et al., 2020; Rakib et al., 2022).

The capability of the proposed model to capture observed MP data has been tested using publicly available data from the DuPage River (USA) (McCormick et al., 2016), the Mignone River (Italy) (Gallitelli et al., 2020), and the Elbe River (Germany) (Scherer et al., 2020). By incorporating these data, the model aims to analyze to which extent advection and dispersion processes control the MP loads observed in the field. The adopted modeling framework has been built to allow its linkage to open-source hydraulic network tools (i.e. MERIT hydro (Yamazaki et al., 2019)) and to account for other transport-transformation processes of MP throughout different riverine environments. Overall, our objective is to understand how river networks control the transport of MP and to provide potential pollution maps showing which parts of the river networks are more critical in terms of MP contamination. Despite its simplifications, the proposed model is intended to serve as an accessible tool that can be used to quantify the distribution of floating MP along streams and rivers and to provide useful contamination assessment.

2. Materials and methods

This section provides a concise description of the methodology used for building the mathematical framework employed in the study. Fig. 1 schematically represent: i) the procedure designed for the estimation of the MP transport in river networks (i.e. input, model and algorithm) and ii) how model outputs can be used to provide maps of potential MP pollution (that may be employed for a preliminary ecological risk assessment).

2.1. Characterization of the river network hydro-morphological structure

River networks are complex and dynamic systems that are influenced by a variety of physical and environmental factors (Rinaldo et al., 2018). Understanding their characteristics is essential for studying the transport of MP. From the mathematical point of view, as illustrated in Fig. 2, river networks consist of a nested structure of sub-basins, reaches, and nodes. Each sub-basin indexed by the variable *i* corresponds to a distinct region of land that drains within the relative *i-th* stream or river reach. Reaches are schematized by segments where MP transport occurs and whose starting and ending locations are represented by nodes through which MP enters and/or mixes along the river network. In the model we assume that MP are injected in the upstream node of each *i-th* reach (major details are reported in Section 2.2). Accordingly, nodes are classified into two types: type 1 nodes or boundary nodes (Green dots in Fig. 2), which receive input only from the drained *i-th* sub-basin (i.e. are upstream ends of reaches with Strahler order 1); and type 2 nodes or confluence nodes (Orange dots in Fig. 2), where two or more reaches converge. Confluence nodes receive input both from the drained *i-th* sub-basin and from the connected upstream reaches. Among all the possible confluence nodes, the proposed model allows for a maximum of 3 reaches per node (typically two upstream and one downstream).

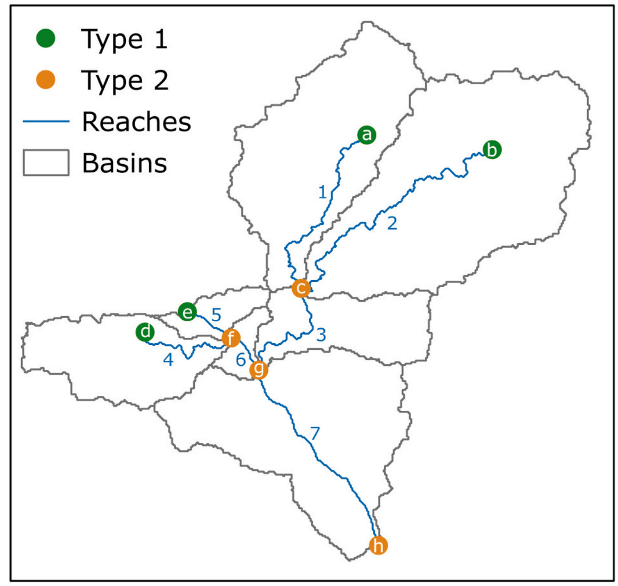

Fig. 2. Example network.

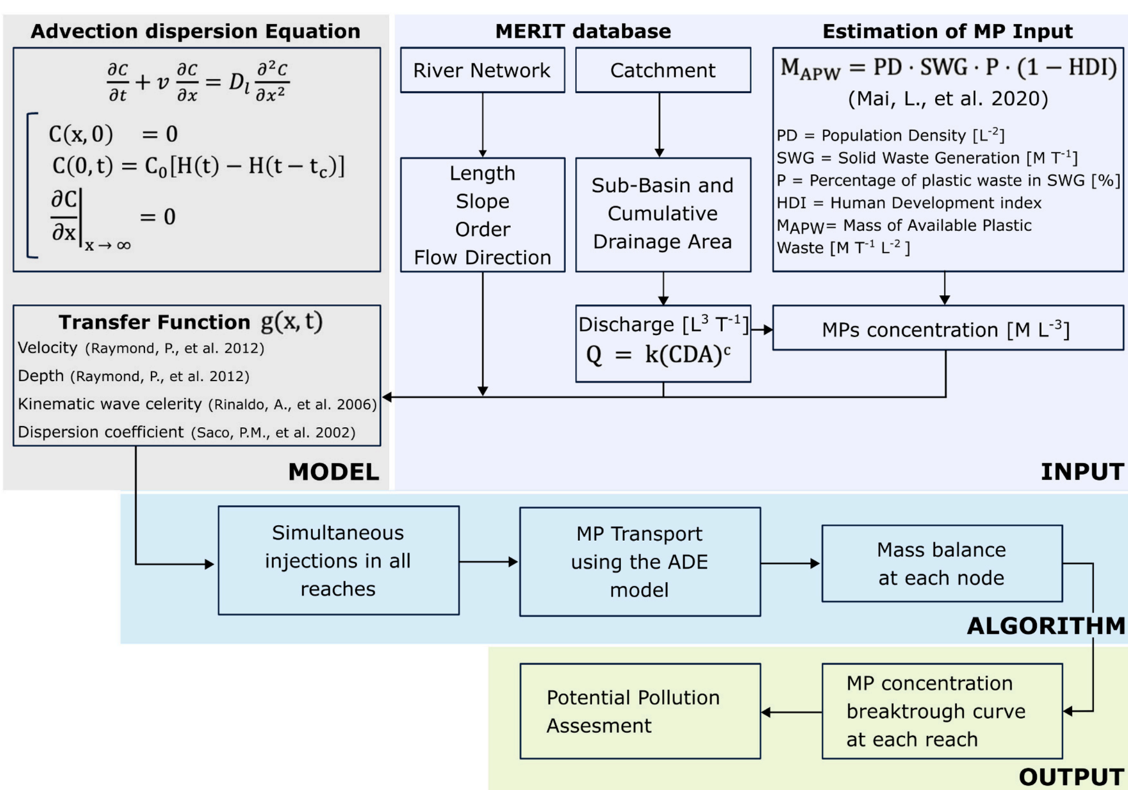

Fig. 1. Overview of the modeling procedure.

To represent the inland fluvial system of the analyzed case studies (Elbe River, Germany (Scherer et al., 2020), Du Page River, USA (McCormick et al., 2016) and Mignone River, Italy (Gallitelli et al., 2020); see Section 3 for major details) we use the MERIT-Hydro dataset (Yamazaki et al., 2019). MERIT-Hydro provides a *"high-resolution global map of river networks"* (see (Yamazaki et al., 2019) for major details). Consequently, since it provides a good approximation of river systems when compared to high-quality dataset (Uuemaa et al., 2020), it has been employed by several authors for different applications (e.g. see applications for the analysis of inundation dynamics (Shin et al., 2020; Cerrai et al., 2020), carbon transport in rivers (Liu et al., 2022b), and tracking of aquatic invasive species (Soto et al., 2023)). It is particularly suitable for our framework as it provides the nested structure described above (i.e. sub-basin-reach-nodes connection) together with information on basin area, Strahler stream order, stream length and slope.

Once fixed the dendritic structure of the river network, an essential parameter to propagate MP is water discharge (Q $[L^3T^-1]$). Q can be measured in the field, retrieved from on-line gauging station data (e.g. United States Geological Survey - USGS) or estimated through the application of rainfall-runoff models (e.g. the GRADES model from the Reach-Hydro database (Lin et al., 2019)). Since the model requires the values of Q along each *i-th* reach of the fluvial system, we utilize data from existing gauging stations to evaluate the coefficients m and k of the power law relationship between Q and the catchment cumulative drainage area (A $[L^2]$) (Galster, 2007; Tucker and Slingerland, 1997). Details on the characterization of the scaling law in Eq. (1) for the analyzed river networks are reported in Fig. S1 of the Supporting Material (hereafter, SM). This modeling choice aim to provide a synoptic characterization of the MP under normal flow conditions (i.e. no flooding or drought):

$$Q = m \cdot A^k \tag{1}$$

The hydraulic characteristics of the river reaches, specifically channel width ($w[L]$), mean flow depth ($d[L]$) and average velocity ($v[LT^{-1}]$) can be derived by using the following formulation:

$$\begin{cases} w = a \cdot Q^b \\ h = c \cdot Q^d \\ v = e \cdot Q^f \end{cases} \tag{2}$$

The hydraulic geometry coefficients ($a = 12.836$, $c = 0.408$, $e = 0.184$) and the exponents ($b = 0.423$, $d = 0.294$, $f = 0.285$) were provided by Raymond et al. (2012).

2.2. Mathematical model

The model used in this work is based on the solution of the classical one-dimensional advection-dispersion equation (Van Genuchten, 1981), employed to model the transport of MP between upstream and downstream nodes of the *i-th* river network reach. The model employs a downstream passing scheme to simulate the movement of MP through the river network:

$$\frac{\partial C_i}{\partial t} + v_i \frac{\partial C_i}{\partial x_i} = D_{L,i} \frac{\partial^2 C_i}{\partial x_i^2} \tag{3}$$

where C_i $[ML^{-3}]$ is the MP concentration along the *i-th* river reach, t $[T]$ is time, x_i $[L]$ is the downstream coordinate along the *i-th* river reach, v_i $[LT^{-1}]$ is the *i-th* mean stream velocity and $D_{L,i}$ $[L^2T^{-1}]$ is the *i-th* longitudinal hydrodynamic dispersion coefficient. $D_{L,i}$ can be estimated as a function of the hydro-geometrical parameters obtained from Eq. (2), the reach slope S_i (from MERIT-Hydro) and the kinematic wave celerity ($v_{w,i}$, $[LT^{-1}]$) according to the formulation proposed by Saco and Kumar (2002):

$$D_{L,i} = \frac{v_{w,i} \cdot h_i}{3 \cdot S_i} \tag{4}$$

with:

$$v_{w,i} = \frac{3}{2} v_i \tag{5}$$

To improve the manuscript readability, the subscript i (identifying the *i-th* reach), was omitted in the rest of the description.

The transport equation (Eq. (3)) is solved analytically by using the Laplace transform technique (Van Genuchten, 1981) under the assumptions that: a) the velocity varies reach by reach as a function of the water discharge but remain constant along each *i-th* reach; b) the settling and re-suspension processes as well as other removal processes that controls the MP transport are balancing each other (Haberstroh et al., 2021). This latter hypothesis is justifiable considering that the focus of the manuscript is on the main processes responsible of the MP fate (i.e. advection and dispersion) under average flow conditions and at the river network-scale (a detailed discussion on possible limits of this assumption are discussed in the Section 3).

Analytical solution of Eq. (3) can be obtained under different initial and boundary conditions (see SM for further details). For the former, we consider a null (0) concentration of MP within the river networks at $t = 0$ (Eq. (6)a). For the latter we model each *i-th* reach as: i) composed by an upstream node in which the release of MP can be represented as a constant pulse release (Eq. (6)b) and ii) long enough to have negligible impact on the concentration gradient at $x \to \infty$ (Eq. (6)c):

$$\begin{cases} C(x,0) = 0 & \text{(6a)} \\ C(0,t) = C_0[H(t) - H(t-t_c)] & \text{(6b)} \\ \frac{\partial C}{\partial x}\Big|_{x\to\infty} = 0 & \text{(6c)} \end{cases} \tag{6}$$

where $[H(t) - H(t - t_c)]$ corresponds to the Heaviside step function mimicking the release of a certain amount of MP (C_0) in the upstream node of the *i-th* reach composing the network and t_c is the time duration of the MP pulse. In order to simulate a continuous and uniform release of MP in our specific scenario, we have partitioned the release over a 24-h time period, while considering the average load value.

The concentration of MP injected in the upstream node of the reach is estimated according to the following formulation:

$$C_0 = \frac{M_{PW} \cdot A_c}{Q} \tag{7}$$

where A_c $[L^2]$ is the area of the drainage basin associated with the *i-th* reach and M_{PW} $[ML^{-2}T^{-1}]$ is the daily mass of generated plastic waste in the study area. This latter quantity is evaluated according to the formulation proposed by Mai et al. (2020) to estimate the release of plastic waste into rivers by accounting for the population that lives in the corresponding drainage basin:

$$M_{PW} = HPD \cdot SW \cdot P \cdot (1 - HDI) \tag{8}$$

where HPD $[capita\, L^{-2}]$ is the human population density obtained from NASA's Data Center's gridded population raster map for each basin (CIESIN, 2018), SW is the solid waste generation $[MT^{-1}\, capita^{-1}]$, P $[-]$ is the percentage of plastic present in the generated solid waste and HDI $[-]$ is the Human Development Index which ranks countries into four levels of human development based on life expectancy, education, and per-capita income (Klugman et al., 2011). SW, P and HDI are parameters unique by country: the former two are extracted from the World Bank's "What a Waste 2.0" report (Kaza et al., 2018); while values of the latter are provided by United Nations (Klugman et al., 2011). HDI index employs the economic growth and the achievements of the population as indicators to characterize the development of countries, which is also correlated to waste management practices (Namlis and Komilis, 2019). All the values used for the computations are reported in Supplementary Table S1.

Under the above described conditions the flux concentration of MP at a given location (x) along the river reach can be calculated using the following convolution integral:

$$C\left(x,t\right) = \int_0^t C_0 \cdot g\left(x, t - t_0\right) dt_0 \tag{9}$$

where $g(x, t - t_0)$ represent the transfer function of the transport problem in Eq. (3) under the initial and boundary conditions in Eq. (6) (Van Genuchten, 1981) (see Eqs. S5 and S9 and the details on its derivation in SM). SM provides the details on how to specialize the transfer function $g(x,t)$ in accordance with the node type (injection or confluence) and reach characteristics. The specialization takes into account two factors: negligible dispersion in reaches with higher Peclet numbers and the input signal of MP after confluence nodes.

2.3. Model implementation

A Python script that implements the modeling framework described above (summarized in Fig. 1) has been developed to predict MP concentration along the river networks as well as the impact of the associated pollution. Data preparation and acquisition were performed using the free, open-source Geographic Information System (GIS) application QGIS (QGIS Developed Team, 2023). Shape files containing the river network structure and the relative sub-basins (obtained from MERIT-Hydro (Yamazaki et al., 2019)) and raster data with information on the population (CIESIN, 2018) were imported and processed in QGIS that allow an efficient and accurate data management. Data are then exported within text files (.csv/.txt) to be read and processed by the developed python code (see example in Supplementary Tables S2, S3 and S4).

To estimate the MP transport, the model starts by automatically identifying type 1 and type 2 nodes. The concentration of MP generated within each *i-th* sub-basin according to Eq. (7) is injected for a constant period of time (t_c) at the upstream node of its corresponding reach. This injection process occurs simultaneously in all nodes of type 1 and type 2, regardless of their location along the network. The transport is then solved using Eq. (9) where the transfer function $g(x,t)$ is defined by Eq. S5. When the injected MP reaches a type 2 node, the concentration from the upstream reaches are mixed together by considering the mass balance (Eq. (10)):

$$C_c = \frac{C_a Q_1 + C_b Q_2}{Q_3} \tag{10}$$

where C_c is the concentration leaving node c, while C_a and C_b are the concentrations from reaches 1 and 2, respectively, that arrive at node c. The discharges of reaches 1 and 2 are denoted by Q_1 and Q_2, respectively, while Q_3 represents the total discharge at node c (i.e. given by the sum of the discharges from reaches 1 and 2: $Q_3 = Q_1 + Q_2$). Subsequently, the MP input to the downstream reach is estimated by subdividing the arriving concentration profile into multiple finite elements that simulate a series of instantaneous injections with a $\Delta t \approx 1\,minute$. The resulting MP concentrations are transported using the transfer function in Eq. S9 and their resulting curves are added up to obtain the breakthrough curve (hereafter, BTC) at the end of each reach. Similar approaches have been used in various studies, and solutions for the advection-dispersion equation for instantaneous injections can be found in literature (Van Genuchten, 1981; Ogata and Banks, 1961; Fan et al., 2015). The output of the procedure is the characterization of the MP BTC at each node and control section of interest.

Before implementing it to analyze MP transport, we tested our framework against the validated advection-dispersion model implemented by Runkel (1996) where two hypothetical examples for a non-reactive solute were simulated at $x = 100$ (Fig. 3a) and $x = 2000\,m$ (Fig. 3b). Fig. 3 shows the comparison between our modeled (black continuous line) and the BTC of a passive tracer obtained by Runkel (Runkel, 1996) (red dashed line). A perfect agreement can be observed, also underlined by the values of the coefficient of determination $R^2_{x=100} = 0.99$ and $R^2_{x=100} = 1.00$ between the two BTC testing the applicability of the proposed model.

Finally the model can be employed to do an assessment of the level of MP pollution of the reaches using the Pollution Load Index (PLI) (Liu et al., 2022a; Xu et al., 2018; He et al., 2020; Rakib et al., 2022). This index compares the concentration of a given pollutant against a predetermined reference condition or baseline (Tomlinson et al., 1980). For a singular MP BTC, the PLI can be calculated as shown in Eq. (11). Based on the resulting value, there are four risk categorizations: a PLI of less than 1 indicates low contamination, between 1 and 3 indicates moderate contamination, between 3 and 6 indicates considerable contamination, and greater than 6 indicates very high contamination.

$$PLI = C_i / C_o \tag{11}$$

where C_i is the concentration of MP in the studied reach and C_o is the baseline concentration of MP (i.e. the lowest observed concentration of MP along the river network).

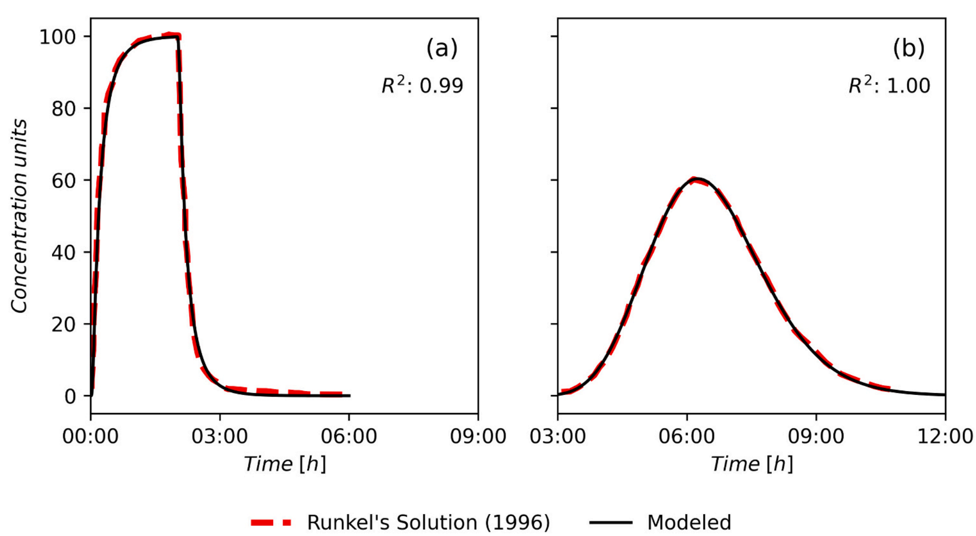

Fig. 3. Comparison between the Breakthrough Curves of a Non-Reactive Solute obtained with the model (black continuous line) and those published by Runkel (1996) (red dashed line) at (a) $x = 100\,m$ and (b) $x = 2000\,m$.

3. Results and discussions

In this section, we present the results of our MP transport model and discuss the implications and its potential uses for pollution assessment. Existing MP models, such as those proposed by Siegfried et al. (2017), Nizzetto et al. (2016b), have primarily been used to provide an estimate of the total yield of MP. The model proposed here aims to predict the concentrations of MP in different sections and points of interest across the entire river system.

The model was applied to three river networks located in different parts of the world and for which data of MP were published. These are (a) the Mignone River near Civitavecchia, Italy (Fig. 4, (Gallitelli et al., 2020)); (b) the DuPage River near Chicago, United States (Fig. 5, (McCormick et al., 2016)), and (c) the Elbe River near Hamburg, Germany (Fig. 6, (Scherer et al., 2020)). Figs. 4, 5 and 6 shows the maps of the three river networks with the measurement points (filled circles on the map) and, for each of them, the points where MP is estimated through the model.

In all the three river networks, and this is especially visible for the Mignone river (Figs. 4), the BTCs at the control points connecting reaches with Strahler stream order $SO = 1$ (i.e. S1, S2, S3, and S4) exhibit the characteristic shape of an injected rectangular pulse evolving over time because of advection and with a negligible contribution of dispersion due to the short length of these reaches. As the MP move downstream, the shape of the BTCs changes according to the structure of the upstream drainage system and the variability of the MP inputs. For example, in Fig. 4, four peaks are clearly visible in the last reach of the Mignone river network (from S5 to S8) corresponding to the MP contributions of the upstream basins. Similar consideration are valid for the BTC represented by the green line in Fig. 5 for the DuPage River. Finally, in the Elbe River (Fig. 6), as two watersheds run independently in the same sub-basin, the BTCs shape reflects a series of combining factors. The green BTC, representing a measuring point close to Hamburg (green dot in Fig. 6), is characterized by a higher concentration of MP due higher population densities ($\sim 610000\,inh$) that reflects in higher MP inputs; while its slimmer shape is given by the size of the watershed that results in shorter travel times. On the other hand, the BTC at Geesthacht (red dot in Fig. 6), exhibits a low concentration of MP reflecting the lower population density ($\sim 12000\,inh$) contributing to MP inputs while the multiple peaks reproduce the contribution of its larger upstream watershed.

Some clear differences in MP concentrations are evident between the three rivers. The Mignone River displays maximum MP concentrations of approximately $700\,\mu g/l$, whereas the DuPage River reaches concentrations of about $\sim 22000\,\mu g/l$ in their most upstream reaches. Despite their relatively similar catchment areas, ($410.71\,km^2$ and $632.75\,km^2$, respectively), their significantly different population densities ($56.7\,inh/km^2$ and $1129.47\,inh/km^2$ respectively) lead to MP concentrations up to 30 times higher in the DuPage River. A similar trend is observed in the Elbe River, where the basins that arrive to the Hafenstrasse (referenced by the green dot in Fig. 6) transport MP from a larger population. Although a larger catchment size can yield higher MP mass, it does not necessarily leads to a proportionally increase in concentration.

We have further evaluated the most downstream reach of each network by analyzing various BTC features, which are frequently used in contaminant and tracer studies (Kwon et al., 2021). These features, detailed in Table 1, and include mean and peak concentrations, the time of peak concentration and the presence interval. Such parameters can give important information about the impact of river networks dendritic structure in controlling MP transport. For instance, the Mignone River and the DuPage River show similar times of peak concentrations, indicating comparable catchment sizes. It is noteworthy that even when networks possess similar sub-basins areas, the concentration is directly impacted by various factors: these include the MP load input from the basin, contributions from upstream sections, effects of mixing at confluence nodes, and distinct hydraulic characteristics of various reaches (e.g.: discharge, velocity). A case in point is the Elbe River (Small Sub-Catchment), which exhibits higher concentrations than the DuPage River, despite having comparable catchment sizes ($532.45\,km^2$ and $632.74\,km^2$ respectively).

The behavior of MP transport is significantly influenced by discharge, which is estimated through a distinctive power-law

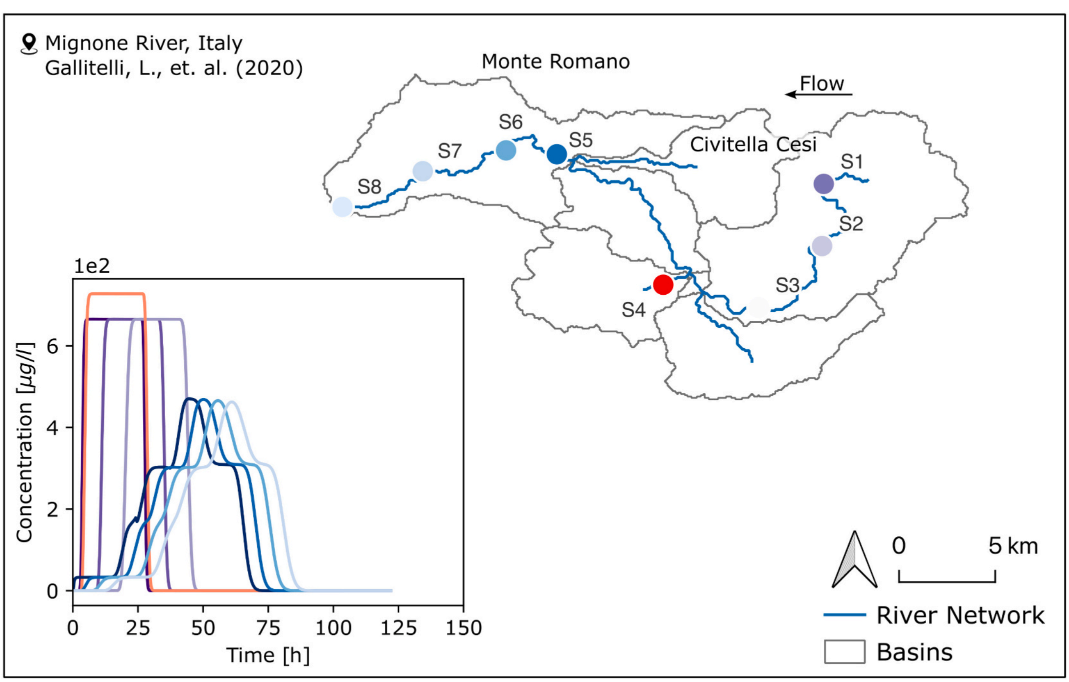

Fig. 4. Model simulation of microplastics in the Mignone River, Italy - Breakthrough Curves and Spatial Distribution. Left: Breakthrough curves showcasing results at measurement points, revealing the dynamic behavior of MP transport. Right: Map of basins and river network, indicating the locations of measurement points (filled circles) found in literature for comprehensive spatial analysis.

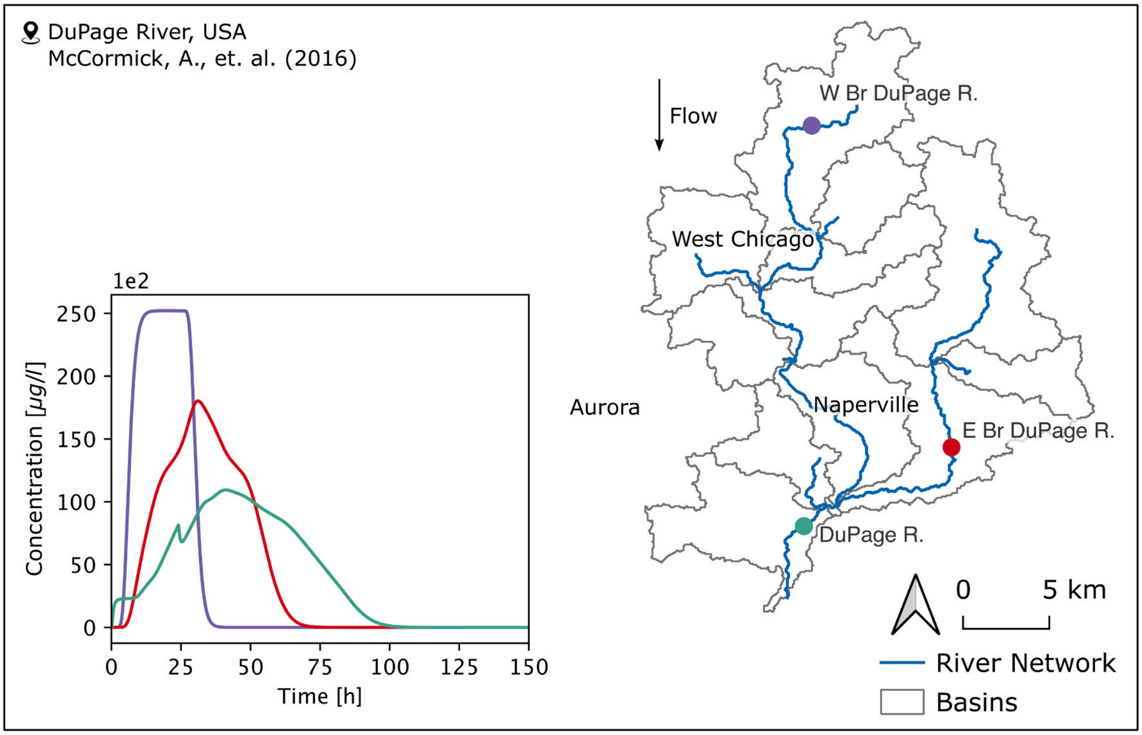

Fig. 5. Model simulation of microplastics in the DuPage River, USA - Breakthrough Curves and Spatial Distribution. Left: Breakthrough curves showcasing results at measurement points, revealing the dynamic behavior of MP transport. Right: Map of basins and river network, indicating the locations of measurement points (filled circles) found in literature for comprehensive spatial analysis.

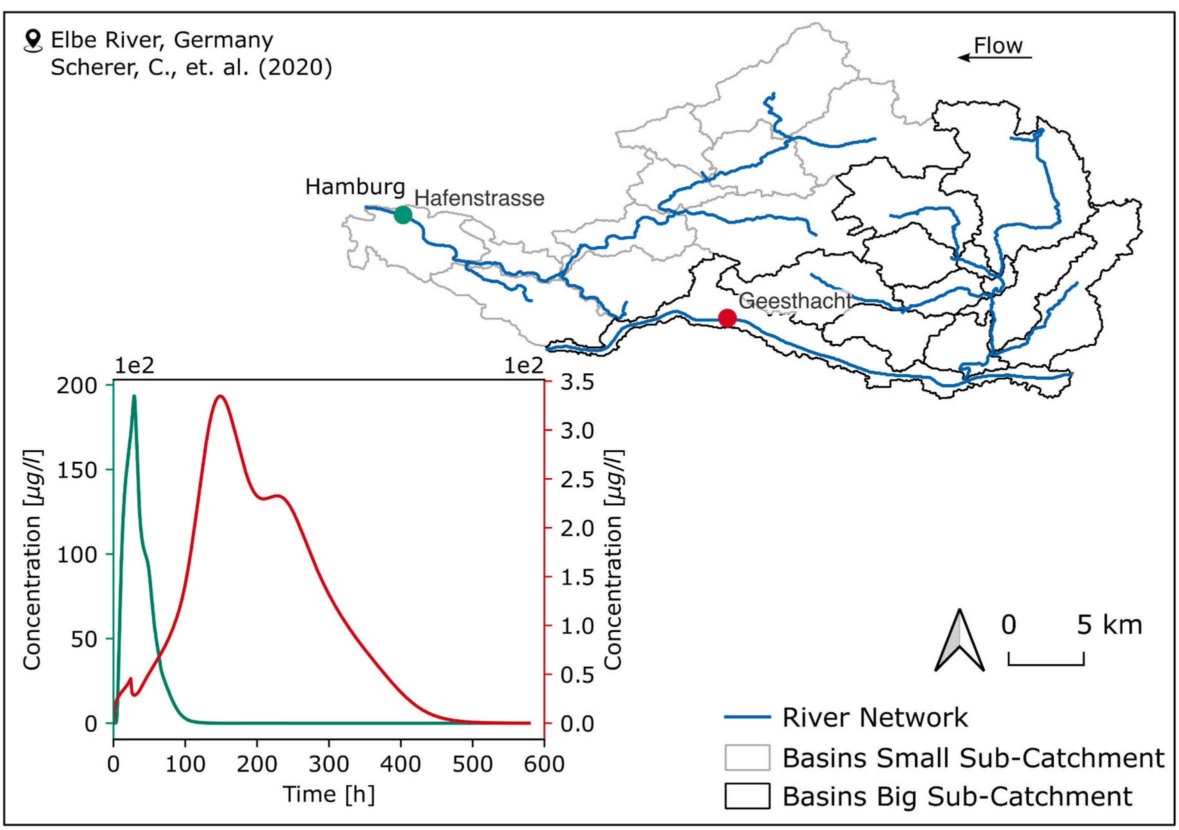

Fig. 6. Model simulation of microplastics in the Elbe River, Germany - Breakthrough Curves and Spatial Distribution. Left: Breakthrough curves showcasing results at measurement points, revealing the dynamic behavior of MP transport. Right: Map of basins and river network, indicating the locations of measurement points (filled circles) found in literature for comprehensive spatial analysis.

Table 1
Breakthrough curve (BTC) features at most downstream reach across all datasets.

	Mean Concentration	Maximum Concentration	Time to peak Concentration	Presence interval	
				Start time	End time
	[$\mu g/l$]	[$\mu g/l$]	[h]	[h]	[h]
Mignone River	117.19	370.91	66.82	1.77	193.32
DuPage River	3912.33	10,826.91	48.62	0.03	578.92
Elbe River	3680.07	19,343.75	28.87	0.83	122.03

relationship for each case study (See Tables S5, S6, S7 and Fig. S1). Uncertainties of power law relationships in streamflow calculations arise from the inherent complexities of natural hydrological systems and the challenges in accurately capturing the dynamics of flow behavior at various scales (Ayalew et al., 2014). Additionally, there are uncertainties originating from the estimation of hydraulic parameters such as river width and depth. These factors are often variable, adding an additional layer of complexity to accurate predictions. However, it is pertinent to note that in instances where these hydraulic parameters are known with certainty, these values can be directly utilized to potentially increase the accuracy of predictions. While quantifying these uncertainties lies beyond the scope of this study, acknowledging their presence is essential for comprehending the limitations and potential variations in the results.

In specific scenarios where streamflow data derived from power-law relationships lead to unusually high values, travel times can be dramatically reduced, resulting in a less evident dispersion effect or rate. This can be observed in a narrower-shaped BTC. It is essential to note that the power-law relationships used in this study, having to represent measurements collected during average flow conditions, are based on averaged seasonal streamflow data, thus neglecting extreme (i.e. flood and drought) conditions.

MP estimations from our model were compared with MP measurements conducted in control sections in all the analyzed basins. Aiming to reduce uncertainties related to the moment or stage at which the MP samples were collected, we decided to use the mean concentration of the resulting BTC for the comparison. Mean concentration was computed by averaging concentration values in the presence interval on the BTC ($C > 0$). Moreover, MP measurements in the field are typically expressed as number of particles per volume of water. Therefore, to make the comparison possible, we used an average particle weight of 962.837 μg (Schmidt et al., 2017). While some of the MP collected data distinguish the shape of the MP particles (i.e. microbeads, foam, pellets, spheres), the transport dynamics related to the shape of the MP particles still remain subject to further investigation. For instance, fibers tend to agglomerate, leading to the formation of larger particles (da Fonseca et al., 2022).

Fig. 7 shows the comparison between the modeled and the measured data. The majority of data points fall within the 95% confidence interval, indicating a significant agreement between model predictions and the measured data. Furthermore, statistical indicators such as the Percent Bias (PBIAS = − 1.3%) suggest a slight tendency for the model to overestimate the observed values, although the deviation from zero is relatively minor (Moriasi et al., 2007). The Nash-Sutcliffe Efficiency (NSE) is 0.70, the coefficient of determination (R^2) is 0.72, and the King-Gupta Efficiency (KGE) is 0.85. These indicators collectively validate the overall performance of our model in predicting "on-average" the MP concentrations measured in real river settings.

It should be stressed that the lack of riverine MP data is a global concern. In most cases their unavailability is related to the fact that there

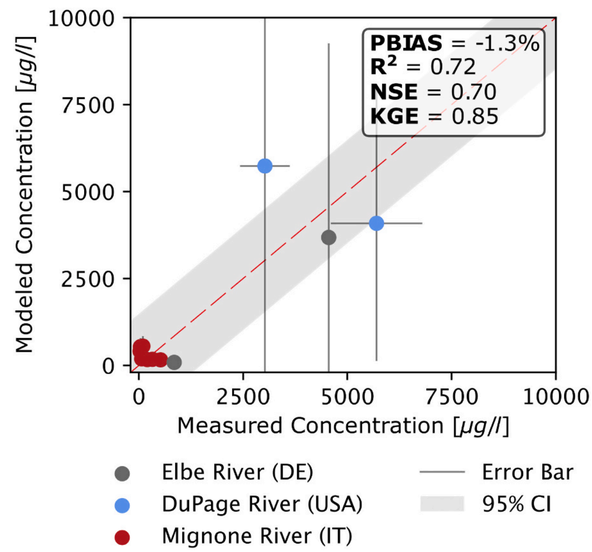

Fig. 7. Measured MP concentration vs Modeled MP Concentration. Dashed red line corresponds to the bisector line.

are no standardized and recognized protocols to measure their concentrations (Müller et al., 2020b). Sampling and analyzing the MP collected in the field, as well as extracting the different MP particles is a challenging process. For most of the cases it requires the combination of different laboratory and image acquisition techniques for their automatic counting and classification. For instance, She et al. (2022) reported a 25 − 50% of spatio-temporal sampling error when using a trawling net. Consequently, it is particularly challenging to compare data from different studies (with all the associated uncertainties) (Li et al., 2020). The non-perfect matching between model results and measured data may be attributed both to the model assumptions (i.e. settling and re-suspension processes are in equilibrium) and to all the uncertainties associated with the model parameterization. Some studies underline that the two transport mechanisms neglected are not in balance (Gerolin et al., 2020; Yang et al., 2021) highlighting a limit of the proposed procedure but also an avenue for further refinement of the model.

Therefore, placing our model alongside others in the literature (Mennekes and Nowack, 2023; Siegfried et al., 2017) is not straightforward because of our assumption to consider in balance sedimentation, re-suspension and other transformation processes. The model does not account of processes that can remove and transform MP during the interaction between water column and the surrounding sediments (Mennekes and Nowack, 2023) or with the lateral river banks (Scherer et al., 2020; Matjašič et al., 2023). Our choice to consider MP transport only within the water column is mainly due to: i) the huge amount of data required to characterize these interactions both in terms of hydraulic (i.e. exchange pathways and their change in space and time) and MP (i.e. size, shape and density of different MP types) data and ii) the impossibility to test and validate possible parameterization because to the lack of standardization procedures for measuring MP across and among these different environments. Moreover, as reported by Mennekes and Nowack (2023) analyzing different scenarios (i.e. interaction between water column and sediments as well as the presence of lakes), streams and rivers are expected to retain ∼ 17% of all input MP. This percentage is strongly influenced by the MP type and in the river networks analyzed in the present study, most of the MP observed are buoyant (polyethylene, polypropylene, polyamide) (Fiore et al., 2022).

Consequently, despite limited data availability for validation, the results proposed in Fig. 7 remains significant for several reasons: the selected data was carefully reviewed for adequacy, and the algorithms of the model were based on established physical theories. The model showed promising performance on the available datasets, demonstrating its potential for other applications. While more data would help strengthen the model's validity, this research represents a valuable step forward in addressing MP transport (by advection and dispersion) in river networks.

3.1. Model application: assessment of MP pollution

Having established the capability of the model in predict the MP transport, its outputs can be utilized to provide a potential pollution assessment using the Pollution Load Index (PLI). Results obtained are shown in Fig. 8. According to the PLI definition (Tomlinson et al., 1980), along the Mignone River network, we observe *Moderate contamination* in most reaches; while moving downstream, the level of pollution decrease, with the most downstream reach classified as having *Low contamination*. This trend can be attributed to the attenuation of MP concentration caused by dispersion processes and the effect of dilution amplified by the increase of water discharge. Along the Mignone river, the effect of MP input (reflecting the degree of human impacts on different sub-basins) is explained by the three upstream basins having the highest population density, which significantly decreases towards the last reach. Additionally, both the MP input and the discharge are proportional to the area of each reach, indicating that the PLI is mostly homogeneous throughout the river. A similar pattern is observed in the DuPage River. Most reaches of order 1 present *Considerable contamination* with a gradual decreases to *Moderate contamination* levels. It is worth to note that the only reach classified with *Low contamination* is located in the sub-basin with the lowest population density (light blue line within the Du Page river network). On the Elbe River, overall results indicate higher contamination levels. Most reaches of stream order $SO = 1$ fall into the category *Very High contamination*, with MP concentration levels decreasing in downstream reaches, leading to *Moderate contamination* in the small sub-basins and *Low contamination levels* in the large sub-basins. It is possible to hypothesise that reaches of $SO = 1$ (typically headwaters) in high populated areas will exhibit higher contamination levels due to the combined effect of smaller water discharge and higher MP input (Ferraz et al., 2020). As observed before, moving downstream the drainage area of the catchment increases, resulting in an increase of the streamflow responsible of a major capability of the river network to dilute pollutants. It should be emphasized that currently, there is no global consensus on a baseline value of concentration (C_o) for MP used in environmental assessment purposes. Consequently, the PLI scores calculated here cannot be directly compared between case studies. In each of the three cases, the minimum concentration estimated by the model is higher than the minimum measured value. This might be attributed to the lack of continuous MP measurements in rivers, for which it is not known the exact value of water discharge during the sample collection. Therefore, our approach estimates a higher baseline concentration that leads to lower PLI values. It is crucial to consider these limitations and to take them into account when interpreting the results and making conclusions based on the application of this model.

4. Conclusions

Representing the natural connectivity between terrestrial and marine environments, river networks play an important role in controlling the fate of MP and consequently the amounts released to seas and oceans. The methodology and the analyses presented in this work intents to integrate the ongoing research on MP by observing the problem at the watershed scale, during average flow conditions and by balancing some of the physical and hydrodynamic processes that induce settling and resuspension of these harmful particles.

Set the spatial and temporal scales, among the different mechanisms (for example atmospheric deposition, WWTPs effluents and stormwater runoff) that controls the injection of MP in fluvial ecosystems, the proposed model estimates the MP load combining all these possible sources through an anthropogenic effect accounted via the population and solid waste generation that characterize the different sub-basins of the analyzed areas.

The developed python script solves the classical advection-dispersion equation along the reaches that compose the river network by treating it as a nested structure (derived from MERIT-Hydro). The nested structure is composed by injection nodes and confluence nodes (where in addition to MP input, mixing occurs) that are connected by

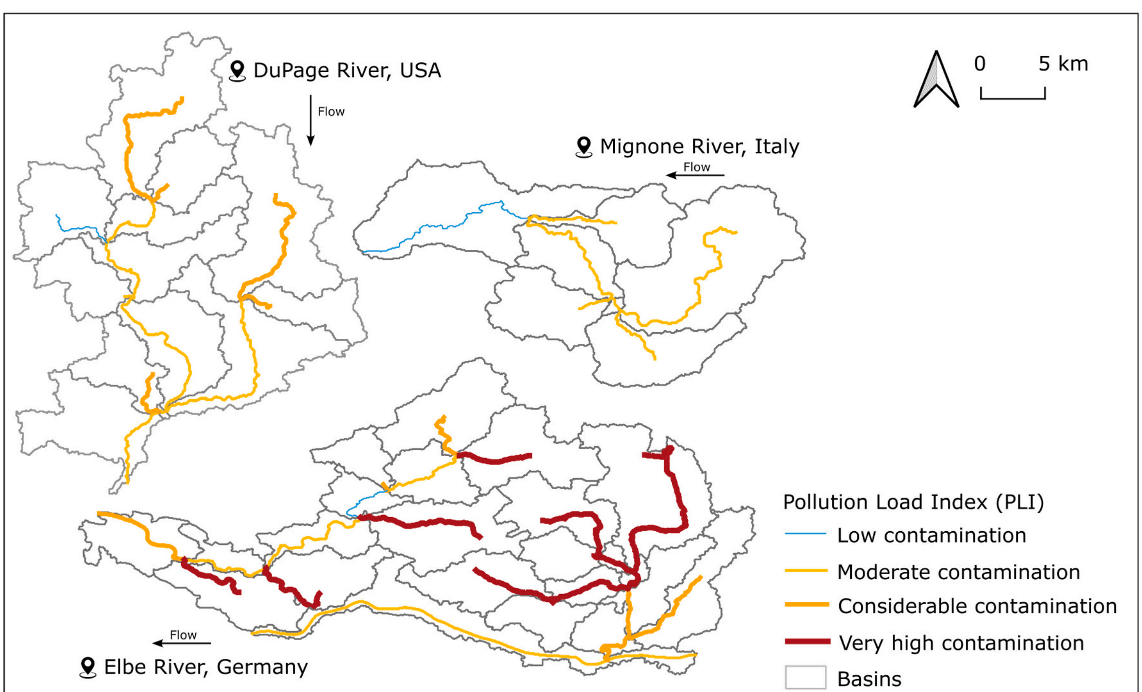

Fig. 8. Maps with potential pollution using the Pollution Load Index (PLI) for the DuPage River (USA), Mignone river (Italy), Elbe River (Germany).

reaches where MP transport occurs. To predict the MP fate, the model estimates the necessary hydrodynamics parameters of the river reaches (i.e. water discharge and hydrodynamic dispersion) by using available gauging station data.

The validity of the model was tested by using available literature data on three river networks located in different parts of the world: Mignone river (Italy), Du Page river (United States of America) and Elbe river (Germany).

Although with some uncertainty related to both the modeled parameters and the collected data, comparison between model predictions (i.e. the average MP concentration obtained from the BTC) and observed data shows a quite good agreement as underlined by some statistical indexes: $NSE = 0.70$, $R^2 = 0.72$ and $KGE = 0.85$.

The model output is also used to perform a potential pollution assessment by using the Pollution Load Index (PLI). This allows to better comprehend how the combination of anthropogenic, morphological and hydrological factors can combine to provide insights into the level of contamination by MP in the different reaches of the river network. Results shows that in all the analyzed systems, headwater streams present a higher degree of contamination that reduces by the dilution effect operated by the increase of water discharge moving along the river network. Evidently, the reduction in the degree of contamination is strongly related to the population (i.e. the mass of MP produces) that occupies the different sub-basins. The model structure was built in a way that it can be further implemented to account for other transport mechanisms (i.e. deposition, resuspension and bioaggregation), the interaction with other emerging contaminants (i.e. pharmaceuticals) and with other riverine connected environments (i.e. hyporheic zone, riparian areas and groundwater).

CRediT authorship contribution statement

N. P. D. A. and A. M. designed the study. N. P. D. A. developed the code and run the simulations. N. P. D. A. and A. M. analyzed data and results and wrote the paper. All authors contributed to the writing of the manuscript and interpretation of the results.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.168227.

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