The Full Multi: An open-source framework for modelling the transport and fate of nano- and microplastics in aquatic systems

Domercq, P.; Praetorius, A.; MacLeod, M.

DOI
10.1016/j.envsoft.2021.105291

Publication date
2022

Document Version
Final published version

Published in
Environmental Modelling & Software

License
CC BY

Citation for published version (APA):
https://doi.org/10.1016/j.envsoft.2021.105291

General rights
It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

Disclaimer/Complaints regulations
If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: https://uba.uva.nl/en/contact, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.

UvA-DARE is a service provided by the library of the University of Amsterdam (https://dare.uva.nl)
The Full Multi: An open-source framework for modelling the transport and fate of nano- and microplastics in aquatic systems

Prado Domercq a,*, Antonia Praetorius b, Matthew MacLeod a

a Department of Environmental Science, Stockholm University, Stockholm, Sweden
b Institute for Biodiversity and Ecosystem Dynamics, University of Amsterdam, Netherlands

A R T I C L E   I N F O
Keywords:
Plastic pollution
Environmental fate modelling
Freshwater
Microplastics
Open source

A B S T R A C T
We present a freely available open-source modelling framework to explore environmental transport and fate of nano- and microplastics in aquatic systems. The “Full Multi” models 1) fragmentation of plastic into a predefined set of size classes, 2) speciation of plastic particles between pristine, heteroaggregated, biofouled, and biofouled & heteroaggregated species, 3) dynamic vertical exchange between water layers and sediment of a freshwater system, and 4) horizontal particle transport by eddy diffusion and advective flow. The Full Multi framework relates emission rates to environmental exposure concentrations while considering the intrinsic properties of plastic particles and variable environmental system properties. The model can be applied to analyse scenarios with different process descriptions, plastic types, emission routes and environmental parameters for hypothesis generation, to identify dominant fate processes, and in hazard and risk assessment. Here, we introduce and illustrate the framework by modelling plastic particles with a range of densities in a generic flowing river system.

1. Introduction
Plastic pollution is found in rivers, lakes and the ocean globally (Peng et al., 2020; MacLeod et al., 2021; Sebille et al., 2015; González-Fernández et al., 2021) and it is present in a variety of sizes, shapes and compositions (Kooi et al., 2021). Plastic debris in aquatic systems is subject to weathering by ultraviolet light, biodegradation and physical stress that results in fragmentation and chemical degradation over time. At the same time, plastic particles can aggregate with suspended particulate matter (SPM) and can be subject to colonization by biofilms. All of these processes affect transport, fate and size distribution of plastic particles, and are likely important for determining their environmental exposure levels (Wagner et al., 2014; Horton et al., 2017; Rochman et al., 2019; Kooi and Koelmans 2019; Rummel et al., 2017).

Also, these processes will occur to different extents depending on the particle characteristics and environmental conditions (Singh et al., 2019). For example, plastic particles with lower density than water might be transported long distances by surface currents, but the same material may sink into sediments and become immobile if it is colonized by a dense biofilm (Lobelle et al., 2021; Hoellein et al., 2019; Semcecesen and Wells 2021; Kaiser et al. 2017; Fazey and Ryan 2016; Ye and Andrady 1991). Large plastic particles that are more dense than water will sink into sediment, however fibres and small fragments of plastic with the same density may remain suspended in water due to slow settling velocities. Changes in the size, shape and effective density of plastic particles over time due to fragmentation, biofouling and formation of aggregates with natural SPM can thus strongly affect the transport and fate of plastic in the environment (Horton et al., 2017). The combined complexity of factors governing these processes restrains our current understanding about these mechanisms, which limits our ability to forecast exposure concentrations for different plastic types and emission scenarios, and to assess potential risks. Therefore, one of the key challenges in plastic pollution is to decipher how their different fate and transport processes compete with each other under different environmental conditions and for different plastic types, so that dominant environmental fate pathways can be identified and targeted for further study (Horton et al., 2017).

Mathematical models have the potential to serve as platforms to integrate information and understanding about the transport and fate of plastics in the environment, and in the face of high uncertainties about appropriate process descriptions, to support hypothesis generation to guide research (MacLeod et al., 2010). Experience with environmental models such as the Water Quality Analysis Simulation Program (WASP) (Di Toro et al., 1983) or the OECD PUV & LRTP Screening Tool.
(Wegmann et al., 2009), demonstrates that, once confidence has been established in a model, it can be used for estimating environmental concentrations for risk assessment or as a basis for formulating monitoring strategies.

In recent years modelling approaches have been developed for quantifying the export of plastics into the oceans (Siegfried et al., 2017; van Wijnen et al., 2019; Lebreton et al., 2017), for assessing the processes that transport or retain plastics in rivers and river catchments areas (Siegfried et al., 2017; Unice et al., 2019; Besseling et al., 2017), and to study the fate of plastic in the ocean (Sebille et al., 2015; Lebreton et al., 2012; Liubartseva et al., 2016; Koelmans et al., 2017). However, up to date, no general framework covering all relevant plastic particle processes (e.g. aggregation, biofouling, fragmentation and degradation), the variability of plastic properties (i.e. size, density and shape) and that is adaptable to different regions and temporal scales has been yet introduced (Alimi et al., 2018). With the aim of filling this gap, here, we present an open-source, flexible multimedia mass-balance transport and fate modelling framework for plastic particles in aquatic environments.

Multimedia mass balance fate and transport models (Praetorius et al., 2012; Meesters et al., 2014; Garner et al. 2017; Liu and Cohen 2014; Knightes et al., 2019) have been recently developed for the study of nanoparticles in the environment. Such models have the potential to be adapted to integrate processes descriptions relevant for plastic using the same principles of nonequilibrium colloidal behaviour, which differs significantly from the behaviour of dissolved chemical contaminants (Praetorius et al., 2014; Hüffer et al., 2017; Alimi et al., 2018). Furthermore, multimedia mass balance models have a modular design that permits adaptation to different regional environments, and flexibility to describe different spatial and temporal scales (MacLeod et al., 2010; Wania et al., 2006). In the fast evolving field of plastic pollution research, where high uncertainties exist for many process descriptions and new knowledge is constantly being generated (Jahnke et al., 2017; Arp et al., 2021), a flexible and adaptable model framework that the scientific community has free access to as open-source software fills a research need.

The Full Multi framework is based on a generic modular unit cell that includes first-order kinetic process descriptions for vertical and horizontal transport, fragmentation, biofouling and heteroaggregation of plastic particles with natural SPM in aquatic systems. The framework can be readily parameterised to describe rivers, lakes or ocean areas at different spatial resolutions and temporal scales. The base version of the Full Multi framework can be extended with new process descriptions or by adding new mass balance compartments as new findings on plastic particle fate processes become available by modifying the mass-balance equations as described in the Full Multi repository user’s manual. The aim is to provide a modelling framework that serves as a standard reference for evaluating the fate of plastic particles using easy-to-run and open access code. The framework is coded in Python and the version described in this paper is accessible via the GitHub repository https://github.com/Nano2PlastProject/TheFullMulti_RIVER.

We illustrate the Full Multi framework by application to describe three types of plastic particles with a range of densities \(d\); polyethylene (PE, \(d = 980 \text{ kg/m}^3\)), polyamide (PA, \(d = 999 \text{ kg/m}^3\)) and polyvinyl chloride (PVC, \(d = 1580 \text{ kg/m}^3\)) in a generic flowing river system. The model scenarios reported in this paper can be replicated using tutorials presented as Jupyter notebooks on the Full-Multi GitHub repository (i.e. FullMulti_RiverModel_Paper_PE.ipynb, FullMulti_RiverModel_Paper_PA.ipynb and FullMulti_RiverModel_Paper_PVC.ipynb).

2. The Full Multi model framework

2.1. Model description

The Full Multi model framework’s name reflects that it is a multimedia mass-balance model that tracks plastic particles in multiple sizes and multiple speciation forms. Environmental compartments within
each unit cell of a Full Multi model are represented as well-mixed boxes and particle size and species-specific transport and fate processes within and between compartments are described using first order kinetics. Some process descriptions in the Full Multi model are taken from the environmental fate and transport model for (inorganic) nanoparticles developed by Praetorius et al. (2012), and additional processes relevant to plastic particles, including fragmentation and biofouling, have been added based on a literature review. The overall mass balance is defined by a system of coupled first-order differential equations. In the default parameterization of the framework described in this paper there are five particle size classes (i.e. 0.1, 1, 10, 100 and 1000 μm particles) and four speciation states (i.e. pristine, heteroaggregated, biofouled, and biofouled & heteroaggregated), thus for each compartment in the modeled system there are (5 × 4 = 20) coupled mass balance equations.

### 2.1.1. The unit cell and the model domain

Models of plastic transport and fate in aquatic systems are built up in the Full Multi framework using a predefined default unit cell that consists of two different types of environmental compartment: water and sediment, that are further subdivided into smaller compartments depending on the modelled system. Applied to describe a generic river system, such as in the default parameterization, the compartments represent: (1) surface water, (2) flowing water, (3) stagnant water and (4) surface sediment (Fig. 1). Compartments are linked to define model unit cells, and unit cells are linked to define the entire modeled domain.

The dimensioning of the compartments and selected number of unit cells will determine the spatial resolution of the model (Table 1) and the connectivity can be adjusted to represent a variety of aquatic systems. For example, a single unit cell representing a lake where particle transport is only studied between compartments in the vertical axes, or several interconnected cells via eddy diffusion representing the global ocean.

In the default generic river parameterization unit cells are connected in series by a unidirectional flow of water in the “flowing water” and “surface water” compartments, as well as bed load transport in the “sediment” compartments. To describe a specific river system, connections between unit cells can also be added to describe tributaries to a main river channel. Each unit cell in a modeled system can be individually parameterised to describe the geographical and geochemical features of the water body.

### 2.1.2. Description of plastic particles

The plastic particles in the Full-Multi framework are represented as a distribution of particles via discrete size classes and described by input parameters specifying diameter, density and shape (Table 2). The default parameterization of the model framework assumes five size classes (i.e. 0.1, 1, 10, 100 and 1000 μm particles), which represents a compromise between computational time/power requirements and resolution of plastic particles at a range of sizes.

The plastic particles can take on four different speciation forms: pristine, heteroaggregated, biofouled, and biofouled & hetero-aggregated. While for the mass balance accounting the particles remain in their originally assigned size class upon speciation (i.e. each particle species is tracked within the size class of the parent pristine particle), effective changes in particles size and density are calculated for each species and accounted for in the calculation of fate processes. For hetero-aggregated and biofouled plastic particles, the effective size and density are calculated based on the equations shown in Table S1, to account for the impact of added volume and different density from SPM and biofilm. For the biofouled particle the calculations are based on a model study by Kooi et al. (2017), where the biofilm layer was assumed to have a thickness of 5 μm and a density of 1388 kg/m³. For the heteroaggregated species the SPM were assumed to have a spherical shape, a diameter of 0.5 μm and a density of 2000 kg/m³ (Praetorius et al., 2012).

### 2.1.3. Fate processes

For each size class of plastic particles in water compartments, the Full Multi model simulates transformation of the particles between the four different microplastic species (i.e. heteroaggregation, aggregate break-up, biofouling and defouling), fragmentation into the next smallest size class, chemical degradation that removes plastic from the system, transport between compartments within a unit cell by settling, rising, mixing, and transport between unit cells by advection (Fig. 1). In sediment compartments the modeled fate processes are fragmentation, degradation, burial into deep, inaccessible sediments, resuspension to the adjacent water compartment within the unit cell, and transport along the river between unit cells via bedload transport (Fig. 1).

The formulation of each of the processes included in the default parameterization of the Full Multi is based on a review of the literature. Some equations were taken from transport and fate frameworks for (inorganic) nanoparticles (Praetorius et al., 2012) based on traditional particle model concepts (e.g. suspended solids and colloids), while other process descriptions such as fragmentation and biofouling have been specified by assuming first order kinetics. A more detailed description is provided below and the equations, with corresponding references and levels of confidence are presented in Table S2. It is worth mentioning that, nano- and microplastics in the environment representing a relatively new area of science, there is still uncertainty about what the main governing processes are. In this sense the process descriptions included in the model framework should be seen as provisional and are intended to be updated and updated by the user community to incorporate new information and understanding as it becomes available.

The fate processes in the Full Multi framework can be broadly divided into three categories: interaction with natural entities, transformation and transport between different compartments.

### 2.1.3.1. Interaction processes (heteroaggregation, break-up and biofouling)

Heteroaggregation, the aggregation of plastic particles with natural SPM, is modelled following classical colloid theory, as discussed in Praetorius et al., 2012, as a pseudo-first order process (Table S2). The rate constant for this process depends on the collision frequency between the two types of particles, the concentration of SPM present in the system and the attachment efficiency for heteroaggregation (q_{agg-het}), describing the probability of heteroaggregate formation upon collision (Praetorius et al., 2020). q_{agg-het} describes the degree of favourable

### Table 1

<table>
<thead>
<tr>
<th>List of parameters required to define each model unit cell and its compartments.</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Unit Cell/Compartments parameters</strong></td>
</tr>
<tr>
<td>Compartments dimensions:</td>
</tr>
<tr>
<td>- Depth (h, in m)</td>
</tr>
<tr>
<td>- Length (l, in m)</td>
</tr>
<tr>
<td>- Width (w, in m)</td>
</tr>
<tr>
<td>- Volume (V, in m³)</td>
</tr>
<tr>
<td>Compartments properties:</td>
</tr>
<tr>
<td>- Shear rate (G, in s⁻¹)</td>
</tr>
<tr>
<td>- Temperature (T, in K)</td>
</tr>
<tr>
<td>- Flow velocity (v_{flow}, in m/s)</td>
</tr>
<tr>
<td>- Concentration of SPM (C_{SPM}, in mg/L)</td>
</tr>
<tr>
<td>Cell connectivity:</td>
</tr>
<tr>
<td>- Discharge (q, in m³/h)</td>
</tr>
<tr>
<td>- Region_I (cell of discharging inflow)</td>
</tr>
<tr>
<td>- Region_J (cell of receiving inflow)</td>
</tr>
</tbody>
</table>

### Table 2

<table>
<thead>
<tr>
<th>List of plastic particles input parameters.</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Plastic particles parameters</strong></td>
</tr>
<tr>
<td>- Density (d, kg/m³)</td>
</tr>
<tr>
<td>- Shape (i.e. sphere, fibre)</td>
</tr>
<tr>
<td>- Diameter (μm)</td>
</tr>
<tr>
<td>- Length a, b and c (for non-spherical particles) (l_a, l_b, l_c, in μm)</td>
</tr>
</tbody>
</table>
interaction between plastic particles and SPM and is influenced by the respective surface characteristics of the colliding particles as well as the water chemistry. In the default parameterization of the Full Multi model, the concentration of SPM is considered constant along the whole system and water chemistry. In the default parameterization of the Full Multi model, due to the lack of experimental data regarding plastic degradation times, long half-lives for degradation rates may be negligible compared to other processes in many systems. However, biodegradable polymers might have shorter degradation rates in deeper waters due to reduced light limiting the growth of the biofilm organisms (Kooi et al., 2017). Hence, biofouling is fastest in the surface water compartment. The values of $t_{\text{biof-growth}}$ assumed in the default parameterization are presented in Table S3 and are based on experimental findings that indicate that biofilm formation takes place within days or weeks (Rummel et al., 2017). Future applications of the model could also consider plastic additives present in the modelled particles as influencing the rate of biofouling (Nelson et al. 2021) as well as different growth rates as a function of temperature, sunlight and water composition.

Defouling, the disintegration of the biofilm layer, can occur due to light limitation, grazing, or dissolution of carbonates in acid waters (Kooi et al., 2017). In the Full Multi framework defouling is represented as a first-order kinetic process characterized by a biofilm degradation time ($t_{\text{biof-degrade}}$). Despite being a potentially relevant fate process, the rate constant for defouling is assumed to be zero in all compartments in the current Full Multi version. Due to high dilution of plastic particles in the environment compared to naturally occurring SPM, homoaggregation is assumed negligible and does not justify the additional computational effort of including a second-order process in the model framework.

### Table 3

<table>
<thead>
<tr>
<th>Input file name</th>
<th>Information type</th>
<th>Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td>MiroplasticsSizeClass.txt</td>
<td>Plastic particles</td>
<td>Characteristics</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Density (d, kg/m$^3$)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Shape (i.e. sphere, fibre)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Diameter (μm)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Length $a$, $b$ and $c$ (for non-spherical particles) ($l_a$, $l_b$ and $l_c$ in μm)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Depth, length and width ($h$, $l$ and $w$ in m)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Volume ($V$, in m$^3$)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Shear rate ($G$, in s$^{-1}$)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Temperature ($T$, in K)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Flow velocity ($v_{\text{flow}}$, in m/s)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Concentration of SPM ($C_{\text{SPM}}$, in mg/L)</td>
</tr>
<tr>
<td>compartmentsGenericRiverSec_prop.txt</td>
<td>Cell dimensions and</td>
<td>properties</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Volume ($V$, in m$^3$)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Shear rate ($G$, in s$^{-1}$)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Temperature ($T$, in K)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Flow velocity ($v_{\text{flow}}$, in m/s)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Concentration of SPM ($C_{\text{SPM}}$, in mg/L)</td>
</tr>
<tr>
<td>process_paramRiver.txt</td>
<td>Fate process parameters</td>
<td>- Attachment efficiency ($\theta_{\text{agg}}$)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Degradation half-life times ($t_{\text{degrade}}$, in days)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Timescale for fragmentation of the 1000 μm size fraction ($\theta_{\text{SPM}}$, in days)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Time of biofilm growth ($t_{\text{biof-growth}}$, in days)</td>
</tr>
<tr>
<td>flow_connectivity.csv</td>
<td>Connectivity between model</td>
<td>- Discharge ($q$, in m$^3$/h)</td>
</tr>
<tr>
<td></td>
<td>cells regarding flow advection</td>
<td>- Region $I$ (cell of discharging inflow)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>- Region $J$ (cell of receiving inflow)</td>
</tr>
</tbody>
</table>

### Fig. 2

Schematic representation of the horizontal profile of the generic river model parameterization of the Full Multi framework in the longitudinal dimension showing all compartments and its dimensioning.
were assumed and specified as being equal for all of the defined compartments. Also, degradation half-lives for pristine plastic particles were assumed smaller than for heteroaggregated and/or biofouled particles, assuming a protective effect of the SPM or biofilm outer layer. Finally, less protective effect of biofilm compared to SPM was assumed (Table S3). Currently, degradation is represented as complete mineralization, but it would be possible to consider the formation of degradation intermediates and their fate in future model implementations.

Fragmentation of plastic particles in the Full Multi is modeled as a size-dependent process based on an estimated rate constant \( k_{\text{frag,gen}} \) for fragmentation of pristine particles in the largest (1000 μm) size class in flowing water. In the default parameterization, we assume fragmentation of pristine 1000 μm particles occurs on a 1-year timescale and set \( k_{\text{frag,gen}} = 1/(1 \text{ year}) = 3.2 \times 10^{-8} \text{ s}^{-1} \). Fragmentation rate constants for all size classes of pristine particles are then calculated by scaling \( k_{\text{frag,gen}} \) assuming fragmentation occurs more slowly for smaller particles in proportion to the ratio of diameters. “Explosive” fragmentation is assumed such that fragmentation produces a number of smaller particles calculated by dividing the volume of particles in the parent size class by the volume of particles in the daughter size class (Table S2). In the default formulation of the Full Multi, fragmentation is assumed to occur unidirectionally from larger to consecutively smaller size fractions only. Thus, for example, particles in the 1000 μm size class fragment into the 100 μm size class only. Fragmentation of plastic particles in the environment is driven by diverse factors such as UV radiation, physical stress induced by water turbulence and wind, and biodegradation (Min et al., 2020). In the default formulation of the Full Multi, fragmentation rates in surface water where UV radiation and turbulence are strongest, are assumed to be ten times higher than in the other water compartments (Table S3). Fragmentation of heteroaggregated particles is assumed negligible in the default model formulation and biofouled particles are assumed to fragment at half the rate of the corresponding pristine plastic particle due to shielding of the particle’s surface from UV radiation and physical stress.

2.1.3.3. Transport processes (advection, bed-load transport, burial, resuspension, settling, rising, mixing). Advective transport of the plastic particles in the moving water compartments, horizontal transport in the sediment bed, vertical mixing of bulk water, burial into deep sediment and resuspension into stagnant water are modelled following the same assumptions and process formulations as in the model developed by Praetorius et al., 2012 describing the fate and transport of titanium dioxide nanoparticles in the Rhine River (Table S2). First order rate constants for settling (or rising) of plastic particles are estimated based on the depth of the compartment of study and the value of settling (or rising) velocity of the plastic particle calculated using Stokes’ law for theoretical settling velocities of a perfect sphere in a fluid with laminar flow (Praetorius et al., 2012) (Table S2). Settling and rising are mutually exclusive, and whether a particle settles or rises is determined by the density difference between the plastic particle and water.
Limitations to this process description have been identified in the literature and involve overestimation of settling velocities of large, low density particles, and underestimation of the influence of particle shape (Waldschläger and Schüttrumpf 2019; Kowalski et al., 2016). Other settling formulations can be implemented in the Full Multi framework by the user if desired.

2.1.4. Mass balance model equations

The mass balance equations for each particle species in each size class form a system of coupled first-order ordinary differential equations (ODEs) (eq. (1)) that are functions of species- and size-specific particle number of plastic particles in each compartment of the model domain and of time. By default, concentrations (eq. (2)) are expressed as number of plastic particles per volume in the water compartments and per dry weight mass in the sediment compartment by converting the volume of sediment into mass using a typical dry sediment bulk density 1.3 g/cm$^3$ (Sekellick et al., 2013). The model allows interconversion between particle number and mass concentration by using the final density and parent size of the particle being evaluated (eq. (3)).

\[
\frac{dN_{\text{Comp-Comp}}^{F,i}}{dt} = q_{\text{Comp-Comp}}^{F,i} \cdot N_{\text{Comp-Comp}}^{F,i}(t) + k_{\text{trans}}^{F,i} \cdot N_{\text{Comp-Comp}}^{F,i}(t) + \sum_{y=1}^{\text{Model Cells}} k_{\text{transport}}^{F,i} \cdot N_{y}^{F,i}(t)
\]

\[
C_{\text{Comp-Comp}}^{F,i} = \frac{N_{\text{Comp-Comp}}^{F,i}}{V_{\text{Comp-Comp}}}
\]

\[
C_{\text{Comp-Comp}}^{F,i} (\text{mg} \cdot \text{m}^{-3}) = C_{\text{Comp-Comp}}^{F,i} (\text{no} \cdot \text{m}^{-3}) \times V_{F,i} \times d_{F,i}
\]

for $i = 1, \ldots, n_{\text{size}}$ and $n = 1, \ldots, n_{\text{Model Cells}}$

where $F$ represents the microplastic speciation form (i.e. pristine, heteroaggregated, biofouled or biofouled & heteroaggregated), $i$ the plastic particle size class, Comp the cell compartment (i.e. surface water, flowing water, stagnant water and sediment) and Cell the specific model unit cell. $q_{\text{Comp-Comp}}^{F,i}$ refers to the input flow of plastic particles of form $F$ and size class $i$, into the compartment and cell specified in particles per minute and $V_{\text{Comp-Comp}}$ is the volume of the compartment within the modelled cell. $N_{\text{Comp-Comp}}^{F,i}(t)$ is the number of particles of form $F$ and size class $i$ present in the compartment and cell specified and $k_{\text{trans}}^{F,i} \cdot N_{\text{Comp-Comp}}^{F,i}$ and

\[
\text{for } i = 1, \ldots, n_{\text{size}} \text{ and } n = 1, \ldots, n_{\text{Model Cells}}
\]
**k^i_{\text{input}}** represent the rate constants for irreversible loss processes (which include degradation, fragmentation from the smallest size class, burial and advection out of the system), the transformation rate constant from any other MP form \(j\) (i.e. heteroaggregation, break-up, biofouling, defouling, and fragmentation) and size class \(i\) and the transport rate constant of the same plastic particle form and size class from the different compartments and/or unit cell, in \(s^{-1}\) respectively. \(V^F_i\) represents the volume of the particle form \(F\) of size \(i\) and \(d^F_i\) its density.

The set of ODEs arranged in matrix form (eq. (4),) is solved in Python with the Scipy.integrate package using the function ODEINT.

\[
\frac{dN}{dt} = N(t) \times M_{\text{FullMulti}} + Q_{\text{input}}
\]

where the vector \(N(t)\) contains the particle numbers of the different plastic particle forms in different size classes for all compartments in the model. The matrix \(M_{\text{FullMulti}}\) contains the rate constants of the mass balance equation eq. (1) and \(Q_{\text{input}}\) is the emission vector.

### 2.2. Model parameterization

The Full Multi framework requires parameters to define the characteristics of the plastic particles, the fate process descriptors, the properties of the modelled system and the connectivity between model unit cells. These input parameters are specified in four input files, whose content is summarised in Table 3 and its modification procedure described in the user’s guide provided in the open access GitHub repository.

Emission scenarios also must be specified to define the \(Q_{\text{input}}\) vector and drive the model calculations. Emissions generally describe the time-variable input flow of plastic particles per minute, but they may also be specified as a pulse or constant emission flow rate. The location or locations where the emissions enter the model domain (i.e. the specific compartments and unit cells receiving emissions) must also be specified. The species and size class of plastic emitted into the system and the simulated time duration and target timestep for the ODE solver are also required inputs.

For the parameterization of the different microplastic fate and transport processes included in the model a series of assumptions about how, when and where the different processes take place in the modelled system are required, and are documented in section 2.1.1 and Table S3. Different degrees of confidence in the formulation of the processes (Table S2) or in the selection of the input parameters (Table S3) are highlighted, where those with low confidence levels correspond to processes where parameters are missing or highly uncertain and/or where process descriptions are based on assumptions that require critical evaluation by further research.

While particle shape is been included as a model input, the fate processes included in the current model version are only parameterised for spherically shaped particles. However, fibre process formulations are previewed for implementation.
3. Illustrative application for a generic river

We illustrate the Full Multi framework by application to describe the fate and transport of plastic particles with a range of densities in a generic river system.

3.1. Framework configuration and parameterization

Our generic river system configuration of the Full Multi framework links model unit cells illustrated in Fig. 1 with uni-directional water flow in the surface water and flowing water compartments, and with bedload transport of sediments (Fig. 2). The modeled generic river is 1000 km long, and is subdivided into 20 unit cells with identical dimensions (50 km length, 80 m width and 5.42 m total depth) and with a water flow velocity of 1.3 m/s (discharge = $562 \text{ m}^3/\text{s}$) in the surface water and flowing water compartments (Fig. 2). The unit cell structure, dimensions and flow velocity were inspired by the model for the Rhine river by Praetorius et al. (2012).

The process descriptions for plastic fate in the generic river system are parameterised according to the assumptions described in section 2.1.1 and are summarised in Table S3. In many cases these parameterisations and process descriptions are provisional and, according to our flexible and open-source modelling approach, they can be modified in future applications of the model as needed for scenario analysis or as new research emerges.

3.2. Scenario analysis for plastic with a range of densities

As an illustrative application of the model framework, we modeled a low density, buoyant plastic, assumed to be polyethylene ($d = 980 \text{ kg/m}^3$), a near-neutrally buoyant plastic, assumed to be polyamide ($d = 999 \text{ kg/m}^3$), and a high-density plastic, assumed to be polyvinyl chloride ($d = 1580 \text{ kg/m}^3$). For each of these plastic types we simulated a constant emission of 100 pristine plastic particles per minute in the 1000 μm size class into the flowing water compartment of the first (i.e., the furthest up-stream) unit cell of the generic river. The model was run to simulate 365 days with a target timestep of 1 h. After the model run information about the speciation of plastic particles, their dynamic exchange between the three water layers and sediment, and fragmentation into the five size classes (i.e. 0.1, 1, 10, 100 and 1000 μm) is produced as output in the form of concentration values of each plastic species in each size class in each compartment of the river over time.

3.3. Results and discussion

3.3.1. Modeled concentrations after 1-year simulation time

Species and size class specific concentrations of plastic particles per compartment along the generic river system after one year of simulation for PE, PA and PVC are shown in Figs. 3-5, respectively. Results are presented as number concentrations of plastic particles (No) per m$^3$ in the water compartments, and per gram of sediment for the sediment compartment. Conversion of the results into mass concentration is possible in the model through estimation of the particle volume and density by taking its shape into consideration.

The distribution, in number concentration, of plastic particles between the different compartments along the whole river system differ strongly as a function of particle density and size (Figs. 3-5). The concentration of low-density pristine PE particles (i.e. 1000 μm free plastic particles) in the flowing water compartment decreases steeply in the first 50 km of the river (i.e., in the first unit cell) due to transfer to the surface water compartment as a consequence of its buoyancy (Fig. 3). As simulated in the model, these particles also quickly aggregate with SPM to form heteroaggregates, which become the...
dominant particle species along the whole river system (Fig. 3 and Table S4). The buoyant low-density PE particles that remain in pristine form or are heteroaggregated are mainly found in the surface water compartment and are mainly found in sizes of 100 and 10 μm. However, a fraction of their heteroaggregated and/or biofouled forms settle down to the sediment and are most abundant in the smallest size bins for the heteroaggregates. This size effect, where the most abundant heteroaggregated particles in the sediment are the smaller size fractions, can be explained by the fact that SPM contributes in higher extent to the final density of the particles when attached to smaller particles (Table S1). As depicted in Fig. 3 and as parameterised in the Full Multi, low-density PE particles remain in the generic river system from upstream to downstream.

The near-neutrally buoyant pristine particles of PA settle into the sediment compartment much more efficiently compared to PE regardless of aggregation state with the biggest size fractions disappearing from the systems through burial at 400–600 km downstream (Fig. 4). However, some PA particles (i.e. those with 1 and 10 μm diameter) remain in the water compartments even far downstream in the river.

Finally, all species of the high-density PVC particles are subject to sedimentation and burial into the deep sediment of the river, with only very low number concentrations of the smallest size fractions being distributed downriver in the sediment compartment (Fig. 5).

3.3.2. Comparing competing processes as half-life times

Since all processes in the model are described using first-order kinetics, they can be compared directly to each other. To facilitate comparison, we calculated half-life times for each process in each compartment of the model as ln2/\( k_{\text{process}} \), where \( k_{\text{process}} \) is the process-specific rate constant in s\(^{-1}\). The half-life time represents the time required for a particle’s concentration to decrease to half its initial value due to the process in question only. Fate processes with the shortest half-life times are the fastest processes, and will be the dominant process pathways that plastic particles will follow in the modelled compartment.

Figs. 6–8 show process half-life times for each plastic particle species and size class for the three different plastic types in form of heatmaps. Green values correspond to short half-life times (fastest, dominant processes) of a few minutes to tens of hours (from lighter to darker green), while the blue colour represents longer half-life times (slower fate processes) ranging from days to months to years (from lighter to darker blue). Grey shading indicates that the process is not taking place for this particular compartment/plastic form/size fraction.
Fig. 8. Heatmap of the transport and fate processes half-lifetimes ($t_{\text{half}}$) of PVC microplastic particles in the simulated generic river system. The $t_{\text{half}}$ of each process for each plastic species (i.e. pristine, heteroaggregated, biofouled or biofouled & heteroaggregated) and size fraction (i.e. 1000, 100, 10, 1 and 0.1 μm) are indicated by a colour ramp from light green representing $t_{\text{half}}$ in minutes to dark blue $t_{\text{half}}$ of years. Grey shading indicates that the process is not taking place for this particular compartment/plastic form/size fraction.

Fig. 9. Relative plastic particle abundance for different particle sizes estimated by Kooi and Koelmans (2019) from sea surface and freshwater surface samples ((a) Enders et al., 2015, (b)Enri-Cassola et al., 2017 (c) Eo et al., 2019), and from our model simulations in the generic river system for PE plastic particles.
and rising half-life times in Fig. 6, are also observed as a function of size and also aggregation state. Settling and rising are formulated as mutually exclusive processes in the model and their half-life times in water compartments are a function of particle size and overall density, which is determined by the density of the plastic particle and the density of SPM for heteroaggregated particles and/or of biofilm for biofouled particles (Table S1). The density of water is specified as 998 kg/m³ in the default parameterization. Thus, as observed in Fig. 6, pristine PE particles have lower density than water and rise rather than settle in the flowing and stagnant water compartments, while heteroaggregated and biofouled PE particles will settle or rise depending on the size of the parent plastic particle. For example, small particles of low-density PE that have been colonized by biofilm or heteroaggregated with particular organic matter have higher bulk density than water (as described in Table S1) which leads to settling rather than rising (Fig. 6). Settling half-life times for the low-density PE particles range from 30 min to a few hours for the smallest heteroaggregated particles and for the biofouled particles. PE half-life times for rising, for those particles with lower density than water, vary from 30 min to several months depending on the size and particle form, and are fastest for the largest 1000 μm size fraction.

Due to the higher density of PA and PVC, no rising to the surface water compartment occurs for any of the particle forms and sizes (Figs. 7 and 8), and the half-life times of settling are again controlled by the particle size. Settling half-life times for PVC and PA particles range from 30 min to a few hours for the largest particles (10–1000 μm), but increase to several months for the smallest PA particles and the smallest, non-biofouled PVC particles, due to higher drag forces.

The half-life times for advection out of each unit cell in surface water and flowing water (7.4 h in our default model configuration) provide a useful benchmark for comparison with other processes in Figs. 6–8. Processes in water with half-life times shorter than advection occur on timescales shorter than the residence time of water in the unit cell, and thus represent important in-cell pathways. In cases where reversible processes are much faster than the particle residence time in the system, they could be parameterised using constant steady-state concentration ratios instead of using a kinetic approach. In this sense, in our default model parameterization for the three plastic types, for example, heteroaggregation and heteroaggregate break-up could be reformulated using steady-state concentration ratios. Mixing between compartments and degradation of the particles are slow processes compared to advection, and thus are not expected to dominate in-cell fate of plastic. In sediments, burial to deep sediment, and resuspension occur on the same timescale, and are faster than down-river transport of plastic with sediments.

### 3.3.3. Model evaluation against field data

The generic river parameterization and constant emissions assumed in our illustrative example does not allow for evaluation of the model against absolute concentrations of plastic particles measured in a real river. However, a degree of evaluation of the model against field data is possible by comparing the modeled size distribution of plastic particles to field measurements. Kooi and Koelmans (2019) collected data from 11 studies that reported plastic particle concentrations in 10 or more size classes and fitted the relative abundance of particles as a function of size with power law regressions. Our modeled relative abundance of PE plastic particles in the three water compartments in the last (i.e. furthest downstream) unit cell of the generic river after 1 simulated year falls within the range of measurements of floating plastic (Fig. 9), which provides a partial validation of the assumptions about fragmentation made in the model.

In terms of distribution of the plastic particles within the different compartments of the generic river system, Table 4 summarises the model estimated particles distribution, integrated over all size ranges and species, as percentage of particles per compartment relative to the total number of particles in the system.

These results are in line with results obtained from field studies where it was found that the higher density polymers prevailed in samples taken from river and marine sediments, while a larger percentage of buoyant polymers were found in surface water samples of fresh waters, effluents and to a lower extent, marine surface waters (Kooi et al., 2021).

### 4. Conclusions

We present a modelling framework to describe the fate of plastic particles in aquatic systems that describes multiple dimensions of particle characteristics (size and speciation) and that can be applied to diverse aquatic systems. We envision this framework as a tool to support hypothesis generation for process-level studies and data analysis, design of field measurement campaigns, scenario analysis and developing bounding exposure estimates for risk assessment.

The calculations for PE, PA and PVC in the generic river system presented in this paper can be replicated by prospective users of the framework by following tutorials in Jupyter notebooks on the models GitHub repository (https://github.com/Nano2PlastProject/TheFullMulti_RIVER). The full open-access model code is also available there and open for continued development.

Our evaluation of the modelled relative abundance of floating plastic particles in our generic scenario against field measurements provides a degree of confirmation that the fragmentation process descriptions in the model are reasonable. However additional performance evaluation and validation should be carried out before the model is used in risk assessment or decision support applications. An interesting feature of the model results illustrated in Fig. 9 is the model’s prediction of low relative abundance of 0.1 μm particles. In their review and meta-analysis of measurement data, Kooi and Koelmans (2019) reported that some datasets showed increasing particle concentrations with increasing size below 20 μm. However, they did not have confidence in this finding due to the methodological challenges of quantifying small plastic particles, and they suggested a practical detection limit of 20 μm. Our generic river modelling with the current process descriptions and parameterization predicts a maximum in relative abundance of floating plastic particles at around 1 μm diameter. That prediction is an example of how the model framework can be used for hypothesis generation to inspire further plastic pollution research.

### 5. Software availability

Name of the software.

The Full Multi Developers.

Antonia Praetorius and Prado Domercq Programming language.

Python 3.7.6
Availability.

The model presented in this paper, the Full Multi, is freely available at https://github.com/Nano2PlastProject/TheFullMulti_RIVER. The documentation of the model together with a guided example in the form of editable Jupyter Notebooks for each of the three scenarios presented (i.e. PE, PA and PVC) are provided in the Github repository including a guide for users in the README.

Declaration of competing interest

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

5. Acknowledgments

Development of The Full Multi was funded by the European Chemical Industry Council (CEFIC) through the Long-Range Research Initiative (LRI) project ECOC48 - NANO2PLAST: Extending nanoparticle models to open-source models of the fate and transport of microplastic in aquatic systems.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envsoft.2021.105291.

References

Sebille, Erik van, Wilcox, Charlotte, Arp, Hans Peter H., Lawrence, Martin, Minko, Nikolai, Hardtste, Britta Denise, van Franeker, Jan A., Erikson, Marcus, Siegel, David, Galganzi, Francois, Law, Kara Lavender, 2015. A global inventory of small floating plastic debris.