Environmental Science Processes & Impacts

Accepted Manuscript

This article can be cited before page numbers have been issued, to do this please use: L. Nizzetto, G. Bussi, M. N. Futter, D. butterfield and P. Whitehead, *Environ. Sci.: Processes Impacts*, 2016, DOI: 10.1039/C6EM00206D.

rsc.li/process-impacts

View Article Online
DOI: 10.1039/C6EM00206D

Environmental impact statement

This study deals with the theoretical analysis of microplastics transport in catchments and analyzes the potential for soils and bed sediments to retain and accumulate these materials. While there is a large amount of concern over the presence of microplastics in the ocean, and considerable effort is in place to investigate the transport and impacts of these materials in the context of marine pollution, so far there has been very little focus on the analysis of their transport over land, and in streams. Terrestrial emissions of microplastics are the largest contributors to the total environmental burden. Here we present the first mathematical model describing the dynamics of microplastic fate in terrestrial environments. The model utilizes meteorological drivers and information on soil and sediment hydrological properties to simulate microplastic behavior as a function of their dimensions and density.

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DOI: 10.1039/C6EM00206D

Oslo, 12/05/2016

A theoretical assessment of microplastic transport in river catchments and their retention by soils and river sediments.

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DOI: 10.1039/C6EM00206D

Abstract

The presence of microplastics (MPs) in the environment is a problem of growing concern. While research has focused on MP occurrence and impacts in the marine environment, very little is known about their release on land, storage in soils and sediments and transport by run-off and rivers. This study describes a first theoretical assessment of these processes. A mathematical model of catchment hydrology, soil erosion and sediment budgets was upgraded to enable description of MP fate. The Thames River in the UK was used as a case study. A general lack of data on MP emissions to soils and rivers and the mass of MPs in agricultural soils, limits the present work to serve as a purely theoretical, nevertheless rigorous, assessment that can be used to guide future monitoring and impact evaluations. The fundamental assumption on which modelling is based is that the same physical controls on soil erosion and natural sediment transport (for which model calibration and validation are possible), also control MP transport and storage. Depending on sub-catchment soil characteristics and precipitation patterns, approximately 16% to 38% of the heavier-than-water MPs hypothetically added to soils (e.g. through routine applications of sewage sludge) are predicted to be stored locally. In the stream, MPs < 0.2 mm are generally not retained, regardless of their density. Larger MPs with densities marginally higher than water can instead be retained in the sediment. It is, however, anticipated that high flow periods can remobilize this pool. Sediments of river sections experiencing low stream power are likely hotspots for deposition of MPs. Exposure and impact assessments should prioritize these environments.

1. Introduction

Plastic litter and microplastics (MP) are global environmental problems. Production of plastic is steadily increasing and currently exceeds 300 million metric tons per year.[1] MPs are generally defined as plastic debris smaller than a few millimeters down to the micrometer range. Primary MPs include micro pellets (such as abrasive microbeads in face scrubber cosmetics or abrasion beads for industrial uses) and plastic pellets used as raw material for injection molding as well as other plastic products with an original size smaller than a few millimeters. Secondary MPs are formed in the environment when plastic litter is fragmented to smaller pieces by weathering or any form of mechanical or chemical stress.

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During usage and wasting a fraction between 1.5 and 4.5% of the global plastic production is released directly to the sea.[2] Marine waters and sediments are important environmental reservoirs of plastic debris and MPs. Impacts on biota and marine environmental quality are well documented[3] with a valued damage for the global economy estimated to be in the range of $ 13 billion per year.[4] Research on MPs has been mainly focused on occurrence and impacts in the marine environment. Despite in-situ formation being generally recognized as an important source of MPs for the oceans,[5] their formation and emission on land has not received sufficient attention, so far.[6, 7]

The lack of scientific attention paid to MPs in land and freshwaters is concerning. For example, between 21% and 42% of the global plastic waste production is stored in landfills on land, often under poorly managed conditions.[2, 7] Here, formation of secondary MPs may occur with higher efficiency compared to the marine environment due to more intense weathering, higher exposure to UV radiation, improper handling and recycling practices. Similarly, considerable sources of secondary MPs are associated with product usage in households, industry and transport. Laundry dust, paint flakes and car tires debris have been indicated as a major source of secondary MPs from land by recent reports.[8,9] MPs from these sources are generally conveyed through canalizations, piping and plumbing (such as sewers) to effluent points along rivers and coastal areas. Considering this, and a number of studies of plastics in estuaries, it can reasonably be hypothesized that terrestrial sources are a relevant, possibly dominant, component of total emission of MPs to the marine environment.[10-13]

The accuracy of MPs emission estimates is currently hindered by lack of data. Specifically, information on MP transport efficiency in runoff and streams is missing. This is despite the large number of qualitative studies of MPs in rivers and sediments.[12,14-16] Similarly, limited assessments of MPs flows from sewage and canalizations, their retention by waste water treatment plants (WWTP) and release by effluents are available.[8, 9, 17] When contaminated effluents are processed by WWTPs with primary and secondary treatment stages, most of the less mobile MPs are apparently retained in the sludge.[18] Sewage sludge is commonly used as fertilizer in agricultural soils.[19, 20] This practice can result in transferring significant loads of MPs to agricultural landscapes with unknown implications for soil quality loss, soil organisms, livestock and human health.[21] From soils, MPs can be further remobilized by runoff and wind erosion. The retention efficiency of soils and river sediment beds has never been investigated in detail.[14, 15, 21, 22] Storage in soils and river sediments may represent important temporary or permanent sinks which delay or prevent MPs from reaching the marine environment. At the same time, their accumulation in these compartments is of concern for the maintenance of good ecological quality.

This paper describes a first conceptual assessment of MP transport in catchments and rivers and focuses on assessing the retention efficiency of soils and river sediments during MP transport toward the sea. The recently developed INCA-Contaminants model[23] and the imbedded sediment transport module based on an upgraded version of the mathematical frame proposed by Lazar et

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al.[24] was used as a quantitative tool to simulate in detail the mechanisms of MP particle storage, entrainment and deposition in soils and streams as a function of hydrologic (precipitation patterns, surface runoff and stream power) and pedological controls.

Due to a lack of empirical data on MP emissions and concentrations in soils and the stream system, the present study is conceived to provide a purely theoretical, nevertheless rigorous, assessment of the MP transport across the pedosphere and hydrosphere. Simulations are performed assuming rough estimates of MP emissions (based on indirect assessments published in recent reports)[8, 9, 25] considering a realistic model scenario (the Thames river catchment in UK).

2. Model description

INCA-Contaminants[23] is a semi-distributed integrated hydrobiogeochemical-sediment transport-multimedia contaminant fate model. The model can be configured to represent realistic catchments by introducing information on river network structure, subcatchments boundaries, slopes and length of river reaches, land use, and soil properties (including organic matter content, and sediment storage). The underlying rainfall-runoff model[26] computes hydrologically-effective rainfall and soil moisture deficit time series that are used to generate daily run-off from the land and river flow predictions. Run-off includes a diffuse component (the outflow of water from individual soil layers) and a surficial component (direct runoff generated from infiltration excess or saturation excess). Hydrological module predictions are calibrated using catchment-specific observations of river flow. The sediment transport module[24] utilizes direct runoff fluxes and predicted stream flow regimes to calculate entrainment and depositions of particles from/to river bed sediments. This module can be used to simulate the transport of natural particles (soil particles and suspended sediments) as well as the transport of artificial particles (such as MPs). In addition, direct effluent inputs can be arbitrarily postulated in multiple locations throughout a river network to simulate emission of particles directly to the stream or sediment bed. This for example can be used to set direct inputs of MPs to the river (e.g. from WWTP effluents).

In order to simulate physical transport of MPs in the catchment and stream, a well-tested mathematical frame used to simulate entrainment of solids from soil and suspended sediment transport dynamics in streams.[24] was used. This frame was modified to accommodate the option of defining and simulating mass budgets of an arbitrary number of particle size classes, as well as allowing MPs to be added to the land surface (for example to simulate application of sewage sludge containing MPs). Finally, a very simple modification was made to allow a MPs store to be fully depleted. Particle size classes in the frame are defined by size ranges (e.g. the range between their minimum (Dlow, m) and maximum (Dup, m) diameter) and user-definable density values ($\rho_s, \mathrm{kg\,m^{-3}}$) (i.e. the density of individual particles). The mass balance of MPs of different size classes is calculated separately.

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MP mobilization from contaminated soils is controlled by precipitation, soil hydraulic characteristics, average slope of the land and the mass of MP in the system. Mobilization is a function of i) the MP pool that is readily available for mobilization (MP_{store}, kg km^{-2}); ii) the overland flow transport capacity for both MP and soil particles (S_{TC}, kg m^{-2} s^{-1}); iii) the detachment/erosion of MPs from soil through splash detachment during rain events (S_{SP}, kg m^{-2} s^{-1}); and iv) flow erosion (S_{FL}, kg m^{-2} s^{-1}). Generic descriptions of these parameters and processes are provided by Lazar et al.[24] In the present version, MP_{store} is user definable and can be specified as an initial condition. Continuous or time-varying inputs of MP (m_{in}, kg m^{-2} s^{-1}) to the land phase can be simulated. S_{SP} depends on rain intensity, duration of rainfall and spatial variation in soil type and canopy cover that can moderate soil erosion. The mathematical formulation adopted to describe S_{SP} for MPs is consistent with that used for soil erosion.[24] S_{TC} represents an estimate of the maximum amount of bulk material that can be transported from the land to the stream by overland runoff including both soil particles and MPs and is calculated using an empirical function dependent on average soil slope in the sub-catchment, water overland run-off and a set of scaling parameters that can be tuned for calibration purposes (as described in Lazar et al.).[24] S_{FL} represents the net detachment of MPs and soil sediments due to flow erosion and depends on S_{SP}, S_{TC} and soil erosion potential.[24] In the version presented here, S_{SP}, S_{FL}, S_{TC} are treated as independent from particle density and sizes.

When simulating mobilization of MPs from individual land use types INCA-Contaminants considers two alternative situations:

i) The MP are added to the pool of movable material by splash detachment but the transport through flow erosion is larger than the total carrying capacity (i.e. $S_{FL} > S_{TC}$), then the mass of sediment of MPs transported to the river over the soil is smaller than S_{SP} and is actually controlled by the total carrying capacity as follows:

$$\frac{dMP_{store}}{dt} = m_{in} + S_{SP} - S_{TC} \qquad 1)$$

In this case the mass of MPs of a given size class *i* (m_{OUT}, kg m^{-2} s^{-1}) delivered to the stream is simply:

$$m_{OUT} = S_{TC} \qquad 2)$$

Although the amount of MP mobilized from the soil storage is unlikely to exceed S_{TC}, it is important to recall that both the mass of MP and natural sediments contribute to filling up the total carrying capacity of the overland flow. The mass of MP transported to the stream is therefore sensitive to the total amount of soil eroded.

ii) Otherwise, with $S_{SP} + S_{FL} < S_{TC}$:

$$\frac{dMP_{store}}{dt} = m_{in} - S_{FL} \qquad 3)$$

and

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$$\mathrm{m_{OUT}} = (\mathrm{S_{SP}} + \mathrm{S_{FL}}) \qquad 4)$$

The original terrestrial sediment component of INCA-Contaminants assumes unlimited availability of sediments for delivery. Concerning MPs, however, it is obviously incorrect to assume an unlimited source. $\mathrm{MP_{store}}$ can eventually be depleted when erosion exceeds inputs over a sufficient amount of time. In order to prevent the soil to deliver an unlimited amount of MPs over time the following simple conditions was included:

$$\mathrm{m_{out}} = \min(\mathrm{m_{out}}, \mathrm{MP_{store}})$$

The in-stream transport model assumes uniformly mixed river segments (reaches) with the following controlling processes: density dependent settling of suspended MPs, size dependent entrainment of MPs from the bed sediment, and, obviously, advection from/to adjacent reaches. Deposition is controlled by a settling velocity u_T (m s^{-1}) that, in turn is a function of the density and dimensions of the MPs:

$$\mathrm{u_T} = \frac{(\rho_s - \rho_f)}{18\mu}\mathrm{g}\mathrm{D}_{\mathrm{med}}^2 \qquad 5)$$

where ρ_f (kg m^{-3}) is the density of water, μ (kg m^{-1} s^{-1}) is fluid viscosity, g (m s^{-2}) is gravitational acceleration and D_{med} (m) is the median diameter of MPs in a given size class *i*. The deposition flux (m_{dep}, kg m^{-2} s^{-1}) of MPs of an individual size class is simply calculated as:

$$\mathrm{m_{dep}} = \mathrm{u_T}\mathrm{MP_{SUS}} \qquad 6)$$

where $\mathrm{MP_{SUS}}$ (kg m^{-3}) is the mass of MPs of size class *i* suspended in the water column at any given time *t*.

The entrainment of MPs from the sediment bank is a function of flow conditions and D as follows:

$$\mathrm{m_{ent}} = \mathrm{a_8}\mathrm{MP_{SED}} \cdot \omega \cdot \mathrm{f} \cdot \mathrm{r_{MP}} \qquad 7)$$

where $\mathrm{m_{ent}}$ (kg m^{-2} s^{-1}) is the entrained flux of a given MP size class; $\mathrm{MP_{SED}}$ (kg m^{-2}) is the mass of MP of size class *i* present in the bed sediment at any given time; ω (J $s^{-1}m^{-2}$) is the stream power per unit of bed surface; f (is the dimensionless friction factor (depending on channel characteristics), a_8 (s^2 kg^{-1}) is a tunable scaling factor and $\mathrm{r_{MP}}$ (dimensionless) is the entrainable fraction of MPs of a given size class *i*. The frame for the calculation of ω and f and underlined assumptions are described in detail in ref.[24] $\mathrm{r_{MP_i}}$ is MP size-dependent as follows:

$$\mathrm{r_{MP}} = \frac{\mathrm{D_{max}} - \mathrm{D_{low}}}{\mathrm{D_{low}} - \mathrm{D_{up}}} \qquad 8)$$

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where D_{max} (m) is the maximum diameter for the entrainable particles and depends on shear velocity, as described elsewhere.[24] In practical terms, equation 4 sets the following scenarios: i) if D_{max} is larger than D_{up} all MPs belonging to class *i* are entrained from the bed sediment; ii) if D_{max} is smaller than D_{low} no MPs of class *i* are entrained; finally, if D_{max} is between D_{low} and D_{up}, a fraction of MPs belonging to class *i* proportional to $D_{max} - D_{low}$ is entrained from the sediment. Equation 4 assumes a homogenous distribution of MPs mass within the interval D_{low} - D_{up}.

The mass budget of each MP size class *i* in the stream/bed sediment system of any given reach can therefore be calculated as follows:

$$\frac{d(MP_{sed})}{dt} = m_{dep} - m_{ent} \qquad 9)$$

$$\frac{d(MP_{SUS})}{dt} = m_{eff} + \sum_u (A \cdot m_{OUT}) + L \cdot W(m_{ent} - m_{dep}) + m_{up} - m_{down} \qquad 10)$$

where, A (m^2) is the surface of each individual land use unit *u* contributing to delivering MPs to the stream; L and W (m) are the length and width of the stream; m_{up} (kg m^{-3} s^{-1}) is the advection of suspended MPs from the upstream reach, m_{down} (kg m^{-3} s^{-1}) is the advection of suspended MPs to the downstream reach (calculated as $m_{down} = MP_{SUS} \cdot Q$, with Q ($m^3$ s^{-1}) being the river water discharge); and m_{eff} (kg m^{-2} s^{-1}) is the MPs discharge from source point effluents in the reach. While not explicitly simulated, sediment burial occurs when long term deposition exceeds resuspension.

As shown in previous publications[23, 27] INCA-Contaminants simulates multimedia fate of contaminants using the underlying hydro-biogeochemical and particle transport modules. It can therefore simulate both MP transport and the fate of MP-bound contaminants. The focus of this initial study is, however, solely on potential transport and fate of "pure" MPs - aspects related to the fate of associated contaminants are not addressed at this stage.

3. Review of MP characteristics and their pathways to the environment

Within INCA-Contaminants, two properties are used to define MPs: dimensions and density (ρ_{MP}, kg m^{-3}). The environmental burden of MPs may include materials made of virtually all sort of different polymers in use. Although MP density may be influenced by, e.g., growth of biofilms or inclusion of metals in polymers, in the present study ρ_{MP} is assumed to be that of pure polymers. Sources and characteristics of MPs by polymer type were summarized based on three recent national emission inventories from the Nordic countries.[8, 9, 25] (Table S1). Density (ρ_{MP}) can range from 160-2200 kg m^{-3}. Lighter-than-water particles are expected to float and not be retained by bed sediments or soils. A number of primary and secondary MPs emitted on land have, however, density values similar to or higher than water (Table S1).

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There is no apparent relation between polymer type and typical size of released MPs. Dimensions depend on formation processes and emission pathways (Table S1). Primary MPs used in personal care products and emitted directly with wastewater effluents are typically in the dimensional scale of 0.005-0.8 mm (interpreted as a diameter of an equivalent sphere). Material emitted from laundry and household dust has been observed to be in the range of 0.01-0.1 mm. Plastic pellets used as raw material for plastic molding are typically between 1-5 mm. Exfoliation of polymer-based paints in urban or industrial environments produces MPs in the typical range of 0.05-1 mm. Degradation, weathering, breaking down of plastic litter in poorly contained landfills or recycling facility can produce MPs in virtually all size ranges (e.g. 0.005-5 mm).[8, 9, 25]

A relevant fraction of the total emission of MPs from land is anticipated to originate in urban and industrial environments through household/industrial effluents and runoff. In most cases these emissions enter the drainage system and are subsequently conveyed to industrial/municipal WWTPs where, based on their characteristics, they will be either retained in the sludge or directly released with treated wastewater. Releases to the environments can therefore be clustered into two emission typologies: i) point source releases and ii) application of MP contaminated sewage sludge on soils or landfills. A recent report[25] showed that up to 99% of MPs in wastewater are retained in the sludge. The recycling of sewage sludge as manure for agricultural soils or the storage in landfills are common practices and can represent an important pathway for the diffuse emission of MPs to the environment. Transport of this material from soil to the stream has never been described, assessed or simulated before.

4. Endpoints of the analysis and simulation strategy

The main scope of the study is to analyze the potential for soil and river bed sediment to delay or prevent transfer of terrestrial MPs to the marine environment. To this end, INCA-Contaminant was setup to represent a realistic scenario (the River Thames catchment) modeled as a main stream divided into several consecutive reaches, each with a sub-catchment encompassing different land use classes. Multiyear meteorological time series were used to facilitate simulation of realistic calibrated hydrological, soil erosion and sediment transport scenarios. Unfortunately there currently are no data of MPs emissions and concentrations to calibrate and assess model performance. The present analysis is therefore conceived to represent a genuinely theoretical mechanistic assessment of MP transport potential. In order to accomplish such an objective two simplified emission scenarios were considered (providing realistic emission scenarios and concentration predictions are not the ambitions of this study).

<u>Emission scenario A:</u> MP from a direct source point effluent. Within this scenario an arbitrary steady flows of 1000 tones y^{-1} of MPs of each selected size class is emitted continuously from a hypothetical source point located in the uppermost reach of the river. This was conceived to emulate emissions from a single wastewater effluent. At the end of the simulation period the

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percentage of the total cumulative MP emission retained by the river bed sediments (namely trap efficiency) was estimated as follows:

$$\text{Trap efficiency} = \frac{\sum_r \mathrm{MP_{sed,t}}}{\int_0^t \mathrm{m_{eff}}\, dt} \cdot 100 \qquad 11)$$

Where t (s) is the time length of the simulation and the numerator represents the sum $\mathrm{MP_{sed,t}}$ across all reaches r.

Emission scenario B: MP from application of sewage sludge on agricultural soils. As described above, the application of contaminated sewage sludge on soils is expected to be a potentially major source of MPs retained during waste water treatment. The sewage sludge inputs to soils were set as follows: 1667 kg km^{-2} during 6 days per each year of simulation (one day each two months) for a total application: 10,000 kg km^{-2} y^{-1}. The content of MP in the sludge was set to 1%. At the end of the simulation period, end points based on the percentage of the total added mass of individual MP classes retained by a) catchment soils and b) the catchment soils + river bed sediments, were estimated as follows:

$$\text{Trap efficiency by soils} = \frac{\sum_{\mathrm{sc}} \mathrm{MP_{store,t}}}{\sum_{\mathrm{sc}} \int_0^{\mathrm{t}} \mathrm{m_{in}}} \cdot 100 \qquad 12)$$

$$\text{Trap efficiency by soils} + \text{sediments} = \frac{\sum_{\mathrm{sc}} \mathrm{MP_{store,t}} + \sum_{\mathrm{r}} \mathrm{MP_{sed,t}}}{\sum_{\mathrm{sc}} \int_0^{\mathrm{t}} \mathrm{m_{in}}} \cdot 100 \quad 13)$$

where the function $\sum_{\mathrm{sc}} \mathrm{MP_{store}}$ indicates sum of store values across all subcatchments *sc*, while the quantity at the denominator simply represents the total mass of MP added through sewage application in the catchment.

Considering the lack of a relationship between size of MPs and type of polymer, simulations for the two emission scenarios were reiterated considering the full range of combinations between D (varying between 0.05 and 0.7 mm) and ρ_{MP} (varying between 1000 and 1300 kg m^3), so as to cover the size and density ranges of most MPs emitted on land (Table S1).

5. Case study scenario, model configuration and calibration

The Thames is the longest river in England, rising in Cirencester to flow into the North Sea while passing through the most urbanised area in the UK, including Greater London. Average flows range from about 1.5m^3 s^{-1} at Cricklade, to around 29.8 m^3/s at Days Weir and up to 65.8m^3/s at its tidal limit at Kingston. Simulations presented here do not include the tidal zone. The River Thames and its tributaries drain a catchment area of approximately 10,000 square kilometres (non-tidal part) in Southern England, with both permeable and impermeable geologies.[28, 29] Land cover is characterised by significant areas of arable agriculture and pasture in the upper parts of

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the catchment, while the urban areas are mainly located lower in the catchment. Significant forest areas are found throughout. Total population in the catchment approaches 10 million.

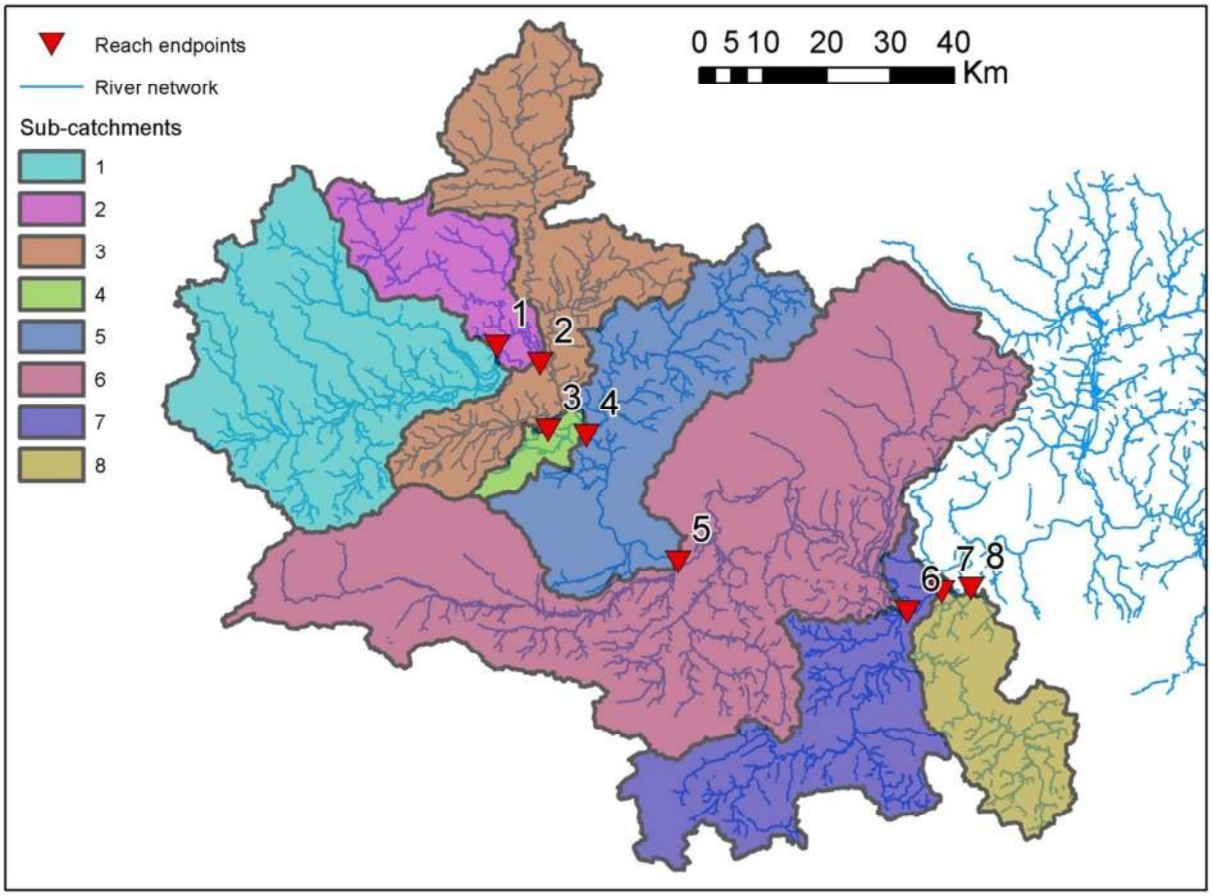

Figure 1 The non-tidal part of the Thames river used in this simulations and the subdivision in subcatchments.

Simulations were performed for the period 2008/01/01-2014/9/30. The model was set up by dividing the Thames into 8 reaches and respective sub-catchments from the source of Thames at Cricklade to its tidal limit at Teddington (Figure 1, Table S2) as described elsewhere. This set-up has been used in many previous applications of INCA-type models to simulate transport of contaminants, sediments and pathogens.[30-32] Daily precipitation and temperature data used as inputs were provided by the UK Met Office referring to all available meteorological stations within the Thames catchment. A spatial average over the catchment was computed for all these variables in order to set up model input files. Mean daily air temperature was computed as the average of minimum and maximum daily values. Data series of hydrological effective rainfall and soil moisture deficit were computed based on these meteorological data using PERSiST[26] an

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evapotranspiration and runoff simulator. After calibration INCA-Contaminants provided a very good simulation of river flows (N-S=0.77-0.82) (Figure 2).

Simulation of soil particle erosion and sediment transport was conducted simultaneously to those of MPs. To this end, 5 sediment size classes were defined representing clay, silt, fine, medium and coarse sand, each with a density of 2400 kg m^{-3}. The Thames is routinely monitored by the UK Environment Agency at multiple sited for a number of parameters including chemical pollutants given serious current and past pollution problems. No monitoring is yet in place for MPs and no MPs emission estimates exist at the moment. Because of this, no direct calibration of the MPs transport module could be performed. Similarly, no assessment of model prediction is currently possible. An indirect calibration of the particle transport module was, however, performed using measurements of natural suspended sediments. The parameters controlling soil erosion/MP mobilization, bed sediment/MPs entrainment and sediment/MPs depositions were adjusted to optimize the fit between time series of simulated and measured data of suspended sediment concentrations in different reaches of the Thames per individual size class. Good fits was obtained after calibration (R^2 = 0.27-0.63) (Figure 2). While this does not guarantee high predictive performance for MPs, the approach adopted is, nonetheless, the only practicable option at present.

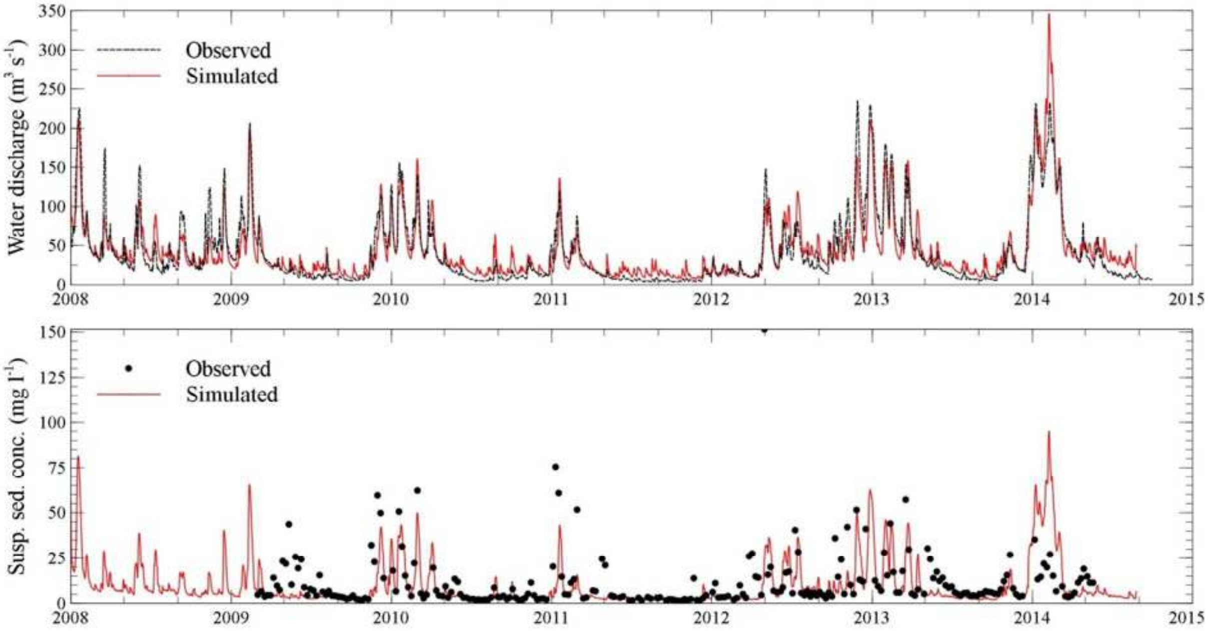

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Figure 2. Comparison between simulated (red lines) and measured (dots) data of river discharge and suspended sediment concentrations in a central reach (Reach 5) of the Thames. Data for other selected Reaches are displayed in Figure S1 and S2.

6. MPs transport by the stream.

Figure 3 shows an example of simulated MPs discharges responding to emission scenario A for a central reach of the Thames. As expected, transport of MPs in the stream is strongly dependent on flow regimes with intense flow and flooding periods causing mobilization of heavier and larger MPs. Discharge peaks coincide with periods of high flow. This behavior is more evident for MPs with intermediate size and density values. The transport of larger (i.e. 0.3-0.5 mm) and heavier-than-water MPs appears to be inhibited during base flow periods. Intense mobilization peaks can, however, be observed during extreme floods. Interestingly this applies also to large MPs with densities similar to that of water (i.e. 1050 kg m^{-3}). In contrast very small MPs (i.e. 0.001-0.005 mm) are transported effectively and have a relatively constant base flow, independent of their densities. These simulations show the potential for a strong hydrological control on the storage of larger and heavier MPs within the stream system. Size (in contrast to density) appears to be a more sensitive parameter influencing MP transport dynamics.

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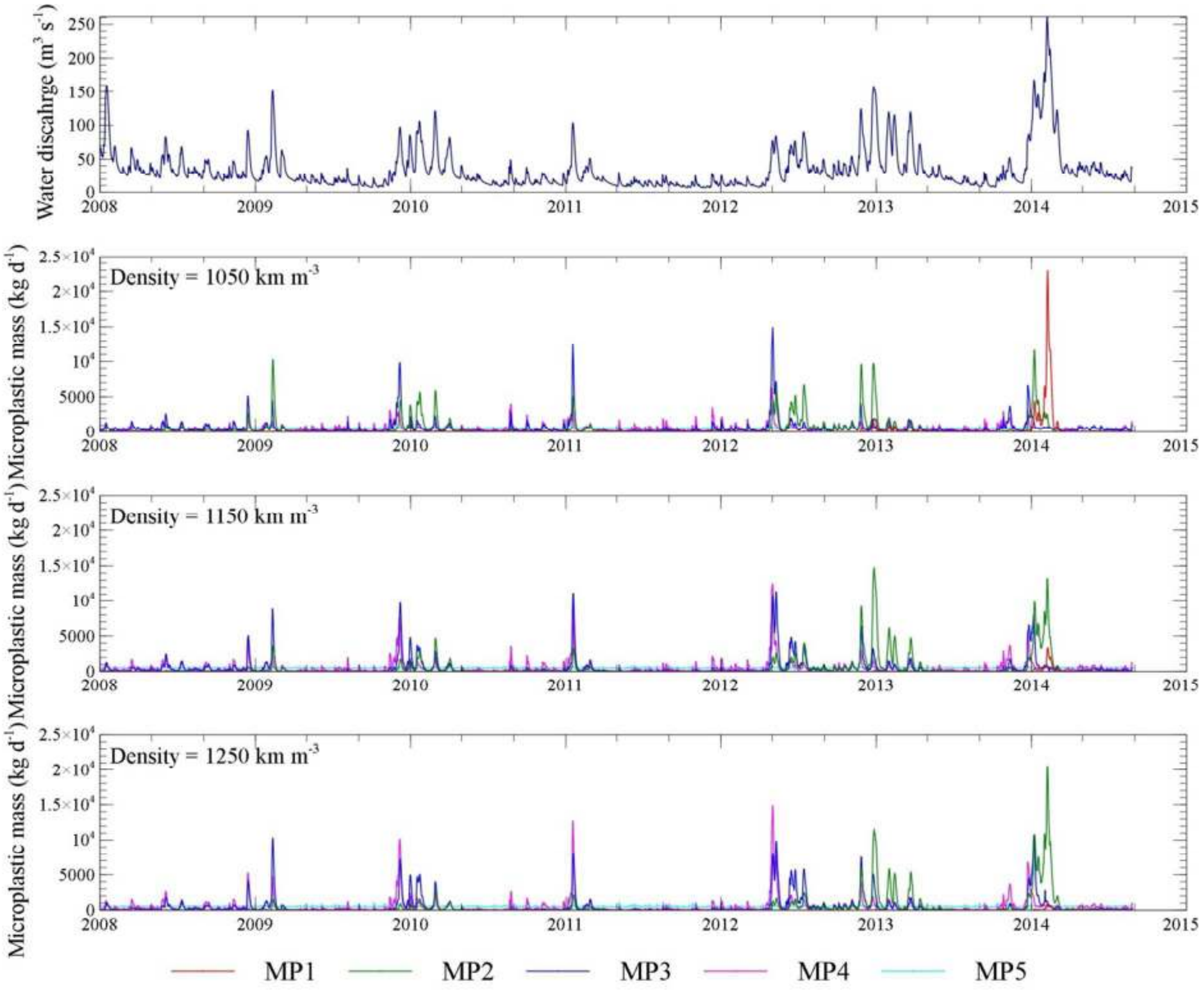

Figure 3. Simulated discharge fluxes for 5 size classes of MPs (MP1: 0.3-0.5 mm, MP2: 0.1-0.3 mm; MP3: 0.05-0.08 mm; MP4: 0.01-0.05; MP5: 0.001-0.005) and selected 3 different density values.

7. MP retention by bed sediment

Figure 4 displays the pseudo-steady state cumulative bed sediment trap efficiency along the river as a function of MP size and density. The figure relates to emission scenario A in which all MPs are emitted from a single effluent point in the most upstream reach. The potential for retention is present in all river reaches for basically all MPs heavier than water. In the upstream reaches (i.e. reaches 1-3), less than 20% of the discharge mass of any type of MP was retained. Cumulative retention efficiency along the full course of the Thames is estimated to sharply increase (especially for larger particles) in downstream reaches, rising to 90-100% in reach 8 for particles with dimensions > 0.2 mm. Total retention efficiency is considerably smaller for smaller particles.

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Variations in retention efficiency are primarily dependent on MP dimensions (with density having a smaller effect). This should not come as a surprise since MP size can range across several orders of magnitude while the variability in density is confined within one order of magnitude for the full range of polymers in use (less than a factor of 2 when considering only heavier-than-water MPs, see Table S1). Despite this, there is obviously a sharp change in parameter sensitivity when the MP density value approaches that of water. Within this domain, a small decrease in ρ_{MP} causes a major decline in retention efficiency for MPs of any size.

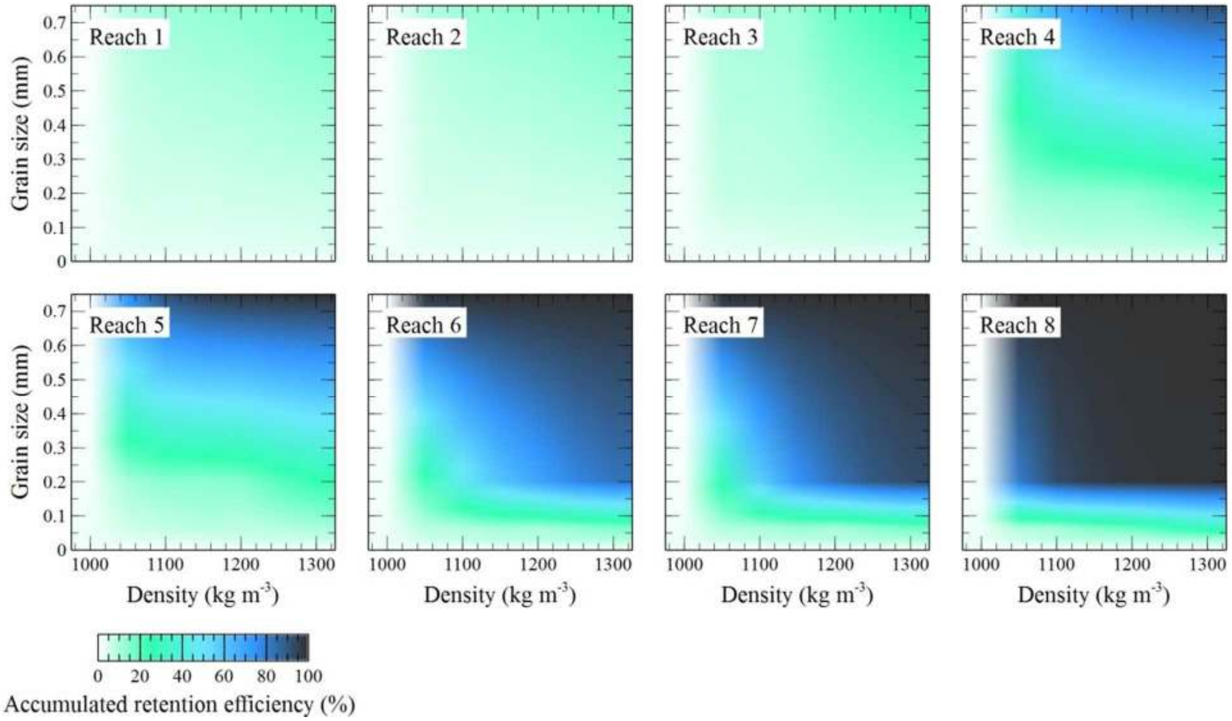

Figure 4. Pseudo-steady state cumulative MP retention efficiency by bed sediment as a function of density and size (emission scenario A).

8. MP retention by soil/stream system

Results for MP-retention efficiency by soil under emission scenario B (application of MP-contaminated sludge) are depicted in Figure 5. Since MPs are added to soils every second month throughout the simulation period, the figure represents the pseudo-steady state scenario. Depending on soil characteristics and precipitation patterns in individual subcatchments, approximately 16% to 38% of the MPs added with the sludge are predicted to be found (at any moment) in the soil. This figure may obviously change when different application rates are used.

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Nevertheless, the simulations show a considerable potential for soils to retain and accumulate MPs.

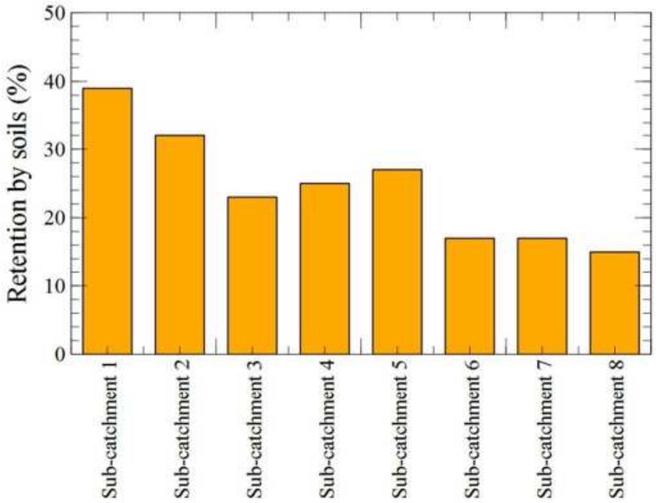

Figure 5. Pseudo-steady state retention efficiency by agricultural soils in individual subcatchments (emission scenario B).

The combined retention efficiency of soils and bed sediment for individual reaches is displayed in Figure 6. This chart is fundamentally different from Figure 4 because it shows diffuse emissions in all subcatchments rather than a single upstream point source. The total retention efficiency of the Thames catchment is represented by the results for the most downstream subcatchment (8). Similar to what was observed for emission scenario A the catchment is expected to effectively retain (in soils and the bed sediment) most of the MPs with density higher than 1050 kg m^{-3} and larger than 0.2 mm. Retention efficiency may, however, vary widely across different subcatchments as a combined result of soil hydraulic characteristics, local precipitation patterns and river hydrology. Low stream power and more permeable soils are obviously associated with predominant storage, retention and deposition. As a results agricultural and forest soils are anticipated to store more MPs than urban soils.

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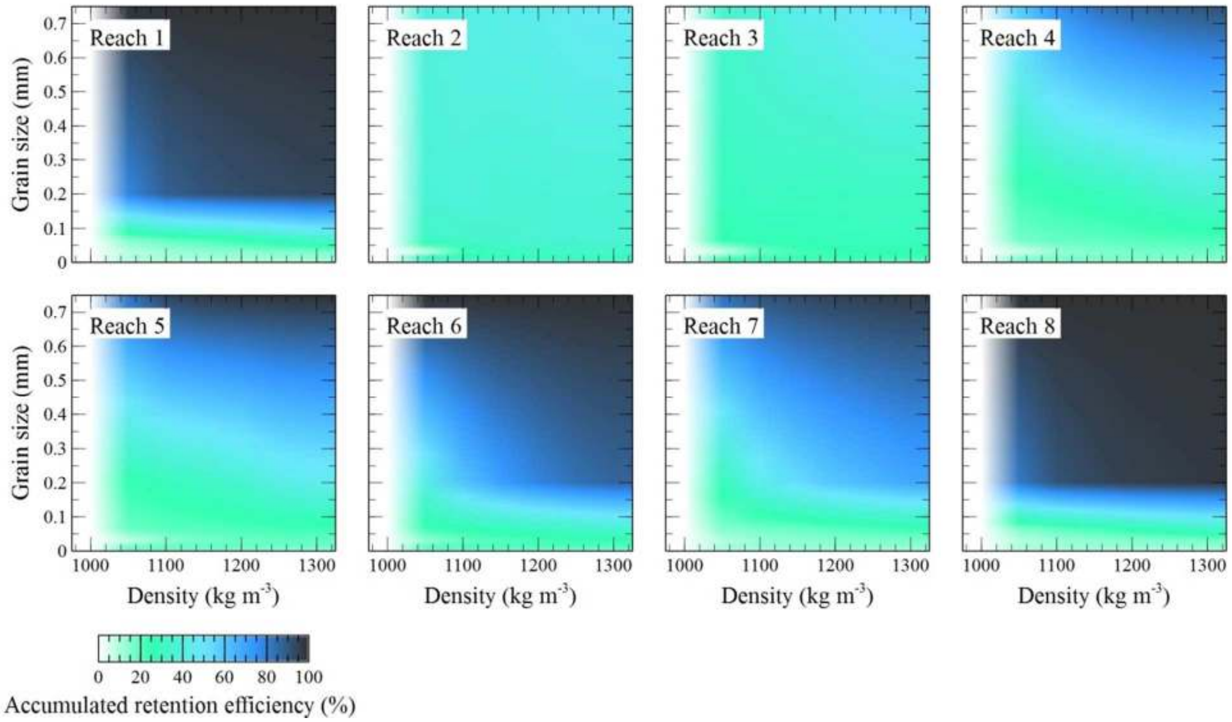

Figure 6. Pseudo steady state cumulative soil+bed sediment retention efficiency for MPs as a function of density and size (emission scenario B).

9. General remarks and limitations of the present assessment

This theoretical assessment anticipates a potential for accumulation in soils of a considerable fraction of the MP emitted on land (e.g. through sewage sludge applications). It also predicts an effective retention of MPs with density higher than water and larger than 0.2 mm, by river bed sediments. Sediments of river sections experiencing low stream power are possible hotspots for the accumulation of plastics. Data from a recent study on MP distribution along river bed sediments[33] (namely the Rhine river in Germany) appear to confirm that river hydrology is an influencial driver of MPs distribution in sediments. As far as we know these are the only available experimental data supporting one of the key finding of our theoretical assessment. Exposure and impact assessment for MPs in river sediments should prioritize these sink areas.

Effective retention in agricultural soils is also of concern considering the possible implications for soil ecosystem functioning, crop production and livestock which may ingest MP while pasturing. Soil amendment practices using sewage sludge should consider these risks and adopt application rates suitable for maintaining MP levels in soil within acceptable safety thresholds.

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Similarly to lighter-than-water particles, MPs smaller than 0.2 mm are predicted to be poorly retained in the catchment (independent of their density) and will eventually be conveyed to the marine environment. This pool includes different types of MPs emitted with city, household and laundry dust, possibly representing a large fraction of the total terrestrial emissions. For these types of MPs, rivers and the terrestrial environment can constitute a significant (possibly dominant) source of MPs found in the Oceans.

There are obvious limitations associated to the present assessment. Unavailability of data for calibrating and assessing model predictions represents, at present, the major difficulty. The soil erosion/particle transport module was calibrated using measurements of natural suspended sediments. Despite the hydro-physical controls over MPs and natural soil particles mobilization and transport are the same, their effectiveness is very likely modulated by the different densities and shapes of MPs. MPs can for example be emitted as microfibers from laundry and household dust. Interaction between particles with elongated shapes and viscous fluids it can actually be very different from that of pseudo-spherical materials. Dedicated experiments and calibration studies are therefore necessary for improving the realism of models.

A second limitation is that our model essentially considers MPs as pure and totally inert polymers. In WWTPs and the environment formation of biofilms on the MPs could results in changing their densities and hydraulic properties, affecting therefore their transport. It can also be hypothesized that smaller particles of some synthetic material could also tend to aggregate in water environments affecting possibly affecting the size distribution and transport behavior while the MPs are moving through the catchment. Similarly, some materials my break down in soils and stream environment resulting in a larger number of smaller MPs. There currently is insufficient information to formulate solid assumptions and attempt a meaningful mathematical description of these processes. A better empirical understanding of MP transport in catchment is therefore necessary before the model can be upgraded to a higher level of detail.

For the further development of predictive tools of MP transport in catchments we envisage the development of new monitoring studies that could provide data on the distribution and characteristics of MPs in sediments and stream water focusing on river sections with contrasting hydrological characteristics. Similarly, experiments that could elucidate the hydrodynamic properties of MPs with different size and shapes are necessary for refining the mechanistic frame.

While it is unrealistic to think about practical remediation actions after MPs have reached the ocean, the only practicable solutions to limit their accumulation in the marine environment are those focusing on management of emissions and transport processes on land. Developing conceptual tools and models to describe these processes at the catchment level is an essential step towards management solutions and preserve a sustainable use of wastewater resources. Limiting emission to the marine environment is pivotal. Nevertheless special attention should be

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dedicated to the analysis of risks and impacts the accumulation of these materials in soils and sediments poses.

Acknowledgements

We are grateful to the UK Environment Agency for making suspended sediment and flow monitoring data available. This paper was conceived during the 2016 NIVA Workshop on Catchment Modelling a.k.a. Science Camp. This work was financially supported by the National Sustainability Programme of the Czech Ministry of Education, Youth and Sports (LO1214) and the RECETOX research infrastructure (LM2015051).

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