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Riverine transport of microplastics from the Dutch border to the North sea

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Client	**Project**	**Attribute**	**Pages**
Yann Friocourt (RWS-WVL) Bert Bellert (RWS-WVL)	11203712-002	11203712-002-ZKS-0004	39

Keywords
Plastic item, advection, deposition, Rhine, Meuse.

Summary
Microplastics are transported with the flow in the river. In this study we modelled the fate of microplastics released at Lobith in the Rhine and at Eijsden in the Meuse, using a depth and width averaged flow model for the Netherlands (*Landelijk SOBEK model*). For the transport of microplastics the processes advection, deposition and hetero-aggregation of microplastics with sediment are included. Overall, the model results suggest that the deposition is small: about 66 to 90 percent of the released microplastics are transported out of the model towards the sea, meaning that 10-34 percent are either deposited to the river bed or are stored in the water column. Resuspension of deposited microplastics was not included in the model. A sensitivity study for which resuspension was included suggests that it is not an important process in the current 1D simulation, since the flow velocities at accumulation areas rarely exceed the critical flow velocity for resuspension. The simulated annual transport of microplastics is higher than estimates based on observations, although sources within the Netherlands are not yet included in the model. This needs to be re-evaluated in the future, after sources of microplastics from within The Netherlands have been introduced in the model.

References
European Maritime And Fisheries Fund, KPP waterkwaliteitsmodelschematisaties 2019.

Version	Date	Author	Initials	Review	Initials	Approval	Initials
0.6	Nov. 2019	Annelotte van der Linden Arjen Markus Frans Buschman		Jos van Gils		Toon Segeren	

Status
final

Contents

1 Introduction

1.1 Background

Based on observations, van der Wal et al. (2015) estimate that in between 20 to 31 tonnes of plastic are transported via the Rhine river to the North Sea annually. Especially microplastics (here defined as particles within the size range 0.05 and 5 mm) may affect human health negatively (Barboza et al., 2018) and also marine and freshwater ecosystems. Important point sources of microplastics in rivers are locations where microplastics are released into the river, such as waste water treatment plants (WWTP). Diffuse sources include the fragmentation of macroplastic items and tire and road wear particles that are flushed into the river (Unice et al., 2019). Once in the river, the different types and sizes of microplastics are transported with the flow. How this transport depends on environmental conditions is largely unknown. Observations to build up this knowledge are scarce.

The European union has put in place the Marine Strategy Framework Directive (MSFD), aiming to maintain or achieve a good environmental status of European seas. An important source is land based litter that is transported via rivers to the sea. Therefore, the implementation for the MSFD includes the development of a monitoring strategy for rivers, aiming to result in an estimate of the yearly load of riverine litter to the sea. The work described in this report is supported by the European Maritime And Fisheries Fund (EMFF). Project 2 of this EMFF project has the aim to develop a standardized monitoring strategy that can be applied to monitor the microplastic concentration in freshwater. The strategy should give a standard for both the sampling and analysis. In order to develop such a monitoring strategy and to help select locations for the monitoring, a model is developed aiming to simulate the transport of microplastics in the rivers Rhine and Meuse. For project 2 of the EMFF project, a roadmap was prepared for the development of such a model (Buschman et al., 2018). The roadmap describes activities up to 2023 in order to have a complete model framework that can be used to simulate the transport of microplastics in Dutch rivers, estuaries, lakes, canals and the sea.

Apart from the EMFF project 2, the Dutch ministry of Infrastructure and Water works is working on a Delta-approach on microplastics. Furthermore, the ministry is preparing pilots in which systems to collect plastic items are being tested and implemented. These initiatives can benefit from modelling the (micro)plastic transport as well. The policy of the ministry is applied by Rijkswaterstaat. Rijkswaterstaat is aiming to develop water quality models that consist of interchangeable building blocks as much as possible, also for modelling plastic transport.

1.2 Problem statement

It is largely unknown how conditions in water bodies affect the processes that result in the transport of microplastics. Observations of microplastic concentrations in the water column are limited. Due to the effort needed to do such observations, usually they are carried out at one location in the water column only and are only repeated a few times. In order to set up a monitoring strategy, a monitoring location needs to be selected carefully. With a model, the spatial and temporal variation of the microplastics concentration can be predicted. In turn, in order to set up a model to simulate the transport of microplastics, observations for verification of the model results are needed.

1.3 Objective

This study focusses on transport of microplastics in the main rivers in the Netherlands: the Rhine and Meuse branches. Rijkswaterstaat and Deltares decided that the focus of this study should be on the larger scale and therefore a depth and width averaged model was used. Hence, this first modelling study does not address the distribution of microplastics in the cross-section of a river.

This study has the following main objective:

> "Determine the annual transport of microplastics in the Rhine and Meuse river branches from the Dutch border to the North Sea from modelling. "

Furthermore, this study describes in addition to the roadmap what modelling functionality is needed to arrive at sound conclusions for the main objective.

1.4 Available observations

For this study, we listed available observations of microplastics concentration (in g/l) within Dutch rivers (Table 1.1). As for other rivers, continuous time series for a year at a single location are not yet available.

Table 1.1: Overview of observations within the Netherlands of microplastic concentration in water (g/l).

Location	**What was monitored (method)**	**Time(s) of observations**	**Reference (remark)**
Lobith/ Bimmen	Microplastic concentration (mg/l) 0.125- 0.25 mm and .25-5 mm (two sieves)	17 observations within January 10th- May 26th, 2014 (average over a weekend)	Urgert, (2015)
Eijsden	*Same*	*Same*	Urgert (2015)
Dommel and outflow point in Maas	Nano- and larger microplastics concentration	2015?	Not yet available; After publication willing to share data (Svenja Mintenig, UU/KWR)
Along German and Dutch Rhine (Rotterdam, Zuilichem, Rees)	Concentration microplastics in particles per l (300 mu mesh)	June/July 2014	Mani et al. (2015)
Rhine (Nieuwe Waterweg, Haringvliet)	Micro- and mesoplastic (5-25 mm) concentration	2014	Van der Wal et al. (2015)
Harbours around Rotterdam (e.g. Londenhaven, Waalhaven) en Tweede Maasvlakte	Pellets type, size and number (using photos of field visits)	Occasionally (6 visits in parts of harbor area briefly reported) in period June-November 2019.	Observations by Piter Hiddema (RWS WNZ) Aim: To find sources of pellets
Cleaning days river shore: Maas, IJssel and Waal	Fraction <10 mm (minimal contribution to weight)	Occasionally in 2015-2018	Schone rivieren/ RWS

Besides the observations listed in Table 1.1, the company Allseas has been monitoring plastics concentration in the Nieuwe Maas. They aim at macroplastics, although particles within a fraction of 1-5 mm are collected and analysed as well.

Since the observations are not aimed at microplastics and microplastics may slip through the nets, these observations are not listed in the table. Similarly, the shoreliner (Tauw, 2017) collected microplastics, although it is aiming to collect larger plastic items. It does not give concentrations and is therefore not shown in the table.
Microplastic concentration (particles per kg dry weight) in river sediment, suspended matter collected in rivers and in sewage water treatment plants (influent, sludge and effluent) were determined by the VU-IVM in the years 2013 to 2015 (e.g. Brandsma, 2013). These results were summarized and put in perspective to observations in other countries by Leslie et al. (2017). Since these studies do not determine the concentration of microplastics in river water in g/l but in number of particles per volume, they are difficult to compare to model results. Therefore, they are not included in the table.

1.5 Approach

For this study the first activities described in the roadmap are carried out, resulting in the first model results of microplastics transport within the Netherlands. The roadmap that was prepared in 2018 for the first part of project 2 of EMFF is summarized in Chapter 2. We focused on the main rivers in the Netherlands: Rhine and Meuse branches (Figure 3.1). Microplastics were released in the main rivers at the Dutch border only (Lobith for the Rhine and Eijsden for the Meuse river). Sources within the Netherlands are not included at this stage, since a careful preparation of these sources would take too much time. This is expected to result in an underprediction of the microplastic transport.

The processes included in the model are advection, deposition and hetero-aggregation (Table 2.1). Resuspension was not included in the model, since there is hardly any information found in literature on the resuspension of microplastics. Without this process the water quality results represent the worst-case estimate of the amount of microplastic that can accumulate on the river bed. To quantify the possible effects of resuspension at least in order of magnitude, a separate simulation was performed. Results of both simulations are presented and discussed in chapter 3.

2 Roadmap for modelling the distribution of microplastics

2.1 Introduction

In Buschman et al. (2018) a roadmap is described how to arrive at a model (set) that can be used to model the transport of microplastics in the main water bodies of the Netherlands. Since that report is only available in Dutch, we summarize the activities of the roadmap here in English. Possibly the roadmap for the development of a model (set) can be optimized, when the first model results are available.

2.2 From conceptual model to effect chain

The transport of microplastics and their distribution depends on many variables. In rivers and estuaries, the flow, point and diffuse sources are important, but also the hydraulic roughness of the river bank, the presence of fish or filter feeders eating microplastics can play a role. Algae can make the particles stickier (biofouling), enhancing the formation of aggregates. Also waves and the presence of (different sizes of) suspended sediment in the water column play a role. The list of variables and processes that potentially play a role in the distribution of microplastics concentration is long. In order to get an overview of the dominant processes and variables, effect chains have been set-up for the different water bodies. Figure 2.1 shows the effect chain for rivers.

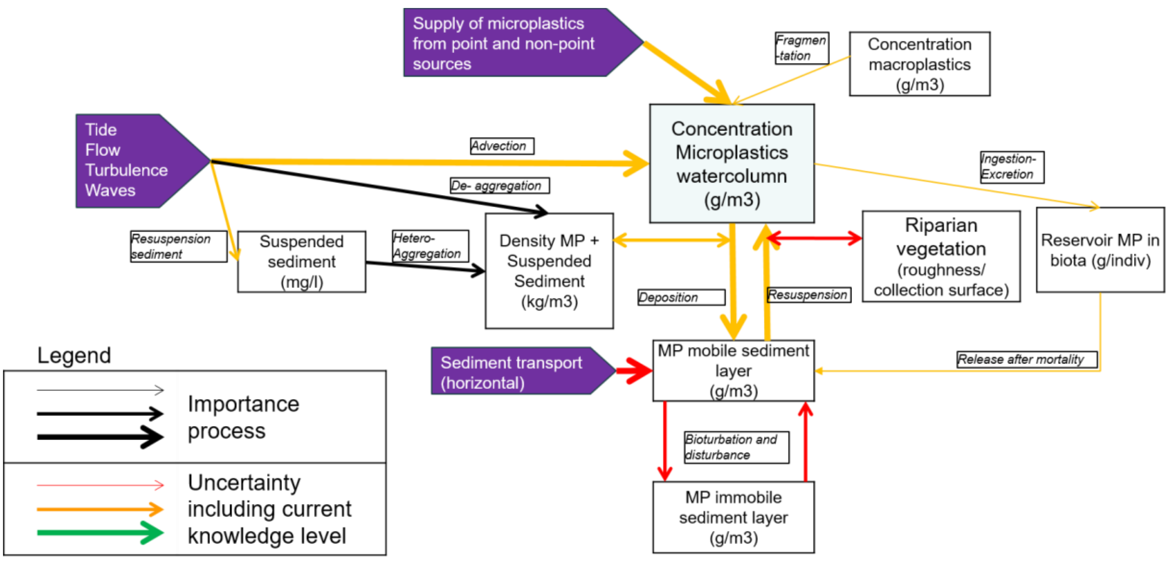


Figure 2.1 Effect chain for microplastic concentration in the water column of a river. As indicated in the legend, a thicker arrow depicts a more important process and a red arrow has high uncertainty (Buschman et al., 2018).

Some of the processes (for definitions see Table 2.1) described are visible in this effect chain. Deposition to the mobile bed and resuspension from this layer occur and some knowledge is available about these processes, which was a reason to color these arrows orange. The impact of vegetation on these processes is largely unknown but is expected to be small in relation to other processes. The dominant process is advection with the flow. Hetero-aggregation with suspended sediment is included as a process with medium importance. Hetero-aggregation usually results in a combined particle that is denser than the plastic particle. Hence, the combined particle is more likely to deposit. This aggregation is accelerated when the water column is salty, as in estuaries. Therefore, salinity is included as an additional variable in the effect chain for estuaries.

For the rest, that effect chain is the same as for rivers (Figure 2.1).

The role of biofouling is not included in the effect chain, because it was believed that the time scale in the river is not that long that biofouling on a plastic particle is changing its stickiness much.

Table 2.1 Definitions of processes relevant for the transport of microplastics

Process	Definition
Advection	Transport along with the flow
Deposition	Sinking of a particle out of the water column to the bed
Resuspension	Uptake of a particle from the bed into the water column
Trapping	Plastic is trapped at obstacles or in vegetation
Ingestion	Eating by organisms (fish, mussels, benthic organisms)
Excretion	Elimination of metabolic waste from an organism
Hetero-aggregation	Formation of flocs consisting of both microplastics and sediment
Homo-aggregation	Formation of flocs of microplastics solely
Biofouling	Formation of an organic layer around a particle that change their characteristics (stickiness, density, size)
Degradation/aging	Changing of the surface of a particle by micro-organisms, UV-light and physical forces such a generated by waves
Fragmentation	Falling apart of a particle by mechanical processes. Fragmentation of a macroplastic can lead to the formation of microplastics.
Bioturbation	Benthic animals can bury microplastics by displacement of sediment
Morphological processes	Large-scale displacement of sediment, such as sand dune migration or dredging can bury microplastics or alternatively bring them to the surface

2.3 Knowledge gaps

Based on the effect chain, our knowledge gaps were identified per category. For the most important processes with a medium uncertainty particularly for rivers, the research questions are:

1 At what critical bed shear stress are microplastics resuspended?
2 How relates trapping in the river bank to conditions, such as river discharge?
3 What is the role of vegetation in trapping?

An additional 14 research questions are formulated for rivers, canals, estuaries, lakes and the sea.

2.4 Two model applications

The first model application concerns the distribution of microplastics along the larger rivers, estuaries and canals of The Netherlands. At discharge peaks the transport may be much higher than in other periods. To study the variation of the transport with river discharge, at least one year needs to be simulated. In rivers a time step of 1 day is sufficient, whereas in estuaries 1 hour is needed to capture the tidal variation. At this large spatial scale and the temporal scales, probably only depth and width averaged modelling (1D) is feasible. This study focused on this type of application.

To make the summary of the roadmap complete, also the second application is mentioned. As a depth and width averaged model cannot capture details such as floodplains or near-shore shallow areas where more sedimentation might occur, a more comprehensive type of modelling is called for to get a better understanding of the system. This approach that could be taken in a follow-up study.

3 Modelling microplastics transport

3.1 Method

3.1.1 General

In this study, we focused on the main rivers in the Netherlands: Rhine and Meuse branches (Figure 3.1). To investigate the transport of the microplastics in these rivers, we used results of the one-dimensional hydrodynamic model Sobek (Advanced Version 2.13.002c) in combination with the water quality software DELWAQ (version 5.08.00.64359M, built 19 July 2019). In this version of DELWAQ the processes of advection, deposition and hetero-aggregation are included.

To model the transport of the microplastics these rivers we made various assumptions:

- Initial concentrations of all substances are zero and no spin-up time.
- The upstream boundary conditions are constant.
- Homo-aggregation of microplastics is negligible compared to hetero-aggregation.
- Trapping of microplastics is negligible.
- Microplastic waste loads are zero.

In addition, resuspension was not included in our model, since there was hardly any information found in literature on the resuspension of microplastics. Without this process the water quality results represent the worst-case estimate of the amount of microplastic that can accumulate on the river bed. Additionally, the absence of resuspension leads to an underprediction of the microplastics concentration in the water column. To study the effect of resuspension in our model we repeated one simulation with a tentative formulation of resuspension (section 3.3).

Homo-aggregation was not included in the model since we assumed homo-aggregation of microplastics is negligible compared to hetero-aggregation, due to low concentrations of microplastics relative to the concentrations of suspended solids. We also neglected trapping of microplastics in vegetation of the river bank, since knowledge about this process is limited.

Waste loads were not included in the model, because this information was not rapidly available. This will lead to an underestimation the microplastics concentration within the Rhine and Meuse rivers. By neglecting microplastic waste loads, the results solely give an indication which part of the microplastics that enter the Netherlands via the large rivers is transported to the North Sea. Another reason for underestimating the annually averaged loads is that we did not apply a spin-up period, such that in the beginning of the simulated year the microplastics concentration starts at 0 mg/l and increases while the initial water is being replaced.

Figure 3.1 Schematization of the Landelijk Sobek model (LSM) version 1.2 and output locations of DELWAQ (circles). Output locations included in the generated river profiles of the Meuse and Rhine are shown in blue and green, respectively. Map was created using ArcGIS 10.6. Labels are assigned to the output locations to increase the readability of the map. The original Sobek IDs and assigned labels are listed in Table 3.1.

Table 3.1 Sobek ID and corresponding label of output locations of DELWAQ. Locations are shown in Figure 1.

Label	Sobek ID
A	224
B	DI_Lekkerkerk
C	DI_RelJerwaard
D	DI_Schoonhoven
E	DI_ZwlJndrecht
F	DO_Evides Scheelhk
G	DO_meas_Crevecoeur
H	KE_C.Gelderlnd_Waal
I	KI_Ashland_O. Maas
J	KI_Kema_NederrlJn
K	KI_Nedri_Maas
L	KI_SABIC_Maas
M	KI_Vogelenz_NdrrlJn
N	MS_008_0
O	MS_016_0
P	MS_022_0
Q	PI_Evides G.vdKerks
R	PI_Kuypers ZGBedr
T	RT_006_0
U	WB_DEMTPCPBSD
V	WB_HARDINXVELD
W	WB_HOEKVHLRTOVR
X	WB_MAASSS
Y	WQ_UNILEVER

3.1.2 Hydrodynamics

For this modelling study, we used hydrodynamic results from version 1.2 of the *Landelijk Sobek Model* (LSM) (Prinsen and Wesselius, 2015). To simulate the plastics transport, we used hydrodynamics results of the year 2014, since for this year there are some observation data available from previous studies (Mani et al., 2015, Urgert, 2015) which we could use to determine the upstream boundary conditions and to validate the model results.

Before using the hydrodynamics for the water quality modelling, we carried out a few tests to investigate the quality of the hydrodynamics of LSM 1.2. We compared the simulated discharges to observation data available at Waterinfo (www.waterinfo.nl) and performed a fraction analysis to identify the area of influence of water entering the model at Lobith and Eijsden. In a fraction analysis, labels are assigned to certain locations in the model. Water that enters the model at a labeled location, will carry this label with it throughout the model network. This way the water can be traced from its source throughout the network. Figure 3.2 and Figure 3.3 show that the Rhine and Meuse fractions are spread over the rivers after 1 month as expected. The Rhine fraction in the Rhine branches is almost 1 and is decreasing towards the sea (including lake IJssel), because Rhine water has not yet replaced the initial water mass.

The simulated discharges compared to measured discharges for 2014 for one location in the Rhine, Driel Boven, and one in the Meuse, Borgharen dorp, are presented in Figure 3.4 and Figure 3.5, respectively.

Besides the overestimations at Driel boven during spring and fall, both locations show a good correspondence between the simulated discharges and observations. Hence, these results indicate that the hydrodynamic model resolved the flow sufficiently well to use it as the base for the transport of microplastics. Therefore, we subsequently coupled the hydrodynamic results with DELWAQ using the Sobek interface.

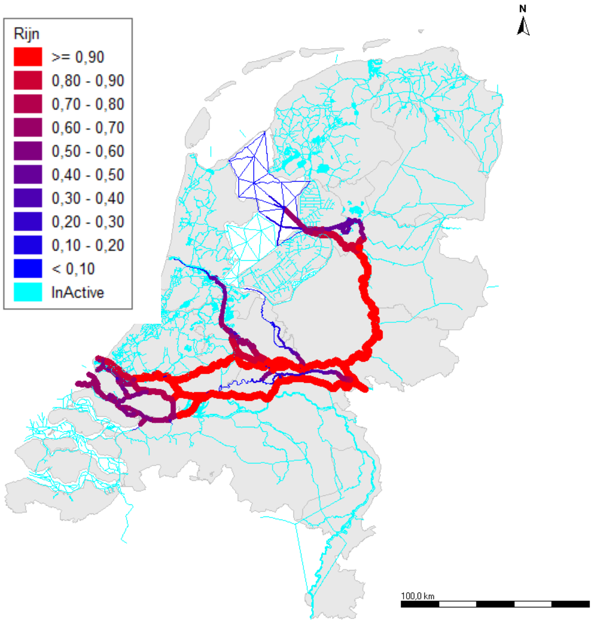


Figure 3.2 Rhine fraction after 1 month simulation (on February 1 2014).

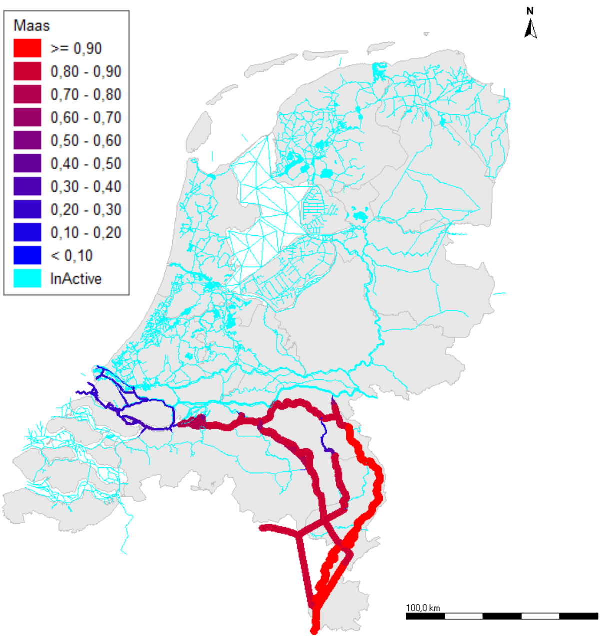


Figure 3.3 Meuse fraction 1 month simulation (on February 1 2014).

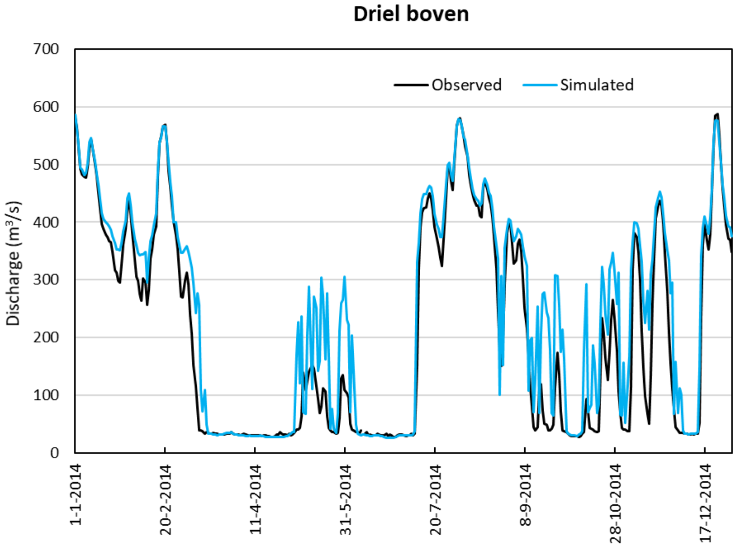


Figure 3.4 Overview of the simulated and observed (black; www.waterinfo.com) discharge in the Rhine at Driel boven (blue) for 2014.

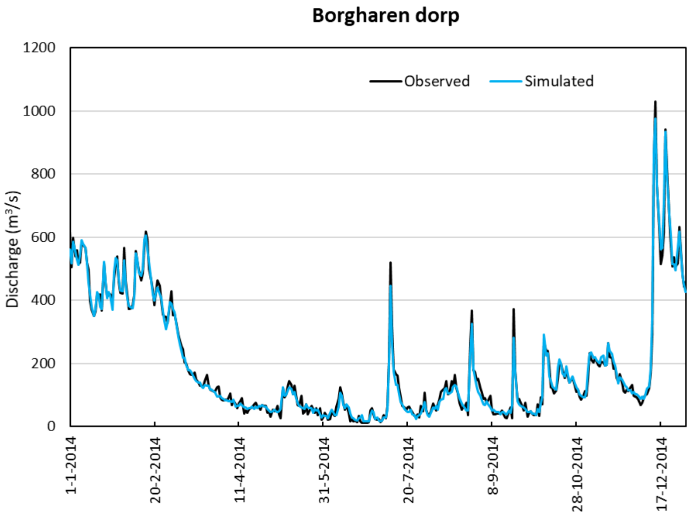


Figure 3.5 Overview of the simulated and observed (black; www.waterinfo.com) discharge in the Meuse at Borgharen dorp (blue) for 2014.

3.1.3 Water quality

3.1.3.1 Microplastics characteristics

Environmental microplastics encompass a wide range of material types with different chemical compositions, shapes, colors, sizes and densities (Thompson, 2015, Mani et al., 2015). Microplastics often occur as unrecognizable fragmented pieces derived from degradation of larger plastic items, making characterization of these particles difficult. Therefore, characterization is usually done based on analysis of polymer type and/or classification of size and/or shape. Due to the lack of a uniform definition and standardized methodologies, microplastics are often classified in different shape categories, e.g. pellets, spherules, fragments (Burns and Boxall, 2018), which makes it difficult to compare results of monitoring studies.

Nonetheless, fragment, fibre, film and foam are predominant microplastics shapes reported in previous studies (Koelmans et al., 2019). Pellets are also frequently detected in riverine surface water samples particularly those made of synthetic polymers such as Polyethylene (PE) and Polypropylene (PP). Both polymers have densities below 1000 kg/m^3 (Koelmans et al., 2019). Since the density of these polymers is so close to that of freshwater, microplastics composed of these materials are buoyant and will therefore get transported over larger distances as compared to heavier particles which will sink to the river bed.

The high diversity in composition makes microplastics complex particles to model. To address this complexity, we characterized microplastics types using discrete size fractions, shape characteristics, and polymer densities. To gain further understanding of the fate of microplastics in rivers in general it is essential to not only focus on one type of microplastics with a specific size, shape and density. Hence, for the modelling we selected a range of size, shape and density combinations which covers the variety of plastic types found in rivers. Within these combinations the shape varied from spherical particles, such as microbeads, to particles shaped as elongated fibres. Furthermore, polymer density varied from 970 to 1300 kg/m^3, representing a wide range of polymer types, with particles with a density of 1000 kg/m^3 and lower also representing non-settling microplastics.

In total, we modelled a set of 24 microplastic particles. The characteristics of the modelled microplastic particles are presented in Table 3.2

Table 3.2

Microplastic particle types with a size of 0.3 mm are included, since observations show that the mass of particles smaller than 0.5 mm (0.05 – 0.5 mm) can be substantial. The shape of the microplastic particles is usually described by the Corey shape factor (S; equation 1), which is defined as:

$$S = \frac{c}{\sqrt{ab}} \qquad (1)$$

where S is Corey shape factor (m), c shortest length of a particle (thickness) (m), b is the intermediate length (m) and a is the longest length (m). For a spherical particle S equals one. Particles shaped as fibres have among the lowest values for S (0.01). The Corey shape factor is used such that the fall velocity of different shapes of a particle can be estimated theoretically.

Fall and rising velocities of the various particles (both microplastics and sediments) were estimated using the empirical relation published by Waldschläger and Schüttrumpf, (2018):

$$D_* = S * d * \sqrt[3]{\left(\frac{\rho_p - \rho_w}{\rho_w}\right)\frac{g}{v^2}} \qquad (2)$$

$$w_s = 0.0025\, D_* \qquad (3)$$

where, D_* is the dimensionless diameter, S is the Corey shape factor, d is the equivalent diameter of the particle, ρ_p is the particle density, ρ_w is the water density g is the gravitational acceleration, v is the kinematic viscosity of water and w_s is the fall or rising velocity of the particle.

Table 3.2 Selected microplastics particles that are typical for rivers, including their characteristics size, density, and shape factor.

No.	Size (mm)	Density (kg/m^3)	Shape factor
1	0.3	1050	1
2	1	1050	1
3	5	1050	1
4	0.3	970	0.01
5	1	970	0.01
6	5	970	0.01
7	0.3	1000	0.01
8	1	1000	0.01
9	5	1000	0.01
10	0.3	970	1
11	1	970	1
12	5	970	1
13	0.3	1000	1
14	1	1000	1
15	5	1000	1
16	0.3	1050	1
17	1	1050	0.01
18	5	1050	0.01
19	0.3	1300	0.01
20	1	1300	0.01
21	5	1300	0.01
22	0.3	1050	0.1
23	1	1050	0.1
24	5	1050	0.1

3.1.3.2 Processes

We used processes available via the Open Processes Library (OPL) of DELWAQ to model the fate and transport of the microplastic particles in river. These processes include:

- Advection;
- Aggregation of microplastic and suspended particles forming hetero-aggregates;
- Deposition of both unattached and aggregation microplastic particles and sediment.

Potentially relevant processes of de-aggregation and resuspension (Figure 2.1) are not included in this model. For the calculation of the deposition and aggregation rate we used the same formulation as was described in Unice et al. (2019). Adsorption of unattached microplastic particles to suspended particulate matter can increase particle size and density. The hetero-aggregation of microplastics with suspended sediments was modelled assuming one single particle of microplastics can attach to one single particle of suspended solids (density 2650 kg/m^3 and a diameter of 0.2 mm).

The aggregation rate was calculated via these equations:

$$k_{i,j} = \alpha K_{i,j} n_j^S \qquad (4)$$

$$\frac{dn_{i,j}^A}{dt} = k_{i,j} n_i^T \qquad (5)$$

where i is the size category for microplastics, j is the size category for suspended solids, n_i^T i is the number concentration of microplastics particles in size category i, n_j^S is the number concentration of suspended solids in size category j, $n_{i,j}^A$ is the number concentration of hetero-aggregates in size category i,j, $k_{i,j}$ is the pseudo-first order rate constant for hetero-aggregate formation (s^{-1}), α is attachment efficiency (unitless); and $K_{i,j}$ is the collision frequency coefficient between suspended solids and microplastic particles (s^{-1}). The aggregation efficiency was adopted from Besseling (2018) and was set at 0.01.

The particle deposition rate is calculated using the equation below:

$$D = \alpha\, \omega_s\, C \qquad (6)$$

where D is the particle deposition rate (kg m^2 s^{-1}), α is the deposition probability (unitless between zero and 1), C is the particle concentration (kg/m^3), and ω_s is the velocity (m/s). ω_s is derived from equation 3. For cohesive fine sediments, α is a function of shear stress. This relationship has been described mathematically by Krone (1962) as $\alpha = 1 - \tau/\tau_c$ where τ is the bottom stress (Pa) and τ_c is the critical stress (Pa). Deposition occurs when the bottom shear stress (τ) is lower than the critical shear stress (τ_c). Shear stress was calculated based on the flow velocity and the Chézy coefficient as described by Unice et al, (2019):

$$\tau = \rho_w g \left(\frac{\bar{V}}{C_{CH}}\right)^2 \qquad (7)$$

where $\bar{V}$ is average flow velocity (m/s), C_{CH} is the Chezy coefficient ($m^{1/2}s^{-1}$), R is the hydraulic radius (m), S is the channel slope (m m-1), ρ_w is the density of water (kg/m^3), g is acceleration due to gravity (m/s^2), and τ is shear stress (Pa). In this study we used a Chezy coefficient of 60 $m^{1/2}s^{-1}$ for the model. The flow velocity is derived from the hydrodynamic results. In theory, a specific critical shear stress for sedimentation could be defined for each microplastic type. However, we used a default value for the critical shear stress of 0.1 Pa for all microplastics, since there is hardly any information found in literature about the influence of microplastics characteristics on this parameter. The horizontal dispersion wat set at 50 m^2/s.

3.1.3.3 *Input data*

The initial and boundary conditions of the various substances are presented in Table 3.3. The initial concentrations of all substances (microplastics and SPM) were set at 0 g/m^3. For this study we focused on two upstream boundaries, namely Lobith (Rhine River) and Eijsden (Meuse River). These boundaries are considered the predominant source of microplastics into the study area. The boundary concentrations of the different microplastic types at Lobith and Eijsden are based on measurements from Urgert (2015).

Urgert, (2015) is one of the few studies that provides information on observed microplastics concentrations (in mg/l) in the water column. Urgert (2015) monitored microplastics (0.125 – 5 mm) concentrations at Lobith/Bimmen and Eijsden from January 10 – May 26, 2014. These observations are plotted against measured river discharges in Figure 3.6 and Figure 3.7.

At Lobith and Bimmen observed concentrations vary between 0.22 to 1.05 mg/m^3 and are on average 0.56 mg/m^3 (Figure 3.6). Figure 3.6 shows no correlation between the microplastics concentration and river discharge at Lobith and Bimmen. Observed concentrations at Eijsden range from 0.03 mg/m^3 (or µg/l) to 0.38 mg/m^3 and are on average 0.14 mg/m^3 (Figure 3.7). In contrast to Lobith/Bimmen, we do observe a positive but weak correlation with the measured river discharge at this measuring location (Figure 3.7). Since the microplastics concentrations were measured over a short period (Urgert, 2015), no reliable pattern can be deduced from them. Therefore, the boundary concentrations at Lobith and Eijsden of the substances representing microplastic were set at 0.56 mg/m^3 and 0.14 mg/m^3.

The concentration of the suspended solids was set at 20 g/m^3 for both upstream boundaries. This value is based on available measurements of suspended matter (SPM) concentrations at Lobith and Eijsden (Appendix 1). The timeseries show that the concentrations SPM are highly variable over time with some extreme outliers. Therefore, we made a rough estimate of the average concentrations at both boundaries and used this concentration as a boundary condition for SPM.

Table 3.3 Input setting for water quality modelling in DELWAQ.

Substance	**Initial concentration (g/m^3)**	**Concentration at Lobith (g/m^3)**	**Concentration at Eijsden (g/m^3)**	**Concentration at other boundaries (g/m^3)**
SPM	0	20	20	0
Microplastics	0	0.00056	0.00014	0

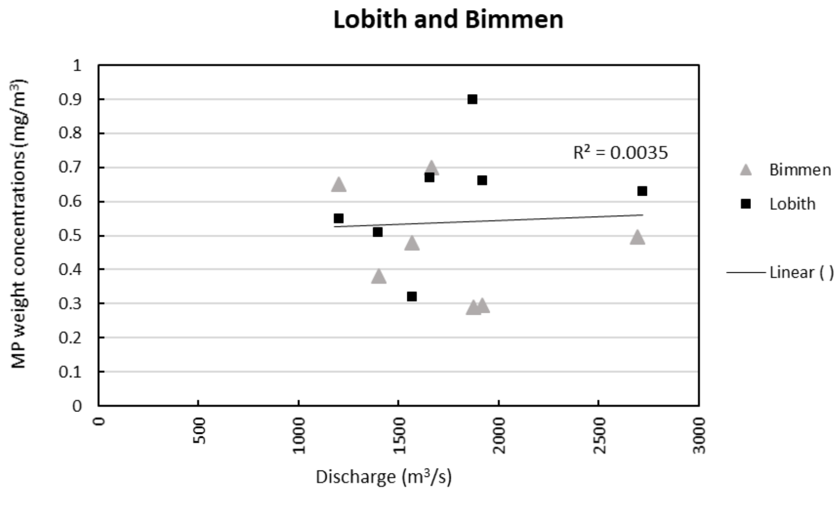


Figure 3.6 Microplastics concentration plotted against discharge measurements at Lobith and Bimmen from Urgert, (2015). Measurements were collected in 2014.

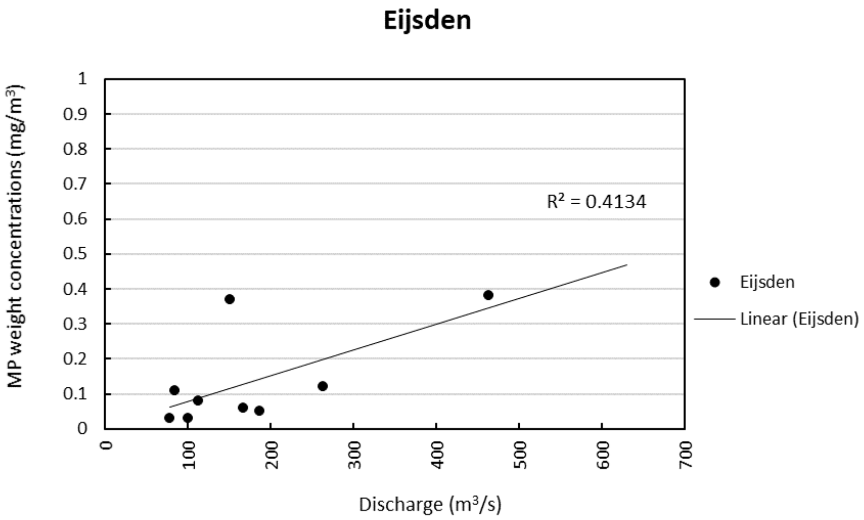


Figure 3.7 Microplastics concentration plotted against discharge measurements at Eijsden from Urgert, (2015). Measurements were collected in 2014.

In our model the microplastics enter the river as unattached microplastic, where they are exposed to several processes, as mentioned in section 3.1.2.1. These processes can lead to the formation of hetero-aggregates and deposition of microplastic on the river bed. In this study three microplastic fractions are distinguished: unattached microplastics, hetero-aggregates (one microplastic particle absorbed to one sediment particle) and deposited microplastics. Once on the bed, all microplastics particles are assumed to be hetero-aggregated with sediment, as the bed contains a large amount of sediment particles.

3.2 Interpretation of results

3.2.1 Effects of microplastics characteristics

To study the effect of microplastic characteristics on the fate of the microplastics in the Meuse and Rhine river, we modelled the transport for 24 different types of microplastics. After entering the river, the microplastics are distributed by the implemented processes over three fractions; unattached microplastics, hetero-aggregates and deposited microplastics. The annual average distribution of the modelled microplastic types is presented in Figure 3.8 and Figure 3.9 for different output locations in the study area. These overviews are created by averaging the concentrations over the calculated period 2014-2015. Figure 3.8 shows the results for three locations along the Rhine. At location 224, near Tiel, the cumulative concentrations of the suspended fractions are similar for all microplastic types, and comparable to the boundary concentration at Lobith. Deposited microplastics are not observed at this location.

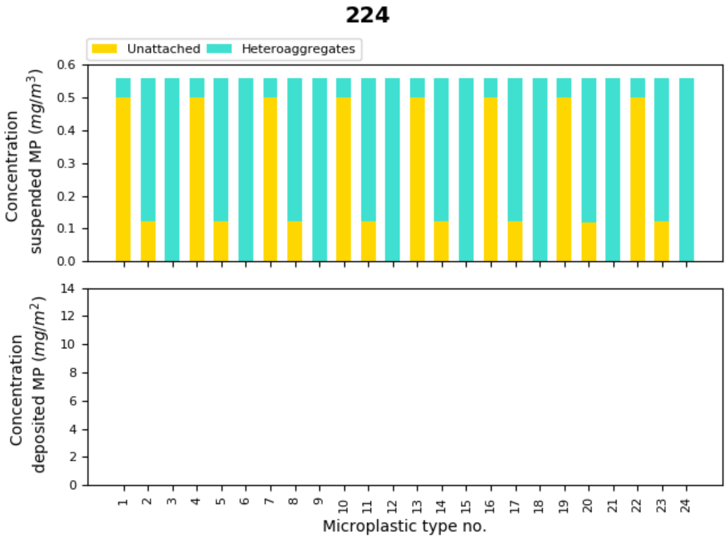
Further downstream at DI_Zwijndrecht, near Dordrecht, the distribution over the fractions shows a different pattern. In contrast to the upstream location, we see a very small decrease in the cumulative concentration of the microplastic fractions in the water column and a simultaneous increase in the annual average concentrations of deposited microplastic for some microplastic types. Especially the larger and “heavier” microplastic, such as Type 3 and 21 show a relatively high deposited concentration. At Hoek van Holland (WB_HOEKVHLRTOVR) the cumulative concentrations of the unattached and aggregated microplastics are lower than at the upstream locations. However, the difference is small. The annual average concentrations of the deposited fraction are also relatively low at this location.

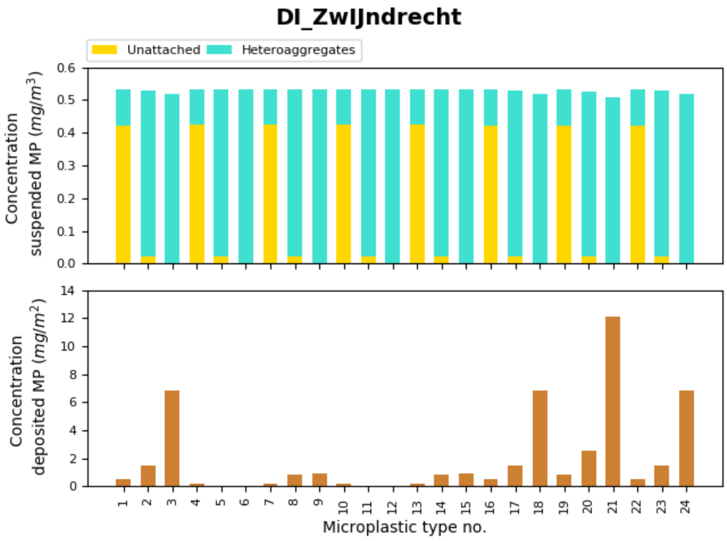
Figure 3.9 reveals the results at three locations along the Meuse. Starting upstream, at MS_008_0, the average cumulative concentrations of the microplastic fractions in the water correspond to the applied boundary concentration at Eijsden. Downstream at DO_meas_Crevecoeur (‘s Hertogenbosch), the annual average unattached and aggregated concentrations are lower. In particular the relatively large and “heavy” microplastic types (type 3, 18, 21, 24) show low concentrations compared to MS_008_0. By comparing the fraction distribution at these two locations we suggest that these low concentrations are related to retention of these microplastic types at upstream location such as MS_008_0. At DO_Evides_Scheelhk (Haringvliet), the cumulative unattached and aggregated microplastics concentrations of all microplastic types appear to be more than twice as high as the concentrations at upstream locations. In addition to these high microplastics concentrations in the water column, this location also shows relatively high concentration of deposited microplastics. The reason for these high concentrations will be discussed in section 3.2.3.

Looking at the fraction distribution of the individual microplastic types, we observe a clear general pattern in both the Rhine and Meuse rivers. In both rivers size seems to play a major role in the distribution of the plastics over the three fractions. A large part of the smallest particles, with a size of 0.3 mm, tends to remain in suspension as unattached microplastics, while the larger particles show relatively high concentration for the aggregated fraction with an increasing shift from the unattached microplastics to hetero-aggregates further downstream of the rivers. Microplastic particles with a size of 5 mm show the highest annual average concentration of hetero-aggregates. Assessing the relation between microplastic size and the unattached/hetero-aggregate ratio in the water column, revealed an evident increase in the unattached/hetero-aggregate ratio for microplastics of 1 mm and larger. Hence, these results suggest that the smaller microplastic particles are less subjected to hetero-aggregation with the suspended solids than the larger microplastics. This observation is not surprising since the chance of a microplastic particle encountering a suspended solid in the water column gets enhanced when the size of the microplastic increases.

Another important feature reflected by these results is that deposition of the microplastic particles depends on both the size of the plastic particles as well as the density. Particles heavier than water can sink to the river bed. Hetero-aggregation will enhance this process, as aggregated particles are likely to have a higher density than the plastic particles alone. Thus, once a microplastic particle is adsorbed to a suspended solid its tendency to sink and deposit on the river bed will increase. Besides being dependent on the particle characteristics, the occurrence of microplastic deposition is also related to the location. Deposition of particles in the water column can occur when the shear stress is lower than the critical value. The shear stress of the water on the river bed depends on the flow velocities. At low river discharge, flow velocities are small, leading to deposition of particles. At increasing river discharge, resuspension of deposited particles may occur. It is important to note that in these results resuspension of the microplastics is not taken into account, leading to accumulation of microplastics at the river bed and an exaggeration of the effect of the deposition process.

It should also be mentioned that inherent in the chosen model approach it is not possible to represent variations in the transport or the processes over the cross-section of the rivers. The shallow areas near the river banks may lead to sedimentation increased in comparison to the main channel, groyne fields greatly impact the flow pattern, which is now assumed to be parallel to the river axis and the flooding of floodplains at high water levels leads significant deviations from the underlying model assumptions. In principle a three-dimensional model of a small stretch of the river can be used to investigate these effects quantitatively. Unfortunately, a representative modelling does require careful consideration.


224
Unattached
Heteroaggregates
Concentration suspended MP (mg/m^3)
Concentration deposited MP (mg/m^2)
Microplastic type no.


Dl_ZwIJndrecht
Unattached
Heteroaggregates
Concentration suspended MP (mg/m^3)
Concentration deposited MP (mg/m^2)
Microplastic type no.

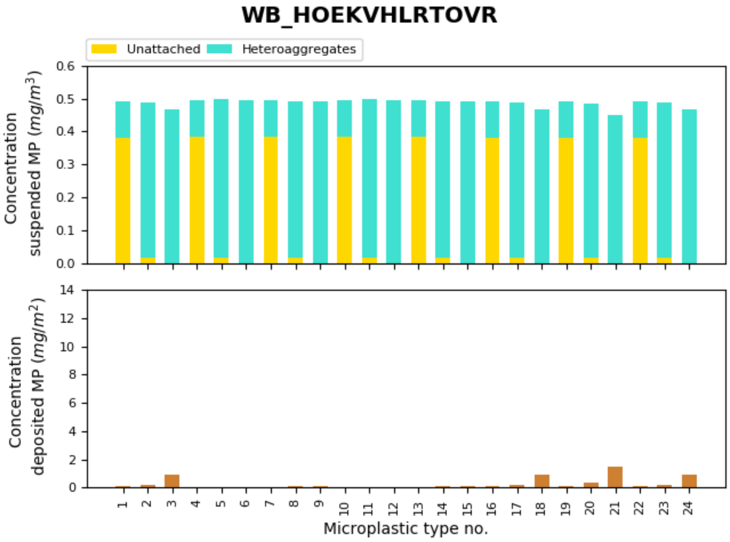
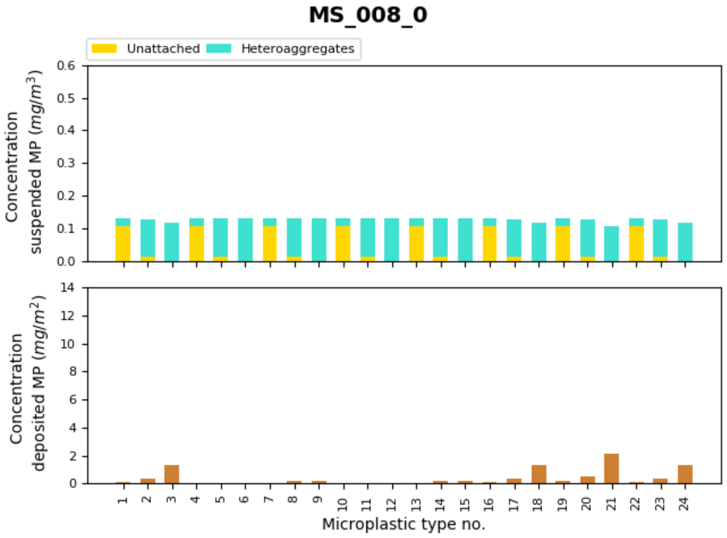
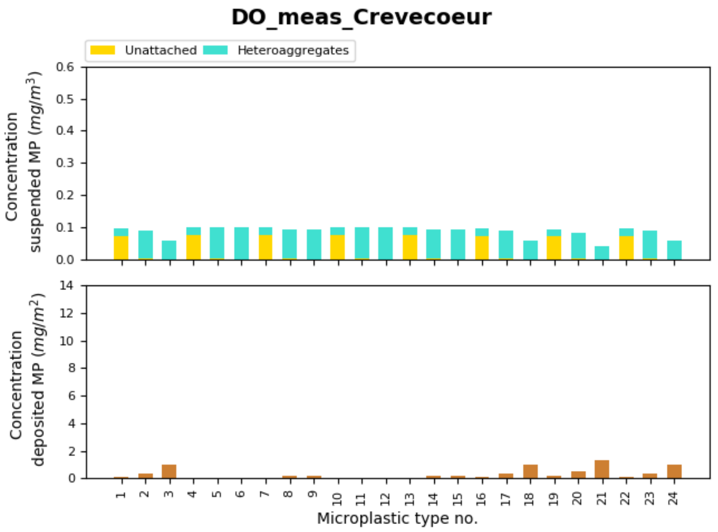


Figure 3.8 Simulated concentrations of the 24 microplastic types in the water column (upper panel) and at the riverbed (lower panel) at 3 output locations in the Rhine. The microplastics concentrations are averaged over time. For every microplastic type three fractions are shown: yellow – unattached suspended microplastic particles, blue – suspended hetero-aggregates and brown - microplastic deposited at the river bed. Note that due to the absence of resuspension the plastics accumulate at the river bed, the concentrations of accumulated plastics at the river bed will remain equal or increase over time. Characteristics of the microplastic types are listed in Table 3.2.


MS_008_0
Unattached
Heteroaggregates
Concentration suspended MP (mg/m^3)
Concentration deposited MP (mg/m^2)
Microplastic type no.


DO_meas_Crevecoeur
Unattached
Heteroaggregates
Concentration suspended MP (mg/m^3)
Concentration deposited MP (mg/m^2)
Microplastic type no.

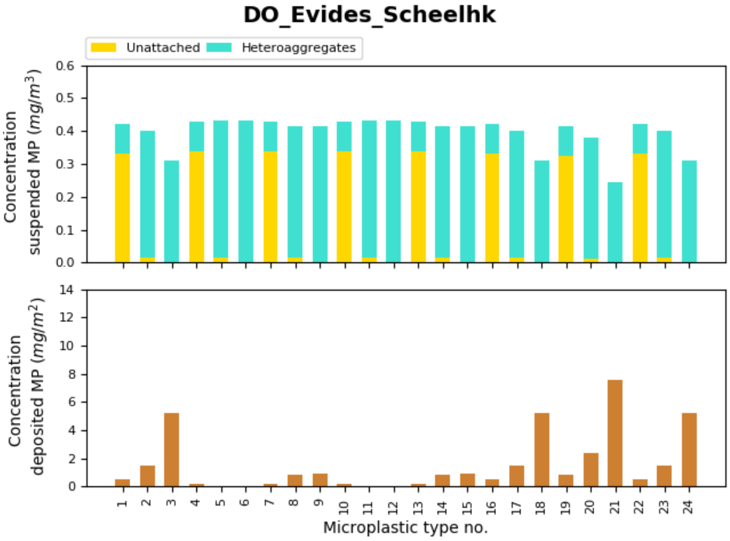


Figure 3.9 Simulated concentrations of the 24 microplastic types in the water column (upper panel) and at the riverbed (lower panel) at 3 output locations in the Meuse. The microplastics concentrations are averaged over time. For every microplastic type three fractions are shown: yellow – individual suspended microplastic particles, blue – suspended hetero-aggregates and brown - microplastic deposited at the river bed. Note that due to the absence of resuspension the plastics accumulate at the river bed, the concentrations of accumulated plastics at the river bed will remain equal or increase over time. Characteristics of the microplastic types are listed in Table 3.2.

3.2.2 Distribution over the microplastics fractions

To obtain a more detailed spatial overview of the distribution of the microplastics over the different fractions (unattached, aggregated and deposited) along the rivers, a longitudinal profile was created. We generated the profile by averaging the concentrations over the modelled period (2014 – 2015). These profiles can also highlight the areas where deposition of microplastic can occur and where microplastics are retained within the Rhine and Meuse river. This will provide more insight on the fate of microplastics in these rivers. Figure 3.10 and Figure 3.11 show the longitudinal profiles of the Rhine and Meuse for three types of microplastics, Type 1, Type 2 and Type 3. These types cover all three size classes modelled in this study and have an equal density (1050 kg/m^3) and shape. We show these three types since the other types of microplastics show similar results.

Regarding the microplastics concentrations several observations can be made from the generated profiles (Figure 3.10 and Figure 3.11, upper panels of the subplots). For the largest particles of 5 mm (Type 3) we observe that almost all unattached microplastic particles are already adsorbed to suspended particulate matter upstream to form hetero-aggregates, in both the Rhine and Meuse river. Looking at the Rhine branch we see that for the smaller plastics with a diameter of 1 mm (Type 2) part of the unattached particles also formed hetero-aggregates upstream, but the concentrations are lower compared to those of the larger plastics (Figure 3.10). Another interesting observation for Type 2 is the decrease in the unattached particles and a rise in the hetero-aggregation further downstream, clearly indicating a shift from one fraction to the other. The formation of hetero-aggregates from the smallest microplastic particles (Type 1; 0.3 mm) is much less pronounced. There appears to be a small drop in unattached microplastic in the water column due to the formation of hetero-aggregates, but overall the microplastic particles tend to remain unattached along the full length of the river.

The longitudinal profile of the Meuse reflects similar patterns (Figure 3.11). For the larger microplastic types, Type 2 (1 mm) and 3 (5 mm), we already observe relatively high concentrations of hetero-aggregates in the water column upstream. For the smallest microplastic type (Type 1; 0.3 mm) we observe relatively constant unattached microplastics concentrations in the water column along the length of the river and no increase in hetero-aggregates of this microplastic type, except at Haringvliet (DO_Evides_Scheelhk.). At this location we observe an evident increase in the concentrations of the unattached microplastics and hetero-aggregates. Furthermore, the larger particles show an increase in hetero-aggregates at this location while the concentrations of the free microplastic particles is near zero. The increase in suspended microplastics concentrations for all three types is probably related to inflowing water of the Rhine bringing in more plastics particles. The results of the fraction analysis support this hypothesis. Figure 3.2 and Figure 3.3 show both the Rhine and Meuse river influence this area.

To investigate the potential retention of microplastics along the rivers, we also added concentrations of deposited microplastics together with the calculated shear stresses to the longitudinal plots (Figure 3.10 and Figure 3.11).
In the Rhine, deposition of the microplastics shows a correspondence with the shear stress. Output locations with enhanced microplastic deposition correspond to areas with a low shear stress. Upstream, annual average shear stresses are high, and no deposition of microplastics occurs.

These high shear stresses are related to the high flow velocities that exceed the settling limit in this part of the Rhine. Further downstream there are some locations that show increased deposited microplastics concentrations, DI_Zwijndrecht, DI_Lekkerkerk and WB_HOEKVHLRTOVR. Deposition of microplastics at these output locations is probably related to the tidal influence at these areas. Here, periods with high river flow velocities alternate with periods of more or less stagnant water, when the tidal inflow is compensated by the river outflow. During these latter periods shear stresses can drop below the critical value (0.1 Pa in this study) and microplastic will get a chance to deposit at the river bed.

Resuspension is not included in our model. The amount of microplastics deposited on the river bed can therefore accumulate over time (see, however, the next section). This means that even if the periods of stagnant water are short and the amount of microplastics deposited during these periods is small. The addition of these small portions can contribute to high concentrations over time. It should be noted that the effect of deposition is exaggerated because resuspension of the microplastics was not considered. Comparing the profiles of the three different size types, we observe higher deposited concentrations for the larger microplastic particles. This also applies for the Meuse. The concentrations of the deposited microplastics of the smallest type are negligibly small. The concentrations increase when looking at the large types. These findings support our hypothesis that the distribution of the microplastics in the rivers and over the different fractions is dominantly influenced by the size of the particles. In contrast to the Rhine, in the Meuse microplastics already deposited upstream. This is likely related to the overall low annual average flow velocities along the Meuse leading to low shear stresses. Locations with high shear stresses show no deposition of microplastics.

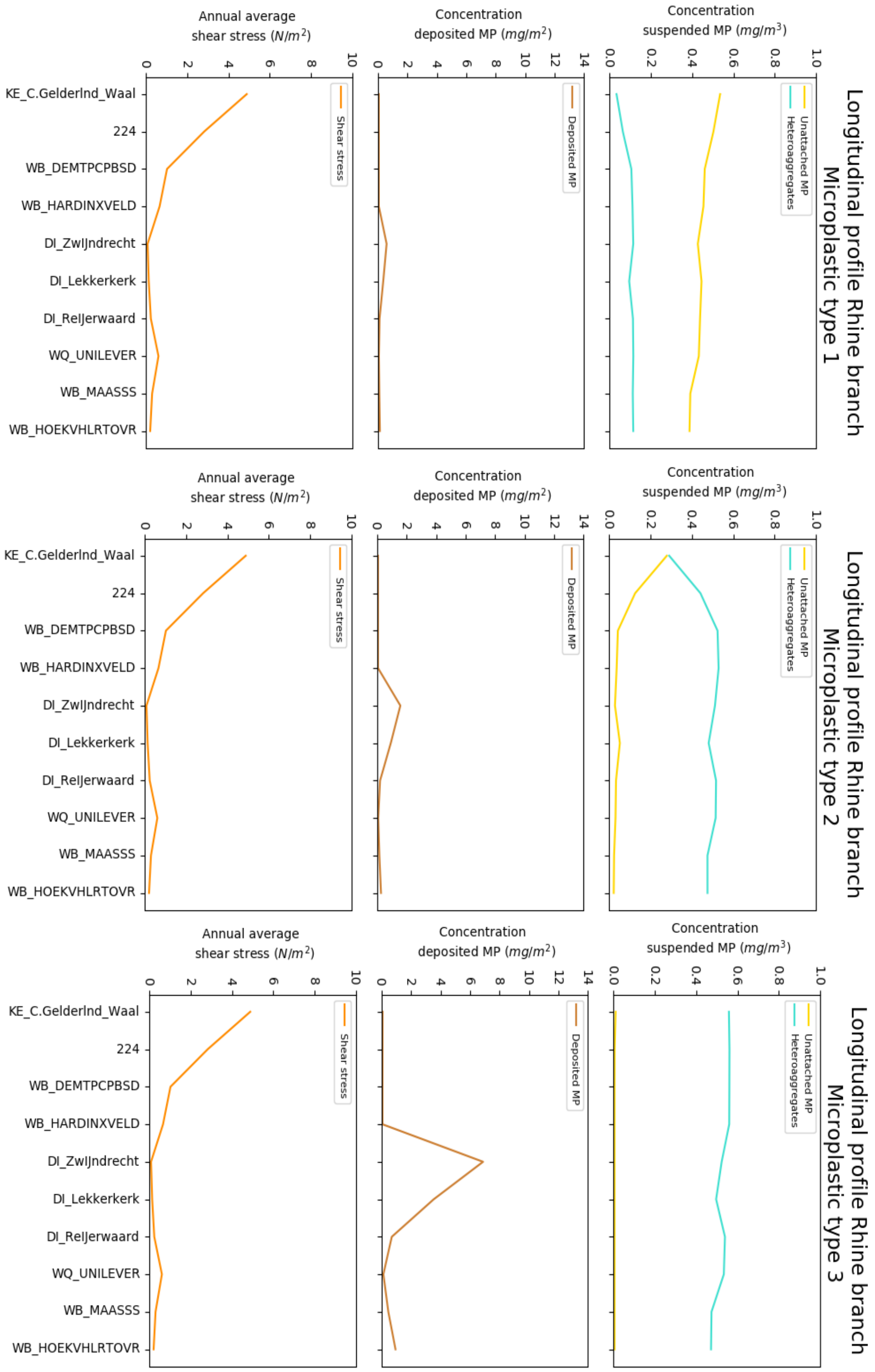


Figure 3.10 Spatial distribution of the microplastic fractions along the Rhine branch for 3 microplastic types averaged over the simulation period 2014 - 2015. The microplastics are distributed by processes over three factions: unattached microplastics, hetero-aggregates (one microplastic particle absorbed to one suspended solid) and deposited microplastics. Microplastic concentrations are given for the water column (upper panel) and sediment (middle panel). The annual average calculated shear stresses are shown in the lowest panel.

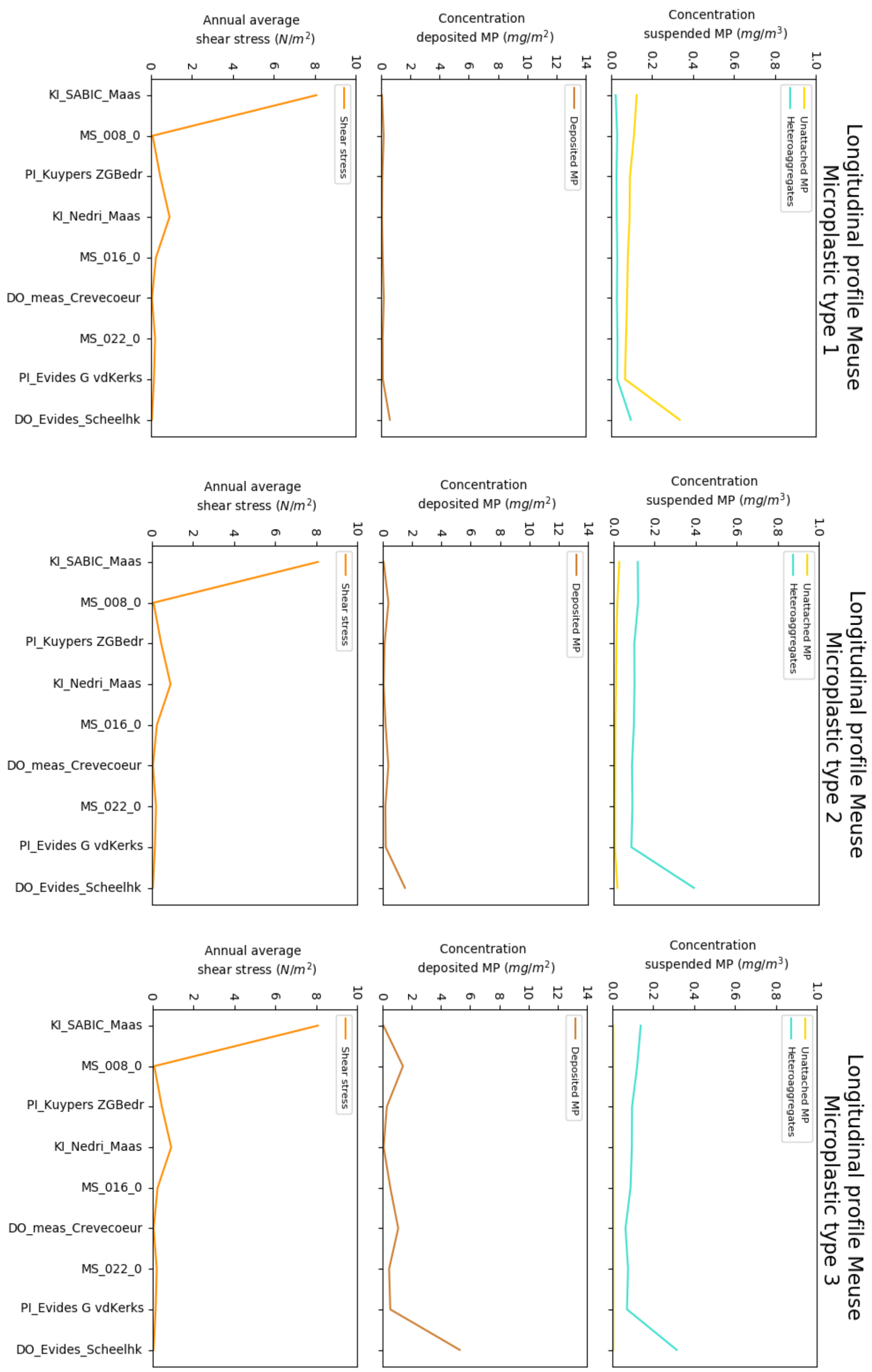


Figure 3.11 Spatial distribution of the microplastic fractions along the Meuse for 3 microplastic types averaged over the simulation period 2014 - 2015. The microplastics are distributed by processes over three factions: unattached microplastics, hetero-aggregates (one microplastic particle absorbed to one suspended solid) and deposited microplastics. Microplastic concentrations are given for the water column (upper panel) and sediment (middle panel). The annual average calculated shear stresses are shown in the lowest panel.

3.3 Estimation of the effect of resuspension

3.3.1 Introduction

Because the modelling is based on a one-dimensional hydrodynamic model, heterogeneous effects from the river banks or floodplains are not incorporated. Especially floodplains, where microplastics may settle during inundation periods, could function as an additional reservoir of microplastics: during the flooding stage microplastics that had sedimented in an earlier period may be resuspended again. It is unknown whether this does indeed occur (whether the flow velocities will be high enough) and whether this would constitute a significant source or not. For this reason, we intend to perform a sensitivity analysis dedicated to the role of floodplains.

3.3.2 Approach

The first step in the sensitivity study was to incorporate the effect of resuspension in the model set-up. In this the general approach that is taken in DELWAQ was followed: resuspension takes place on the bottom material as a whole, when the shear stress exceeds a certain threshold. The resuspension rate depends on the shear stress excess and on the availability of bottom material, but not on the amount present (zeroth-order process, the Partheniades formulation).

To get a first impression of the relative importance of resuspension the microplastics model was extended with this process with the following process parameters:

- Critical shear stress for resuspension: 0.4 Pa. This is a fairly low value, indicative for freshly deposited material. For a typical smooth bottom (Chézy coefficient of 55 $m^{1/2}/s$) resuspension would occur at a flow velocity of 0.34 m/s.
- Resuspension rate coefficient: 100 $g/m^2/day$. This is a rather difficult parameter that generally requires calibration. However, to give an impression of what this value means: The rivers Rhine and Meuse are in the order of 5 to 10 m deep. The given value means that in one day 100 g/m^2 can resuspend, if that material is available on the bottom and if the shear stress is high enough. For instance at a shear stress of 0.8 Pa (flow velocity of 0.5 m/s) the resuspension transport is 100 $g/m^2/day$. Then for a 10 m deep river the concentration can increase with 100 / 10 = 10 g/m^3 = 10 mg/l in a day. This is in the order of the fluctuations one can see in measured concentrations of suspended sediment.

3.3.3 Results and discussion

For one set of microplastics (types 2, 5 and 14) the calculation described previously has been rerun with this process included.

The calculations show that for the above parameter settings no net deposition occurs for some output locations (Figure 3.12). Several of these are in the Rotterdam Harbour, so the influence of the tide may play a role, but surprisingly the output location at Hoek van Holland in the river mouth shows a steady increase instead. Other output locations where significant resuspension occurs are located in the Meuse in Limburg.

In many other output locations the effect of resuspension is marginal (Figure 3.13). The bottom material can be seen to increase steadily there, as would be expected if there is no resuspension, only sedimentation, and for short periods only to decrease slightly. This indicates that the flow velocity is only large enough to cause resuspension during short intervals. Material that is deposited onto the bottom will in general remain there. Note that also for the simulation without resuspension a small decrease occurs in July 2014.

This is related to the water level variation and the related changes in the bed surface: at high water level the surface is relatively large, which leads to a lower concentration per m^2. For floodplains the situation may be different, as the filling and emptying can be accompanied by locally large velocities. This, however, remains to be examined.

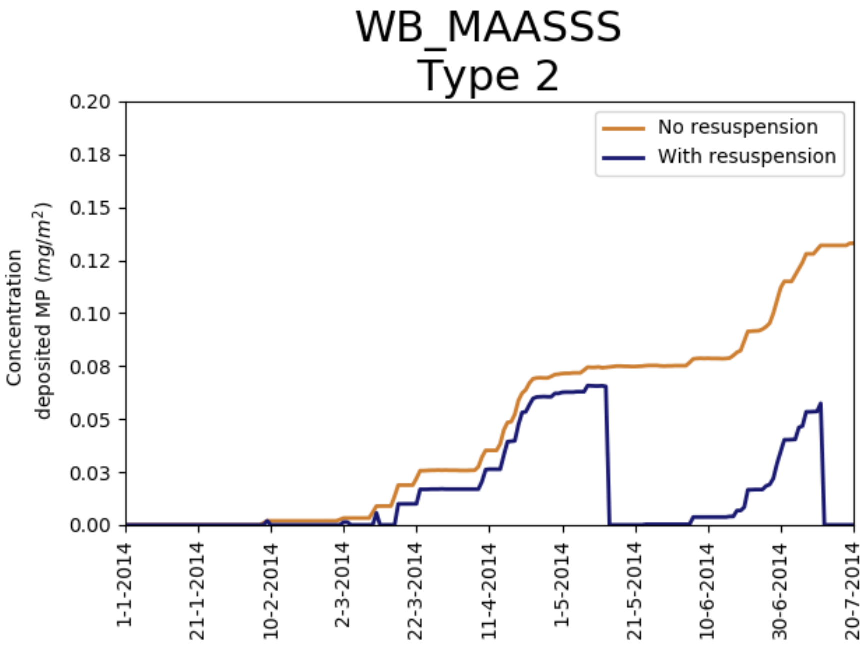


Figure 3.12 Timeseries of the deposited fraction of microplastic type 2 at a location in the Rhine, showing both the simulated concentration after implementing resuspension and without.

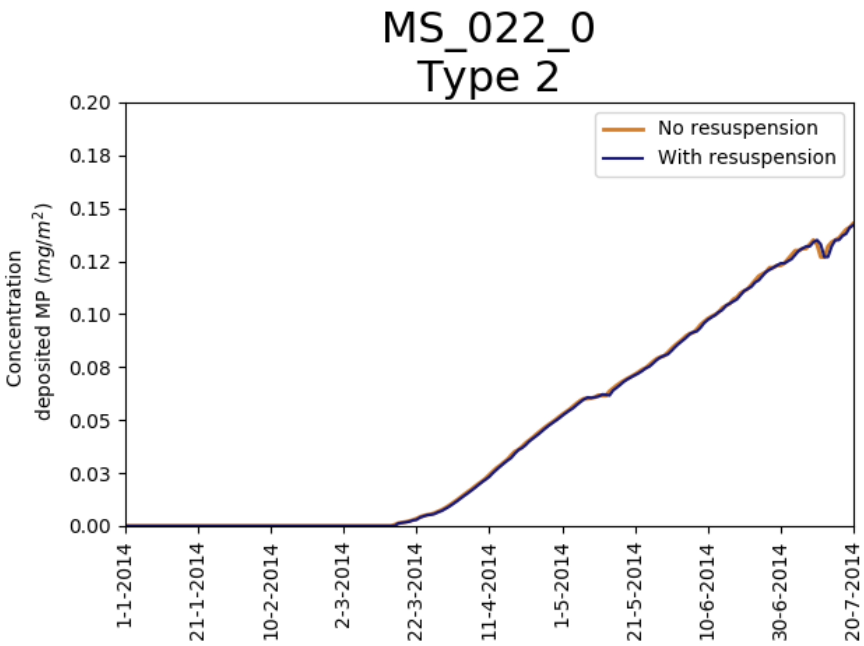


Figure 3.13 Timeseries of the deposited fraction of microplastic type 2 at a location in the Meuse, showing both the simulated concentration after implementing resuspension and without.

3.4 Verification

3.4.1 Overall results

We averaged the simulated concentrations of all 24 microplastic types, to obtain an overall view of the fate and transport of microplastics in the Rhine and Meuse. Information about the abundance of the different microplastic types in these rivers is lacking. Therefore, we assumed that the contribution of the different types to the upstream boundary condition is equal (expressed in mass, not in particle counts). The overall results are shown in Figure 3.14.

The general overview shows that in the Rhine the concentration of unattached microplastics decreases downstream, while the concentration of hetero-aggregates increases (Figure 3.14A). The cumulative concentration of the microplastics in the water column (unattached and hetero-aggregates) does not show strong variations along the river. Figure 3.14A shows that along the Rhine roughly 70% of the suspended microplastics occurs as hetero-aggregates. Furthermore, we observe that upstream concentrations of deposited microplastics are equal to zero due to relatively high bottom shear stresses related to high flow velocities. This indicates that deposition processes are not of major importance at this part of the Rhine. At downstream locations, like Zwijndrecht, deposition of microplastics does occur. Here, deposition of microplastics is likely related to the tidal dynamics.

In the Meuse, the concentration of the suspended microplastics also remains nearly constant along the length of the river, except at Haringvliet (DO_Evides_Scheelhk) (Figure 3.14B). As mentioned before, the prominent increase here is related to the influence of the Rhine in this area. In the Meuse, 70% of the microplastics in riverine water column is present as hetero-aggregates. In contrast to the Rhine, deposited microplastics can be found at multiple locations along the Meuse, both upstream and downstream.

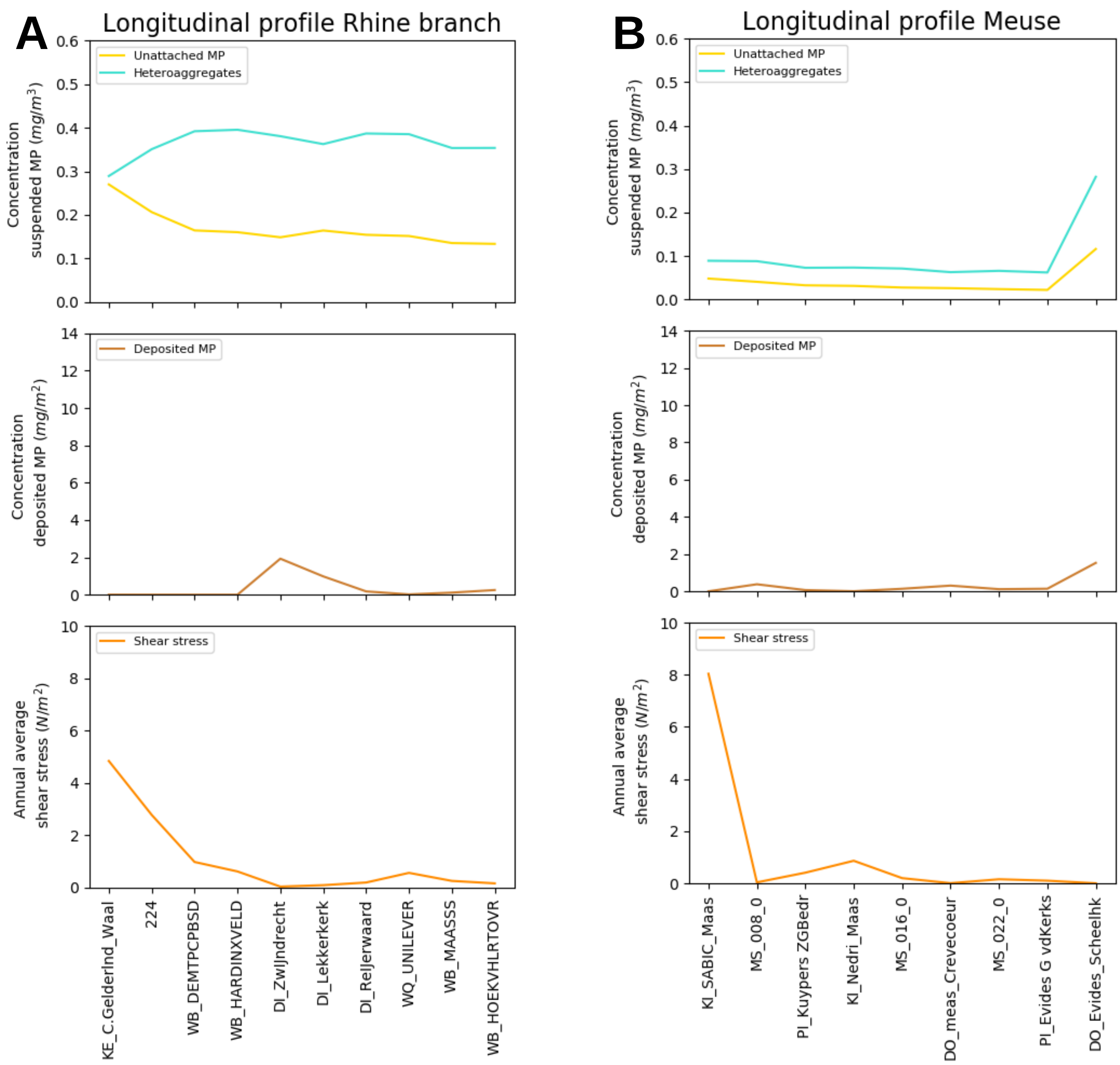


Figure 3.14 Overview of the spatial distribution of the microplastic fractions along the Meuse over the simulation period 2014 - 2015. Concentrations are calculated by averaging the concentrations of the individual microplastics types. Microplastic concentrations are given for the water column (upper panel) and sediment (middle panel). The annual average calculated shear stresses are shown in the lowest panel.

The concentrations of the different fractions shown in Figure 3.14 indicate that a large part of the microplastics that enter the Rhine and Meuse at Lobith and Eijsden get transported to the North Sea.

To investigate how many microplastics eventually get transported to the sea, we compared the inflowing transport of microplastics with the outflowing transport using the mass balances for the total model area generated in DELWAQ. Table 3.4 shows that about 66 to 90 percent of the microplastics that enter the rivers at Lobith and Eijsden are transported to the sea at the downstream boundaries. The percentage that remains in the model is either in the water column (free or aggregated; 4 – 11 percent) or deposited to the bed (0 – 30 percent). Therefore, these results suggest that deposition does not play a major role in the current 1D model.

Table 3.4 Mass balance results of modelled microplastic types for the total model area. The mass fluxes are presented in tonnes. Mass balance output is integrated by DELWAQ over the complete specified output period. We verified if all the segments were included in the mass balances to ensure that the mass balance was generated for the total model area.

Microplastic type no.	Transport In (tonnes/year)	Transport out (tonnes/year)	Deposited (tonnes/year)	In system (tonnes/year)
1	35.7	30.9	1.4	3.4
2	35.7	29.6	3.2	2.9
3	35.7	25.6	8.2	1.9
4	35.7	31.5	0.5	3.7
5	35.7	32.3	0.0	3.4
6	35.7	31.8	0.0	3.9
7	35.7	31.5	0.5	3.7
8	35.7	30.4	2.1	3.2
9	35.7	30.4	2.1	3.2
10	35.7	31.5	0.5	3.7
11	35.7	31.8	0.0	3.9
12	35.7	31.8	0.0	3.9
13	35.7	31.5	0.5	3.7
14	35.7	30.4	2.1	3.2
15	35.7	30.4	2.1	3.2
16	35.7	30.9	1.4	3.4
17	35.7	29.6	3.2	2.9
18	35.7	25.6	8.2	1.9
19	35.7	30.5	2.0	3.3
20	35.7	28.5	4.7	2.6
21	35.7	23.4	10.7	1.6
22	35.7	30.9	1.4	3.4
23	35.7	29.6	3.2	2.9
24	35.7	25.6	8.2	1.9

3.4.2 Comparison with other studies

Although we expect that the model underpredicts microplastics concentration and transport, we compare the results with observations to verify that they are in the same order. However, published and publicly available observations within the Netherlands are scarce. Furthermore, our model is based on various assumptions of which the consequences are not well known, making verification of the model difficult.

Urgert (2015) is one of the few studies that have reported on observations of microplastics in the Rhine and Meuse. However, we have already used these observations as boundary conditions for our model and could therefore not use these observations for our model verification. Other recent observations are published by Mani et al. (2015). Mani et al. (2015) have determined concentrations of microplastics at 11 sampling sites along the German and Dutch Rhine during June to July 2014.

They found variable concentrations and different types of microplastics, in the size range 300 µm to 5 mm, along and across the river, with a prominent drop in concentrations in the Dutch part of the Rhine, at Rotterdam and Zuilichem. Observed concentrations at these sites are listed in Table 3.5. Mani et al. (2015) measured on average 5.7 microplastics particles per m^3 at Zuilichem and 1.5 microplastic particles per m^3 at Rotterdam.

We compared these observed concentrations to our simulated concentrations to verify our results. However, the microplastics concentrations in their study are reported in particles per m^3 and no sizes or densities were reported. In order to compare our results to the observations of Mani et al. (2015), we converted the modelled concentration in mg/m^3 to number of microplastics particles per m^3. We estimated the number of microplastic particles following equation 1, using the modelled concentration and mass of the microplastic particles.

$$number\ of\ microplastic\ particles\ m^{-3} = Conc_{modelled}/m \qquad 1)$$

$Conc_{modelled}$ is the cumulative concentration of the microplastic fractions in the water column (unattached, hetero-aggregates). The mass of the particles was estimated based on the volume and density of the microplastic particles as follows:

$$V = \frac{4}{3}\pi r^3 \qquad 2)$$
$$m = V * \rho \qquad 3)$$

Table 3.5 Comparison of simulated results (in mg/m³ and number of microplastic particles (MP)/ m³) with observations data ((size range 0.3 mm to 5 mm; number of MP/ m³) at Rotterdam and Zuilichem. Observation data is derived from Mani et al. (2015).

Sampling site	**Sobek location**	**Observed concentration (number of MP/ m³)** *Mani et al. (2015)*	**Simulated concentration (number of MP/ m³)** *This study*	**Simulated concentration (mg/ m³)** *This study*
Zuilichem	WB_DEMTPCPBSD	5.7	3.5	0.56
Rotterdam	DI_Lekkerkerk	1.5	3.4	0.53

Since details on size and type of the collected microplastics are not provided by Mani et al. (2015), we compared the observations with the average concentration of all 24 modelled microplastics types at these sites. We used an average radius and density of 1.05 mm and 1080 kg/m^3, respectively. The resulting estimated and original concentrations are in listed Table 3.5. Based on equation 1 to 3, the simulated concentrations are approximately 3.5 microplastic particles m^{-3} at Zuilichem and 3.4 microplastic particles m^{-3} at Rotterdam. These values are in the same order as the concentrations observed by Mani et al. (2015). However, contrary to our results, the results of Mani et al. (2015) show lower concentrations at Rotterdam compared to Zuilichem. In their study, Mani et al. (2015) show a prominent difference in concentration across the river at Zuilichem. At the right bank, concentrations are comparable to concentrations found at Rotterdam, while measured concentrations at the left bank are more than four times higher. Mani et al. (2015) argued that concentrations at Rotterdam and the right bank of Zuilichem deviate from the measured concentrations upstream, along the German part of the Rhine. Mani et al. (2015) suggested that the decreasing microplastics concentrations at Rotterdam and Zuilichem (right bank) might be related to a retention process.

However, results of a more recent study on riverbed microplastics concentrations in the Rhine contradict this hypothesis (Mani et al. 2019). They stated that riverbed concentrations at upstream sites are not lower than the estuary Rhine riverbed concentrations. Furthermore, concentrations measured by Urgert (2015), which we used as boundary concentrations, are in the same order as concentrations measured by Mani et al. (2015) in the Dutch part of the Rhine. The high observed concentrations at Zuilichem might be related to a microplastics emission source to the Rhine at the left bank, leading to the high observed concentrations in this part of the river. Such waste load sources are not included in our model. Hence, this could be an explanation for the deviation between the observed and simulated concentrations at Zuilichem. Despite this deviation, comparison with the observations indicate that the model can roughly approximate the observed concentrations. However, it is important to emphasize that this comparison is solely based on data of one observation study. The lack of data from observations currently limits further verification of the model. More data is required to validate the credibility of the model results.

To put our results in more perspective we compared our results to other modelling studies, such as that by Nizzetto et al. (2016). They modelled the transport of microplastics in the Thames River Catchment and observed that size (in contrast to density) appears to be a more sensitive parameter influencing microplastic transport dynamics. The authors hypothesize that in the river stream microplastics <0.2 mm are generally less well retained, regardless of their density. Larger microplastics with densities marginally higher than water can instead be retained in the sediment. However, high flow periods can remobilize this pool. Sediments of river sections experiencing low flow velocities are likely hotspots for deposition of microplastics. Nizzetto et al. (2016) further emphasize the importance of further development of new monitoring studies that could provide data on the distribution and characteristics of microplastics in sediment and stream water focusing on river sections with contrasting hydrological characteristics. The findings of Nizzetto et al. (2016) are in accordance with our results. They support the hypothesis that size, as well as local hydrodynamics play an important role on the fate of microplastic particles in rivers.

In total, about 35.7 tonnes of microplastics are imported in the model based on the constant concentration and the discharge timeseries. About 66-90 percent of this is transported to the sea at the downstream boundaries, which is 23.4-32.3 tonnes. Van der Wal et al. (2015) estimate that in between 20 to 31 tonnes of plastic (microplastics and plastics with a length in between 5-25 mm combined) are transported via the Rhine river to the North Sea annually. The contribution by weight is larger for plastics with a size in between 5 and 25 mm (van der Wal et al., 2015). Hence, in our study with the concentration of microplastics at the border based on available observations, the model overestimates the transport of microplastics.

3.5 Discussion

3.5.1 Effect of neglecting microplastic sources within the Netherlands

In our model we neglected the contribution of microplastics derived from sources within the Netherlands. Sources of microplastics into fresh water include; wastewater effluents, mismanaged plastics wastes and run-off from land-based sources. Land-based sources of microplastics can for instance originate from road surface run-off of tyre wear debris. Verschoor et al. (2016) estimated that, in the Netherlands, 1,800 tonnes microplastics resulting from tyre and road wear enter the water each year. Microplastic fibres derived from textiles due to washing are also recognized as a source of microplastics into the aquatic environment.

Not considering important point and diffuse sources within the Netherlands as tire and road wear particles and effluent of WWTP leads to an underestimation of the microplastics concentration in the modelled rivers. Our current model solely gives an indication which part of the microplastics that enter the Netherlands through the large rivers is transported to sea.

Following the approach of Siegfried et al. (2017) we made a rough estimate of the amount of microplastics entering the aquatic environment via WWTP effluent. This estimation is based on the estimated amount of microplastics produced per person and the count of people connected to the centralized sewage system combined with the sewage treatment efficiency. In the Netherlands, WWTPs remove 95 percent of microplastics from the water (Siegfried et al. 2017). Hence, 5 percent of the microplastics entering a WWTP are discharged to surface waters where a part gets deposited on the riverbed along the way to the sea. Based on several studies, Siegfried et al. (2017) estimated the annual microplastic waste generation per person at 0.39 kilograms per person per year of which half originates from tyre and road wear particles (TRWP). TRWP occur as point-sources as well as diffuse sources (e.g. particles released from road surface). For this estimation, TRWP from diffuse sources are neglected, In the Netherlands, nearly all households are connected to a sewage system. Based on these values we estimate a yearly microplastics flux of 329 tonnes to the surface waters in the Netherlands via WWTP effluent. This is about ten times the weight of microplastics transported into the Netherlands at Lobith and Eijsden.

3.5.2 Optimizing the roadmap

The modelling software used in this study uses a so-called Eulerian approach where the presence of microplastics is expressed as a concentration in g/m^3 and the particulate character of the plastic particles is implicitly captured in processes. The particles may sink and settle to the bottom or may float to the surface, but this is formulated in terms of processes working on a substance homogeneously distributed over the water volume. This continuum approach is generally useful, but it has several drawbacks. First, the numerical solution method associated with this approach uses a single value as the concentration of plastic particles in a grid cell. The resolution is therefore limited by the size of the grid cells. This is not a significant limitation, if the particles are spread over a large enough area. It may, however, hinder the modelling in the vicinity of the sources, such as the outflows of wastewater treatment plants. Secondly, within this approach it is difficult to consider aspects of aging, influence of the wind on the spreading of floating plastic particles and entanglement in vegetation.

Such aspects, some of which are of particular importance for larger plastic items, are better covered by a Lagrangian approach that considers the plastics' particulate character. Particle tracking, as implemented in the Delft3D-PART model, is a more suitable approach, then. It is capable of tracking particles within the grid cells, so that even with a relatively coarse model a detailed picture of the spreading can be obtained. Each particle is tracked individually throughout the calculation, so that its age is known – the moment in the calculation minus the time it was released. In Delft3D-PART the wind is included as influencing the transport of particles near the surface. Entanglement could be modelled fairly simply via a probability that a particle gets "stuck" or "unstuck" (if stuck, it will not move until it gets unstuck) or in a more sophisticated way by considering the size of the particle and the characteristics of the vegetation. Other processes that affect the particulate character can be implemented in a similar way.

One point to note: a “particle” as tracked via Delft3D-PART (or for that matter any particle tracking model) does not necessarily mean an individual (plastic) particle, but rather a typical particle. (In that respect the approach is akin to the continuum approach.)

To put it concisely:

- A continuum approach, such as used by DELWAQ, is useful for small particles where the transport is due to water flow or gravity (sinking or floating).
- A particle tracking approach, such as used by Delft3D-PART, is useful when the mechanisms of transport involve the particulate character, such as with larger items.

4 Conclusions and recommendations

4.1 Conclusions

1) This study presents the first model results on the fate and transport of microplastics in the Rhine and Meuse branches.
   a) Microplastics were released in a depth and width averaged model of Dutch river and canal branches at Lobith in the Rhine and at Eijsden in de Meuse.
   b) Processes of advection, deposition hetero-aggregation of microplastics with sediment are included.
   c) We found that in both rivers the microplastic particles of 0.3 mm show much less hetero-aggregation than the microplastics of 1 and 5 mm.
   d) From the released microplastics about 10-34 percent remain in the model after one year of simulation. This suggests that the role of retention due to deposition in the current 1D model is not major.
2) Resuspension was not included in this model, because required process parameters concerning microplastics are unknown.
   a) The absence of resuspension could lead to an underprediction of the microplastics concentration in the water column.
   b) A sensitivity study on the effect of resuspension showed that its effect is smaller than expected. Periods when shear stress exceeds the critical value in accumulation areas are short. This suggests that the role of resuspension in the Rhine and the Meuse is minor.
3) Comparison of the modelling results with in situ observations in the Rhine showed that:
   a) the modelled concentrations are roughly in the same order as the measured concentrations (Mani et al., 2015).
   b) The annual simulated microplastics transport is higher than estimated by van der Wal et al. (2015).
   c) Because the sources of microplastics are not included in the model, it was expected that the simulation results would be lower than the observations, which is not the case.
   d) The lack of annual observation time series limits further verification of the model.
4) Sources of microplastics within the Netherlands are neglected in our current model.
   a) We estimated that the input of microplastics from point sources can be as high as ten times the weight of the microplastics transported into the Netherlands.
   b) The estimate is based on several assumptions. Therefore, the exact contribution of microplastics from point sources remains uncertain.

4.2 Recommendations

For future work we recommend:

1 Add the microplastics emissions in the Netherlands to the model.
2 Include resuspension in the model, although its effect seems limited.
3 Examine the effect of deposition and resuspension of microplastics in floodplains on the transport of microplastics along the deeper part of the river. For this the approach of following particles (such as with Delft3D-PART), in addition to modelling concentrations (DELWAQ), will be beneficial.

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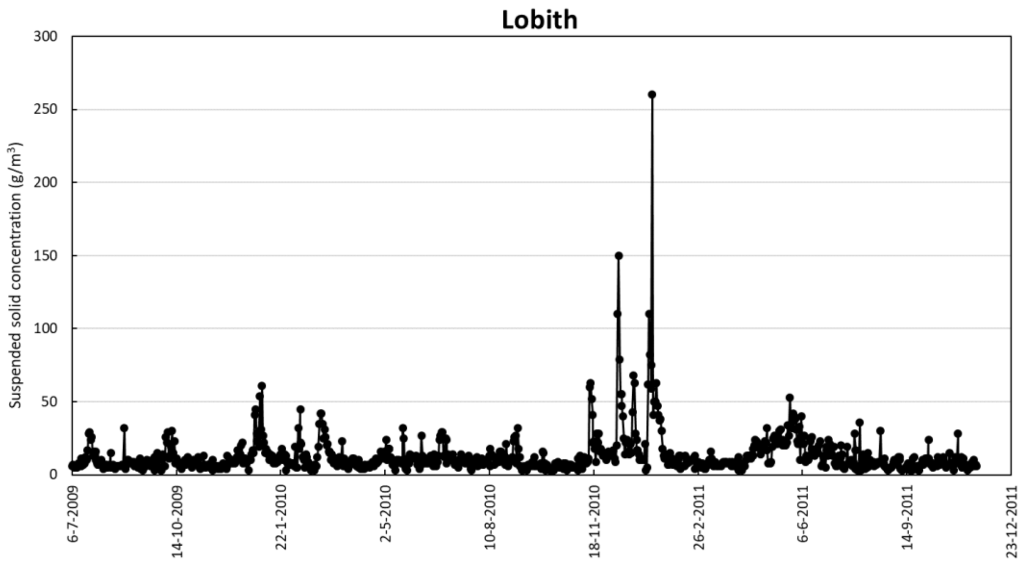
A Appendix 1: suspended sediment concentration observations



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