MODELLING THE MICROPLASTIC FLUX FROM WASTEWATER-TREATMENT PLANTS TO THE DUTCH WADDEN SEA

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Environmental Systems Analysis

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Summary

The possible threat of microplastics to biota is of increasing scientific concern, as microplastics are ingested by a wide variety of marine organisms and potentially pollute the environment. Additionally, this microplastic threat is poorly studied, especially when compared to macroplastics.

My study aims to quantify the flow of microplastics to reduce the knowledge gap. I focus on the microplastics that enter Dutch river segments through wastewater-treatment plants and eventually emerge in the Dutch Wadden Sea. River segments are a part of a river. With every place where two or more rivers come together, a new river segment arises. First, the sources and relevant processes are identified, and afterwards the flow is quantified and modelled.

The identification of the sources and processes, and the modelling is done through a literature review. The fraction of microplastics that enters and is removed in a wastewater-treatment plant, depends on source properties and the plant's treatment system.

A new model quantified the flow of microplastics. This model uses a generic water-quality model developed from Water Framework Directive Explorer with set parameters to identify the travel time in Dutch river segments. Travel time is then combined with the settling-coefficient K and an initial flow from different plants to determine the microplastic mass flux and concentrations in each river segment. The highest mass flux stem from cosmetics, followed by car tires, and cleaning agents. Paint and coatings have the lowest mass flux. Stavoren, IJsselmeer, Markermeer, Randmeren and Flevoland are areas with high concentrations and flux to the Dutch Wadden Sea.

The results of this study are uncertain. Four sources were selected, based on the available data and their contribution to microplastics. However, the wastewater of washing machines also contributed much. This source is ignored due to lack of data. The total mass flux is therefore probably higher than my estimates. This model was likely the best choice for this study and estimate the mass flux and concentration of microplastics. Other literature showed, however, a much lower mass flux. This discrepancy cannot be explained. Although the model used a decay rate, I used a settling rate. The properties of the microscopic particles had to be estimated and these estimations are probably inaccurate. Despite these shortcomings this model suited best with the currently available data to quantify the microplastics in Dutch rivers and the Wadden Sea. This study therefore likely achieved the best possible results. The study's results are a first major step to quantify the flow of microplastics in Dutch rivers and the Wadden Sea. These results reduce the knowledge gap on microplastics. This report serves as a basis for further studies on microplastic fluxes in the Netherlands.

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1. Introduction

1.1 Background

Large-scale production of plastics began in the 1950s. Currently over 300 million metric tons is manufactured globally each year. Between 1960 and 2000 the world production of plastic resins increased 25-fold, while the recovery of the material remains below five percent. Between 60 and 80 percent of the marine litter is now plastic (Moore, 2008). Despite the benefits of plastic, which are durability, low-cost and widespread use, there is a down-side of plastics as well. First, it is increasingly used to manufacture single-use products where after the plastic is thrown away. Secondly plastics are commonly present in seas and marine organisms. Lastly the durability of plastics makes it highly resistant to degradation (Andrady, 2011; Cole et al., 2011; Shim & Thomposon, 2015). This lead to an increasingly threat to marine life, ecosystems and potentially human health (SEAS, 2017). Microplastics are easier to ingest by marine biota, harder to remove than macro plastics and under-studied compared to macro plastics. Those are the reasons that this report focuses on microplastics only.

This study defined microplastics as particles smaller than 5 mm in diameter (Arthur et al., 2009; Auta et al., 2017; McCormick et al., 2014; National Oceanic and Atmospheric Administration, 2014; Wright et al., 2013).

Microplastics may be ingested by a range of organisms including plankton, fish, birds and even mammals. This results in accumulation throughout the food web (Wright et al., 2013). Beside the danger of ingestion, plastic contains multiple chemical additives (Dekiff et al., 2014) and absorbs organic contaminants from their surroundings (Bakir et al., 2012). These compounds can transfer to organisms through ingestion so microplastics act as a vector for other organic pollutants (Zarfl & Matthies, 2010). It is currently unclear whether the ingestion of non-polluted microplastics have any significant adverse health effects on biota (Cole et al., 2011). Microplastics have the potential to deliver concentrated POPs, mainly those picked up from sea water, to organisms. According to Andrady (2011) there is an urgent need to assess the future impact of increasing microplastic levels on the world's oceans.

Despite the available data about marine microplastics, research about microplastics in freshwater systems is still lacking (Wagner et al., 2014). Data about microplastics in the marine environment is for example available for the coast of China, the Indonesian Archipalego, between Hawaii and California (Sherman & van Sebille, 2016), the North Sea (Dubaish & Liebezeit, 2013) and the Pacific Ocean (Desforges et al., 2014). One of the seas where data is currently lacking is the Wadden Sea. Additionally, the quantity of microplastics entering the sea from land is currently unknown (Jambeck et al., 2015). This study tried to quantify both the microplastics in Dutch freshwater systems and the part in the Netherlands that goes to the Dutch Wadden Sea (DWS). In addition, the DWS must deal with ecological vulnerability (Lahr et al., 2007), which is the degree to which the sea is (and remains) disturbed by a certain stress factor (Faber et al., 2004; Offringa & Lahr, 2007).

1.2 Focus of this study

This study focuses on microplastics from point sources. Four sources are selected: car tires, cosmetics, cleaning agents, and paint and coatings. These sources are selected based on the available data and their contribution to the total microplastic flux. These sources, especially car tires, can enter the sea through different pathways. My study specified the pathways of sources that pass wastewater-treatment plants (WWTPs) and enter rivers and eventually seas.

The lack of data about the Wadden Sea was the main reason to focus on the Wadden Sea. To make it specific and measurable, I chose to focus only on the Dutch part of the Wadden Sea.

The Netherlands is partly connected to the North Sea as well, but there is much less data available on microplastics in the DWS. To calculate the flux and concentration of microplastics from WWTPs first all 355 WWTPs in the Netherlands were used. Afterwards 32 WWTPs were selected since those led to the DWS. This method helped to compare the microplastic of the Netherlands to the mass flux that entered the DWS.

The combination of a consumption-based and hydrological model will form the basis in this study. Data is available about the average number of microplastics in the sediment in the DWS, but comprehensive data is still lacking. The literature showed that the concentration of microplastics in the DWS was higher than in the North Sea (Leslie et al., 2013). This study takes the first step in providing insight in sources, flows and concentrations of microplastics in the DWS.

1.3 Aim of this study

This study aims to quantify the flow of primary microplastics that enter the DWS via WWTPs to reduce the current knowledge gap. To quantify the flow six research questions (RQs) are answered:

RQ1: What are the relevant sources of microplastics that enter WWTPs via sewage systems?

RQ2: Which processes are used in WWTPs to remove particles from wastewater in the Netherlands?

RQ3: What happens to the particles that enter the rivers after treatment?

RQ4: Which models are available to model the flow of microplastics in rivers and seas?

RQ5: Which model is most suitable to the aim of this study?

RQ6: What are the results of the model?

1.4 Reading guide

In chapter 2 the methodology used during this study is explained. Sources and pathways were identified by a literature review, as well as a model. The most important sources, processes in the chosen pathway and the model equations can be found here. The application of this methodology led to some results. Chapter 3 shows that the highest fluxes were found near IJsselmeer, Markermeer and Randmeren. You can also find here where the largest concentrations were found. These results are compared to literature which showed that these results are reliable. In chapter 4 the results were put in perspective by a discussion. Here was stated that despite the uncertainties at this moment no better outcome could have been reached. The conclusion in chapter 5 shows that this study is a major step in quantifying the flow of microplastics in the Netherlands and reducing the knowledge gap. This study can suit as a basis for other studies about microplastics in Dutch rivers and the Wadden Sea.

2. Methodology

A combination of qualitative and quantitative research was used in this study. I chose for a literature review to identify the current gap between available and missing knowledge and data. Relevant microplastic sources were identified and the processes in WWTPs, and a model was identified. Data from literature was used as input for this model. The quantitative results comprise the outcome of the model.

2.1 Literature study

At the start of the literature study key words were chosen. This helped to look for specific data. Google Scholar has been used as searching machine to find the literature. Table 1 shows the key words for each subject.

Table 1 Key words used per subject to conduct the literature study

Subject	Key Words
Sources	Sources Microplastic DWS
	Sources Microplastic Wadden Sea
	Sources Microplastic Water Netherlands
Pathways	Fate and transport of microplastics
	Fate and transport of microplastic in DWS
	Microplastics from WWTPs to aquatic environment
Models	Microplastic ocean models
	Models Wadden Sea
	Modelling microplastic marine environment

2.1.1 Four sources to focus on

Four sources have been found in literature as relevant for this study: cosmetics, cleaning agents, paint and coatings, and wear and tear of car tires (van Wezel et al., 2016; Verschoor et al., 2016). The sources are selected based on their contribution to the microplastic load and the available data to calculate with.

Cosmetics

Microbeads is a term used by industry to refer to microplastic particles (Napper et al., 2015). Polyethylene represent 93% of these microbeads in the size range of 450 to 800 µm. (Gouin et al., 2015). Other types of microbeads are for example polypropylene, polystyrene and polymethylmethacrylate (Leslie, 2014).

Cleaning agents

In cleaning agents microplastics are used as an abrasive (Leslie et al., 2012). The use of cleaning products increased by 10% between 2008 and 2013 (Nederlandse Vereniging van Zeepfabrikanten, 2013).

Paint and coatings

Paints are likely to contribute to microplastic emissions (van Wezel et al., 2016). For example, microplastics can end up in wastewater after cleaning used equipment.

Car tires

Car tires release plastic particles through mechanical abrasion. In contrast to the early days, when only natural rubber was used, currently a mix between natural and synthetic rubber is used for car tires. Synthetic rubbers consist of polymers made from petroleum, sulphur (1% to 4%), zinc oxide (1%) and carbon black (22% to 40%) (Kole et al., 2017).

2.1.2 A quantification of the two groups in the Netherlands

Now that the different sources are defined, the emissions from each source will be quantified. This quantification will be the first input for the model. Emissions from cosmetics, cleaning agents, and paint and coatings are quantified together as consumer products. Emissions from car tires are calculated separately.

Consumer products

The consumer products are quantified using average data from literature (van Wezel et al., 2016). Table 2 shows the predicted environmental concentrations.

Table 2 Predicted environmental concentrations of microplastics in sewage-treatment plant effluents (Reprinted from Wezel, Caris & Kools 2016)

	Predicted environmental concentrations
General	
- *Removal during WWTP treatment (1 - R_{stp})*	0.10
- *Amount of wastewater produced (WW_{inh}) (L capita^{-1} d^{-1})*	116.90
Cosmetics (emission factor 1)	
- *Concentration of microplastic (mp) in product (C_{mp}) (g g^{-1})*	0.04
- *Daily usage of product (Up_{rod}) (g capita $^{-1}$)*	4.80
Cleaning agents (emission factor 1)	
- *C_{mp} (g g^{-1})*	0.25
- *U_{prod} (g capita $^{-1}$)*	0.68
Paint and coatings (emission factor 0.005)	
- *C_{mp} (g g^{-1})*	0.20
- *U_{prod} (g capita $^{-1}$)*	5.70

Table 3 shows the microplastic concentration in kilograms per year for each group in the average scenario. The emissions are calculated with the following equation:

$$E_X = \frac{(U_{prod} \cdot C_{mp})}{1000} \cdot Inhabitants\ 2015 \cdot 365 \cdot emission\ factor \qquad \text{Eq (1)}$$

E is the total emission from source x (kg/y);

U_{prod} is the daily usage of product (g/capita^{-1}); and

C_{mp} is the concentration of microplastic in product (g/g^{-1}).

Table 3 Microplastic emissions of the three consumer products in kg/y in 2016

	Emissions (tons/year)
Cosmetics	1,246
Cleaning agents	1,051
Paints and coatings	35

Car tires

Another source that is recently recognized as responsible for microplastic particles is the by-product of city dust and road wear, such as asphalt and car tires. (Kole et al., 2015; Sundt et al., 2014). Table 4 shows the origin of wear dust from car tires in the Netherlands.

Table 4 Origin of wear dust from car tires (Kole, Löhr, & Ragas, 2015)

Category	***Emission factor (kg/10^6 km)***	***Amount of km driven (10^6 km)***			***Emissions (tons/y)***
		In town	Out of town	Highway	
Passenger car	100	20,876	36,472	45,349	6,263
Truck	600	406	525	1,434	659
Tractor	495	274	867	3,418	762
Other traffic					1,084
Total					8,768

Comparing consumer products and car tires

Figure 1 shows the percentage of how much each source contributes to the total emissions.

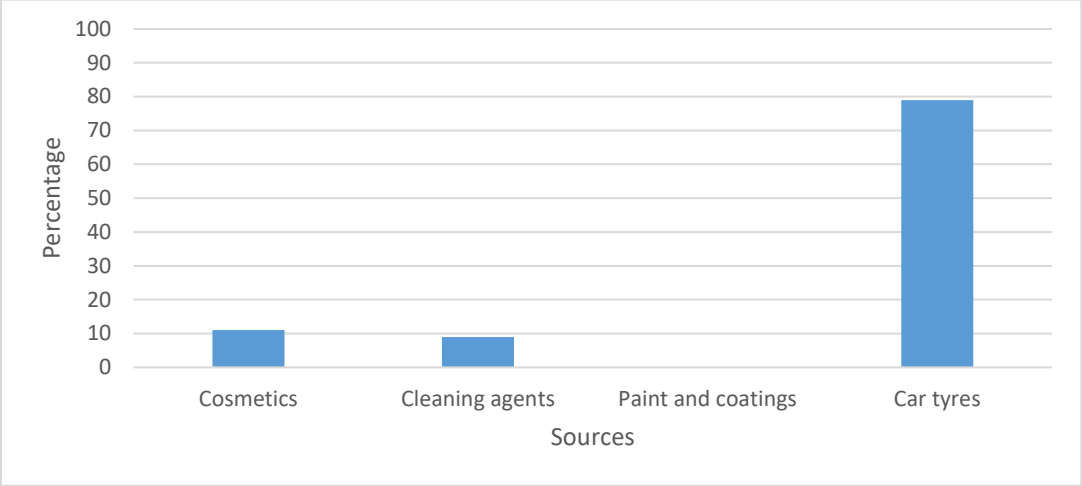

Figure 1 Percentage of contribution of each source to the total emissions

Figure 1 clearly shows that car tires emit the largest emissions of microplastics. Cosmetics and cleaning agents are almost the same whereas the contribution of paint and coatings seems negligible.

2.1.3 Pathway of microplastics

The pathway that has been studied is from WWTPs to the DWS. WWTPs are frequently suspected to be a pathway of microplastics to the aquatic environment (Besseling et al., 2017; Carr et al., 2016; Ziajahromi et al., 2017) and few quantitative data of microplastics transported by rivers to seas is currently available (Duis & Coors, 2016).

The microbeads present in cosmetics and cleaning agents enter the marine environment through domestic drainage systems (Murphy et al., 2016). The water from these systems will go via sewers to the nearest WWTP for treatment. The microplastics that originate from the wear of car tires, and paint and coatings partly end up in WWTPs via sewer systems.

Processes in WWTPs in the Netherlands

Microplastics can possibly end up in the effluent or settle with the sludge. Two different installations can be used in Dutch WWTPs. These are an active sludge installation (Leslie et al., 2012) and a membrane bioreactor (MBR) (Leslie et al., 2017; Leslie et al., 2012).

The active sludge installation exists of two compartments. In the first compartment bacteria flakes are floating and wastewater is led through these flakes. The bacteria break down the organic material under aerobic circumstances. The second compartment is where the sludge

will sink at the end of the process (http://www.saniwijzer.nl/content/content.asp?menu=1030_000000_000000_000000).

The MBR combines the active sludge installation with membrane filtration. A MBR exists of two compartments as well. Wastewater in the first compartment gets the same treatment as in the active sludge installation. In the second compartment are membranes submerged. Sludge and other organic material are removed by those membranes (http://www.saniwijzer.nl/content/content.asp?menu=1157_000000_000000_000000).

The active sludge installation let about 1.5 particles more pass through compared to the MBR. Research about sewage sludge pointed out that a variety of microplastics were present between 1 and 40 µm. A filter of 0.7 µm caught these microscopic particles. This study showed that almost all particles disappears with the primary and secondary sludge to the processing of the sludge. Currently it is unknown what happens afterwards (Leslie et al., 2012).

After treatment in WWTPs the effluent enters the river that is connected to the WWTP. Netherland has 355 WWTPs with 17,675 connected river segments. River segments are a part of a river. At every spot where two or more rivers emerge, a new river segment arises. Microplastic particles that are not removed during the treatment will remain in the effluent and thus enter rivers.

Processes in Dutch river segments

Different aspects influence how microplastics act in rivers. These are among others physical characteristics of the particles and characteristics of the water system and its surroundings.

Physical characteristics of the particles

The settling of particles in the sediment of rivers is influenced by five aspects. The density and radius of the particle, the density and viscosity of the water, and the acceleration of gravity. When the density of the particle is smaller than water the particle will float, if the density of the particle is higher the particle will sink over time (Langelaan et al., 2014).

Characteristics of the water system and its surroundings

The flow rate, water levels, wind and obstacles determine the quantity and distribution of the plastics in freshwater. Still waters can act as sinks for plastics and in flowing waters the plastic can be transported to other rivers, lakes and/or seas (Tysmans & Timmermans, 2014). Due to vertical flow velocity differences and turbulence, plastics that are sunk can be swirled up again and transported elsewhere (van den Brink, 1995).

2.2 Potential models

The first model that was found was a nutrient budget model in the Western Dutch Wadden Sea (Jung et al., 2017). This model used long-term field observations of nitrogen and phosphorus that came into the Wadden Sea via different pathways. The time series of nutrient concentrations from the water quality monitoring database were used (DONAR, http://www.watergegevens.rws.nl). This model was not selected based on two reasons. First, the model was based on long-term observations, while this study had to be finished in five months. Secondly, the model used datasets that were not available for microplastics.

Another potential model was about modelling marine surface microplastic transport (Sherman & van Sebille, 2016). This model could not be used as well, because it was based on long-term calculations. But an even greater shortcoming was that calculations were based on a

removal sink of plastics smaller than 20 cm. That was a huge difference with the focus in this study: particles smaller than 5 mm.

Other models were too difficult (Koelmans et al., 2017), did not cover the right context (Gräwe et al., 2016) or used too complicated programmes (Jahnke et al., 2016).

Eventually a model was chosen that was originally used to calculate pharmaceuticals from WWTPs to freshwater systems (Coppens et al., 2015). The equations used for that model could be adjusted to calculate the load of microplastics and the needed data was available. Additionally, it was user friendly and could be applied in the short time available.

2.3 Selected model to quantify the flow of microplastics

Here the eight equations are described that were used to quantify microplastics from WWTPs to the DWS. The input for the different parameters is given as well.

First the emission for each microplastic source is calculated based on the emission per source, the fraction that enters the WWTP, the capacity of the WWTP and the removal in the WWTP:

$$W_{Xi} = \frac{(M_X \cdot 1000000)/(365 \cdot 24 \cdot 60 \cdot 60)}{IE_{tot}} \cdot f_{Xau} \cdot f_{WWTPi} \cdot IE_i \qquad \text{Eq (2)}$$

W_{Xi} is the emission of microplastic source X from WWTP i (g/s);

M_X is the emissions per source (tons/yr);

IE_{tot} is the total capacity of the 345 STPs in inhabitant equivalent;

f_{Xau} is the fraction that enters the WWTP after use;

f_{WTTPi} is the fraction that passes WWTP I; and

IE_i is the capacity of WWTP i in inhabitant equivalent.

The emissions per source in kilograms per year and the fractions were found in literature for the four sources (Kole et al., 2015; van Wezel et al., 2016). The total capacity of the WWTPs in inhabitant equivalent was taken from literature as well (Coppens et al., 2015). Table 5 shows the data for each parameter for each source. The capacity of each WWTP was also given in separate Excel files.

Table 5 Data used for each parameter in equation 2

Symbol	***Explanation***	***Cosmetics***	***Cleaning agents***	***Paint and coatings***	***Car tires***
M_x	Emissions (tons/yr)	1,246	1,051	35	8,768
f_{eau}	Fraction entering the WWTP after use	1	1	0.005	0,15
f_{WWTPx}	Fraction that passes WWTP	0.10	0.10	0.10	0.09
IE_{tot}	Total capacity in inhabitants' equivalent	24,300,000	24,300,000	24,300,000	24,300,000
IE_i	Capacity of WWTP i in inhabitants' equivalent	-	-	-	-

Secondly, the travel time in the river is calculated from two matrices that were the result of a model calculation by Deltares using the Water Framework Directive Explorer with the Deltares Water Quality (DWaq) model. This model was run for a wet period of three months in 1998. The matrices show the mass fluxes of a conservative tracer and a non-conservative

tracer for each river segment and each WWTP. Both was put in the river at the location of each WWTP with a concentration of 1000 g/s. The non-conservative tracer has a set decay variable (k) of 0.005. Using both matrices, the travel time can be calculated using a modified standard first-order decay equation. The travel time in the river is essential to determine the settling of the microplastics.

$$T_{i,j} = -\frac{\ln(\frac{F_{nc,i,j}}{F_{c,i,j}})}{k_{nc}} \quad \text{Eq (3)}$$

$T_{i,j}$ is the overall travel time (d);

$F_{nc,i,j}$ is the non-conservative tracer without decay (g/s);

$F_{c,i,j}$ is the conservative tracer with decay (g/s); and

k_{nc} is 0.005 d^{-1}.

After the travel time was calculated, the settling viscosity is quantified. It is based on the density of particles and fluid, the dynamic viscosity of water, the gravity on earth and the radius of each particle:

$$v = \frac{2}{9} * \frac{(\rho_{nX} - \rho_f)}{\mu} * g * {R_X}^2 \quad \text{Eq (4)}$$

v is the settling viscosity (Stokes Law) (m/s);

ρ_{pX} is the density of particle X (g/cm^3);

ρ_f is the density of water (g/cm^3);

μ is the dynamic viscosity (g/cms);

g is the gravity (cm/s^2); and

R_X is the radius of particle X (cm).

The data used for each parameter can be found in Table 6.

Table 6 Data used for each parameter in equation 4

	ρp	ρf	μ	g	R	*Results*
Cosmetics	0.95	1				0
Cleaning agents	1.50	1	0.0147	980.7	0.00025	4.63E-06
Paint and coatings	2.25	1	0.0147	980.7	0.00005	4.63E-07
Car tires	1.20	1	0.0147	980.7	0.00025	1.85E-06

My literature review showed that 93% of all microplastics in cosmetics is polyethylene (Napper et al., 2015). An assumption has been made that the microbeads in cosmetics only exist of polyethylene. The density of polyethylene depends on the kind of polyethylene, i.e either high density polyethylene or low density polyethylene (http://www.plasticmoulding.ca/polymers/polyethylene.html). I assume a density of 0.95 g/cm^3. The density of water is around 1 g/cm^3. This means that the microbeads float, since the density of the particle is lower than the density of water.

The density of microplastics in cleaning agents has been estimated at 1.5 g/cm^3 (Scudo et al., 2017). The dynamic viscosity is a standard value as well as the gravity on earth. The radius of cleaning agents has been estimated at 0.00025 cm. The particles in paint and coatings are epoxy resins (Ahmad et al., 2005). The density of epoxy resins is 2.25 g/cm^3 (Electrolube), with an estimated radius of 0.00005 cm. Microplastic particles originated from car tires have an estimated density of 1.2 g/cm^3 (Verschoor et al., 2016). The radius of wear and tear from car tires has been estimated at 0.00025 cm (Pant & Harrison, 2013).

The fourth step was to calculate the river depth for each river segment. This was based on the river discharge that was given by provided Excel files:

$$Z_i = 0.34\ {Q_i}^{0.341} \quad \text{Eq (5)}$$

Z_i is the river depth (m); and

Q_i is the river discharge (m^3/s^{-1}).

The settling coefficient is then calculated based on the settling viscosity (equation 4) and river depth (equation 5):

$$k = \frac{v}{Z_i} * 24 * 60 * 60 \quad \text{Eq (6)}$$

k is the settling coefficient (day^{-1})

The k value that has been calculated in equation 6 can be used here to calculate the mass flux of each source. The conservative tracer was given by the Excel files and the travel time was calculated in equation 3.

$$F_{X,i,j} = F_{c,i,j} * \exp(-k_X * T_{i,j}) \quad \text{Eq (7)}$$

$F_{x,i,j}$ is the mass flux microplastic source X from WWTP i in river segment j (g/s); and

k_X is the settling coefficient of microplastic parti6les (d^{-1}).

The mass flux of the microplastics for each source are calculated by first dividing the emissions from each WWTP by 1000 and then multiplying it with the mass flux calculated in equation 7. Then all the those fluxes were added up for each river segment to come to M_X,j.

$$M_{X,j} = \sum_i \frac{W_{X,i,j}}{1000} * F_{X,i,j} \quad \text{Eq (8)}$$

$M_{X,j}$ is the mass flux of microplastic source X in river segment j (g/s)

The last step is to calculate the concentration of each microplastic source X in each river segment j. This is done by dividing the mass flux, calculated with equation 7, by the given discharge for each river segment.

$$C_{X,j} = \frac{M_{X,j}}{Q_j} \quad \text{Eq (9)}$$

$C_{X,j}$ is the concentration of microplastic source X in river segment j (g/m^3);

Q_j is the local water discharge (m^3/s).

3. Results

3.1 Microplastic flux in the Netherlands

3.1.1 Cosmetics

Figure 2 shows the mass flux of microplastics from cosmetics in the Netherlands. The y-as shows the river segments, 16025 river segments were used in total. In only 691 of the 16025 river segments was a mass flux found. The x-as shows the mass flux in each river segment, which was for cosmetics between 0.0 and 1.1 g/s. The total flow of microplastics originated from cosmetics is 25.95 grams per second. You can see in the figure that the highest mass flux originated from river segment 481. This is the same for each source. This segment is located near Haringvliet. The y-as is sorted from highest mass flux to lowest mass flux. This means that the river segments are sorted in a way that first the segment with the highest mass flux is shown (481), followed by the second highest flux (325) and so on. This is the same for all sources.

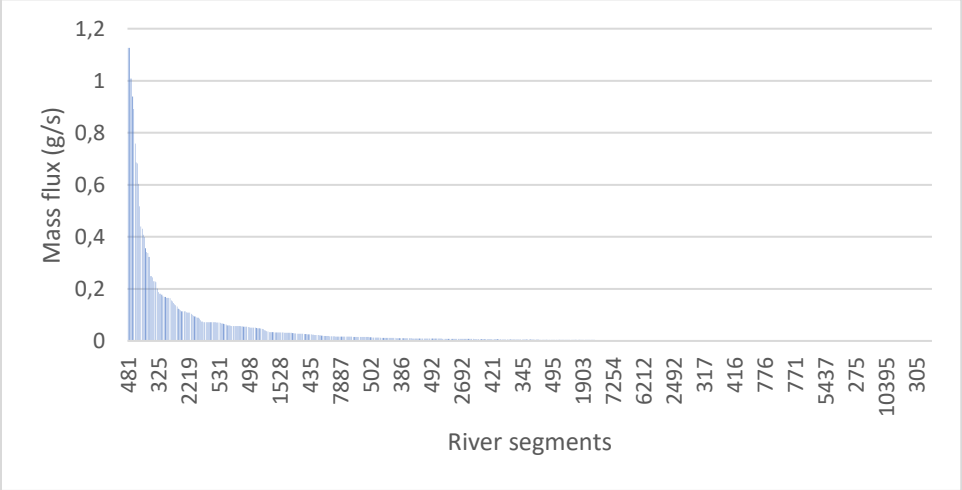

Figure 2 Mass flux of microplastics from cosmetics per river segment

3.1.2 Cleaning agents

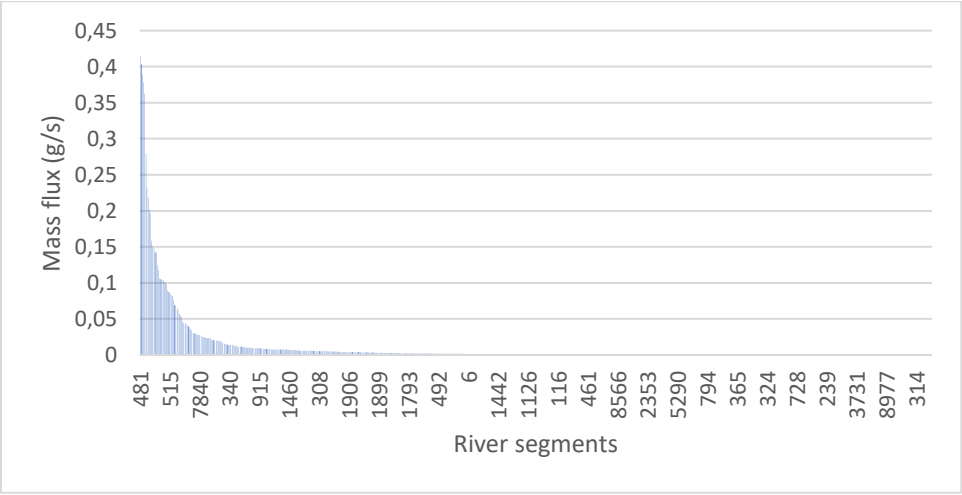

Figure 3 Microplastic flow from cleaning agents

Figure 3 shows the mass flux of microplastics from cleaning agents. The mass flux in each river for cleaning agents was between 0.00 and 0.41 g/s. The total flow of microplastics

originated from cleaning agents is 7.91 g/s. There are some differences between cosmetics and cleaning agents. For cosmetics high fluxes were found near Geleenbeek and Hollandse IJssel, while high fluxes from cleaning agents were found near Eemskanaal and Eem.

3.1.3 Paint and coatings

The mass flux from paint and coatings was much lower than the previous two sources and was between 0.000 and 0.028 g/s. Figure 4 shows the mass flux of microplastics originated from paint and coatings. The total flow of microplastics from paint and coatings was calculated at 0.58 g/s for the Netherlands. High fluxes here were found near Westervoort and Groningen.

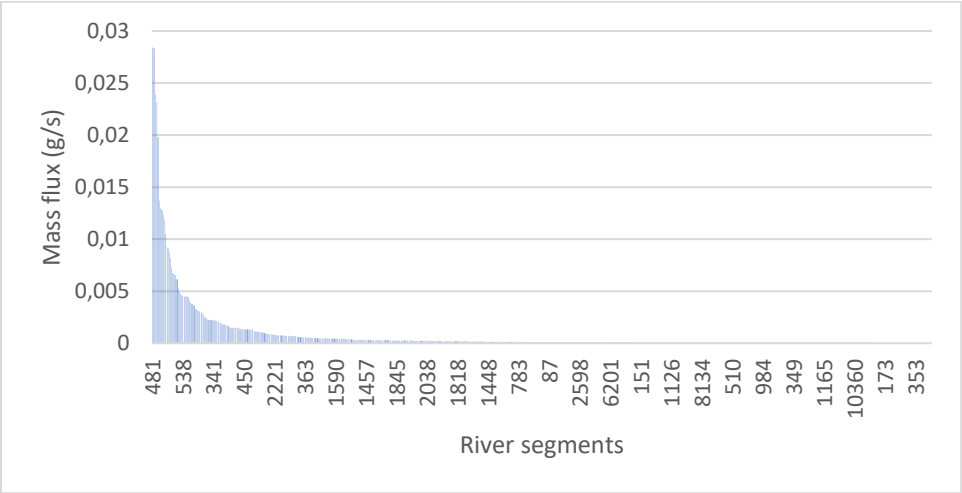

Figure 4 Microplastic flow from paint and coatings

3.1.4 Car tires

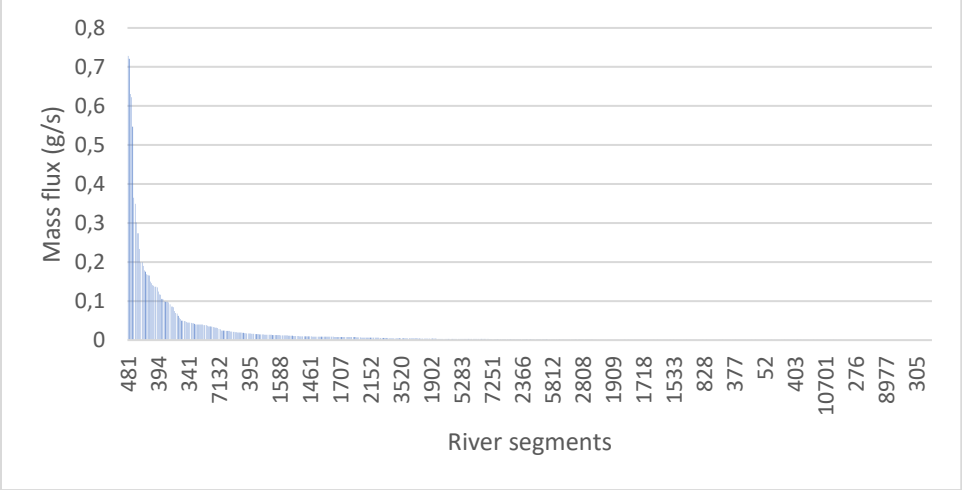

Figure 5 Microplastic flow from car tires

The last mass flux was the one of microplastics originated from car tires, as shown in Figure 5. The mass flux of microplastics from car tires lays between 0.00 and 0.72. This total flow was calculated at 12.82 g/s. The highest fluxes here were found near Overijsselse Vecht and Groningen.

The emissions from the four sources showed that car tires were by far the highest (Table 4), followed by cosmetics, cleaning agents and eventually with a small contribution paint and

coatings (Table 3). The figures here showed that the mass flux from cosmetics was much higher than the flux of car tires. There are two explanations. First, where all the microplastics from cosmetics enter the WWTPs, only a part of wear and tear of car tires end up in WWTPs. Additionally, while microscopic particles of cosmetics float, the particles from car tires will sink over time. Paint and coatings are again the smallest contributor compared to the other three sources. This was expected, both because the emissions of paint and coatings were smallest as well as because only a low percentage of microplastics in paint and coatings end up in the WWTPs.

3.2 Microplastic concentration in the Netherlands

3.2.1 Cosmetics

Figure 6 shows the concentrations of microplastics originated from cosmetics in the Netherlands. The legend shows that the concentration has a range between circa 0 and higher than 1 g/m^3 per river segment. The legend shows six colours, with six different ranges. Most particles were between 0.0 and 0.2 g/cm^3 (purple), followed by the range of 0.02 to 0.05 g/cm^3 (blue) and 0.1 to 0.5 g/cm^3 (yellow). A few red spots can be found in the figure that show the highest concentrations. The average concentration of microplastics is 0.05 g/m^3. Only one spot, near Stavoren, had a concentration around 7.0 g/m^3. In the rest of the Netherlands the concentration was circa 1.0 g/m^3 and lower. The high concentration from Stavoren enters Lake IJssel so this is a possible threat to the DWS.

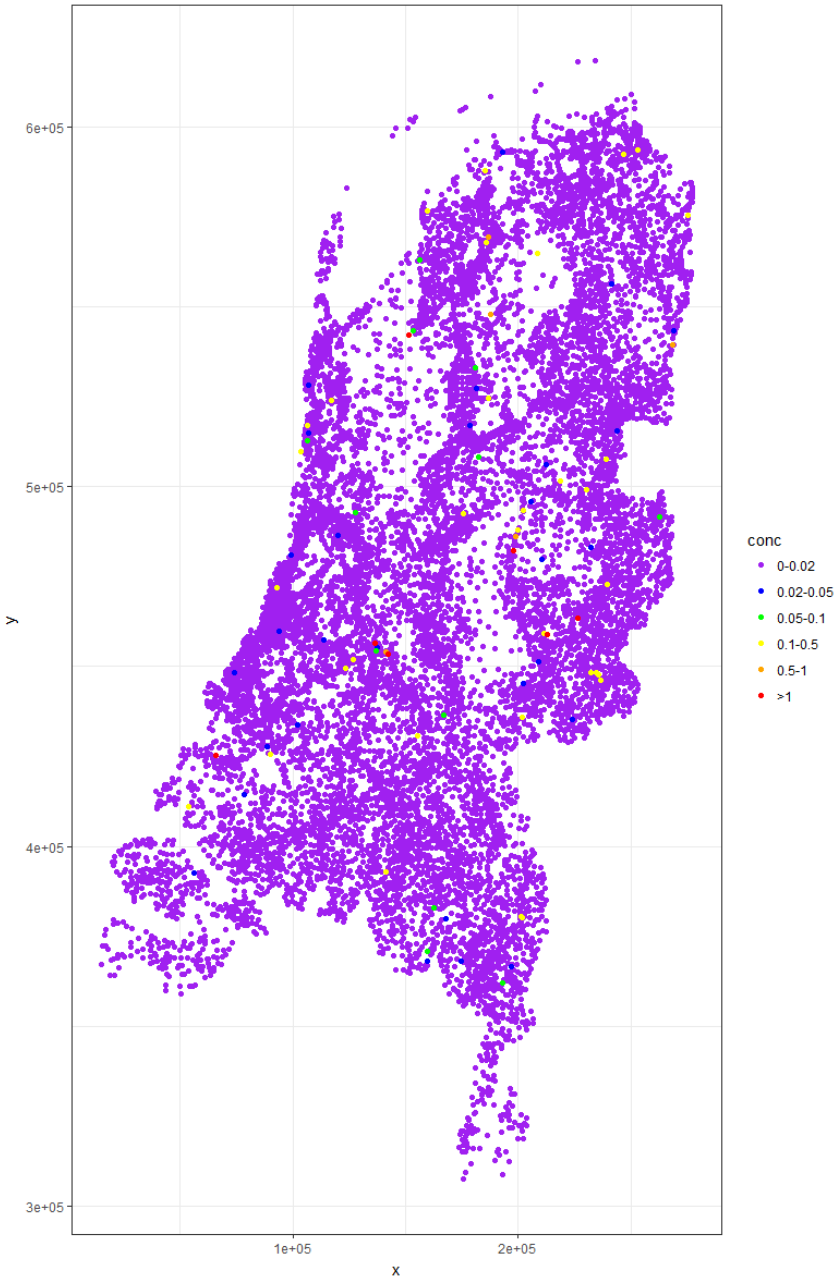

Figure 6 Concentrations of microplastics from cosmetics in the Netherlands

3.2.2 Cleaning agents

Figure 7 shows the concentrations of microplastics originated from cleaning agents in the Netherlands. The legend shows that the concentration has a range between circa 0 around 0.80 g/m^3. The highest concentration of microplastics from cleaning agents was calculated near Utrecht. Beside the range from 0 to 0.00005 (purple), the most common concentration was between 0.0005 and 0.05 (green).

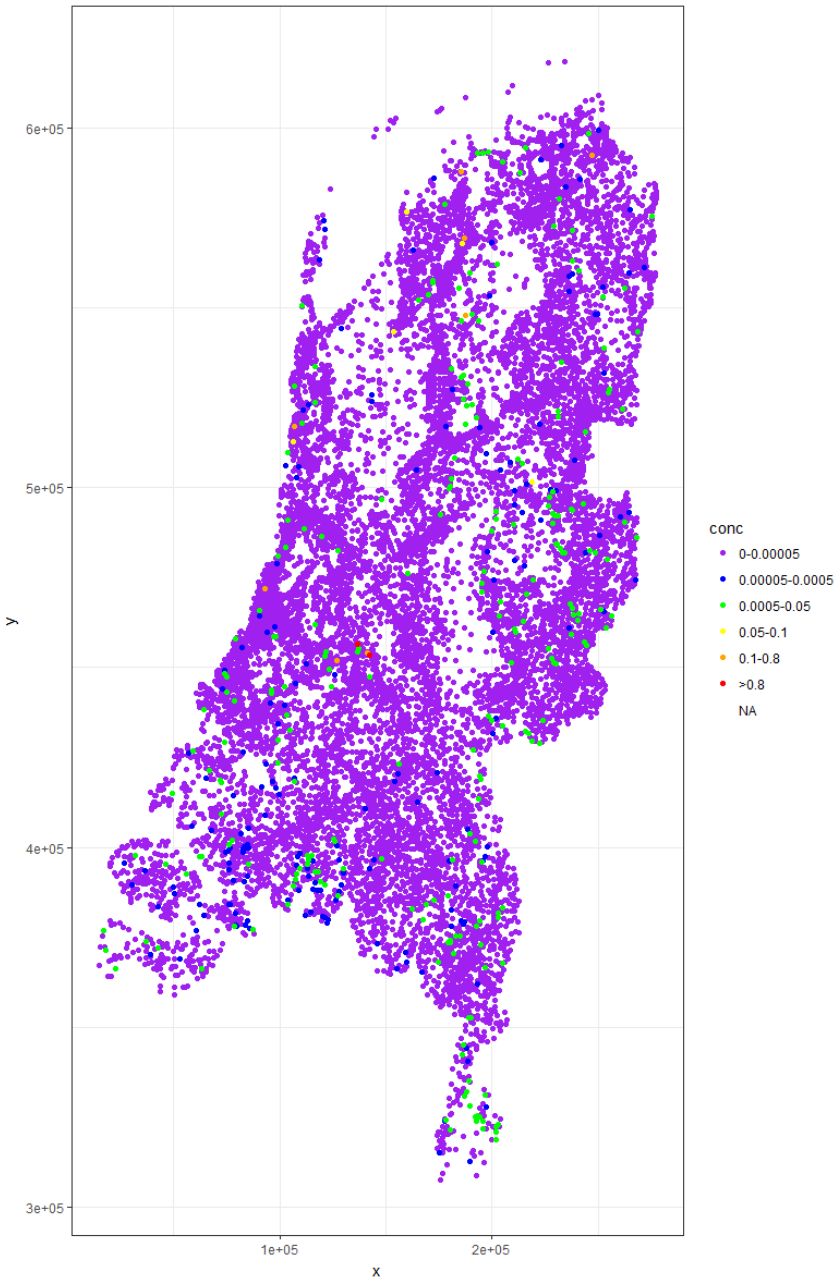

Figure 7 Concentrations of microplastics from cleaning agents in the Netherlands

3.2.3 Paint and coatings

Figure 8 shows the concentrations of microplastics originated from paint and coatings in the Netherlands. The legend shows that the concentration has a range between 0 and around 0.035. Here the highest concentration was again found near Utrecht. Beside the concentration

between 0 and 0.0000005 (purple), most concentrations were calculated between 0.000005 and 0.00005 (blue) and between 0.00005 and 0.0005 (green).

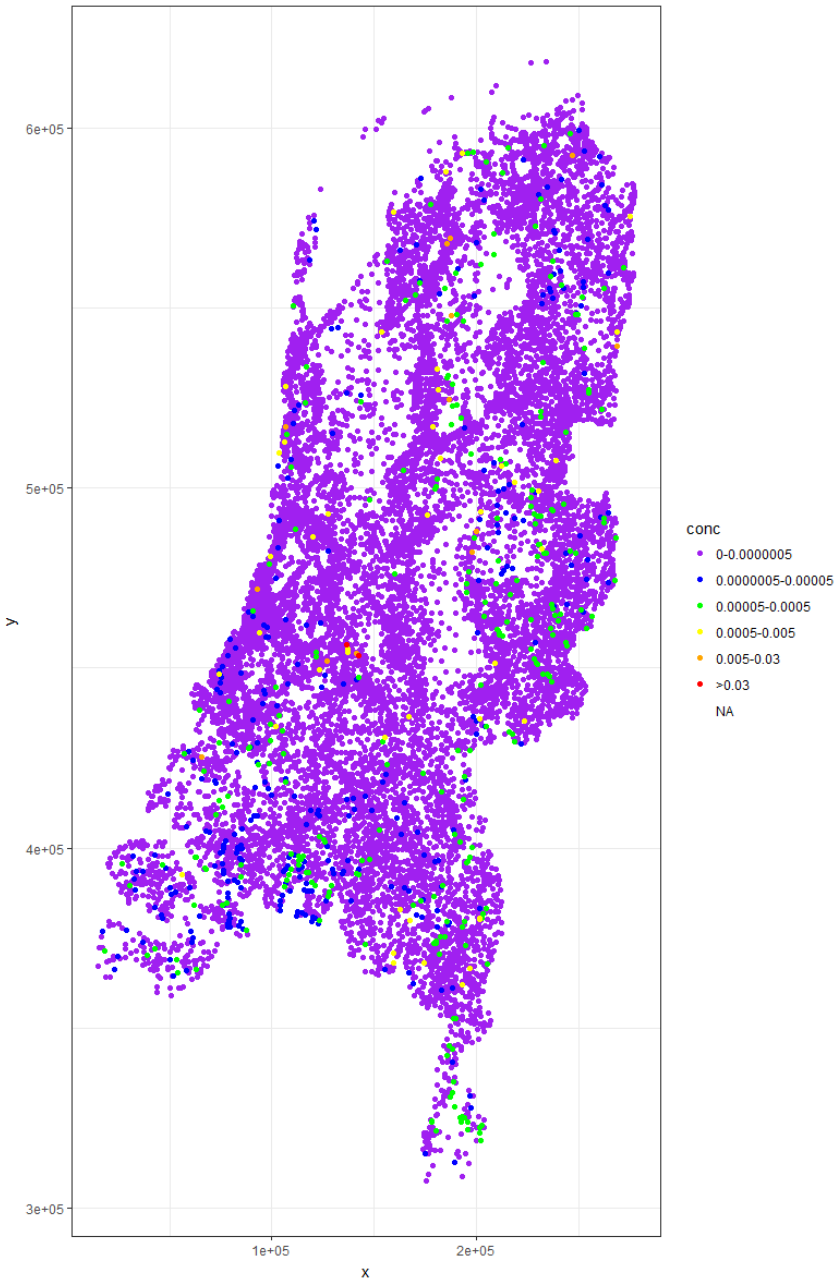

Figure 8 Concentrations of microplastics from paint and coatings in the Netherlands

3.2.4 Car tires

Figure 9 shows the concentrations of microplastics originated from car tires in the Netherlands. The legend shows that the concentration has a range between 0 and around 0.6. Here the highest concentration was calculated near Roosendaal. Even though the concentration is based on the inhabitant equivalent, the concentration for each source is high in a different place. Only for cleaning agents and paint and coatings the highest concentrations were found in the same area. This is caused by the different settling coefficient of each source. Most concentrations were found in the lowest range again (0 to 0.0005, purple). Other concentrations were mostly between 0.0005 and 0.005 (green) and between 0.005 and 0.05 (green).

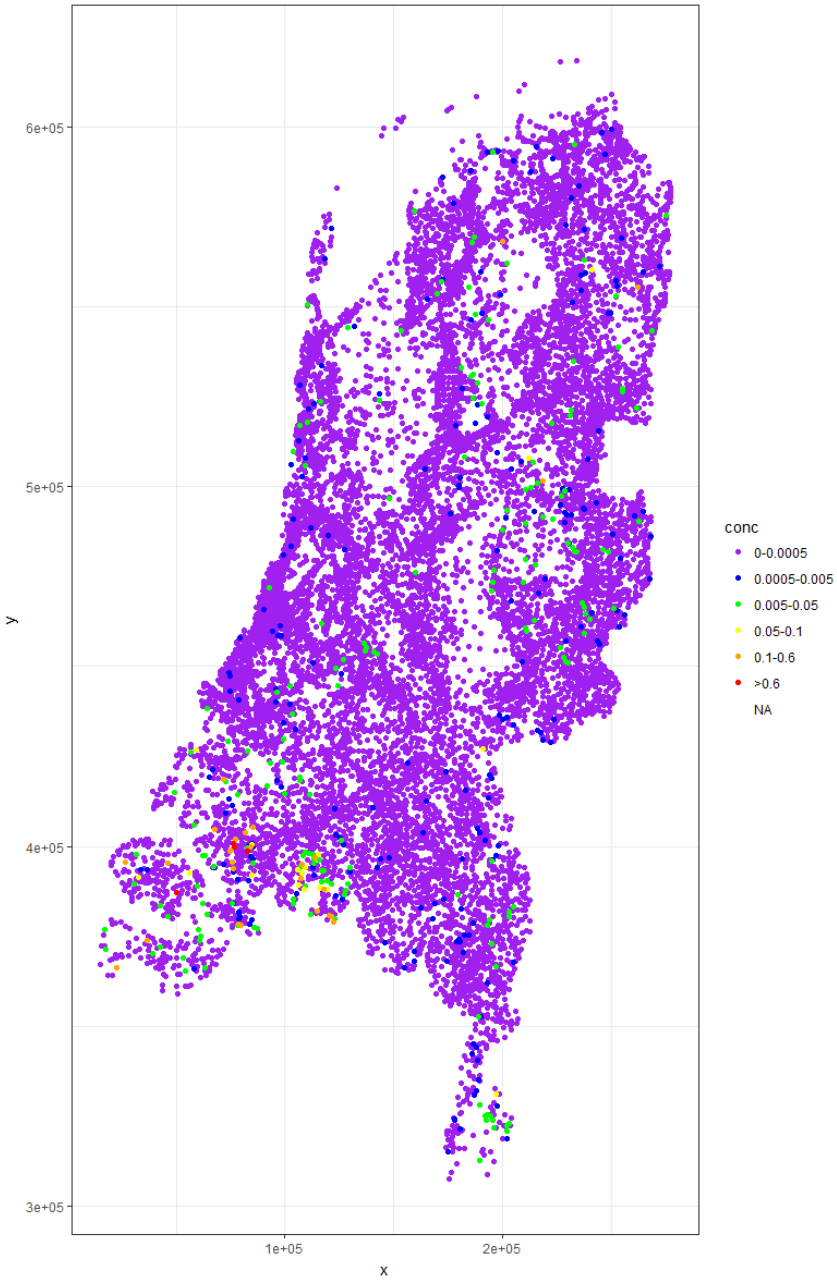

Figure 9 Concentrations of microplastics from car tires in the Netherlands

In the Southern California coastal waters, a concentration of 0.020 g/m^3 was found with a 0.333 mm mesh net. In the California Current a concentration of 0.003 g/m^3 was found and in the Southeast Bering Sea and near the US West coast a concentration of 0.209 g/m^3 (Wang et al., 2016). These concentrations are reasonably similar compared to cleaning agents, paint and coatings, and car tires. Only the concentrations originated from cosmetics are far higher than what was found in other areas.

3.3 Fraction of microplastics in the Netherlands to the DWS

32 of the 355 WWTPs are assumed to be connected to rivers that end up in the DWS. This resulted in a calculation of a mass flux of 1.83 g/s that end up in the DWS. Figure 10 shows a comparison between the mass flux in the Netherlands and DWS for each source.

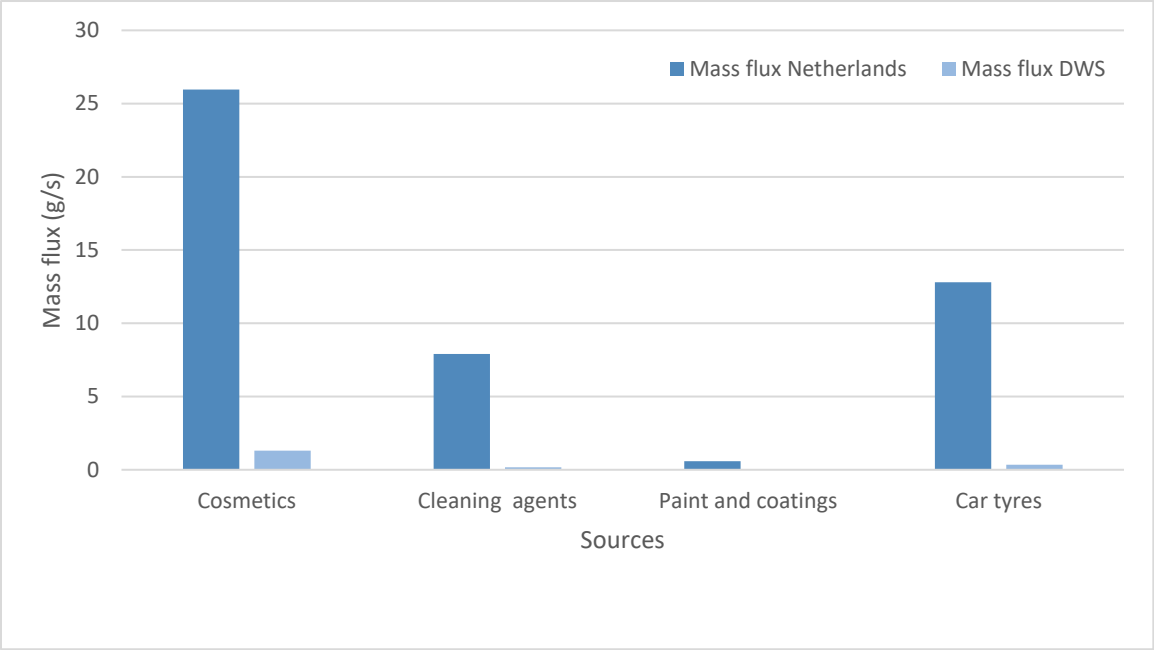

Figure 10 Comparison of the mass flux (g/s) in the Netherlands and the DWS

The total mass flux in the Netherlands has been calculated at 47.26 g/s. This means that only an estimated 3.87% end up in the DWS.

For cosmetics the highest mass fluxes that led to the DWS were found near the Afsluitdijk (Lake IJssel), Friese Boezem and Randmeer (Lake IJssel). For cleaning agents, the highest fluxes were found near Flevoland, Tochten and Lage Vaarten. The same high fluxes were found here for car tyres. The highest mass fluxes from cleaning agents were found near Flevoland, Friese Boezem and Tochten. The WWTP that contributed most to the mass flux to the DWS was the WWTP in Zeewolde and in St-Annaparochie. The microplastics that enter the DWS from Zeewolde travel via Lake Marker and Lake IJssel. The microplastics from St-Annaparochie enter the DWS via Zwarte Haan, Wadden Sea – Afsluitdijk and Fries Kleigebied. Mitigation measures will be most important here to reduce the microplastic mass flux as fast as possible. Currently it is unclear whether this mass flux to the DWS is high or low.

3.4 Comparing the results to available studies

I compared my results with those by Siegfried et al. (2017) who calculated an export load of 1.1 kilotons of microplastics per year to the North Sea. 1.1 kilotons are the same as 1.1 e^9 grams. The mass flux to the DWS was 1.83 g/s which is the same as 57.7 e^6 g/y. The total mass flux in the Netherlands was calculated at 47.26 g/s which is the same as 1.5 kilotons per year. This is close to the calculation of Siegfried et al., (2017) at 1.1 kilotons per year that end up in the North Sea. This means that the calculated load in this study is probably a realistic amount. Part of the 1.5 kilotons per year in the Netherlands will settle, but how much is currently unclear.

4. Discussion

To conduct this study both qualitative and quantitative data is used. One of the first steps of this study was to decide on which sources to focus. Four sources were selected based on their relevance and available data. A missing source appeared to be microplastics originated from washing clothes. It is seen as an important source of microplastic pollution and enters rivers via WWTPs and other sources. (Browne et al., 2011; Napper & Thompson, 2016). I have chosen to ignore this source for now, since currently too little information is available to include this source in the model (Browne et al., 2011).

I moreover chose to only focus on point sources that enter the marine environment through WWTPs. For example, part of the microplastics originating from car tires enter the marine environment through runoff or via atmospheric transport. This means that not the total microplastic contribution from car tires is calculated but only the part that enters the sea via WWTPs. The consequence is that the actual load is expected to be higher. Future studies should include diffuse sources as well. When appropriate information about the properties of microplastics from washing clothes is available, this source can be included in other studies as well.

When the sources and pathway were selected a model had to be selected. Some models seemed useful, but they all had shortcomings. The models were too complex or needed too much data. Such data was unavailable or not available for a longer period. Based on the available time and data a model eventually was chosen. The model that has been chosen was a model originally used to model pharmaceuticals in freshwater systems. My model focuses on the mass flux and concentration of microplastics from WWTPs to the sea and is seen as a first step in quantifying the flow in the Dutch rivers and the Wadden Sea. This model was the best choice at this moment. The advantages of this model were that the needed data could be found; that it was easy to apply; it met the aim to gain insight in the current flow of microplastics in Dutch river segments and the DWS; and it was a first step in closing the knowledge gap.

The emissions were calculated by using data from a literature review (van Wezel et al., 2016). The calculation by using the data from van Wezel et al. (2016) was far higher than data found by Verschoor et al. 2016. For example, the emissions from cleaning agents were calculated at 1,051 tons/year while Verschoor et al. (2016) stated that cleaning agents only emitted 2.6 tons/year. There is chosen for the article from van Wezel et al. (2016) because the data there seemed more comprehensive. The total emissions from car tires were calculated at 8,768 tons based on van Kole et al. (2015). Other studies stated that the total emissions from car tires were 17,300 tons (e.g. Verschoor et al., 2016). The differences between the emissions are caused by different ways of calculating the emissions using different data sets. My results seemed reliable after comparing it to the results from Siegfried et al. (2017).

After the emissions from the different sources were calculated, the mass flow could be calculated. In the pharmaceutical model a decay-rate was used, but this was not applicable to the calculation of microplastics. It can namely take decades for plastics to degrade. Instead the settling of microplastics was calculated by using the Stokes Law. Some estimations were made here. 93 percent of the microplastics in cosmetics are represented by polyethylene (Gouin et al., 2015). This study estimated that the only microplastics in cosmetics are polyethylene. Two kinds of polyethylene exist: low-density polyethylene (density between

0.91 and 0.93 g/cm^3) and high-density polyethylene (density between 0.94 and 0.97 g/cm^3). Here the density is assumed at 0.95 g/cm^3. The diameter of microplastics from car tires is mostly smaller than 0.008 cm. This study estimated the microplastics at a diameter of 0.005 cm, resulted in a radius of 0.00025 cm. The density of the resins used in paint and coatings was estimated at 2.25 g/cm^3 based on literature (https://datasheet.octopart.com/ER2074RP250G-Electrolube-datasheet-537902.pdf). These assumptions are based on the little literature available.

Despite the uncertainties that occurred during this study, at this point no better outcome could have been reached. When I started this study, almost no specific data on the amount of microplastics that enter the sea via WWTPs, was available. With the available data the best possible result has been achieved with this study.

A follow-up study could for example add all the other relevant sources that were ignored. Additionally, diffuse sources should be identified as well since now only point sources were calculated. Other pathways should be calculated as well to get a more comprehensive picture of the total microplastic pollution in the Netherlands. My study is a first step in gaining insight in the total microplastic flow in Dutch rivers and the Wadden Sea.

After the total flow of microplastics from WWTPs to the DWS is known, studies can start to focus on the effects of microplastics. For example, the effect on organisms, the environment, the water quality and perhaps there is even an effect on humans. This would give more insight in the overall problem of microplastics. Furthermore, more research about the effects could focus on mitigation measures. Since this study pointed out that cosmetics are by far the most problematic source a study could identify how this problem could be reduced.

5. Conclusion

The aim of this study was to quantify the flow of microplastics that enter the DWS via WWTPs to decrease the knowledge gap. This was done by answering six research questions.

First, the relevant sources had to be selected. Through literature review four sources were selected in this study: cosmetics, cleaning agents, paint and coatings, and car tires. These sources were considered most relevant, given the known use of microplastics in view of desired product properties. Additionally, enough data was available about the sources to model them. The highest emissions in the Netherlands come from car tires. They are followed by consumer products, then cleaning agents and the emissions of paint and coatings were almost negligible. After the specific emissions from the sources to the WWTPs were calculated this sequence changed a bit. Cosmetics, cleaning agents and car tires were almost the same, paint and coatings was by far the smallest emitter again.

Secondly, the processes used in WWTPs had to be identified. Microplastics from consumer products enter WWTPs via sewers after use. From car tires microplastics enter sewers as well due to wear and tear. Depending on the kind of source a fraction of the use of microplastics will enter the sewer and thus the WWTP. The processes in a WWTP have an impact on the microplastic load that remain in the effluent. The active sludge installation captures less microplastics than the membrane bioreactor.

The third step was to identify what happens to the particles that enter the rivers after treatment. The fraction that remains in the effluent enters a river segment, where microplastics can either sink or float. Different aspects have an influence on if they sink and how fast they sink. This study looked at the settling of particles and the characteristics of the rivers. The settling depends on the density and radius of the particle, the density and viscosity of the water, and the acceleration of gravity. The particles are furthermore influenced by the discharge and depth of the rivers.

After the processes were identified, a model had to be selected to quantify the flow of microplastics. After identifying available models, eventually a model was chosen that originally was used for pharmaceuticals. The new model uses a generic water-quality model developed from Water Framework Directive Explorer with set parameters to identify the travel time in Dutch river segments. The travel time was then combined with the settling-coefficient K and an initial flow from different plants to determine the microplastic flux and concentration.

This model let to the results of this study. The mass flux appeared to be largest for consumer products, followed by car tires. Cleaning agents had the third highest mass flow and paint and coatings had the lowest mass flow. The average concentration that was calculated was again highest for cosmetics, followed by car tires, then cleaning agents and again the lowest for paint and coatings.

This study is a first step in closing the knowledge gap about microplastics in Dutch rivers and the Wadden Sea by quantifying the flow. Due to current available data the best possible result has been achieved with a model that was applicable. This study can be used as basis for further identification of the microplastic flow and threats in the Netherlands.

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