BTO 2019.006 | Mei 2019

BTO report

Microplastics in (sources for) drinking water

BTO 2019.006 | Mei 2019

BTO

Microplastics in (sources for) drinking water

BTO 2019.006 | April 2019

Project number 402045/061

Project manager Jos Frijns

Client BTO Vewin - Beleidsonderbouwend Onderzoek

Quality Assurance Thomas ter Laak

Author(s) Patrick S. Bäuerlein Stefan A.E. Kools

Sent to This report is distributed to BTO-participants. The report is public as of May 2019.



More information dr. Stefan A.E. Kools T +31 30 606 9539 E stefan.kools@kwrwater.nl

KWR PO Box 1072 3430 BB Nieuwegein The Netherlands



Nederlandse samenvatting

Microplastics in (sources for) drinking water

Over microplastics is nog veel onbekend, maar te verwachten is dat de kleinere deeltjes (nanoplastic) ook het drinkwater zullen bereiken

Auteur(s) dr. Patrick Bäuerlein en dr. Stefan Kools

Microplastics zijn plastic deeltjes kleiner dan 5 mm en nanoplastic is nog vele malen kleiner. De vraag is wat bekend is over deze deeltjes in water. KWR heeft in een Engelstalige rapport gegevens samengebracht uit de wetenschappelijke literatuur over microplastics in afvalwater, oppervlaktewater, grondwater en drinkwater. Uit deze literatuur blijkt dat meetmethoden nog sterk ontwikkeling zijn. Ondanks het feit dat resultaten lastig te vergelijken zijn is het beeld dat microplastics algemeen voorkomen in het milieu en oppervlaktewater (inclusief Nederlandse bronnen voor drinkwater). Ook in drinkwater is nog weinig gemeten, maar enkele studies tonen aan dat ook hier microplastic is aangetroffen (met lagere gehaltes in drinkwater uit grondwater). De betekenis voor de gezondheid van mensen is nog niet goed duidelijk. Over de nanoplastics ontbreekt nog elk inzicht in voorkomen en risico's. Er zijn te weinig gegevens over het voorkomen en de effecten van nano- en microplastics in milieu en water voor een goed onderbouwde risicoschatting en voor ontwikkeling van verwijderingsmethoden. Dat zal ook nog wel even duren. De onderzoekers adviseren drinkwaterbedrijven zich te blijven inzetten voor een gestandaardiseerde meetmethode en die eerst projectmatig en later meer routinematig in te zetten voor monitoring in bronnen en geproduceerd drinkwater. In afwachting van aanvullende Nederlandse meetgegevens kunnen de gegevens uit het buitenland gebruikt worden als achtergrondinformatie, zoals samengebracht in dit rapport.

Microplastics everywhere

High amounts of microplastics have been found not just in the sea and on beaches, but also in rivers and soils around the world, demonstrating how pervasive this modern pollution is. Sources include leakage from landfills, plasticulture, littering, and sewage sludge. Data from (1).



Grote hoeveelheden microplastics zijn niet alleen in de zee en op het strand aangetroffen, maar ook in rivieren en bodems over de gehele wereld. Emissiebronnen zijn stortplaatsen, 'plasticulture' (het gebruik van plastic in de landbouw), zwerfvuil en zuiveringsslib. Data afkomstig uit [2]. Credits: GRAPHIC: N. DESAI/SCIENCE.

Microplastics worden gedefinieerd als plastic deeltjes kleiner dan 5 mm. Door verwering kunnen hieruit nanoplastics ontstaan: deeltjes kleiner dan een um (micrometer: een duizendste millimeter) of zelfs kleiner tot een nm (nanometer: een miljoenste millimeter). In het milieu worden verschillende soorten plastic deeltjes gevonden: polypropyleen (PP), nylon, polystyreen (PS) en polyethyleen (PE). Veel methoden om deze deeltjes op te sporen en hun hoeveelheden te bepalen zijn nog in ontwikkeling. De meeste meetmethoden kunnen nu alleen deeltjes aantonen van 10 micrometer of groter. De huidige manier van monstername, monstervoorbewerking en analyse is arbeidsintensief. Ook is nog nauwelijks ringonderzoek uitgevoerd met standaarddeeltjes, zoals voor andere laboratoriumanalyses In lopend onderzoek wordt wel al gekeken naar betere meetmethoden.

Focus op deeltjes in drinkwater(bronnen)

In eerdere studies ging de aandacht specifiek naar deeltjes in zee. In deze literatuurstudie is vooral gekeken naar concentraties in afvalwater, oppervlaktewater en grondwater (omdat deze direct gerelateerd zijn aan drinkwaterproductie, zie figuur) en naar concentraties in geproduceerd drinkwater. Aan de hand van reviews in tijdschriften en gerichte zoektermen in wetenschappelijke databases van artikelen zijn een aantal studies verzameld. Enkele (buitenlandse) studies wijzen op een algemeen voorkomen van lage gehaltes microplastics in drinkwater uit oppervlaktewater (1-470 deeltjes/L), en op nog lagere gehaltes in drinkwater dat geproduceerd is uit grondwater. De hoogste concentratie op vijf verschillende locaties was 0.003 deeltjes/L in de watermeter. Hun exacte herkomst is nog onduidelijk. In het water uit de kraan zijn geen deeltjes gevonden.

Data nog te onbetrouwbaar voor risicoschatting

Verschillende experts stelden dat het aantal documenten op dit gebied exponentieel groeit, maar dat de kennis niet in hetzelfde tempo groeit. Zo is vooral onduidelijk wat de precieze (dagelijkse) inname van microplastics is, terwijl dit soort gegevens voor nanoplastics zelfs totaal ontbreekt. Kennis over blootstelling en inzicht in de mogelijke effecten zijn essentieel voor het inschatten van gezondheidsrisico's. Zo is in diermodellen wel enig bewijs gevonden dat plastic deeltjes ontstekingsreacties kunnen veroorzaken, maar het is nog onduidelijk hoe deze studies zich vertalen naar mensen en de blootstelling via (drink)water. Slechts weinig studies richten zich op de kleinere microplastics en nanoplastics. Wel wordt duidelijk dat hoe kleiner de deeltjes, hoe groter de aantallen in het water. Juist in de kleinere fracties ontbreekt nog veel inzicht. Over microplastics en nanoplastics is nog veel onbekend, maar te verwachten is dat nanoplastics ook het drinkwater zullen bereiken. Ook bestaat veel aandacht voor een bijkomend aspect: de mogelijkheid dat chemicaliën op en in plastic deeltjes kunnen bijdragen aan de blootstelling aan deze stoffen. Hiervoor ontbreken momenteel betrouwbare schattingen. Daarnaast is de rol van de vorm van de deeltjes nog grotendeels onbekend.

Samenvattend zijn de huidige data te onbetrouwbaar, wat een duidelijke behoefte aan verbeterde bemonsterings- en detectiemethoden, blootstelling- en effectstudies onderstreept. In afwachting van aanvullende Nederlandse meetgegevens kunnen de gegevens uit het buitenland gebruikt worden als achtergrondinformatie, zoals samengebracht in dit rapport.

Vooruitzicht en handelingskader

De sterk toegenomen media-aandacht voor micro- en nanoplastics zal de publieke opinie blijvend beïnvloeden, ook al ontbreekt inzicht over aanwezigheid en risico's. Drinkwaterbedrijven wordt aangeraden hierover transparant te communiceren en dus ook de onzekerheden te benoemen. Over het plasticsvraagstuk lijkt in de politieke en publieke opinie een redelijke mate van consensus te bestaan, wat momentum geeft naar beleid en actie. Zo is bijvoorbeeld nauwelijks sprake van ontkenning of plastic denial, zoals wel bestaat op het gebied van klimaatverandering en de rol van mensen daarin. Er bestaat consensus dat economische bedrijvigheid zorgt voor een constante emissie van plastics. Het is een realistische verwachting dat het nog enige tijd zal duren voor een volledig beeld is ontstaan van de risico's van plastic deeltjes in het milieu en in water in het bijzonder. Diverse actoren in de waterketen, zoals waterschappen en drinkwaterbedrijven,

zouden vanuit de groeiende aandacht en het voorzorgprincipe technische aanpassingen kunnen overwegen om deeltjes (nog beter) te verwijderen. Het is daarom voor de korte termijn aan te bevelen dat drinkwaterbedrijven (blijven) inzetten op het ontwikkelen van meetmethoden. Vanuit de inventarisaties naar de aanwezigheid van de deeltjes zou later ook monitoring kunnen volgen. Verder is het aan te bevelen om het uitwisselen van gegevens van de verschillende onderzoeksgroepen te stimuleren, zodat de gegevens onderling vergelijkbaar worden. Met het verbeteren van de meetmethoden zal ook meer goede informatie beschikbaar komen over de mogelijkheden voor verwijdering van plasticdeeltjes tijdens de (drink)waterbehandeling.

Rapport

Dit onderzoek is beschreven in *Microplastics in* (sources for) drinking water (BTO 2019.006)

Summary

General

It becomes more and more clear that plastics can be found in all parts of our environment. All plastics in the environment result from various sorts of human activity such as waste (litter), industrial activities, agriculture applications and household use. The most common types of plastics that have been found in the environment are polyethylene (PE), polypropylene (PP), polystyrene (PS) and PET. Plastics appear in many forms and sizes as can be learned from the fast-growing literature. Plastic particles smaller than 5 mm are commonly defined as *microplastics* and even smaller particles below one micrometre are referred as *nanoplastics*. Note that 1 micrometre is a thousandth of a millimetre and 1 nanometer another thousand times smaller (10⁻⁹m). This report gathered the findings in the literature on these micro- and nanoplastics with special focus on water and drinking water.

Sampling and measurement

Currently, there are no universally accepted protocols on how to take samples and measure them. Also, the way the findings are reported differ from publication to publication. This impedes comparison of different data sets. Furthermore, there are none or few interlaboratory studies that would enable to evaluate the quality of the currently developed analytical methods, as is common for more classical chemical and microbial analyses. Due to these reasons and due to the fact that particle measurement is still time-consuming as well as laborious, routine measurement is not yet done on a wider scale.

The difficulty of detecting micro and nanoplastics is that these particles may be found everywhere so that contamination may occur during sampling, treatment or analyses. In some cases (especially in drinking water samples) the reported concentration is close to the limit of quantification (LOQ of particles per litre). The LOQ is important because detecting one particle is technically possible but the blanks need to have less particles than the actual sample to be precise on the concentration in the samples when reporting. Note that the limit of detection (LOD) means the lower limit of the particle size, meaning the lowest size range that can be detected.

Waste water

Particle concentrations in influent and effluent can vary from 300 to 1000 p/L and ca. 1 to 50 p/L, respectively. This means that waste water treatment plants (WWTP) remove between 90 to 99% of the plastic particles. Yet WWTPs may discharge billions of particles each day, up to several grams per day. For this estimate it is assumed that roughly 300,000 m³ sewage water per day (size of a modest to large treatment plant) is treated and that only particles larger than 20 micrometre (μ m) are taken into account (equivalent to one thousandth of a millimetre). It is safe to assume that the amount of particles getting into the environment is significantly higher when all smaller particles are also included. This can mean that up to 1000 times more particles could be released, but more reliable estimates are not available.

Surface water

In rivers, particle numbers as high as 22,000 p/m³ can be found. Particle numbers between 100 and 3000 p/m³ are detected and the numbers are dominated by the lower size particles. Close proximity to a sewage treatment plant usually results in a higher particle concentration, indicating that sewage is a major point pathway for these emissions. Microplastic can also be found in the sediments in rivers. There, between 200 and 2000 p/kg dry soil have been detected, which translates into 1 – 8 mg of plastic per kg dry soil. The Dutch project "Technologies for the Risk Assessment of Microplastics (TRAMP) is currently ongoing that will provide more insights into the occurrence in the Meuse basin. This project aims to not detect microplastics but also nanoplastics, using a recent developed protocol.

Groundwater

Only one study so far deals with the presence of plastic particles in groundwater (as expected) with lower concentrations than other waters. In nine ground water samples between 0 to 7 particles per m³ were found.

Tap water and bottled water

Research dealing with the presence of microplastics in tap water is yet quite limited. Only a few publications (scientific papers as well as reports) exist. According to these publications, the particle number in drinking waters ranges between no detection of particles (LOD >10 μ m) to 300 - 600 particles per litre (LOD >1 μ m). It was noted that) the reported concentration is close to the limit of quantification (LOQ of particles per litre). However, it is quite clear from the reports that the size strongly influences the number of particles. Here, smaller particles are generally detected in higher numbers, but smaller particles are more difficult to detect.

It is also important to notice that water treatment plants for tap water have a removal efficiency of roughly 80%. In treated water particles concentrations (>1 μ m) between 300 and 700 particles per litre can be found. In raw water the concentrations vary between 1500 and 3600 particles per litre. Presence of particles in treated water , may be the result of abrasion from pipes in the distribution nets, but this is yet not clear. Here, more detailed studies are needed to get a better insight and a better idea of the uncertainties around these figures. This may eventually result in the necessity to change treatment methods.

Bottled water was also analysed for the presence of plastic particles. Here, the numbers vary between 2700 to 6330 particles per litre for particles larger than 1 μ m. In another study only particles larger than 5 μ m were counted, resulting in 11 to 50 particles per litre. This indicates that there is more plastic in bottled water than in tap water. This may be *e.g.* due to the fact that plastic shrew caps are being used to seal bottles. These are prone to releasing plastic particles which will partly end up in the water. Also here, more detailed studies on the exact numbers, sources of plastics and uncertainties are needed.

Hazards and risks

Based on the first observations in the scientific reports, the exposure of organisms and humans to plastic particles in the environment cannot be neglected. However, several experts stated "*The number of papers is growing exponentially in this field, but knowledge is not growing at the same rate*" [3]. This finding is underlined by the fact that is yet unclear what the daily intake of microplastics is and that for nanoparticles this type of data is lacking totally. The knowledge on exposure and effects is essential for estimating health risks. There is some evidence that plastic dust causes inflammatory reactions in animal models, but it is unclear how these studies relate to humans and how it specifically relates to plastics in drinking water. Chemicals on and in plastic particles may contribute to exposure via water. Yet, the impact that the shape of a particles has, is yet also unknown for microplastics. In conclusion, current input on exposure and effects are very limited. The current outcome is too unreliable to perform risk calculations. This underlines a clear need for improved sampling and detection

Current outlook

methods, exposure quantifications and effect studies.

It can be expected that the increased media attention will permanently influence public opinion about plastics. Drinking water companies can respond to this by providing data, while they are more and more able to collect these data themselves. Additionally, communicating in a transparent manner about the uncertainties is necessary, especially in the case of water, food and human health.

Moreover, there seems to be a consensus and momentum for action. There is no *plastic denial* on this subject, in contrast to climate change, for example. It is clear to everyone that human activities are the only source of the plastics in the environment. Economic activity causes a constant emission to the environment and this may put also the water sector under (increased) pressure. It is also realistic that it takes some time before all information is available to provide a risk analysis of plastic particles in the environment and waters in particular.

In all, not only the water sector but also citizens and policymakers think about taking further actions, most likely without waiting for detailed risk assessments.

Contents

Summary 2				
Contents 5				
1	Introduction	6		
1.1	General	6		
1.2	Plastic production and the environment	7		
1.3	From ocean to land	7		
1.4	Media attention and the water sector	8		
1.5	This report	9		
2	Plastics types and analyses	10		
2.1	Plastic types	10		
2.2	Detection of microplastics	11		
2.3	Improving methods and data availability	12		
3	Waste water	13		
4	Riverine samples	15		
5	Lake samples	19		
6	Groundwater	21		
7	Drinking water	22		
7.1	Bottled water	22		
7.2	Tap water	25		
7.3	Plastic in tap water in Norway and Denmark	27		
7.4	The Guardian as conducted by Orb Media	28		
7.5	Research in the Netherlands	29		
8	Hazards and risk assessment	30		
8.1	Risk assessment	30		
8.2	Chemicals related to plastic	30		
8.3	Plastics in drinking water	31		
8.4	Conclusions	32		
8.5	Outlook	32		
9	Literature	33		
Attacl	nment I - Locations and concentrations of plastic			
(in se	lected literature)	36		

1 Introduction

1.1 General

Over the last few years, the issue of plastics in the environment have been a growing concern to the public and the scientific community [4-9]. In 2016 the world-wide plastic production has reached 335 million ton per year of which 60 million tons are produced in Europe and global production is still mounting (Figure 1). Half of this—3900 Mt—was produced in just the past 13 years [2]. Much of this plastic is produced for packaging and will outlive its usefulness quickly after production. Then, it will become waste that is often neither fully collected nor recycled. It is estimated that in 2010 4.8 to 12.7 million ton of the produced plastic were released into the oceans by 192 coastal countries [10].



Figure 1: Global and European plastic production (in million metric tonnes per year) . Source via this <u>link</u>, as seen in January 2019:

Currently it is estimated that the amount of plastic ending up the oceans will have increased by an order of magnitude in 2025 if we continue to produce and handle plastic waste like this (Figure 2). This means the total amount of plastic in the ocean might rise even further. The most common reason for plastic showing up in the environment is mismanaged waste, illegal dumping and littering followed by abrasion of plastic, as depicted in Figure 1. In this graph, the trend in research is showing opposite from the flow of plastic as the first findings of the plastics in the environment were at the open sea on the so-called North Pacific central gyre, a place where plastic was accumulating [11]. Later, this fact became noted by the author as the 'Plastic Soup'.



Figure 2: Cumulative plastic waste generation and disposal (in million metric tons). Solid lines show historical data from 1950 to 2015; dashed lines show projections of historical trends to 2050, as taken from [2]

1.3 From ocean to land

Yet, it is more and more clear that plastic will not only litter the oceans, but it may affect animals and plants in other aquatic environments too. Plastic litter has appeared almost everywhere in the environment and that they may pose a threat to flora and fauna [6, 12-14]. Additionally, it will undergo changes. Plastic particles may break down into smaller particles until they are as small as a few millimetres. These smaller plastic particles are considered to be microplastics. If they further break down to reach a size below ca. 100 nm they sometimes labelled as nanoplastics]. Plastic in general has shown to have a direct effect on marine organisms and animals by entanglement and uptake from large particles in (marine) mammals [15]. Smaller particles are suspected to *e.g.* disrupt the function of the guts [14].

Microplastics everywhere

High amounts of microplastics have been found not just in the sea and on beaches, but also in rivers and soils around the world, demonstrating how pervasive this modern pollution is. Sources include leakage from landfills, plasticulture, littering, and sewage sludge. Data from (1).



Figure 3: Figure taken from [1]: High amounts of microplastics have been found not just in the sea and on beaches, but also in rivers and soils around the world, demonstrating how pervasive this modern pollution is. Sources include leakage from landfills, plasticulture, littering, and sewage sludge. Data from [2], credits: GRAPHIC: N. DESAI/SCIENCE

1.4 Media attention and the water sector

All this attention on the issue of plastics in the environment led to (micro)plastics being put on the policy agenda on several national and international governments (e.g. Dutch Parliament (*"Kamerbrief over maatregelen tegen microplastics"*, June 2018 via <u>link</u>) and the Dutch Health Council [16], but also the European Union (Oct. 2018, via <u>link</u>). Especially due to media attention in recent years, people at home are also increasingly aware that the use of plastic is problematic, not just from an environmental point of view but also because single-use plastic is a squander of resources.

Therefore, it can be expected that the water utilities and policy makers (among which the Dutch and Flemish) will be confronted with questions regarding this matter from customers and policy makers may respond too. To answer adequately to these questions, a clear overview of the current state of affairs is necessary. This means it is important to know the prevailing state of tap water (sources) and to identify sources of (micro) plastics.

Just recently, by the end of 2018 and early 2019, several key reports came out that were taken into account, including those specific on drinking water occurrences [17-19] and the state of the current scientific debate by SAPEA, directly feeding European policy bodies [3].

1.5 This report

Aim of this study is to present an overview of the current knowledge regarding microand nanoplastics in waters, sources for drinking water as well as tap water. Therefore, this report will focus on the presence of microplastics in fresh water systems. Information on the presence of plastic in the marine environment is exhaustively reported on here [20-22]. In the chapters to come firstly publications dealing with plastics in various water sources will be discussed. The last chapter will deal with the presence of plastic in drinking water (tap and bottled water) and identify some conclusions and outlook for the future

2 Plastics types and analyses

2.1 Plastic types

The most common types of (nano- or micro-) plastics are shown in Table 1. Each type of plastic has a different field of employment. Therefore, some plastics are more prone to show up in the environment than others. Its application and characteristics of the plastic will also determine how plastic will spread and may be found in the environment.

Table 1: Common types of plastics and their application

Chemical structure	Most common use
	Packaging, furniture, bags, beads
	Tableware, foam peanuts, packaging
	Pipes, caps, containers, straws
	Bottles, Fibres
	toothbrush, fishing nets, clothes
	Wraps, pipes, floor
$\begin{bmatrix} CI \end{bmatrix}$	Foam, elastomers, paints
	$\begin{bmatrix} Chemical structure \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\$

Environmental detection of microplastics is comprised of four equally important parts.

- 1) sampling,
- 2) sample treatment,
- 3) measurement and
- 4) data evaluation.

Each of these steps has a significant impact on the final data. If one of these steps is not executed carefully, the final data can be difficult to interpret. The problem is that for none of these steps, a standard (protocol) has been developed. Such as protocol would need to describe exactly how samples (depending on the type of sample) have to be taken, treated, measured and how the data needs to be reported (*e.g.* in particles per litre).

At first, sampling, then (2) sampling treatment and (3) measurements are timeconsuming (yet), laborious and therefore still far-off being called routine. This means that at the moment there is no unified method that would be acceptable for routine measurements. Currently, in the Netherlands, such sampling protocol is developed in close collaboration between academia, water sector and laboratories (among which KWR Watercycle Research Institute, water laboratories and drinking water companies within the Joint Research Program BTO (Bedrijfstakonderzoek).

Additionally, for (3) measurement and (4) data evaluation a range of techniques are being employed, which all produce different data output. Measuring the same sample with two different techniques will result in two different results and yet difficult to compare. This will become apparent when looking at the techniques, which are currently being used and still further improved. In Table 2 these analytical techniques are being presented and Appendix I presents an overview of results in the literature with differing units. However, each technique has its advantages and disadvantages to provide specific information on plastic particles in the environment. The ultimate consequence is that in many cases reports and scientific publication cannot be compared easily, hence data evaluation (4) is difficult, also for the purpose of reviewing this data for this study.

Most commonly used techniques at the moment are Fourier transformation infrared (FT-IR) microscopy, attenuated total reflection infrared (ATR-IR) and optical microscopes. These methods cover different size ranges of plastic particles. The result of this is that different numbers of plastics can be found. The outcome of the analysis by various techniques can differ by three orders of magnitude. In combination these techniques are able to detect particles larger than 20 µm. However, often only one of these techniques is being used. Other methods, including thermal gravimetric analysis (TGA) and pyrolysis do not give information on the particle number but are able to report on the amount of plastic and specific types of plastic. This makes it even more difficult to compare data. Currently, a lot of work is being done on improving these techniques and to increase the comparison and reliability.

2.3 Improving methods and data availability

From our studies on the different references, we have encountered that thorough reporting on specific sampling conditions and how the sample work up has been done is needed (including procedural blanks) to ensure that data can be compared or discrepancies can be explained. A useful tool for that is to have round robins or interlaboratory studies. These would be extremely helpful to assess the quality of data from the laboratories. However, there are yet no validated and standardised methods, let alone sufficient reference materials. Therefore, for the time being comparison of data from different studies is still difficult and the use of reference samples or materials is recommended. Yet, we present an overview of data in the following chapters.

Table 2: Most commonly employed methods to detect (micro)plastics.

Technique	Size range	Additional information
FT-IR microscopy [23] Raman microscopy [24]	10 to 500 μm 2 - 500 μm	Type of plastic, particle number, shape Type of plastic, particle number, shape Type of plastic, particle number, shape
an optical microscope [23] TGA – analysis [25] Pyrolysis [23]	any any	Type of plastic, particle number, shape Type of plastic, amount of plastic (mass) Type of plastic

3 Waste water

Several studies look at the microplastic contamination of influent and effluent of waste water treatment plants (WWTP). For example, researchers aimed to get information on the removal efficiency of WWTP with regard to microplastics [26]. Here, they measured particle concentrations in influent and effluent and found an average concentration of 650 p/L and 2 p/L, respectively (Figure 4). This translates into a removal efficiency of more than 99%. However, as this particular WWTP discharges each day 270,000 m³ of treated waste water, this means that still each day 540,000,000 p/m³ are released into the environment, from this plant alone. Under several assumptions, for example that particles are perfectly round and that the majority is LDPE (low density PE) with a mean diameter of 20 μ m this would result in about 2 g/d of plastic still coming from the plant. Of course, this figure is far from a precise measure but gives an idea of the total mass outflow.





For a study in the Netherlands Leslie *et al.* looked for plastic particles (10 to 5000 μ m) in the influent, effluent and sludge samples from several WWTP (Heerenvliet, Houtrust, Amsterdam and Westpoort). They found that the effluents of these four WWTP contained between 39 to 58 p/L, while the particle number in influent of Heerenvliet was 68 p/L and 910 p/L in the WWTP Westpoort. The influent of Amsterdam was not measured. In sludge of Westpoort and Amsterdam they found between 510 and 760 p/kg (wet weight), respectively.

For his master thesis Erich analysed effluent from two different WWTPs that discharge into the river Meuse [27]. All particles larger than 300 μ m were analysed. In the WWTP in Maasbommel he found 46 particles per m³ (1 PP and 45 PE). In the effluent of the WWTP in Oijen between 211 and 1487 particles per m³ were found. The vast majority were PE followed by PP.

In another study Mintenig et al. found in 12 different WWTPs particle numbers between 0-50 particles per m³ (> 500 µm) and 10-9000 particles per m³ (< 500 µm) [28]. The most frequent plastic type was PE. Also fibres, predominately polyester (PEST), were found in the effluent. Their numbers ranged from 90 to 1000 per m³. These numbers can be translated into an annual discharge of about $9x10^7$ to $4x10^9$ particles and fibres per WWTP (see Figure 5).



Figure 5: Synthetic fibres in treated waste water. A) Percentage composition of synthetic polymers, B) Annual load, C) Number of fibres per cubic meter. Source: reference [28].

4 **Riverine samples**

One of the first comprehensive studies that tried to give an insight into the presence of plastic pollution in the two major rivers (rivers Rhine and Meuse) in the Netherlands was conducted by W. Urgert in 2014 and 2015 [29]. It was found that in the size range of 0.125 – 5 mm the average concentration for plastic in the river Meuse was 0.14 mg or 9.7 particles per m³. In the river Rhine on average 0.56 mg or 56 particles per m³ were detected. The types of plastics varied in chemical composition and shape. Figure 6 shows a few examples of found plastic particles.



Figure 6: Clockwise: Plastic films found near Lobith, plastic spherules found near Lobith, miscellaneous plastics particles found in the river and transperant spherules found in the Rhine.

At about the same time researchers in Germany wanted to know how much plastic can be found in the sediments of the rivers Rhine and Main in Germany in the state of Rhineland Palatinate [30]. Therefore they took samples at different location to get an insight into the special distributions of plastics along these rivers. Figure 7 shows the sample locations and the amount of plastic that had been found. A closer look at the data shows that the largest size fraction contributes most to the total weight of plastic, whereas the largest number of particles stems from the smallest size fraction. Furthermore, it is interesting that the site with the most particles is not in the vicinity of a sewage treatment plant and sites that are close to or downstream of a sewage treatment plant do not contain significantly more plastics than others sites. This is remarkable as sewage treatment plants are known to be sources of microplastics [31, 32].



Figure 7: Top: Sample location along the rivers. Grey area's are ubranised or settlement areas. Arrows indicate sewage treatment plants. Middle: Mass fractiot of plastics subdivided into different size categories. Bottom: Numerical abundance of plastics particles subdivided into different size cateogies [30].



Figure 8: (A) Microscopic image of the 63–200 μ m size fraction. Particles were measured, and the size is displayed in micrometers (μ m) in parentheses (one micrometer is one thousand of a millimeter). The particles are classified as fragments (F), spheres (Sp), and fibers (Fi). (B) Composition of separated microplastics by shape. Results are displayed as the average abundance of each size fraction of randomly picked sediment samples. Numbers of particles (n) classified for each size fraction were 382 (63–200 μ m), 434 (200–630 μ m), and 210 (630–5000 μ m). (C) Average relative abundance of polyethylene (PE), polypropylene (PP), polystyrene (PS), polyamide (PA), and other polymer types identified in sediments of all sampling sites. Data are plotted as the abundance of weight and as the abundance of particle numbers [30].

For this research also the types of plastics were identified, both chemical composition and the type of plastic (sphere, pellet, fibre). Taking a closer look at particle numbers reveals that polystyrene outnumbers the other types of plastics by far. In case that weight is decisive, PE outweighs the other types of plastics significantly. There is also a difference noticeable when one is looking at the shape of the plastic and the size fraction. In the smaller category the beads/spheres dominate, where as in the larger fraction fragments and pellets are the largest group.

In another publication researchers report on particle concentrations between 5-55 mg/m³ (100–1300 p /m³) in riverine sediment and 2-70 mg/m³ (10 – 650 p/m³) in the river itself [33]. The most abundant plastic types in the sediment and the water were PE and PP, both 29%.



Figure 9: Typical microplastic categories in the Rhine. Left: Duisburg sample consisting of 65% opaque spherules, further fragments and fibres, bar: 2 mm. (a/b) transparent spherules with gas bubbles, polymethyl-methacrylate (Zuilichem), bars: 1 mm; (c/d) opaque spherules, polystyrene (Duisburg, Rees), bars: 500 µm [34].

5 Lake samples

In 2018 Sighicelli *et at.* conducted a survey to get information the presence or absence of microplastics in three lakes in Italy, Largo Maggiore, Iseo and Garda. The highest number of plastic particles (300 μ m - 5mm) found was 57,000±36000 and the lowest 4000±2700 p/km². On average the three lakes have the same abundance of particles (15,000 - 40,000 p/km²). As an example of the diverse units in which plastics are reported, this is another example deviating from the concentrations as earlier stated).

A closer look at these particles reveals that the majority of particles that can be found are polyethylene- (40 - 50%) and polystyrene-based (9 - 25%). These two types of plastic already account for more than 50% of all plastics found. Categorising the particles also reveals that the largest fraction is fragments followed by microbeads.

In another study in the UK researchers took sediment samples in the urban lake Edgbaston Pool in central Birmingham. They found 250-300 particles per kilogramme dried sediment. The most common particles were fibres and films. For this research they also looked at plastic debris (Figure 10). In Figure 11 it can be seen that the sites with the highest debris amount coincide with the sites with the highest abundance of microplastics in sediment. If the microplastics are categorised by their shape, the by far largest two fractions are fibres and films. Incidentally also fragments and foam particles can be seen. The debris contain e.g. bottle caps, plastic bags, bottles, syringes and ropes.



Figure 10: Clockwise from top left. Photograph of the Edgabston Pool looking north; a Mute swan's (Cygnus olor) nest at the southern end of the lake incorporating plastic debris; retrieval of a heavily biofouled plastic bag retrieved from the northern end of the lake during sampling; accumulation of debris at the southern end. [35]



Figure 11: Top: Macroplastic distribution in Edgbaston Pool. (a) Number of debris items found at each sampling location and (b) abundance divided by debris type. Bottom: Microplastic concentrations (number particles/100 g dried sediment) in the surface sediments of Edgbaston Pool. (a) Microplastic films and (b) fibres.

6 Groundwater

The amount of plastic in groundwater is low.

In recent research, several water samples along the tap water production line were sampled and probed for the presence of microplastics [18]. In total 24 water samples were taken of which 9 were groundwater samples. The number of microplastic particles ranged from 0 to 7 per m³. In none of the tap water samples plastic particles were detected. Only in the water meter between 1 and 3 particles per m³ were found. These particles can be originate from material used in the distribution network.



Figure 12: Microplastics identidied in (1) raw water, (2) water treatment plant outlet, (3) water meter and (4) a selected household tap. In Holddorf also 3 different groundwater wells were sampled. [18]

Sources for these microplastics in the water meter could be – due to abrasion - pipes and other plastic base materials used in the water treatment process [18].

A possible source for plastic in groundwater was given recently in two studies about earthworms [36, 37]. The invertebrates seem to be able to transport microplastics to greater depths and may influence groundwater resources. The plastic particles can be incorporated into the soil via cast or burrows. As a consequence the chance for plastic to appear in groundwater could be rising.

7 Drinking water

It is essential to include particles smaller than 10 μm into the research.

7.1 Bottled water

Recently, two scientific studies dealt with the presence of plastic particles in bottled water. For this research the groups of Oßmann *et al.* [38] and Schymanski *et al.* [39] investigated various types of bottles (single use, reusable and glass bottles). Oßmann *et al.* could show that in the size range from about 1.5 μ m to more than 10 μ m between 2649±2857 and 6292±10521 plastic particles per litre can be found (Figure 13). It can be noted that error bars exceed the values, resulting in errors below zero. This indicates the uncertainty.

What can be learned from these graphs is that the particles *smaller than* 5 µm make up between 80 to 95% of the total number of particles, confirming the observation as earlier stated (the smaller the particles to detect, the higher the numbers will be). With regard to the type of plastic, PET does -not unsurprisingly- constitute the largest group in PET bottles, whereas PE is the main plastic in glass bottles. The origin of PE in glass bottles is most likely the (plastic) screw cap.





The group of Schymanski *et al.* found that in the size range of 5 μ m to more than 100 μ m between 14±14 to 118±88 particles per litre can be found (Figure 14). This is in accordance with the findings of Oßmann *et al.* In their study particle larger than 5 μ m represent 1.7% and 5% of the total particle number, which translates into about 50 to 300 particles per litre. Also in the study by Schymanski *et al* the most abundant plastic is PET in PET-plastic bottles. In this study, however, PET is also present in glass bottles in the same amount as PE, which implies that in that case probably PET screw caps have been used.

When these two studies are compared, the necessity to include particles as small as possible in the research is emphasised. The smaller the particle size is, the larger the particle number gets. Therefore, it can be assumed that the number of particles smaller than 5 μ m, will outnumber the larger particles.





Figure 14: (Top) Mean microplastic content (> 5 μ m) of water from different packaging. Different letters above the error bar indicate significant difference; same letter indicate no significant difference (p < 0.05). Error bars represent ±1 standard deviation. (Bottom) Polymer distribution of the microplastics found in water from different packaging types. [39]

7.2 Tap water

So far merely three peer-reviewed scientific study deal with the presence of plastic particles in drinking water. The study by Mintenig *et al.* was already mentioned in section 5 [18], as it also covers groundwater. In this study particles larger 20 μ m were of interest. In none of the tap water sample plastic particles were found. Merely, in the water meter in some of the samples about 1 particle per m³ was found. The particle size ranged from 50 to 150 μ m. These authors stated that "*the abrasion of plastic equipment used during water purification or transport is a likely explanation for the plastic particles detected in water samples*" [18].

In another study by Kosuth *et al.* particle number concentrations (100 μ m - 5 mm) between 0 and 61 p/L were found [40]. This translates into an average of 5.45 particles per litre. The majority (98.3%) of these particles were fibres. For this research 159 samples of tap water from various countries were analysed. The highest number of plastic particles per litre were found in samples from Uganda (26), Ecuador (24), Lebanon (21) and the USA (33). In European tap water the lowest numbers were found (1-8 particles per litre). In this study no information on the type of plastic is given, though.

A third study took a closer look at a water treatment plant (WTP) [19]. Pivokonsky *et al.* found that in treated drinking water (outflow of the WTP) between 338 ± 76 and 628 ± 28 particles per litre can be found (Figure 15). The largest fraction by number is the size fraction between 1 and 10 µm. In comparison only small amounts of particles between 10 and 50 µm can be found. Larger particles are entirely absent. These larger particles are still present in untreated drinking water, however. A comparison of raw (inlet WTP) and treated drinking water shows that the particle number is reduced by 83% on average and that the smallest particles will be least removed. The most abundant plastics in treated and untreated are PET, PP and PE (Figure 16). This study also emphasises the need to include the size range below 10 µm when analysis water samples. Especially when the treatment efficiency of WTP is to be assessed as it can be suspected that particles even smaller than 1 µm will also pass through the WTP. Yet, more studies are needed to assess the treatment options for drinking water production more clearly.



Figure 15: Size distribution of microplastics detected in a) raw and b) treated water as taken from [19] Legend: WTP 1 = coagulaltion/flocculation and sand filtration, WTP 2 = coagulaltion/flocculation, sand and granular activated carbon filtration, WTP 3 = coagulation/flocculation, flotation, sand and granulated activated carbon filtration.



Figure 16: Material composition microplastics in raw and treated water. [19]

7.3 Plastic in tap water in Norway and Denmark

Two other non-peer review studies have looked into the presence of plastic in tap water in Norway and Denmark. In Norway the research institute Niva found that in 24 drinking water samples, taken right after the last treatment step, only in one sample an average of 2.4 particles (> 100 μ m) per litre were found [41]. In the other samples the amount of plastics was below the LOQ. When tap water samples were taken from the distribution system in 5 of the 24 samples an average between 2.0 and 3.7 plastics per litre were found. The researchers point out, however, that there is chance that these samples were contaminated due to a prolonged contact with the surrounding air.



Figure 17: Map of the 17 sites selected for analysis of microplastics in Danish drinking water. [42]

The scientists from Aarhus University collected 17 water samples (Figure 17) directly at the tap to look for microplastics larger than 100 μ m [42]. They found that 16 of these 17 samples contained microplastic numbers below the LOQ. Merely one sample showed a particle number slightly above the LOQ. Additionally, they also collected three samples, which were checked for the presence of particles larger than 5 μ m. Also here the particle number was below the LOQ. The results from both studies (Norway and Denmark) coincide as the authors of the Danish study state themselves.

7.4 The Guardian as conducted by Orb Media

Another non-peer reviewed study that got a lot of attention by the media, was published in The Guardian and conducted by Orb Media. In this study several tap waters from around the world were analysed for the presence of plastic (Figure 18). The particle number spanned from 1.9 in Europe and Indonesia to 4.8 in the USA.



Figure 18: Amount of plastic in tap water per litre according to OrbMedia, see link

7.5 Research in the Netherlands

Several actors in the water sector are committed to deliver data on the presence of plastics and risks. Water boards and Rijkswaterstaat are aiming at monitoring plastics, including microplastic in estuaries (e.g. Haringvliet), major Dutch rivers, tributaries and lakes. The parties not only conduct studies in waters, but also focus on sediments. Studies of these are carried out by Institutes as Deltares, Wageningen Marine Research (WUR), KWR Watercycle Research Institute and several academic groups such as Wageningen University (prof. Bart Koelmans, see [47]) and Vrije Universiteit Amsterdam; (e.g. Dr. Heather Leslie, e.g. see [43]. Many of these groups aim for harmonising results and methods, both national with NEN as internationally by linking to ISO and CEN. A complete overview of these studies is beyond the scope of this study.

We shortly highlight a Dutch project from Utrecht and Wageningen University as this is has resulted in sampling and analysis protocols for use in the Netherlands [44]. The full name is "Technologies for the Risk Assessment of Microplastics, abbreviated by TRAMP. Next, this project includes sampling locations that relate to sources for drinking water from surface waters in the Meuse region (Figure below). In all, the TRAMP project aims at (a) developing technologies to detect nano- and microplastics in environmental samples, (b) developing technologies to assess fate, hazards and effects of plastic in the freshwater environment including the possible abatement options, and (c) providing a prospective assessment of the present and future risks of plastic in the freshwater environment. The results from these studies come available in 2019 and will include sampling station in the Meuse region (see figure).



Figure 16: Sampling stations in the TRAMP project, as sampled in 2018 (source: Svenja Mintenig, PhD candidate Utrecht University).

8 Hazards and risk assessment

8.1 Risk assessment

As with other chemical and microbial threats to the human health, the risk assessment combines a hazard and exposure evaluation. On the hazards side, the effects of plastic particles are mostly studied on ecological endpoints. Human health risk assessment for nanoplastics has not yet been published. Here, reliable data on the concentrations of nanoplastics in environmental compartments are limited. Therefore, exposure can also not be quantified. On the hazards or effects side, there is only some data available, but the criticism is on the lack of clear dose-effect relationship. Furthermore, the limited studies use synthesized nanoplastics, most often nanosized polystyrene, and it is unknown how well these represent nanoplastics that occur in the environment [45].

8.2 Chemicals related to plastic

Microplastics contain additives, compounds that are added to give certain desired characteristics, e.g. hardness or flexible properties. Flame retardants may be added to be heat and fire resistant. Next to the additives, microplastics are known to sorb compounds with some focus on Persistent, Bioaccumulative and Toxic compounds (PBT compounds). These compounds may be sorbed from water. Then, the particles act as carriers and/or vectors to biota via uptake (see [46] and references therein), while some authors state that this route is very limited [47]. Examples of compounds that are studied are PAHs, DDT, PFOA, and DEHP. In a recent article, a first attempt is made to perform estimations from exposure via drinking water to diverse chemicals from in particles [17]. Concentrations in drinking waters as calculated in their studies would contribute to a relative small fraction (<5%) of the total dietary intake of environmental contaminants and plastic additives.

8.3 Plastics in drinking water

As shown in this report, microplastics (with lower size limits of 10-100 µm) have been detected in bottled and tap drinking water. It is therefore clear that humans get microplastics via their diet. A summary of data on drinking water sources is stated in Table 3. Yet, based on a low number of studies on the diet components, the contribution of drinking water seems very limited (e.g. [17]). Furthermore, the quality of studies that detected particles in biota or drinking water is also low, which makes it difficult to draw firm conclusions.

In all, this means that we have no full and balanced view about the occurrence of microplastics in food and drinking water. Nanoplastics are likely to be present in higher numbers but to our knowledge occurrence of microplastics in components of the human diet is not yet established As for nanoplastics in drinking water and food, there is no information at all. This means that currently there is insufficient data to assess risks based on reliable exposure and effect data.

Table 3: Comparision of microplastics abundance in drinking water. Taken from [19] and extended.

Sample source	Size range of analysed particles	Microplastic abundance (particles L^{-1}) F		Reference
		Mean	Range	-
Tap water Norway	>10 µm	< LOQ	< LOQ	[41]
Tap water Denmark	>100 µm	< LOQ	< LOQ	[42]
Tap water Germany	>20 µm	< LOQ	< LOQ	[18]
Mineral water from returnable plastic bottles	>5 µm	118 ± 88	28-241	[39]
Mineral water from single-use bottles	>5 µm	14 ± 14	2-44	[39]
Mineral water from glass bottles	>5 µm	50 ± 52	4-156	[39]
Mineral water from beverage cartons	>5 µm	11 ± 8	5-20	[39]
Mineral water from single-use PET bottles	>1 µm	2649 ± 2857	90-9311	[38]
Mineral water from reusable PET bottles	>1 µm	4889 ± 5432	0-11,301	[38]
Mineral water from glass bottles	>1 µm	6292 ± 10,521	813-35,462	[38]
WTP1 raw water	>1 µm	1473 ± 34	1383-1575	[19]
WTP1 treated water	>1 µm	443 ± 10	369-485	[19]
WTP2 raw water	>1 µm	1812 ± 35	1648-2040	[19]
WTP2 treated water	>1 µm	338 ± 76	243-466	[19]
WTP3 raw water	>1 µm	3605 ± 497	3123-4464	[19]
WTP3 treated water	>1 µm	628 ± 28	562-684	[19]
Tap water Europe	unknown	3.4	-	[48]
Tap water USA	unknown	9.6	-	[48]
Tap water Europe	>100 µm		1-8	[49]
Tap water USA	>100 µm		33	[49]

8.4 Conclusions

In due time, it is expected that more data becomes available to determine the relative importance of intake via drinking water and other foodstuff in comparison to inhalation for example. Based on the first observations in the scientific reports, the exposure of organisms and humans to plastic particles in the environment cannot be neglected. However, several experts stated "*The number of papers is growing exponentially in this field, but knowledge is not growing at the same rate*" [3]. This finding is underlined by the fact that is yet unclear what the daily intake of microplastics is and that for nanoparticles this type of data is totally lacking. At this moment, the same lack of information is on the (drinking) water treatment options while it is expected that studies will become available.

The current increased attention on plastics in science and public debate is clear, while the data to perform a reliable and evidence-based risk assessment is yet limited. The current lack of insight into the risks does not mean that the risk is absent, especially on the nanoplastic. We simply cannot state this with sufficient certainty. This underlines a clear need for further method development, especially validation of the different methods to quantify exposure and effect. Also, the application of current developed protocols from sampling to detection and data analysis need to be applied to achieve a better understanding on the occurrence and variations.

8.5 Outlook

As can be expected, the increased media attention will influence the public opinion about plastics. All kind of organizations play a role in understanding the plastics and give a basis for effective policy interventions as the growing pressure on water resources is eminent. In the near future, the role for drinking water utilities may be to provide sound data, as soon as it will become available. In this manner, utilities may play the role of information source when customers have questions on occurrences, effects and risks. An important and interesting conclusion in light of drinking water production by the SAPEA working group was that communicating in a transparent manner about the uncertainties in the scientific evidence is a safer approach than assuming a lack of risk, especially in sensitive domains such as food and human health [3].

The issue of plastics in the environment has a special dimension. More specific, humans alone are the reason why plastics end up in our environment. There seems a consensus and momentum for action and no evidence of 'plastic denial' (as opposed to climate change denial) [3].

To end, drinking water companies may underline the fact that economic activity drives a constant emission of plastics to the environment and that this will put a pressure on the water sector (as comparable with other activities and pressures). Moreover, an understanding and risk perception of the issue for society will fuel citizens and policy makers to take further actions, most probably also without information on specific and detailed risk assessments. Not only the media and public but also politics may develop as scientific reports become more and more available. BTO 2019.006 | Mei 2019

9 Literature

- 1. Rochman, C.M., *Microplastics research—from sink to source*. Science, 2018. **360**(6384): p. 28-29.
- 2. Geyer, R., J.R. Jambeck, and K.L. Law, *Production, use, and fate of all plastics ever made.* Science Advances, 2017. **3**(7): p. e1700782.
- 3. SAPEA, *A Scientific Perspective on Microplastics in Nature and Society*. 2019, SAPEA, Science Advice for Policy by European Academies: Berlin.
- 4. Silva, A.B., et al., *Microplastics in the environment: Challenges in analytical chemistry A review.* Analytica Chimica Acta, 2018. **1017**: p. 1-19.
- Klein, S., et al., Analysis, Occurrence, and Degradation of Microplastics in the Aqueous Environment, in Freshwater Microplastics : Emerging Environmental Contaminants?, M.
 Wagner and S. Lambert, Editors. 2018, Springer International Publishing: Cham. p. 51-67.
- 6. Li, J., H. Liu, and J. Paul Chen, *Microplastics in freshwater systems: A review on occurrence, environmental effects, and methods for microplastics detection.* Water Research, 2018. **137**: p. 362-374.
- 7. Prata, J.C., *Microplastics in wastewater: State of the knowledge on sources, fate and solutions*. Marine Pollution Bulletin, 2018. **129**(1): p. 262-265.
- 8. Burgess, R.M., et al., *Microplastics in the aquatic environment—Perspectives on the scope of the problem.* Environmental Toxicology and Chemistry, 2017. **36**(9): p. 2259-2265.
- 9. Dumichen, E., et al., *Fast identification of microplastics in complex environmental samples by a thermal degradation method.* Chemosphere, 2017. **174**: p. 572-584.
- Jambeck, J.R., et al., *Plastic waste inputs from land into the ocean*. Science, 2015.
 347(6223): p. 768-771.
- 11. Moore, C.J., et al., *A Comparison of Plastic and Plankton in the North Pacific Central Gyre.* Marine Pollution Bulletin, 2001. **42**(12): p. 1297-1300.
- von Moos, N., P. Burkhardt-Holm, and A. Köhler, Uptake and Effects of Microplastics on Cells and Tissue of the Blue Mussel Mytilus edulis L. after an Experimental Exposure. Environmental Science & Technology, 2012. 46(20): p. 11327-11335.
- 13. Wan, J.-K., et al., *Distribution of Microplastics and Nanoplastics in Aquatic Ecosystems and Their Impacts on Aquatic Organisms, with Emphasis on Microalgae*, in *Reviews of Environmental Contamination and Toxicology Volume 246*, P. de Voogt, Editor. 2019, Springer International Publishing: Cham. p. 133-158.
- 14. Jin, Y., et al., *Impacts of polystyrene microplastic on the gut barrier, microbiota and metabolism of mice.* Science of The Total Environment, 2019. **649**: p. 308-317.
- Ryan, P.G., Ingestion of Plastics by Marine Organisms, in Hazardous Chemicals Associated with Plastics in the Marine Environment, H. Takada and H.K. Karapanagioti, Editors. 2019, Springer International Publishing: Cham. p. 235-266.
- 16. Netherlands), G.H.C.o.t., *Briefadvies Gezondheidsrisico 's van microplastics in het milieu*. 2016. p. 6.
- 17. Eerkes-Medrano, D., H.A. Leslie, and B. Quinn, *Microplastics in drinking water: A review and assessment*. Current Opinion in Environmental Science & Health, 2018.
- 18. Mintenig, S.M., et al., *Low numbers of microplastics detected in drinking water from ground water sources.* Science of The Total Environment, 2019. **648**: p. 631-635.
- 19. Pivokonsky, M., et al., *Occurrence of microplastics in raw and treated drinking water.* Science of The Total Environment, 2018. **643**: p. 1644-1651.
- Van Cauwenberghe, L., et al., *Microplastic pollution in deep-sea sediments*. Environ. Pol., 2013. 182(0): p. 495-499.
- 21. Hidalgo-Ruz, V., et al., *Microplastics in the marine environment: A review of the methods used for identification and quantification.* Environ. Sci. Technol., 2012. **46**(6): p. 3060-3075.

- Andrady, A.L., *Microplastics in the marine environment*. Marine Pollution Bulletin, 2011.
 62(8): p. 1596-1605.
- 23. Mintenig, S., et al., *Closing the gap between small and smaller: Towards an analytical protocol for the detection of Micro- and Nanoplastics in freshwater systems*, in *MICRO 2016*. 2016: Lanzarote.
- 24. Käppler, A., et al., *Identification of microplastics by FTIR and Raman microscopy: a novel silicon filter substrate opens the important spectral range below 1300 cm-1 for FTIR transmission measurements.* Analytical and Bioanalytical Chemistry, 2015.
- 25. Majewsky, M., et al., *Determination of microplastic polyethylene (PE) and polypropylene (PP) in environmental samples using thermal analysis (TGA-DSC).* Science of the Total Environment, 2016. **568**: p. 507-511.
- 26. Talvitie, J., et al., *How well is microlitter purified from wastewater? A detailed study on the stepwise removal of microlitter in a tertiary level wastewater treatment plant.* Water Research, 2017. **109**: p. 164-172.
- 27. Erich, M., *Microplastics determination in freshwater samples*, in *Freshwater and Marine Biology*. 2018, University of Amsterdam: Amsterdam. p. 22.
- 28. Mintenig, S.M., et al., *Identification of microplastic in effluents of waste water treatment plants using focal plane array-based micro-Fourier-transform infrared imaging.* Water Research, 2017. **108**: p. 365-372.
- 29. Urgert, W., Microplastics in the rivers Meuse and Rhine. 2015, Open Universiteit.
- Klein, S., E. Worch, and T.P. Knepper, Occurrence and Spatial Distribution of Microplastics in River Shore Sediments of the Rhine-Main Area in Germany. Environmental Science & Technology, 2015. 49(10): p. 6070-6076.
- 31. Kay, P., et al., *Wastewater treatment plants as a source of microplastics in river catchments*. Environmental Science and Pollution Research, 2018. **25**(20): p. 20264-20267.
- 32. van Wezel, A., I. Caris, and S.A.E. Kools, *Release of primary microplastics from consumer products to wastewater in the Netherlands*. Environmental Toxicology and Chemistry, 2016.
- Rodrigues, M.O., et al., Spatial and temporal distribution of microplastics in water and sediments of a freshwater system (Antuã River, Portugal). Science of The Total Environment, 2018. 633: p. 1549-1559.
- 34. Mani, T., et al., *Microplastics profile along the Rhine River*. Scientific Reports, 2015. **5**: p. 17988.
- 35. Vaughan, R., S.D. Turner, and N.L. Rose, *Microplastics in the sediments of a UK urban lake*. Environmental Pollution, 2017. **229**: p. 10-18.
- 36. Rillig, M.C., L. Ziersch, and S. Hempel, *Microplastic transport in soil by earthworms*. Scientific Reports, 2017. **7**(1): p. 1362.
- 37. Huerta Lwanga, E., et al., *Incorporation of microplastics from litter into burrows of Lumbricus terrestris.* Environmental Pollution, 2017. **220**: p. 523-531.
- 38. Oßmann, B.E., et al., *Small-sized microplastics and pigmented particles in bottled mineral water.* Water Research, 2018. **141**: p. 307-316.
- 39. Schymanski, D., et al., Analysis of microplastics in water by micro-Raman spectroscopy: Release of plastic particles from different packaging into mineral water. Water Research, 2018. **129**: p. 154-162.
- 40. Hermsen, E., et al., *Quality Criteria for the Analysis of Microplastic in Biota Samples: A Critical Review.* Environmental Science & Technology, 2018. **52**(18): p. 10230-10240.
- 41. Uhl, W. and C. Svendsen, *Mapping Microplastics in Norwegian Drinking Water A summary of results and evaluation of suspected health risks*. 2018, Niva, FHI, Norsk Vann.
- 42. Strand, J., et al., *Analysis of microplastics particles in Danish drinking water* 2018, Aarhus University, DCE. p. 34.
- 43. Leslie, H.A., et al., *Microplastics en route: Field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota.* Environment International, 2017. **101**: p. 133-142.

- 44. Mintenig, S.M., et al., *Closing the gap between small and smaller: towards a framework to analyse nano- and microplastics in aqueous environmental samples.* Environmental Science: Nano, 2018. 5(7): p. 1640-1649.
- 45. Gigault, J., et al., *Current opinion: What is a nanoplastic?* Environmental Pollution, 2018. **235**: p. 1030-1034.
- 46. Wang, F., et al., *Interaction of toxic chemicals with microplastics: A critical review.* Water Research, 2018. **139**: p. 208-219.
- 47. Koelmans, A.A., et al., *Microplastic as a Vector for Chemicals in the Aquatic Environment: Critical Review and Model-Supported Reinterpretation of Empirical Studies.* Environmental Science & Technology, 2016. **50**(7): p. 3315-3326.
- 48. Carrington, D., Plastic fibres found in tap water around the world, study reveals, in TheGuardian.
- 49. Kosuth, M., S.A. Mason, and E.V. Wattenberg, *Anthropogenic contamination of tap water, beer, and sea salt.* PLoS ONE, 2018. **13**(4).
- 50. Lechner, A., et al., *The Danube so colourful: a potpourri of plastic litter outnumbers fish larvae in Europe's second largest river*. Environ Pollut, 2014. **188**(0): p. 177-81.
- 51. Dris, R., et al., *Microplastic contamination in an urban area: a case study in Greater Paris.* Environmental Chemistry, 2015. **12**(5): p. 592-599.
- 52. Sighicelli, M., et al., *Microplastic pollution in the surface waters of Italian Subalpine Lakes*. Environmental Pollution, 2018. **236**: p. 645-651.
- Fischer, M. and B.M. Scholz-Böttcher, Simultaneous Trace Identification and Quantification of Common Types of Microplastics in Environmental Samples by Pyrolysis-Cas Chromatography-Mass Spectrometry. Environmental Science & Technology, 2017.
 51(9): p. 5052-5060.
- 54. Ziajahromi, S., et al., *Wastewater treatment plants as a pathway for microplastics: Development of a new approach to sample wastewater-based microplastics.* Water Research, 2017. **112**: p. 93-99.
- 55. Besseling, E., E.M. Foekema, and A.A. Koelmans, *Verkennend Onderzoek Microplastic in het beheersgebied van Waterschap Rivierenland*. 2014, Wageningen University. p. 1-18.
- 56. Loder, M.G.J., et al., *Enzymatic Purification of Microplastics in Environmental Samples*. Environmental Science & Technology, 2017. **51**(24): p. 14283-14292.
- 57. Dubaish, F. and G. Liebezeit, *Suspended Microplastics and Black Carbon Particles in the Jade System, Southern North Sea.* Water, Air, & Soil Pollution, 2013. **224**(2): p. 1352.

Attachment I - Locations and concentrations of plastic (in selected literature)

Table Annex I-1: Selection of river water and sediment samples in NW-Europe with focus on Rhine & Meuse basins and reported information on detected plastic particles (concentrations, density). For more details, see references [xx]. Note that units differ among references. In case the concentrations is given in "per kg" this means per kg dry weight, unless it is stated otherwise (ww, wet weight). *This list is not intended to be complete.*

Type of water [reference]	Lower size class	Moderate	size class	Higher size class
River samples	125 – 5000 µm			< 5000 µm
River Meuse [29]	0.14 mg/m ³			0.14 mg/m ³
	9.7 p/m³			9.7 p/m ³
River Rhine	0.56 mg/m ³			0.56 mg/m ³
	56 p/m³			56 p/m ³
River samples	300 – 5000 µm			<5000 µm
Rhine (CH) [34]	1000 - 3000 p/m ³			1000 - 3000 p/m ³
Rhine (FR)	1000 - 2000 p/m ³			1000 - 2000 p/m ³
Rhine (DE)	1000 - 22,000 p/m ³			1000 - 22,000 p/m ³
Rhine (NL)	3000 - 9000 p/m ³			3000 - 9000 p/m ³
River samples	300 – 5000 µm			<5000 µm
Meuse [27]	1 - 8 p/m³			1 – 8 m ³
River samples	20 – 200 µm	200 - 630 µm	630-5000 µm	<5000 µm
River Rhine sediment [30]	1 - 8 mg/kg	5 - 100 mg/kg	10 - 900 mg/kg	10 - 1000 mg/kg
	200 - 2000 p/kg	100 - 1000 p/kg	9 - 500 p/kg	200 - 50000 p/kg
River Main sediment [30]	2 - 6 mg /kg	2- 60 mg/kg	50 - 600 mg/kg	50 - 600 mg/kg
	600 - 700 p/kg	80 - 600 p/kg	30 - 70 p/kg	800 - 1000 p/kg
River samples	10 – 5000 µm	10 - 300 µm	300 - 5000 µm	< 5000 µm
Meuse Eijsden [43]	1400±520 p/kg	40 %p	60 %p	
Rhine Lobith	4900±540 p/kg	30 %p	70 %p	
Rhine Bimmen	1700±390 p/kg	40 %p	60 %p	
Urban canal	100±49 p/L	61 %p	39 %p	
Urban canal sediment	2071±4061 p/kg	41 %p	59 %p	

Table Annex I-2: Selection of water and sediment samples from European rivers and lakes and reported information on detected plastic particles (concentrations, density). For more details, see references [xx]. Note that units differ among references. In case the concentrations is given in "per kg" this means per kg dry weight, unless it is stated otherwise (ww, wet weight). *This list is not intended to be complete.*

Type of water [reference]	Lower size class	Moderate size class		Higher size class
River samples	55 – 5000 µm			< 5000 µm
River Antuã Water [33]	2 - 70 mg/m3			2 - 70 mg/m3
	10 - 650 p/m3			10 - 650 p/m3
River Antuã sediment [33]	5 – 55 mg/m3			5 – 55 mg/m3
	100 - 1300 p/m3			100 - 1300 p/m3
River samples	500 – 5000 µm			
Danube 2010 [50]	937±8543 p/1000 m ³			
	11±44 g/1000 m ³			
Danube 2012	55±75 p/1000 m ³			
	2±3 g/1000 m ³			
River samples	1000 – 5000 µm			
Seine (Port van Gogh) [51]	0.280 p/m ³			
Marne (Chemine de Halage)	7-108 p/m³			
Seine (Le parc de Belloy)	36-72 items/m ³			
River samples	10 - 5000 µm	>80µm	>300µm	
River Seine [51], see below		0.03-0.1 p/L	0.0005 p/L	
Lake samples	300 – 5000 µm			< 5000 µm
Largo Maggiore [52]	40,000 p/km ²			40,000 p/km ²
Largo Iseo	40,000 p/km ²			40,000 p/km ²
Largo Garda	25,000 p/km ²			25,000 p/km ²
Lake samples	500 – 5000 µm			< 5000 µm
Urban lake sediment [35]	250 - 300 p/kg			
Lake samples	< 300 – 5000 µm			
Lake Bolsena [53]	2.22 p/m ³			

Table Annex I-3: Selection of waste water treatment plants and reported information on detected plastic particles (concentrations, density). For more details, see references [xx]. Note that units differ among references. In case the concentrations is given in "per kg" this means per kg dry weight, unless it is stated otherwise (ww, wet weight). *This list is not intended to be complete*.

Type of water [reference]	Lower size class	Moderate size class		Higher size class
Waste water treatment	10 - 5000 µm			< 5000 µm
Effluent Heerenvliet	58±29 p/L			
Influent Heerenvliet	68± 27 p /L			
Effluent Amsterdam	60± 45 p/L			
Sludge Amsterdam	760 p/kg ww			
Effluent Westpoort	39 p/L			
Influent Westpoort	910 p/L			
Sludge Westpoort	510 p/kg ww			
Effluent Houtrust	55±15 p/L			
	20 - 5000 µm	Rel. Abund. (>30	0, 100-300, 100-20 µm)	
Influent (Finland) [26]	650 p/L	20%, 40%, 40%		
Effluent (Finland)	2 p/L	2%, 28%, 70%		
	60 – 5000 µm			
Effluent [54]	1 p/L			
	10 - 5000 µm	>80µm	>300µm	
Influent [51]	260-320 p/L			
Effluent	14-50 p/L			
River Seine, as above		0.03-0.1 p/L	0.0005 p/L	
	20 – 500 µm	>500 µm	Comment	
Effluent [28]	0-50 p/m ³	10-9000 p/m ³	Average of 12 WWTPs	
	>1 µm		Comment	
WTP raw water [19]	2297±189 p/L	1383 - 4464 p/L	Average of three WTP	Mainly < 10µm
WTP treated water [19]	470±38 p/L	338-628 p/L		Mainly < 10µm
	50 µm – 1 mm	>1mm		
Rural area (river) [55]	1.29±0.56 p/10 m ³	-		
Urban proximity (river)	0.32±0.58 p/10 m ³	0		
Effluent	21.61±18.60 p/10 m ³	5.40 p/10 m ³		
WWTP proximity (river)	0.32±0.56 p/10 m ³	0		
Effluent & river samples	300 – 5000 µm			<5000 µm
Meuse [27]	1 - 8 p/m ³			1 - 8 m ³
Effluent	50 - 1500 p/m ³			50 - 1500 p/m ³

Table Annex I-4: Selection of drinking water related samples and reported information on detected plastic particles (concentrations, density). For more details, see references [xx]. Note that units differ among references. *This list is not intended to be complete.*

Type of water				
	>100 µm	10 - 100 µm	Comn	nent
Tap water (Denmark) [42]	< LOQ	< LOQ	Not peer-reviewed research	
Tap water (Norway) [41]	< LOQ			
	size class unknown		Not peer-review	wed research
Tap water Europe [48]	3.4 p/L			
Tap water USA [48]	9.6 p/L			
	>100 µm			
Tap water Europe [49]	1-8 p/L			
Tap water USA [49]	33 p/L			
	20 – 5000 µm			< 5000 µm
Groundwater [18]	0 - 7 p/m ³			0 - 7 p/m³
Water meter	1 - 3 p/m ³			1 - 3 p/m ³
Tap water	0 p/m ³			0 p/m³
	1.5 – 300 µm [38]	5 – 100 µm [39]		
PET bottles	2649±2857 p/L	14±14 p/L - 118±88 p/L		
Glass bottles	6292±10521 p/L	50±52 p/L		

Table Annex I-5: Selection of North Sea (marine waters) and reported information on detected plastic particles (concentrations, density). For more details, see references [xx]. Note that units differ among references. *This list is not intended to be complete and presented here for sake of comparison to fresh water systems (see Tables Annex I-1 to I-4).*

North Sea	20-5000 µm		
Bremerhaven [56]	0.05 to 4.42 p/m ³		
	80 – 5000 µm	80 – 5000 µm	
Jade System [57]	64±194 particles/L	88±82 fibres/L	

Type of water [references]	Most abundant plastic (%) Weight.	Most abundant plastic Numerical
Rhine/Main [30]	PE(49), PP(26), PS(10),PA(3)	PS (54), PE(23), PP(15), PA(2)
Largo Maggiore [52]	n.a.	PE(48), PP(17), PS(15)
Largo Garda [52]	n.a.	PE(45), PS(24), PP(22)
Largo Iseo [52]	n.a.	PE(41), PS(27), PET(9), PP(5)
Effluent (Maarten)	n.a.	PE(95), PP(3)
River Antuã [33]	n.a.	PE(29), PP(29), PS(9), PET(9)
PET bottles [38]	n.a.	PET(74), PP(10)
PET bottles	n.a.	PET(78), PP (7)
Glass bottles [38]	n.a.	PE(46), PP(23), PS+Butadiene(14)
		PE(35), PET(33), PA(8)
WTP raw water [19]	n.a.	PET (20-70), PP (15-25), PE (0-20%)
WTP treated water [19]	n.a.	PET (25-60%), PP (10-35), PE (0-25%)

Table Annex I-6: Types of plastic found in various samples as reported in selected references [xx].