



The influence of discharge on the observed plastic concentration in the Dutch Rhine Delta

Results of the KOR-net fishing in 2023 during high discharges. Two peaks in concentration were observed during a rising discharge.



Colophon

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Preface

This report was written as a graduation internship for the Earth, Surface & Water Master in Utrecht. The graduation internship took place at Rijkswaterstaat Oost-Nederland within the department Netwerkontwikkeling en Visie (NOV) and team water quality and Ecology under the Supervision of Margriet Schoor.

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Abstract

Since the introduction of plastics in the early 20th century it has proven to be very beneficial to society. However, plastics have a enormous negative impact on the environment since it is a very durable material and stays in the environment for hundreds to thousands of years. An important transport route of plastics are rivers, which eventually flow into the oceans. Most research on plastics are focused on floating and marine plastics. Barely anything is known about the amount of plastic in the river Rhine, since most studies of the Rhine focused on floating plastics. To be able to develop a representative monitoring strategy, the driving processes of plastic transportation should be taken into account. With that information, the source of plastics can eventually be determined and mitigations techniques for reducing inputs can be made. To achieve this, samples were taken of the plastic in the Rhine in 2023 during high discharges since previous sampling only took place during relatively low discharges. The location of the sampling was on the border with Germany and the Netherlands, in a river bend. Considering the high discharges, sampling only took place in the middle and at the surface of the water column on the right side of the river, the outer bend. 72 Samples were collected during this period by trawls hanging from a ship. The captured particles were first counted and categorized according to the River OSPAR-method in order to get a view on what kind of plastics are present in the river. The number concentrations (pieces/1000m³) were calculated for the vertical positions in the water by correcting for the local sampled volume of water during each survey. From all counted and sorted plastic particles, plastic polystyrene pieces of soft plastic accounted for 83.5 percent. These plastic particles were mostly clear pieces of film, which can originate from food packaging, plastic bags or other objects. Significant differences were found in the plastic concentrations in the vertical position of the river. The surface of the water column contained significantly more plastic pieces than the middle, as categories such as styrofoam, hard pieces of plastics and cotton swabs were found mainly at the surface of the water column. The plastic concentration over time did not follow the same curve as the discharge over time. The discharge only rose during the sampling period, where the concentration of plastics showed two peaks, with the second peak occurring just before the peak discharge. The riverbanks were flooded during the entire sampling period and could thus have been a major source of plastics. Whether these plastics were deposited earlier on these riverbanks, or were littered by people is unknown. It is recommended to explore the different heights of the riverbanks in combination with the water level and peaks of the plastic concentrations.

Samenvatting

Sinds de introductie van plastic aan het begin van de 20e eeuw heeft het bewezen zeer nuttig te zijn voor de samenleving. Plastics hebben echter een enorme negatieve invloed op het milieu omdat het een zeer durabel materiaal is en honderden tot duizenden jaren in het milieu blijft. Een belangrijke transportroute van plastic zijn rivieren, die uiteindelijk uitmonden in de oceanen. Het meeste onderzoek naar plastic is gericht op drijvend plastic en plastic in de zee. Er is nauwelijks iets bekend over de hoeveelheid plastic in de Rijn, omdat de meeste onderzoeken naar de Rijn gericht waren op drijvend plastic. Om een representatieve monitoringstrategie te kunnen ontwikkelen, moet er rekening worden gehouden met de verschillende processen die plastic transport beïnvloeden. Met die informatie kan uiteindelijk de bron van het plastic worden bepaald en kunnen mitigatietechnieken worden ontwikkeld om de toevoer te verminderen. Om dit te bereiken werden in 2023 monsters genomen van het plastic in de Rijn tijdens hoge afvoeren, aangezien eerdere bemonsteringen alleen plaatsvonden tijdens relatief lage afvoeren. De bemonsteringslocatie bevond zich op de grens met Duitsland en Nederland, in de buitenbocht. Gezien de hoge afvoeren vond de bemonstering alleen plaats in het midden en aan het oppervlak van de waterkolom aan de rechterkant van de rivier. 72 Monsters werden er in deze periode verzameld met netten die aan een schip hingen. De gevangen plastic deeltjes werden eerst geteld en gecategoriseerd volgens de OSPAR-methode om een beeld te krijgen van het soort plastic dat in de rivier aanwezig is. De plastic concentratie (stuks/1000m³) werd berekend voor de verticale posities in het water. Van alle getelde en gesorteerde deeltjes bestond 83,5 procent uit zacht niet identificeerbare plastic polystyreen stukjes plastic. Deze plastic stukjes waren meestal doorzichtige stukjes folie, die afkomstig kunnen zijn van voedselverpakkingen, plastic zakken of andere voorwerpen. Er werden significante verschillen gevonden in de plasticconcentraties in de verticale positie van de rivier. Het oppervlak van de waterkolom bevatte significant meer plastic deeltjes dan het midden, aangezien categorieën zoals piepschuim, harde stukken plastic en wattenstaafjes voornamelijk aan het oppervlak van de waterkolom werden aangetroffen. De plastic concentratie over de tijd volgde niet dezelfde curve als de afvoer over de tijd. De afvoer steeg tijdens de bemonsteringsperiode, waarbij de plastic concentratie twee pieken vertoonde, met de tweede piek vlak voor de piekafvoer. De rivieroever stonden gedurende de hele bemonsteringsperiode onder water en kunnen dus een belangrijke bron van plastic zijn geweest. Het is niet bekend of deze plastic stukjes eerder op deze oevers zijn afgezet of door mensen als afval zijn achtergelaten. Het wordt aanbevolen om de verschillende hoogtes van de rivieroever in combinatie met de waterstand en de pieken van de plasticconcentraties te onderzoeken.

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1. Introduction

Since the introduction of plastic in the early 20th century it has quickly become evident how beneficial plastics are from previous materials that were used to make appliances and other basic necessities (Napper & Thompson, 2020). The production of plastics has increased from 1.5 million tons in 1950s to 390 million tons in 2022 due to its versatility, durability and low production costs (Berry et al., 2023). The success of plastics has shaped the modern society and numerous benefits are in particular evident in healthcare, transport, construction and packaging (Plastics Europe, 2022). The durability of plastics is however also a downfall, it is non-biodegradable and the longevity is estimated at hundreds to thousand years (Berry et al., 2023). Many of the plastics used by people everyday are single use plastics, like packaging materials, and end up in the marine environment (Jambeck et al., 2015). Around 75 percent of all marine litter are plastics which accumulate on beaches, at the sea surface, in the deep sea and in arctic sea ice as well as accumulation on land and in freshwater habitats (Napper & Thompson, 2020). Weathering of plastic debris can cause fragmentation into particles that marine invertebrates ingest, particles too small to trace to its source and particles being very challenging to remove from the open oceans or rivers. This indicated that the most successful mitigation techniques must reduce inputs (Jambeck et al., 2015). The amount of plastic waste that is accumulating all over the world indicates that plastic pollution can become an environmental hazard and a threat for ecosystems, aquatic life and human health (Thompson et al., 2004).

An estimated 8 million tons of improperly disposed plastic waste makes their way into the oceans each year, and there is evidence that this number is rising. The North Pacific Ocean gyre, also known as the Great Pacific Garbage Patch, is one of the places where plastic garbage is accumulating due to winds and surface currents (Eriksen et al., 2014). The Great Pacific Garbage Patch is estimated to carry 45-129 tons of plastic garbage and is exponentially growing (Lebreton et al., 2018). Most of the plastic waste in the oceans comes from inland sources and is emitted from rivers and coastlines (Napper & Thompson, 2020). However riverine plastic transport is understudied in comparison to marine plastic litter which emphasizes the importance of increasing the global knowledge on plastic pollution in freshwater environments (Blettler et al., 2018). The first studies on riverine plastic waste were conducted only in the early 2010's, so conducted studies are all relatively new (Moore et al., 2011; Morritt et al., 2014; Yonkos et al., 2014). The majority of the research focused on particles in the surface layer of the water column (Kuizenga et al., 2023; Vriend et al., 2020) and/or microplastics (Klein et al., 2015; Mani et al., 2015; Rocha-Santos et al., 2015; Yonkos et al., 2014). In 2021, van Emmerik visually counted floating plastics and extrapolated the floating plastic data to the entire water column to calculate the total plastic transport, he used a correction factor of 1.25-1.5. Using a correction factor is very cost-efficient but can cause large errors as under dynamic conditions no depth variability can be found (Hohenblum, 2015).

The discharge of a river is dependent on multiple variables and differs spatially. In general, water near the stream bank or stream bed moves more slowly due to friction than water in the middle. The amount of friction that slows the flow depends on the roughness of the bed and the channel shape, a wider shallow channel has a larger wetted perimeter and therefore a slower flow than a semicircular channel (Marshark, 2015; Park & Ahn, 2019). Velocity distributions are thus different in a spatial and temporal sense. Variations in time are caused by the effects of turbulence and can cause local velocities much higher or lower than the time-averaged velocity. Variations in depth and width will form a velocity profile and the velocity gradient determines the shear stress that is exerted on the bed. Downstream

variations of the flow velocity, which commonly show minor changes or a slight increase, are caused by a decrease in channel roughness and an increase in hydraulic efficiency which counteracts the decrease in channel slope (Charlton, 2007).

The flow velocity variations in the cross section of a river are partially caused by meandering of a river channel. The highest flow velocity in a meander bend is towards the outside curve and as the water flows towards the outside wall it will start to follow a spiral path where the surface water moves towards the outer bend and the flow at the bottom moves towards the inner bend. This spiral flow is known as helical flow and is pictured in the figure below (Marshark, 2015; Park & Ahn, 2019).

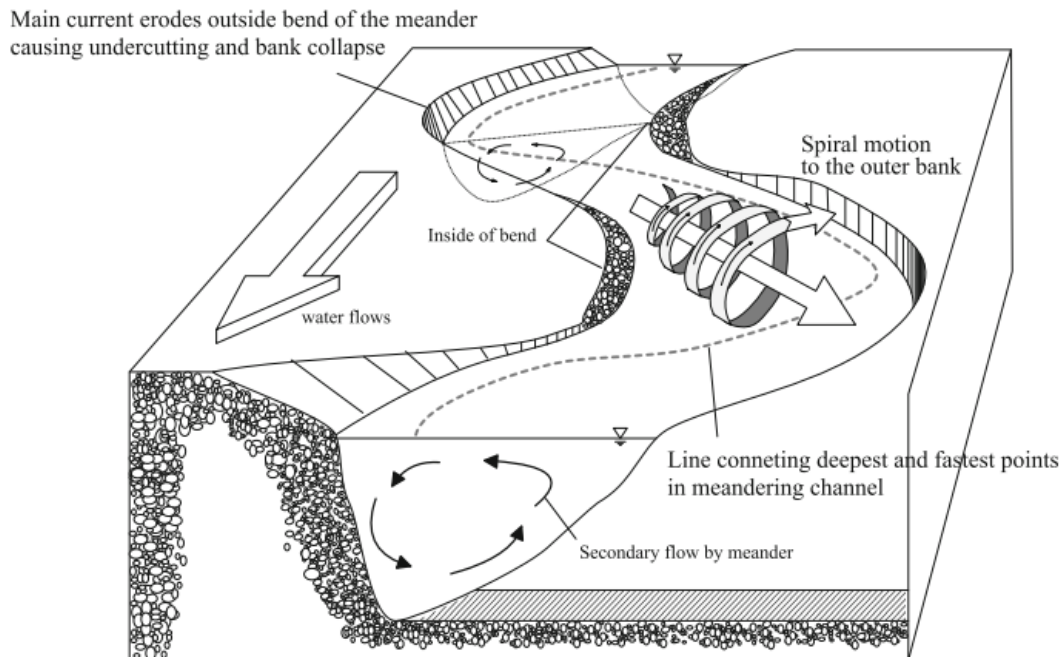


Figure 1 - A meander bend in a river. Arrows indicate the direction of the waterflow (Park & Ahn, 2019).

To eventually reduce the amount of plastics present in the river Rhine, it is important to know more about the characteristics of the plastic and the driving processes of plastic transportation. With that information, the source of plastics can be determined and mitigation techniques of reducing inputs can be made (Jambeck et al., 2015). Little is known about the driving processes of plastic transportation in rivers. Van Emmerik et al., (2022) researched the role of hydrology, e.g. discharge, on floating plastics in the Rhine-Meuse Delta in a spatial extent. This study only focused on floating plastics, which were visually counted from bridges along the Rhine and Meuse. They found discharge is an important driver of floating plastic transport, however the exact relationships vary strongly per location per river (van Emmerik et al., 2022). The direct relationship between the discharge of a river and the plastic concentration is unknown. Collas et al., (2021) found no relationship between these two variables in the Rhine and the Waal, which was possibly the result of only small variations in the discharge during the monitoring period (1218-1593 m³/s at Lobith).

Rijkswaterstaat has measured the plastics in the Dutch rivers from 2018, and since 2021 they used thralls hanging from a ship to catch plastics at different positions in the river. From 2018 till 2021 they used two different nets for the plastic sampling, stow net fishing and 'larvae nets' (Collas, Oswald, & Verberk, 2021). From 2021 and onwards they used 'KOR-nets', which will be explained further in the methodology. This research will build on the previous knowledge that was gathered during the earlier research. However during the previous

sampling years, the discharge of the Rhine was relatively low (1600-3100 m³/s) so more information is needed about the plastic distribution at higher discharges (4000-5000 m³/s). The aim of this study is to determine if the plastics particles in the Dutch Rhine have a correlation with the discharge. Samples were taken in January of 2023 during high discharges and are the main focus of this study.

Research question:

What is the correlation between the plastic particles in the Dutch Rhine river and higher discharges?

Sub-questions:

What kinds of different plastics are found in the high discharge samples, and how does their distribution vary with respect to vertical positioning within the river?

How does the distribution of plastic categories found in 2023 differ from the distribution of plastics found in 2021 and 2022?

What is the spatial and temporal variation in the concentration of the plastic particles from the high discharge samples?

How does the concentration of plastic particles of the high discharge samples compare to lower discharge samples from previous years?

2. Fundamentals of sediment transport

The physics behind plastic movement in water is still relatively unknown. The section below gives an overview of the general flow characteristics and the principles behind sediment transport in a fluvial system. The fundamentals of sediment transport will be linked with plastic transport, to gain more insight. Afterwards an explanation of plastics in a fluvial system will be given with the focus on the input, storage and transport.

2.1 Sediment supply

The main sources of sediment supply are materials that are transported from hillslope erosion and bank erosion. The supply of fine sediments, that mainly comes from bank erosion, controls the rate of suspended sediment transport which implies that the supply of fine sediments have a larger influence on the sediment concentration than the flow conditions of the channel. The rate of supply is dependent on multiple variables and varies during individual events, between events, seasonally and annually. These variables are; the intensity of the rainfall, the shape of the hydrograph, the antecedent circumstances and the vegetation growth. Higher discharges are related with higher concentrations of suspended sediment, which is caused by increased erosion on channel banks and hillslopes subjected to rainfall. However, there is no straightforward relation between the discharge and suspended sediment in a river (Charlton, 2007; Dean, et al., 2016). One phenomenon that can occur is called hysteresis and occurs when at the same discharges on the rising and falling limb of a hydrograph the sediment concentrations differ. This hysteresis effect will be explained later on.

2.2 Sediment entrainment and transport

There are different mechanisms that control sediment transport and deposition in rivers and understanding them is fundamental. River flows are mostly unsteady, three-dimensional, involve different phases of interactions and are in a state of turbulent motion (Shams, Ahmadi, & Smith, 2002).

Rivers transport sediment in different ways, the total volume of sediment carried by a river is called sediment load. Sediment load consists of three components; bed load, suspended load and dissolved load (Marshark, 2015). These different ways of transport are shown in figure 2. Transport of sediment in rivers depends on the balance between resisting and driving forces. The driving forces that act on a particle consists of a lift and drag force. The pressure difference above and below the particle, which is explained by the Bernoulli principle, is what generates the lift force. The drag force is oriented in the same direction as the stream flow and is the effect of the flow of fluid on the object (Charlton, 2007). Sediment transported as bed load bounce or roll along the stream floor and are generally gravel size and larger, it is a sporadic movement and caused by variations in bed shear stress. Finer bedload will move in saltation which consist of a series of short jumps. Lift and drag forces move the saltating grains from the bed in a steep angle, the lift force will then decrease and the drag force will carry the grain downstream following a shallow trajectory towards the bed. Suspended load moves downstream supported by turbulence which prevents the object from rising and settling. Dissolved load consists of the ions of dissolved materials (Charlton, 2007; Marshark, 2015).

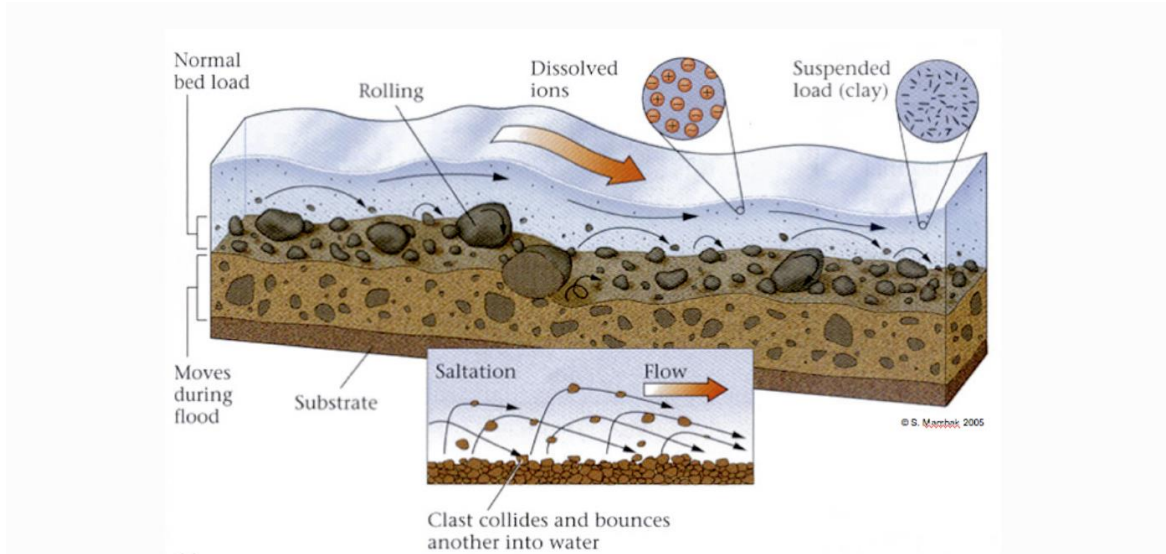


Figure 2 - Different ways that rivers transport sediment (adapted from: Marshak, 2015).

Plastics move in a same way as sediment trough a river. They can either be in suspension, rolling along the bottom of the river of be afloat at the surface of the water column. Where most river surveys focused on floating plastics, Valero (2022) characterized a near-surface plastic flow where floating plastics and plastics in suspension were coexisting. This study only focused on near surface plastics which protrude the water surface when moving and not on plastics completely in suspension. However, the findings are of great importance as they infer that suspended plastic transport in the remaining water column can be explained by suspended-particle theories. The balance of vertical forces acting on a plastic element anywhere in the upper water column is explained by Newton's second law and is pictured in figure 3:

$$\sum F_z = M \frac{du_{p,z}}{dt} \quad (1)$$

With M the total mass of the volume (water, air and plastic) and $u_{p,z}$ is its instantaneous vertical velocity at time t. The sum of the gravity, turbulence and surface tension forces include:

$$\sum F_z = F_\rho + F_w + F_b + F_\sigma \quad (2)$$

With F_ρ including the weight and buoyancy forces, F_w the force due to the plastic coupling with the water flow, F_b the added buoyancy force due to attached bubbles and F_σ the surface tension force existing when the plastic sticks out from the water surface. The water coupling force (F_w) is assumed to be more or less equal to the drag force ($F_w \approx F_D$) (Valero et al., 2022). The weight and buoyancy forces can be estimated through the Archimedes principle:

$$F_\rho \equiv \rho_w g V_b - \rho_p g V_p \quad (3)$$

With ρ_w being the water density, ρ_p being the particles density, V_b and V_p the submerged volume and the particle volume respectively and g the gravity acceleration. This insinuates that if the particle is fully submerged, only the density of the particle influences this buoyancy force. A plastic object that is fully submerged and has a lower density than water will therefore

have a negative buoyancy and will likely sink, not taking the added buoyancy from bubbles into account. However, the buoyant force of bubbles (F_b) is seemed to have a minor effect on the total buoyant forces (Valero et al., 2022).

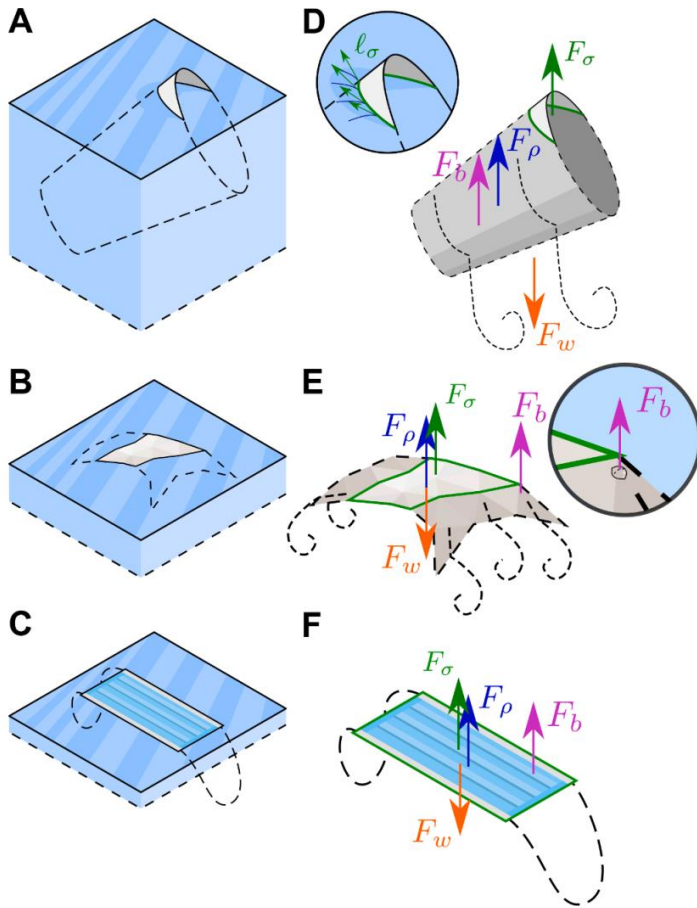


Figure 3 - Forces driving the transport of plastics in river flows. The different objects: A. Plastics cup, B: plastic film, C: face mask, D,E and F for the forces acting on the corresponding plastic element (Valero et al., 2022).

The ratios of the dominant forces are influenced by different transport regimes. One of these regimes is particles that are in suspension which is explained by the ratio of turbulent to buoyant forces, which can be expressed by the Rouse number:

$$\beta = \frac{w}{\kappa u^*}$$

With w being the rising velocity of the suspended particles, $\kappa(=0.41)$ the von Kármán constant and u^* the flow shear velocity. Positively buoyant particles have a positive rising velocity and hence $\beta > 0$ (Valero et al., 2022).

Another relationship of dominant forces can be explained by surface tension (F_σ). Particles in surfaced transport are influenced by buoyancy, turbulence and surface tension. This surface tension becomes more important when the particle is small or when the flow velocity decreases. Plastics which are more creased will have a stronger surface tension since they have a larger interfacial contact line (Valero et al., 2022).

2.3 Sediment deposition

There is a deposit of sediment when there is a reduction in the transport capacity of the flow. This is mainly due to a decrease in the slope of the streambed, reduction in discharge, increase in boundary resistance, obstructions to flow or an increase in the channel width which creates more friction between the streambed and the water. The process of deposition takes place on a small scale, for individual grains, but can create large depositional forms on different spatial scales. Deposition of suspended sediment takes place when the fall velocity is higher than the turbulence forces. The size of the sediment is an important factor for deposition, a larger particle will settle at much higher flow velocities than smaller particles which will only deposit at very low velocities. Thus, coarser sediment will settle much further upstream than fine sediment which creates sediment sorting along the length of a river. As said earlier, the flow velocity is highest in the outside of a meander bend which means that coarser sediment will deposit in the inner bend and create point bars (Charlton, 2007; Marshark, 2015).

2.4 Hysteresis

Erosion and sediment transport processes are important contributors to river dynamics. So identifying sediment sources can contribute to a broader understanding of the connectivity of hillslopes to the river channel (Malutta, Kobiyama, Chaffe, & Bonumá, 2020). A hysteresis analysis will allow to analyze the relationship between discharge and sediment transport during flood peaks, which are important periods for the transport of sediment in a catchment (Lloyd et al., 2016).

The definition of hysteresis is; the relationship between discharge and concentrations of sediment (Lloyd et al., 2016). This means that during the same discharge in the rising and falling limb of a hydrograph, the concentration of suspended sediment is different. The knowledge about this extends multiple decennia and can be caused by the availability of sediment, like sediment depletion and supply (Asselman, 1999; House & Warwick, 1998). Porter (1975), observed higher concentrations of suspended solids during the rising limb and smaller concentrations of suspended solids during the falling limb of hydrographs and attributed this to depletion of grainy material during storms. Webb and Walling (1985) investigated the hysteresis effects of dissolved inorganic nitrate in a catchment during storms and found evidence for both the dilution and accumulation effects of the nitrate. An increasing river discharge resulted in dilution which is attributed to the dilution of solutes in the base flow, which is defined as continuous run-off consisting of delayed sub-surface runoff and groundwater (Webb & Walling, 1985). Increasing solute concentrations during the rising limb of the hydrographs can be contributed to the flushing of sub-surface reservoirs due to a higher flushing rate and former conditions. Surface runoff, in the form of loose material, plays a significant role when concentrations of nitrates increase with discharge, this is also the case with sediment supply from surface runoff and increasing sediment concentrations (Porter, 1975; Webb & Walling, 1985). The analysis of hysteresis patterns can be used to link the source areas and flow pathways in a complex drainage system from the temporal variations in sediment and nutrient transport to streams. A sediment source that is close to the monitoring point will create sediment concentrations to increase more rapidly than the discharge. The opposite will happen when the sediment source is farther away from the monitoring point, and thus a lag in increase of the sediment concentration is monitored (Lloyd et al., 2016).

2.4.1 Hysteresis patterns

Hysteresis patterns can be observed by plotting discharge and concentration data, which will show a loop from which the shape is dependent on the response between the discharge and water quality variables (Lloyd et al., 2016). There are a lot of different types of hysteresis and

the patterns are a result of different spatio-temporal distributions of rainfall, temperature, erosion rate, seasonal change, soil moisture, travel distance of the eroded sediment and geomorphologic characteristics. Williams (1989), has classified 5 prevalent hysteresis patterns: a single line, clockwise, anti-clockwise, single line plus a loop and figure eight. These patterns are shown in figure 4, in their respective order.

A single line (Type I in figure 4) occurs when the relationship between the suspended sediment concentration and the discharge is the same in the rising and falling limb of the hydrograph. This is a pattern that specifically occurs when the sediment transport velocity and the discharge wave's travel duration are the same (Williams, 1989; Yang & Lee, 2018). It is formed by fine suspended sediment and is caused by an uninterrupted sediment supply. The occurrence of this type of hysteresis is quite scarce since sediment supply is generally exhausted during an event (Malutta et al., 2020).

A clockwise loop (Type II in figure 4) is the most common type of hysteresis. The relationship between the suspended sediment concentration and the discharge is larger in the rising limb of the hydrograph than in the falling limb (Malutta et al., 2020). This means that the sediment peak arrives earlier at a given cross section of the river than the discharge peak (Williams, 1989). The cause for this, the lower suspended sediment concentration in the falling limb, is most likely the depletion of sediment that is available for transport (Malutta et al., 2020). Another cause can be the sediment supply areas being close to the river and the sediment supply which originates from tributaries to the source being short (de Boer & Campbell, 1989). The counterclockwise loop (Type III in figure 4) is the opposite of the clockwise loop and occurs when the sediment peak arrives later than the discharge peak (Yang & Lee, 2018). This can have multiple causes such as the discharge wave traveling faster than the sediment wave, the flood wave traveling faster than the average flow velocity or a more distant sediment source, upstream tributaries and a delayed sediment supply by these tributaries (Malutta et al., 2020). A counterclockwise loop is more likely to form when the sediments originate from channel erosion and not from hillslope erosion (Pietroń, Jarsjö, Romanchenko, & Chalov, 2015).

The single line plus a loop (Type IV in figure 4) combines the single line and the subsequent loop. This loops indicates that the travel time of the flow is different from the sediment travel time. The single line shows that at the beginning and the end of the hydrograph the discharge and sediment concentration are directly related. When the discharge rises more, the loop will form which indicates that the discharge and the sediment concentration at the middle of the hydrograph are not synchronized. This type of hysteresis can have a lot of different causes, for example extremely dry conditions (Malutta et al., 2020; Yang & Lee, 2018).

The figure eight pattern is the combined pattern of the clockwise and counter-clockwise loop. The counterclockwise loop is present at the lower part which means a delayed sediment concentration in comparison with the discharge at the beginning of the hydrograph. After that the clockwise loop is present in the higher part and sediment concentrations are higher than the discharges (Williams, 1989; Yang & Lee, 2018).

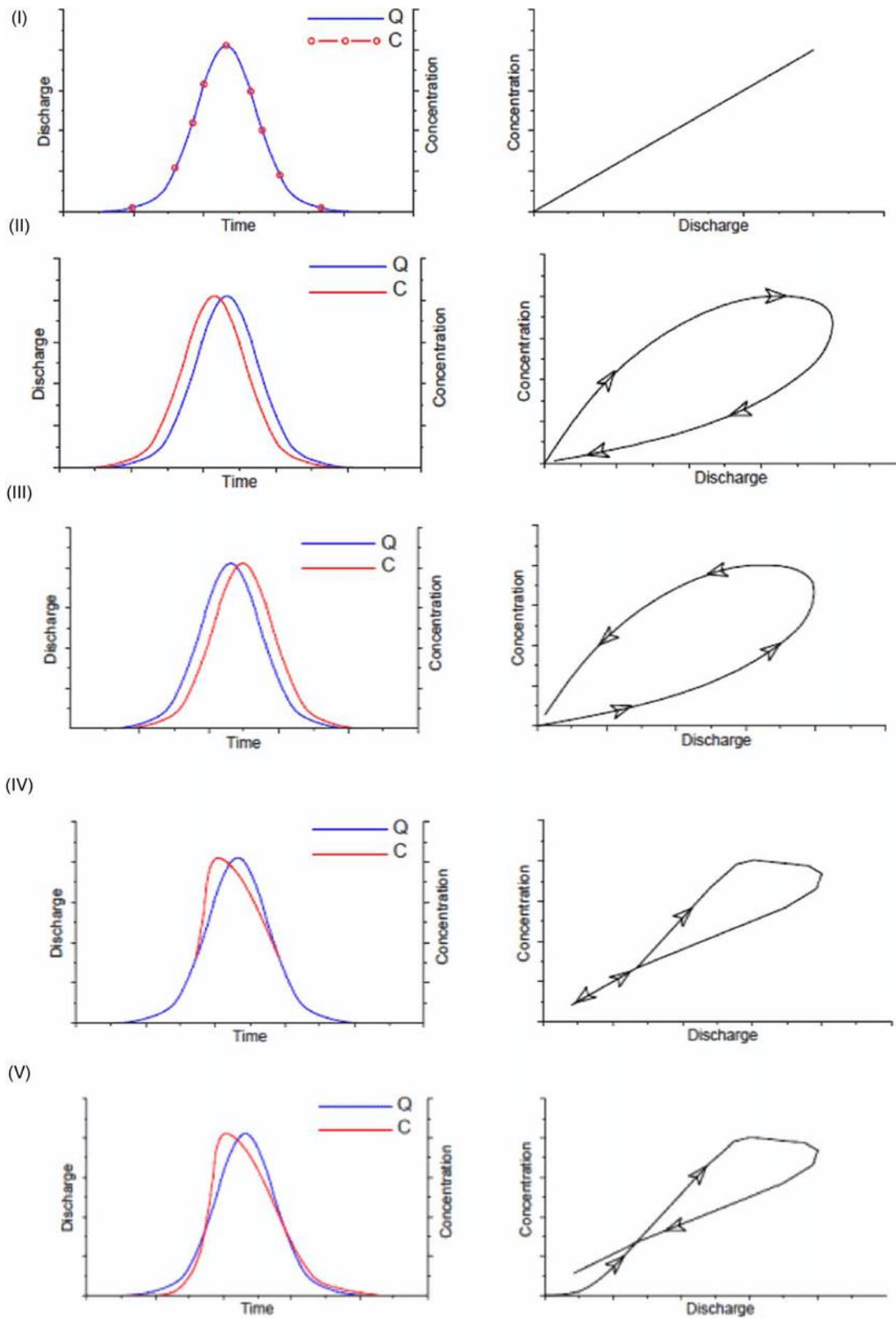


Figure 4- 5 Different hysteresis patterns. Q is the discharge of the river (m^3/s) and C is the sediment concentration. I) A single line pattern. II) A clockwise pattern. III) A anti-clockwise pattern. IV) A single line plus a loop. V) a figure eight patten. (Williams, 1989).

2.4.2 Hysteresis in the Rhine river

Hysteresis has been reported in the Rhine river basin. The suspended sediment behavior in the tributaries of the Rhine is related to the transport of the suspended sediment. The relation of the suspended sediment concentration and the discharge in the main channel of the Rhine is a bit more complex. The first flood in the hydrological year always resulted in the steepest increase in sediment concentration. However, during floods later in the year, concentrations were much higher than earlier floods which suggests that this sediment originated from a different source (Asselman, 1999; Kleinhans et al., 2007).

For single floods, different hysteresis patterns were observed, which are shown in figure 5. A counter clockwise hysteresis loop was observed mostly during summer periods. No hysteresis was also observed relatively often during the summer, when upstream tributaries have an equal supply in sediment. A moderate clockwise hysteresis pattern was observed when sediment concentrations increase gradually with increasing discharge, which often occurred in the winter when most sediment was supplied by the Mosel. A pronounced clockwise hysteresis was observed when the sediment concentrations increased very quick. This is related to early sediment supply by the tributaries that are located just upstream of the measurement location or erosion (Asselman, 1999; Kleinhans et al., 2007).

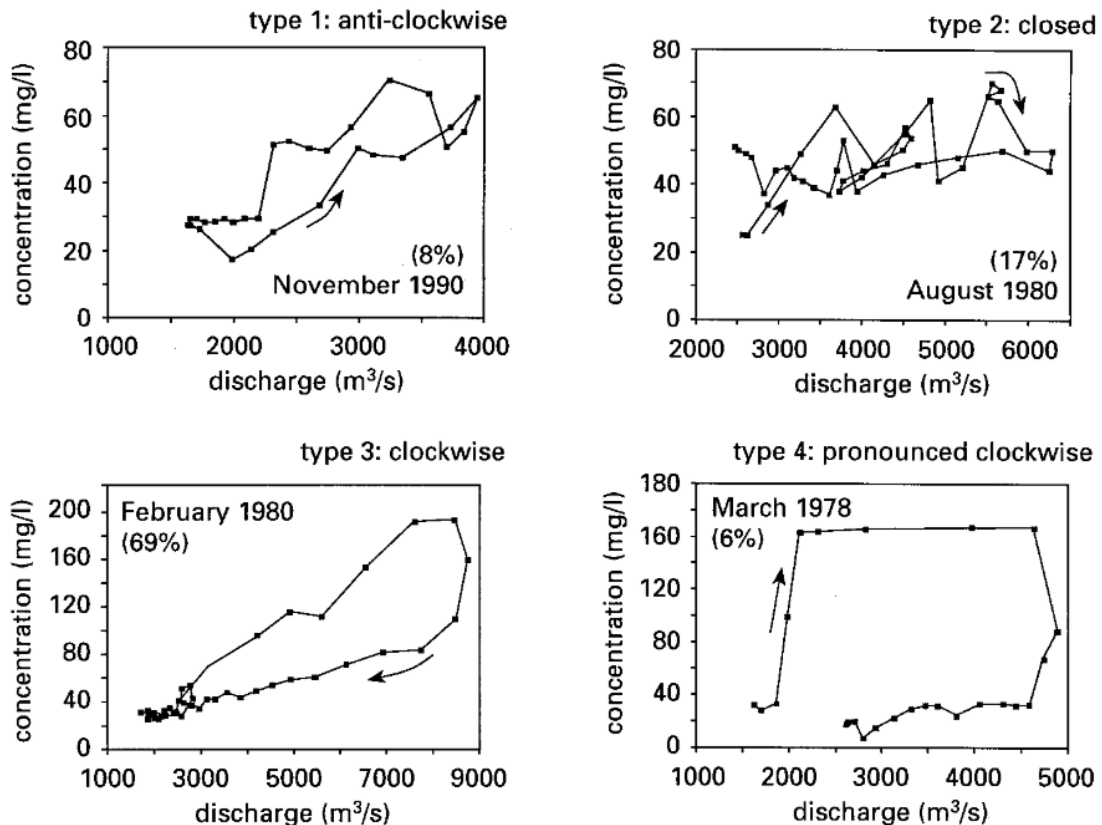


Figure 5- Different types of hysteresis patterns observed in the Rhine for sediment concentrations in mg/l. The percentage of floods with a maximum discharge exceeding 4000 m³/s are given (Asselman, 1999).

2.5 Plastics in rivers

Sediment supply, transport and deposition in fluvial systems is thoroughly investigated and known. However, how plastic moves in freshwater systems is still relatively unknown. In this segment we will discuss the information that is known about plastics in freshwater systems. The figure below shows the different ways plastics can be incorporated in a river system. This is quite similar to sediment in rivers with a few differences; there are floating plastics due to their buoyancy and plastics can end up in biota. Most studies are focused on floating plastics (van Emmerik & Lange, 2021; Vriend et al., 2020) and plastics on riverbanks (van Emmerik & Schwarz, 2020), however more recent studies have also shown insights in vertical distribution of plastics in the water column (Collas et al., 2021). Data of the vertical distribution of plastics in the Rhine showed no significant differences in the plastic concentration between the bottom, middle and surface of the water column. However, in the Waal there was a depth gradient in the plastic concentration, the highest concentrations of macroplastics were found at the surface which descended to the bottom. Mesoplastics were found more abundant at the bottom and decreased in concentration towards the surface of the water column. These differences in vertical concentration were attributed to local hydrodynamics (Collas et al., 2021).

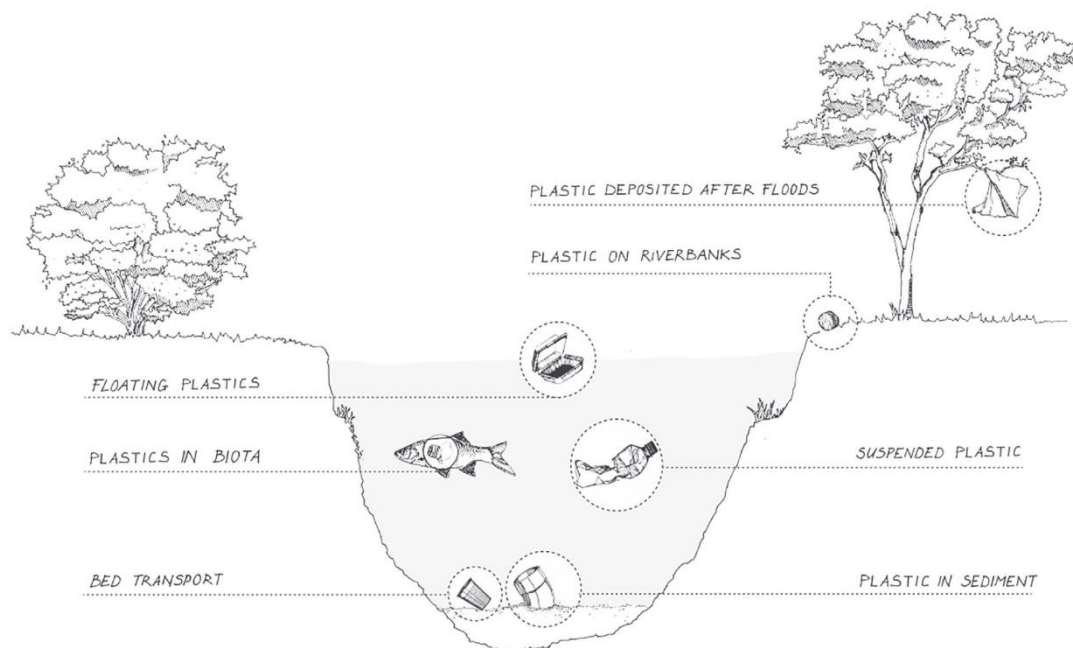


Figure 6 Different ways plastic are found in a freshwater system (van Emmerik & Schwarz, 2020)

2.5.1 Origin of plastics

The source of plastics in fluvial systems is directly related to human activity, since there is a high correlation with urbanization, waste management, wastewater treatment and population density (Best, 2018). Plastic can end up in river by dumping or natural transport processes, like wind, river geomorphology and rainfall induced surface runoff (Bruge et al., 2018; Haberstroh et al., 2021; Moore et al., 2011). In urban areas, plastic is often spilled on the ground which can end up in rivers by the wind (Bruge et al., 2018). Rain events will cause surface runoff which may carry plastic debris into nearby streams (Moore et al., 2011). However, in the Rhône a delay in the plastic transport peak was measured several days after rainfall events, which suggests that this plastic originated from upstream (Castro-Jiménez et al., 2019).

2.5.2 Course of plastics

The course of plastics in fluvial systems is dependent on three different processes; the transport, accumulation and degradation of plastics. The transport of plastics is influenced by river hydrodynamics and particle properties. The highest flux of floating plastic is found to be in the middle of the river, where flow velocity is highest (van Emmerik, Strady, Kieu-Le, Nguyen, & Gratiot, 2019). Nevertheless, shipping can influence the abundance of floating plastic towards the sides of the river channel, and in a river bend the plastics are found more towards the outer bend due to higher flow velocities (Collas et al., 2021). Wind speed and direction affects floating plastics, which can deposit plastic litter on the riverbanks or cause floating plastics to move faster (van Emmerik, Strady, et al., 2019). Plastics are not only located at the surface of the water column but are vertically distributed in the water column. This vertical distribution can be caused by turbulence and causes plastics to move from the surface to the stream floor and back. Plastic properties also influence the vertical mixing; higher density plastics tend to sink easily. Foil with a high surface area to mass ratio, tend to be affected by surface pollution and therefore sink more easily and smaller plastic with a high surface area to mass ratio tend to be more affected by vertical transport due to turbulence (van Emmerik & Schwarz, 2020). The plastic flux can increase significantly during floods which was studied by van Roebroek et al., (2021). They found that during floods with a return period of 10 years, plastic can be remobilized and add to the already present plastic in the water, which can increase the plastic concentration in tenfold (Roebroek et al., 2021). They did however not take into account that the plastic can be supply-limited and the plastic concentration will decrease because of that (van Emmerik, Loozen, Van Oeveren, Buschman, & Prinsen, 2019). Thus the relationship between the discharge and the plastic concentration is not always so straightforward and sometimes not even found (van Emmerik, Loozen, et al., 2019).

The accumulation of plastics can be in the river sediment, riverbanks or floodplains of a river. During floods, plastic can be deposited on the riverbanks or floodplains. Subsequently, after floods, plastics can be mobilized from those locations into the water column (van Emmerik & Schwarz, 2020).

Degradation also plays a role in the course of plastics as macroplastics can degrade into smaller micro and nano plastics. Degradations of plastics can be thermal and UV as well a mechanical degradation that are caused by contact with other plastics or objects (Andrady, 2015; van Emmerik & Schwarz, 2020).

3.The Rhine river

The Rhine is a major River in Europe and enters the North sea through the Rhine-Meuse Delta. Six countries border the river and industry is densely located along the river banks (Mani et al., 2015). Figure 7 shows the Rhine basin where it originates in the Swiss Alps and then flows through Liechtenstein, Austria, France, Germany and ending in the Netherlands (Frings et al., 2019). The mean discharge of the Rhine is around 2300 m³/s and the total length is 1230 kilometers (Ionita et al., 2012). The Rhine River has been intensively modified by humans starting with the Lower Rhine in the late 19th century (Pfeiffer & Ionita, 2017). The Rhine river is in disequilibrium, which means that large parts of the river are prone to erosion or sedimentation. Where other rivers have erosion upstream and deposition downstream, the Rhine has net deposition upstream and net erosion downstream. A large part of this disequilibrium is due to human interference, like dredging and a discontinuous sediment flux (Frings et al., 2019).

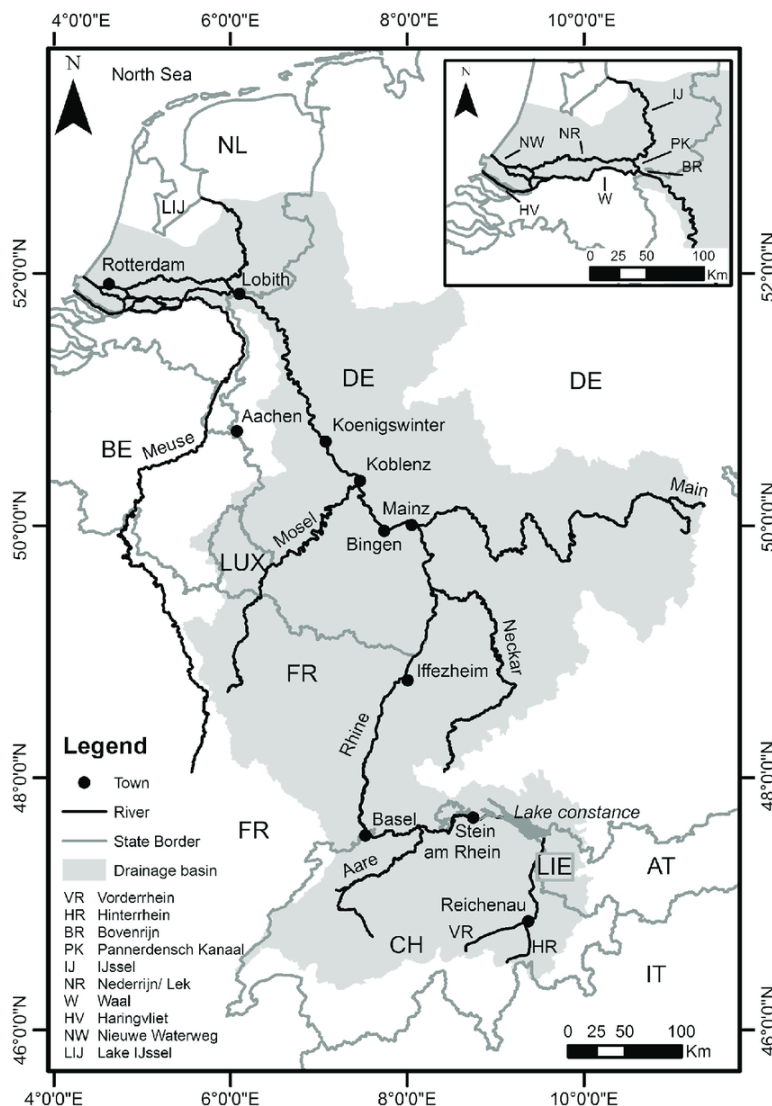


Figure 7- The Rhine basin, originating in the Swiss Alps and ending in the Netherlands where it enters the sea (Frings et al., 2019).

3.1 Study area

The study area is located on the border between the Netherlands and Germany, where the Rhine enters the Netherlands close to Lobith (visible on figure 7). Figure 8 shows the sampling location which is on the Dutch part of the Rhine. The black line shows the fairway and the sampling location is situated on the right side of the fairway, which is also the outside of the bend. There is a gauging station located in Tolkamer, which is a few hundred meters to the west of the sampling location. Information about the water level and discharge at Lobith was retrieved from Rijkswaterstaat (Rijkswaterstaat, n.d.). This data was used to determine different relationships between the concentration of plastic and the discharge. When the water level reaches approximately 1000 centimeters above NAP at Lobith, the groynes overflow and at approximately 1100 centimeters above NAP the water reaches the floodplains.

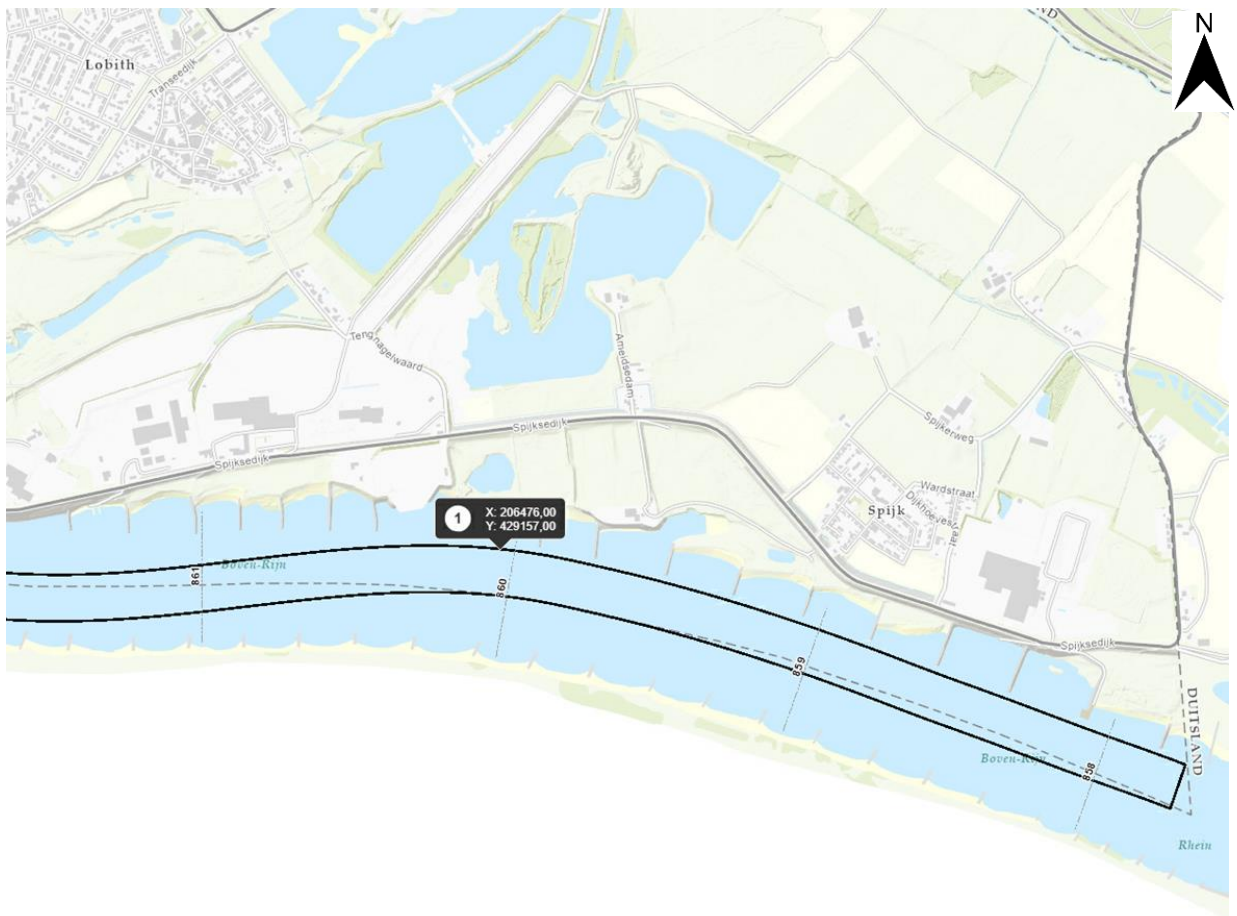


Figure 8 - Locations where the samples were taken. The black lines indicate the fairway.

4. Methodology

4.1 Sampling

Plastic monitoring was performed on four consecutive days in the middle of January of 2023 near Lobith on the Rhine. Samples were taken with two 4 m² trawls hanging from the port and starboard sides of a ship, see figure 9. The trawls, called KOR-nets, have a mesh size of 6 millimeters and are explicitly made for this purpose. The mesh size of 6 millimeters will primarily capture meso- and macro-plastics whereas microplastics will not be captured and are accordingly not included in the analysis.



Figure 9 - From left to right: The Stern, the ship from which the samples were taken. The trawl (4x1m) hanging on the side of the ship, both on port and starboard. The trawl being lowered into the water to collect plastic samples.

In January, 72 samples were collected in the Rhine near Lobith, during this period high discharges were measured at the gauging station near Lobith. The discharge ranged between 4300 and 5100 m³ s⁻¹ and the water depth ranged between 1170 and 1260 centimeters (both measured at Lobith gauging station, waterinfo.rws.nl). Considering the high discharge, samples were only collected from the right side of the cross-section of the river and only at two different depths; at the surface layer and in the middle layer, seen in figure 10. Samples near the bottom were not achievable because the KOR-net would not stay near the bottom during the high discharges as it needed more weights to maintain this height. The duration of the sampling ranged from 10 minutes to 40 minutes, most samples were 30 minutes in the water. These times were modified by the amount of organic material and plastic that was collected in the KOR-nets. On board of the ship all the fish were put back into the river and, if possible, most of the organic material was separated from the plastic. This was accomplished by multiple people using tweezers to get all the plastic pieces.

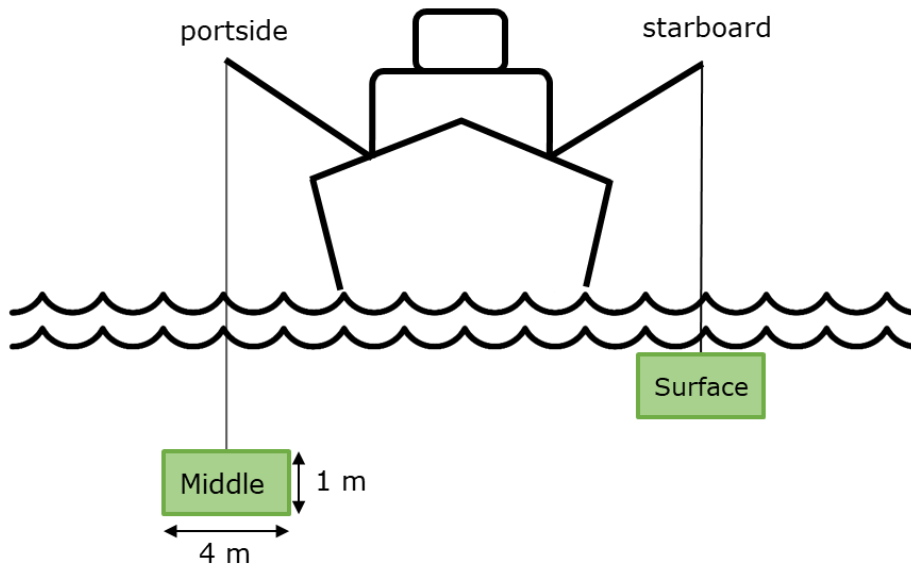


Figure 10 - Method of sampling from the ship, trawls (4x1m) located at the surface (on the port side of the ship) and at the middle (on the starboard side of the ship) of the water column.

The local flow velocity of the river was measured by performing Acoustic Doppler Current Profiler measurements (ADCP). An ADCP transmits sound waves which then scatter back from particles within the water column, the shift in frequency (called the Doppler shift) will then determine the flow velocity (Holdaway, Thorne, Flatt, Jones, & Prandle, 1999). This method will be able to determine location specific flow velocities in the river and will be used to determine the volume of sampled water during the surveys and ultimately be used for the analysis of the plastic concentrations.

4.2 Counting and sorting of plastics

Most of the organic material was separated from the plastic on board of the ship, with the remainder being sorted out in the laboratory. Here, the plastics particles were cleaned and separated into groups of mesoplastics (5 mm – 25 mm) and macroplastics (>25 mm) based on their longest length. The plastic pieces were then counted and categorized using the River OSPAR-method. This is a list of more than 100 specific items divided into 9 main categories adapted from the marine OSPAR list that is used for beach litter cleanups to better suit the Dutch riverine litter. The main categories are; plastic, rubber, textile, paper, wood, metal, glass, sanitary and medical items (van Emmerik, Vriend, & Roebroek, 2020). Most of the plastic is identified into categories independent from the classification of the size, for instance string/cord and food packaging. However, there are three categories; soft plastic/polystyrene pieces, hard plastic/polystyrene pieces and unidentified styrofoam pieces which make a distinction in size between meso- and macroplastic in their category. More knowledge about the language, brand, expiration date and other important information was also documented. Afterwards, the plastic pieces from a sample were put onto A4 papers, trying to lay the pieces as flat as possible and photographed from straight above with a ruler next to it. The photographs were taken with a 26 mm lens from approximately the same height every time. An example of a picture is showed below in figure 11. Post hoc, pictures of each sample were inspected and categories of some plastic pieces were changed which mostly were not recognized as certain categories at first. For example, the water filters/biofilms were initially not recognized and were now put into their corresponding category.



Figure 11 - An example of plastic pieces on a paper. These were macro plastic pieces from the sample 47 OPP SB RO

4.3 Data analysis

The data of the counted and categorized plastic pieces per sample were then entered separately for meso- and macroplastics into a database (Microsoft Excel) and subsequently analyzed using R version 4.2.3 (R Core Team, 2023). To answer the objective about the plastics that are found in these samples, the number of plastics found in a category were added for meso- and macro. The relative proportion of each meso- and macroplastic category was then analyzed, together with the total amount of plastics in a category.

To answer the objective about the spatial and temporal variation of the concentration, first the measurements of the ADCP were used to ultimately calculate the concentration in items per m^3 . By combining the measured flow velocities in the net, measured using the aforementioned ADCP, with the surface area of the net (m^2) taking the sampling duration into account, the sampled volume was derived. Subsequently, the found plastic items were divided by sampled volume yielding a plastic concentration (items/ m^3). The volume and concentration were derived for each individual sample category combination.

A generalized linear model (GLM) was used to analyze the effect of different explanatory variables on the plastic concentration (Dobson & Barnett, 2008). These variables include categorical variables: 'vertical position' (middle and surface), 'side of the ship' (port and starboard), and the continuous variables: 'water level' (at Lobith), 'discharge net' and 'discharge Lobith' (the discharge at Lobith retrieved from Rijkswaterstaat). First the distribution of the plastic concentration data was determined in order to define a correct regression model. A Cullen and Frey graph, shown in appendix A, provides more insight in the

possible distribution of the data (Etemadidavan & Collins, 2020). This graph plots the kurtosis against the square of skewness and displays different possible distributions, for example Weibull, gamma and lognormal distributions. To compare the possible distributions, the plastic concentration data was fitted to these distributions. A Kolmogorov-Smirnov test was performed to find the goodness of fit of the distributions of the concentration data (Berger & Zhou, 2014). Subsequently, several general linearized models with the distribution of the data are fitted to examine the effect of the different explanatory variables on the plastic concentration. The best model selection was based on the lowest Akaike's information criterion (AIC) value in combination with the highest Nagelkerke R^2 -value. A stepwise variable selection was used to find the best fitted model. Together with a manual model selection, the best model was found. The stepwise model selection included an forwards and backwards selection. Forwards selection starts with an empty model and adds variables stepwise while comparing the models. Backwards selection starts with a model that includes all variables and removes them one by one while comparing the models. This stepwise approach was first done without interactions and afterwards with interactions. These interactions were only included in the model if they significantly improved the model. After this stepwise approach, a manual model selection was also conducted, to reduce the number of model possibilities a stepwise approach can miss. A Chi-squared test was used to analyze the difference in deviance between the models and the selection for the best model was based on this. A Tukey post hoc analysis was executed on the best model to determine the differences of the means (Lenth, 2016; Ruxton & Beauchamp, 2008).

The plastic concentration data did not include the specific data of the meso- and macro concentration as this concentration is the total concentration not taking the size of the plastic into account. Therefore a GLM was used to determine the effect of different variables on the meso- macroplastic concentration. The same explanatory variables were used and the same procedure was followed as the analysis of the plastic concentration.

A hysteresis analysis was conducted on the plastic concentration data. The individual concentration of the most abundant categories were also plotted against the discharge, this displays the different influences the discharge has on the single categories.

To get an insight in how the concentration of plastic particles of this sampling year compares to the concentration of previous sampling years, this data was consulted. This included data from 2021 and 2022 at the same location near Lobith as this year. In the previous years, sampling took place at 3 widths of the river: left bank, middle and right bank and at three depths: surface, middle and bottom. However, since the sampling of this year only took place on the right bank of the river and only at the surface and middle of the water column, only this data was used in the comparison. The table below gives the general information of the sampling days of 2021 and 2022, the information about 2023 is also added for comparison. It is noted that the number of samples is around 5-6 times less in 2021 and 2022 than the number of samples in 2023.

	2021	2022	2023
<i>Time period</i>	12-04-2021 till 19-04-2021	06-04-2022 till 20-04-2022	15-01-2023 till 18-01-2023
<i>Sampling days</i>	6	6	4
<i>Total number of samples</i>	60	71	72
<i>Number of samples of the right bank; middle and surface of water column</i>	12	15	72
<i>Discharge in m³/s</i>	1600-1900	1600-3100	4300-5100
<i>Water level (cm + NAP)</i>	852-895	838-1035	1184-1258

Table 1 – The general information about the sampling in 2021, 2022 and 2023

5. Results

This chapter will include the results of the sampling during high discharges. The number of plastics and the different kinds of plastics were calculated and their vertical distribution was determined. The plastics that were found during this sampling year were then compared to the categories found in 2021 and 2022, mainly looking at the different distribution of categories. After that the concentration of the plastic particles was calculated and statistical tests showed different significant variables that have influence on the concentration. The concentration of the plastic particles over time was calculated as well as the concentration of the plastic particles over the discharge. A complete hysteresis analysis could not be performed since the sampling only took place during the rising discharge and for hysteresis you need data during a complete peak in discharge. The concentration data of 2023 was then compared to the data of 2021 and 2022, which occurred during lower discharges, to look at the influence of a higher discharge on the concentration of plastic.

5.1 Discharge and water level at Lobith

The sampling period took place between the 15th and 18th of January in 2023. The figure below shows the discharge at Lobith, taken from the gauging station situated there. The black square indicates the sampling period, which is during a rising discharge. The samples are consequently only taken before and at the peak discharge. During the sampling the discharge increased, however a small stagnation of the increase is observed at 16:00 on 15 January till 2:30 on 16 January. No samples were taken during this time period. After the sampling period the discharge descended again as well as the water level. The water level increased from 1190 centimeters above NAP to 1260 centimeters above NAP during the sampling period. As follows, the groynes and floodplains were both already submerged during the sampling period in January.

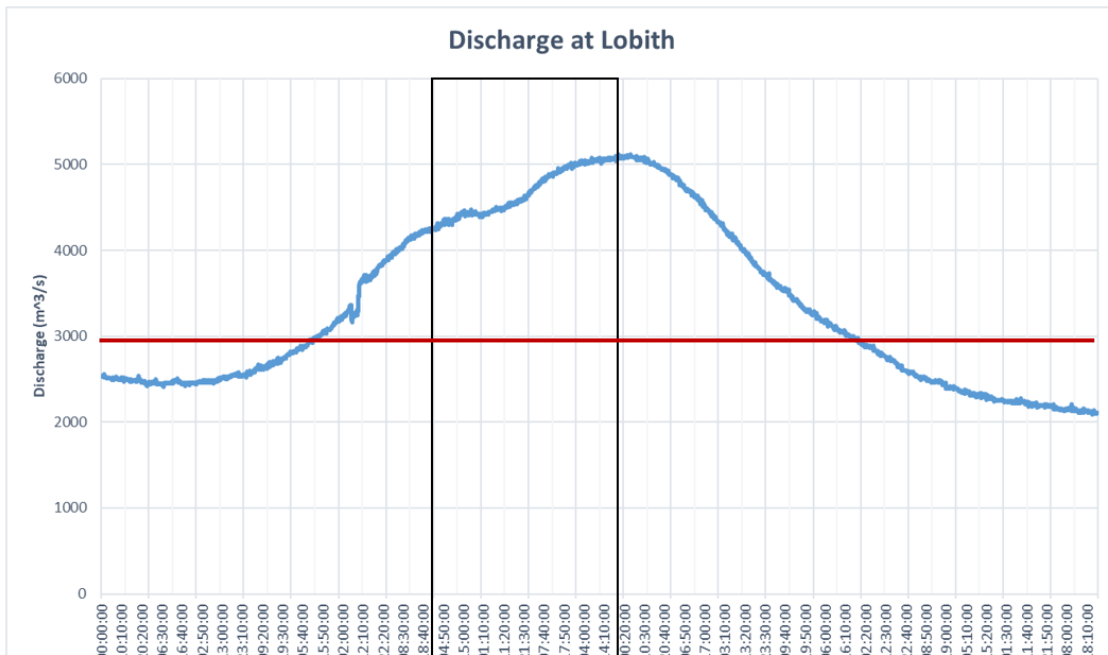


Figure 12 - The discharge at Lobith before, during and after the monitoring period (waterinfo.rws.nl). The black square indicates the monitoring period, the red line indicates when the groynes are overflow.

5.2 OSPAR-categories

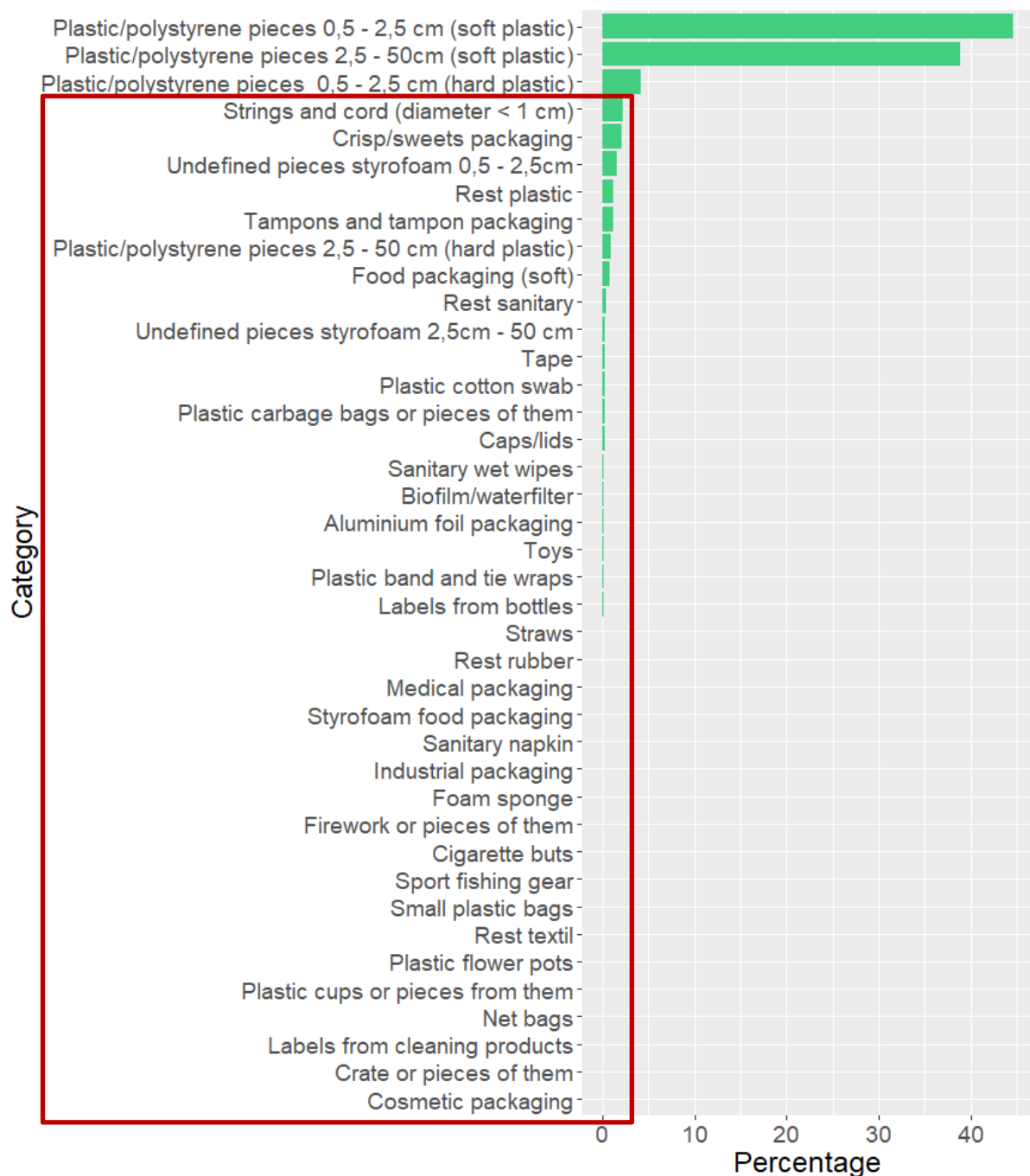
In total 6745 pieces of plastic were counted and categorized, and 40 different categories were found in the samples. The percentage of macroplastics was 48% and of mesoplastics was 52%. 83.5% Of the plastics found were plastic/polystyrene pieces of soft plastic for meso and macro combined. These unidentifiable pieces were mostly clear pieces from which the original purpose of the product could not be verified. The unidentifiable plastic pieces are sorted into two different categories, meso (<0.5-2.5cm) and macro (>2.5cm), from which the mesoplastics were found 1.14 times more than the macroplastics. Table 2 shows the number of plastic pieces per category, as well as the percentage.

OSPAR name	Count	Percentage
Plastic/polystyrene pieces 0,5 - 2,5 cm (soft plastic)	3014	44.59899379
Plastic/polystyrene pieces 2,5 - 50cm (soft plastic)	2629	38.90204202
Plastic/polystyrene pieces 0,5 - 2,5 cm (hard plastic)	282	4.172832199
Strings and cord (diameter < 1 cm)	146	2.160402486
Crisp/sweets packaging	137	2.02722699
Undefined pieces styrofoam 0,5 - 2,5cm	108	1.598105949
Rest plastic	82	1.213376739
Tampons and tampon packaging	78	1.154187629
Plastic/polystyrene pieces 2,5 - 50 cm (hard plastic)	58	0.858242083
Food packaging (soft)	53	0.784255697
Rest sanitary	27	0.399526487
Undefined pieces styrofoam 2,5cm - 50 cm	20	0.295945546
Plastic garbage bags or pieces of them	15	0.22195916
Tape	15	0.22195916
Plastic cotton swab	15	0.22195916
Caps/lids	14	0.207161882
Sanitary wet wipes	9	0.133175496
Biofilm/water filter	7	0.103580941
Aluminum foil packaging	6	0.088783664
Toys	5	0.073986387
Plastic band and tie wraps	4	0.059189109
Labels from bottles	4	0.059189109
Medical packaging	3	0.044391832
Straws	3	0.044391832
Rest rubber	3	0.044391832
Industrial packaging	2	0.029594555
Firework or pieces of them	2	0.029594555
Foam sponge	2	0.029594555
Styrofoam food packaging	2	0.029594555
Cigarette butts	2	0.029594555
Sanitary napkin	2	0.029594555
Crate or pieces of them	1	0.014797277
Plastic cups or pieces from them	1	0.014797277
Net bags	1	0.014797277
Small plastic bags	1	0.014797277
Sport fishing gear	1	0.014797277
Plastic flower pots	1	0.014797277
Labels from cleaning products	1	0.014797277

Rest textile	1	0.014797277
Cosmetic packaging	1	0.014797277

Table 2 - The number and percentages of plastic pieces per category.

Most plastics were soft plastic/polystyrene pieces, the order of the abundance of the categories that were found is seen in figure 13. The third abundant category was hard plastic/polystyrene pieces 0,5-2,5cm with 4.2%. 34 Crisp/sweets packages had identifiable languages, German was found most (n=32) and 2 pieces were Swiss, only 3 pieces had a readable expiration dates (25-3-2022, 1-11-2023 and 1-1-2020). Food packaging was also found reasonably abundant, 0.8%, which only had 10 pieces with recognizable languages: German (n=8), Dutch (n=1) and French (n=1) and one expiration date of 2018.



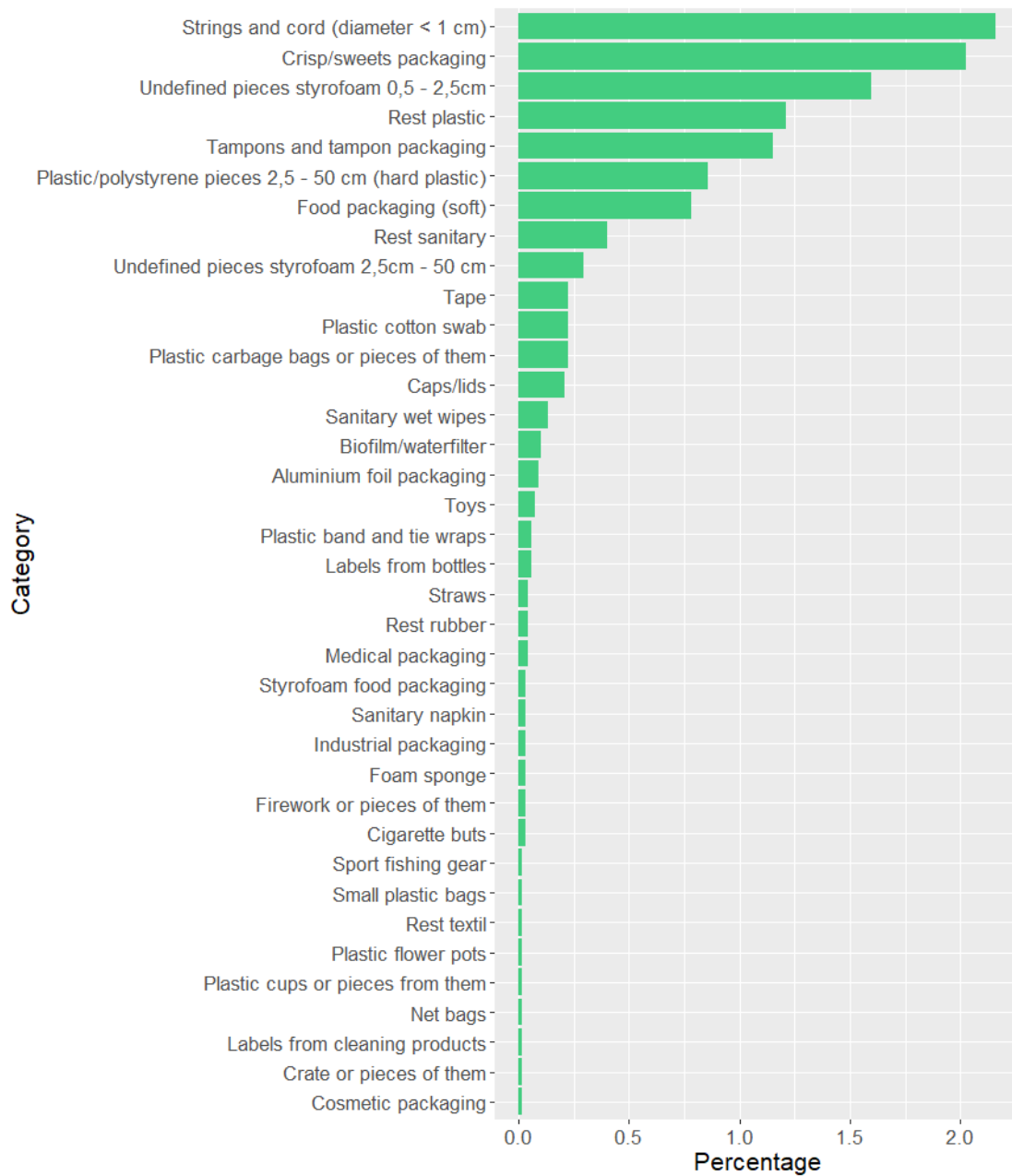


Figure 13 -Top: The relative abundance of plastic categories expressed in percentages according to the River-OSPAR method from the samples of January 2023. (n=6745). The red circle is pictured in the bottom picture.

5.2.1 Categories per vertical position

To get an insight to how different categories are found per vertical position in the water column, the figure below was made. This figure shows the relative abundance of plastic categories, with the abundance separated for the middle and the surface of the water column expressed in percentages. Almost for all categories, relatively more pieces were found on the surface of the water column. However, there are a few exceptions, plastic garbage bags and sanitary wet wipes were found more abundantly in the middle of the water column. Some rare

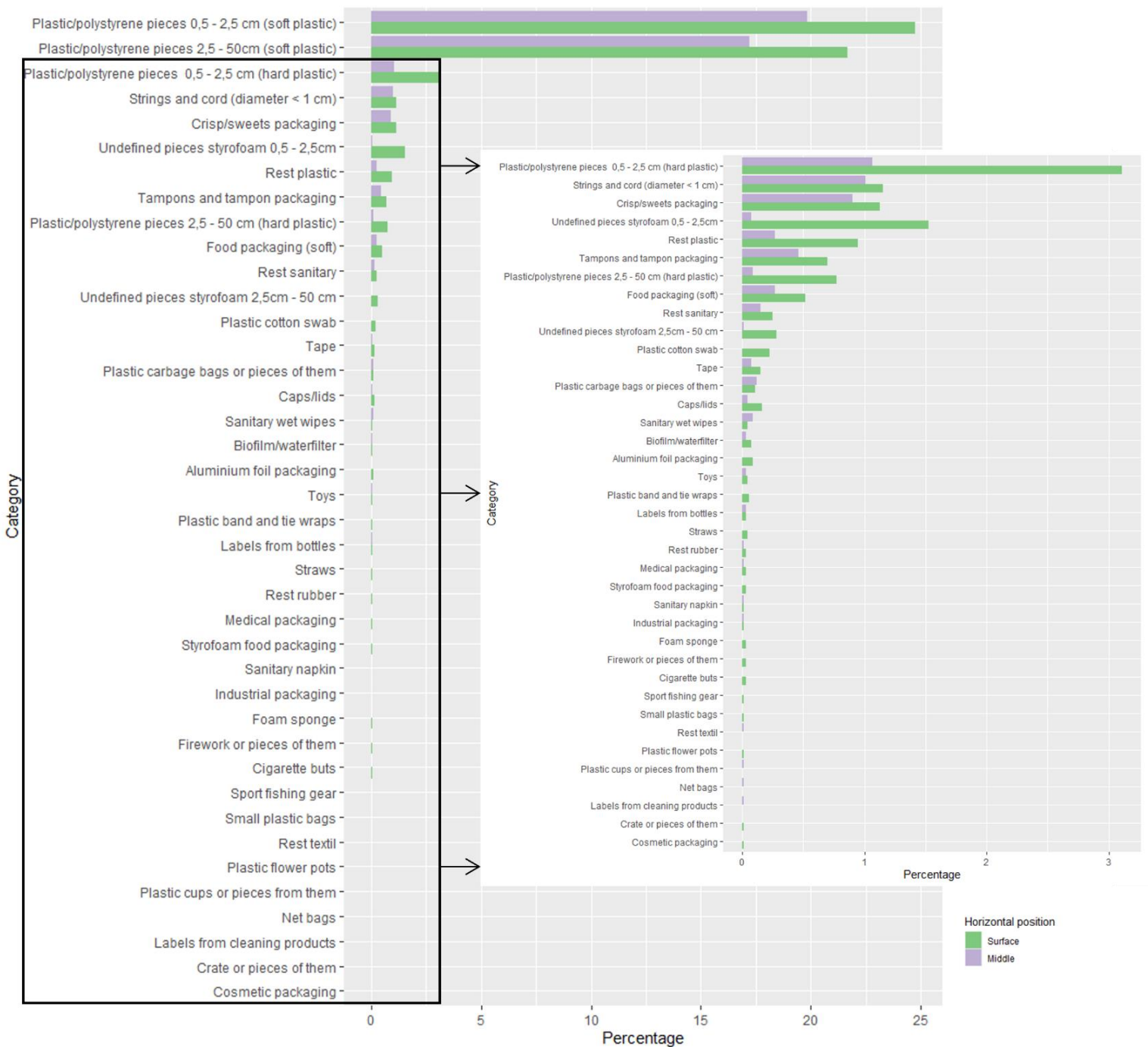


Figure 14 – The abundance of plastic categories, with the abundance separated for the middle and the surface of the water column expressed in percentages.

categories (e.g. cigarette butts, straws, pieces of fireworks, band/tie-wraps) were predominantly located in the surface of the water column.

The categories with more than 8 pieces in total were put into a separate figure, figure 15. The abundance of the plastic pieces per vertical position in a category was calculated, which clearly showed some large differences in abundance.

The most striking category is plastic cotton swabs, which were only found at the surface of the water column, which is a result of their density which causes them to float ("Cotton Bud

Survey - Clean Ocean Foundation,“). 95 Percent of the unidentified styrofoam pieces, for both macro- and mesoplastic, are located at the surface of the water column. Likewise 90 percent of hard plastic/polystyrene pieces (2,5-50 cm) are found in the surface of the water column and 74 percent of large hard plastic/polystyrene pieces (2,5-50cm) are also found at the surface. Note that the categories below soft food packaging, in the figure below, have less than 30 pieces in total and that it can not be assumed that the vertical position of the pieces is normally distributed.

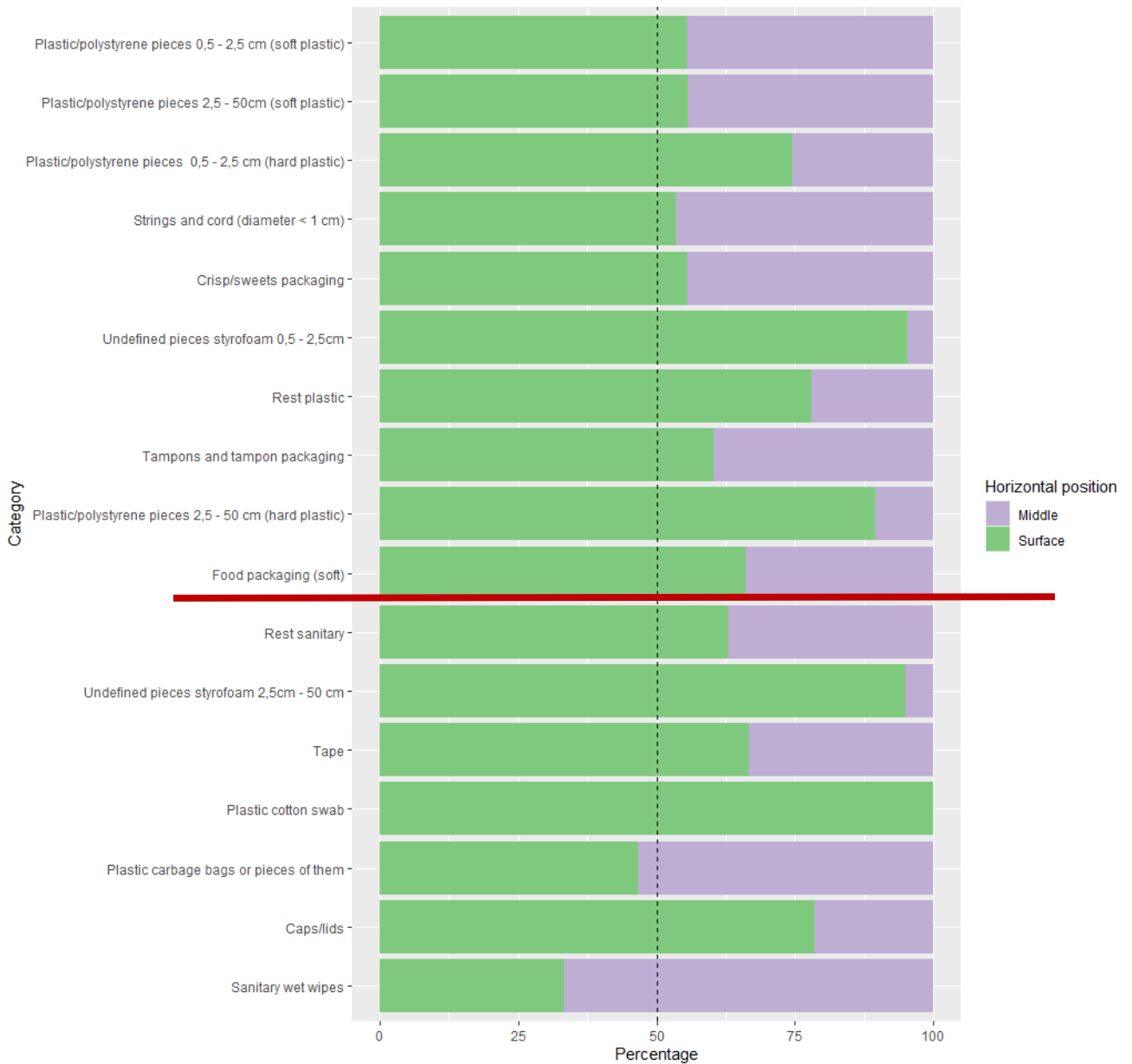


Figure 15 - The percentage of plastic pieces per category found in the middle or at the surface of the water column. The categories below the red line have less than 30 items.

5.3 Categories compared to previous years

Table 3 shows the most abundantly found plastics in 2021, 2022 and 2023 (data of 2021 and 2022 retrieved from Rus, 2022). 83.5% Of the plastic pieces collected in 2023 were unidentifiable soft plastic pieces, compared to 72.8% in 2021 and 72.1% in 2022.

A considerable difference between the different years is the abundance of sanitary wet wipes, which only contributed to 0.1% of the pieces in 2023 and around 6.5% and 3.8% for 2021 and 2022. A possible explanation for this phenomenon is the fact that no bottom samples were taken in 2023 and the sanitary wet wipes were mostly found in the bottom samples of 2021 and 2022 (Rus, 2022). The string and cord (diameter <1cm) category was also less abundant in 2023 compared to the other 2 years.

Category	2021	2022	2023
Plastic/polystyrene pieces 0.5-2.5cm (soft plastic)	49.5%	38.0%	44.6%
Plastic/polystyrene pieces 2.5-50cm (soft plastic)	23.2%	34.1%	38.9%
Plastic/polystyrene pieces 0.5-2.5cm (hard plastic)	3.2%	2.9%	4.2%
String and cord (diameter <1cm)	6.5%	6.0%	2.2%
Crisp/Sweets packaging	1.3%	1.3%	2.0%
Styrofoam pieces 0.5- 2.5cm	0.0%	2.2%	1.5%
Rest plastic	0.7%	0.7%	1.2%
Tampons and tampon packaging	0.5%	1.6%	1.2%
Plastic/polystyrene pieces 2.5-50cm (soft plastic)	0.5%	0.7%	0.9%
Food packaging	1.2%	0.8%	0.8%
~~~~~			
Sanitary wet wipes	6.5%	3.8%	0.1%
Cigarette butts	0.5%	1.6%	0.03%

*Table 3 - Most abundant categories found in 2023. The bottom two categories were not found abundantly in 2023 but were found abundantly in 2021 or 2022.*

### 5.4 Concentration

The distribution of the concentration data was found to be Gamma-distributed by the Kolmogorov Smirnov test (p-value=0.97) and a log-normal distribution also fitted the data well (p-value=0.95). This resulted in a model selection of GLM's with gamma distributions and log-links. The stepwise variable selection resulted in the best model of the plastic concentration based on the variables: 'Water level at Lobith', 'Vertical position', 'Discharge at Lobith' and 'Discharge net' with a significant interaction between the water level and discharge at Lobith (table 4). The AIC of the model was 304.5 and the R² was 0.492.

Explanatory variable	Df	Deviance	t-value	Pr(p-value)
Water level	1	1.33	-3.656	0.001
Vertical position	1	0.66	3.798	0.0003
Discharge net	1	0.21	-2.244	0.03
Discharge at Lobith	1	0.34	4.586	2.07e-5
Waterlevel:Discharge Lobith	1	1.01	-4.261	6.6e-5

*Table 4 - Parameters of the GLM*



The total plastic concentration differences between the middle ( $\mu_{1/2}=7.4$  particles/1000 m³) and surface of the water column ( $\mu_{1/2}=9.0$  particles/1000 m³) was found to be significant (t-ratio=-3.2, p-value=0.002), shown in table 5). The concentration of meso- and macroplastics were also calculated separately and are displayed in figure 16. The results from the best regression model included the variables 'vertical position' and 'size of plastic'. Other variables were not included since only the influence of the vertical position on the concentration of meso- and macro plastics was considered. The table below shows the results from the least square means test, which included a pairwise comparison of the vertical position and the size of the plastic (meso- or macroplastic). For both meso- and macroplastic, significant differences were found between the middle and the surface of the water column. Where the middle in both cases included lower plastic concentrations. Since the GLM did not show a significant relationship between the size of the plastic (meso- or macroplastic) on the concentration, no post hoc analysis was conducted.

Concentration	Location	Mean (particles/1000m ³ )	Statistical test between middle and surface	
			t-ratio	p-value
Total	Middle	7.4	-3.2	0.002
	Surface	9.0		
Meso-plastic	Middle	3.84	-3.34	0.001
	Surface	4.71		
Macro-plastic	Middle	3.60	-3.34	0.001
	Surface	4.28		

*Table 5 - Results of the least square means test, with a pairwise comparison of the vertical position and the size of the plastic.*

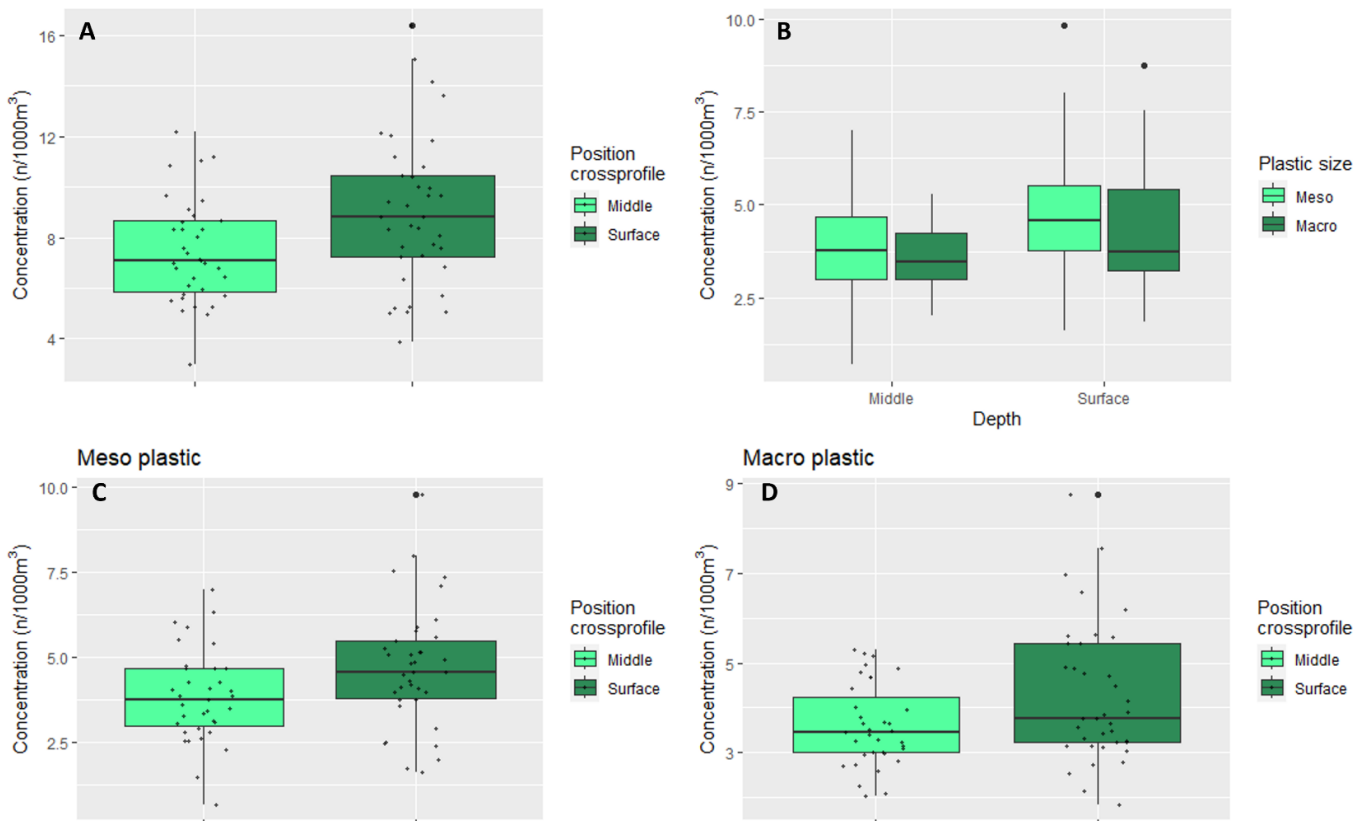


Figure 16 - A: Total plastic concentration, B: Total plastic concentration per depth for meso- and macroplastic separated. C: Mesoplastic concentration per depth, D: Macroplastic concentration per depth.

#### 5.4.1 Plastic concentration over time.

The plastic concentration follows a distinct pattern over time. A post hoc analysis of a least square means test showed the significant relation between the concentration and water level (t-ratio=-3.66, p-value=0.001) and the significant relation between the concentration and the discharge at Lobith (t-ratio=4.59, p-value=0.00002). This has a logical explanation as both the discharge and the water level follow the same rising curve during the sampling period and are thus considerably the same. This is shown in figure 17, where the discharge and water level are plotted over time.

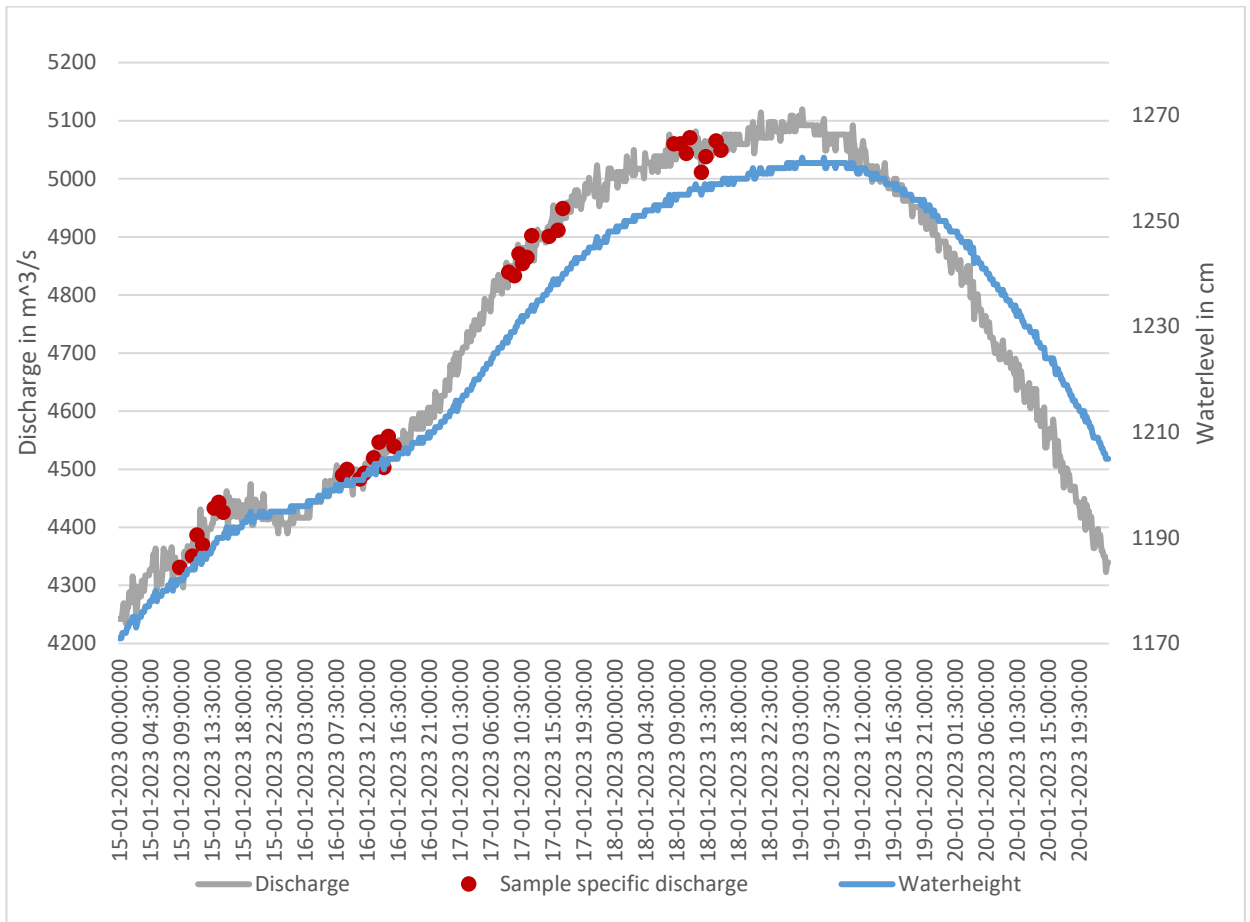


Figure 17 - The discharge and water level at Lobith over time. The red dots indicate the sample specific discharge.

Figure 18 shows the plastic concentration ( $n/1000m^3$ ) over time. Regression lines were fitted per day and per depth, since there was a significant difference between the plastic concentration between the middle and the surface. The lines are fitted per day to emphasize the changes during a sampling day and not the changes over the entire sampling period.

The concentration of the plastics does not follow the same curve as the discharge of Lobith over time, shown in figure 17, as this discharge only rises from the first day to the last. The figure shows the specific discharge at Lobith during the sampling, and the discharge before, during and after the sampling. The rise in discharge is the highest between the 16th and the 17th of January, with a rise of 300  $m^3/s$  in 15 hours.

It is seen that on the first day the plastic concentration rises both in the middle of the water column as at the surface, however the surface concentration rises more than the middle, from which the latter only has an outlier in the concentration in the middle of the day. The plastic concentration on second day, 16th of January, descends drastically in the middle and at the surface of the water column, which is not in line with the rising discharge. On the 3rd day a rise in plastic concentration in the middle and surface of the water column is clearly seen which corresponds to the rising discharge. The last day is again different since the plastic concentration lowers again and the discharge has only stopped rising.

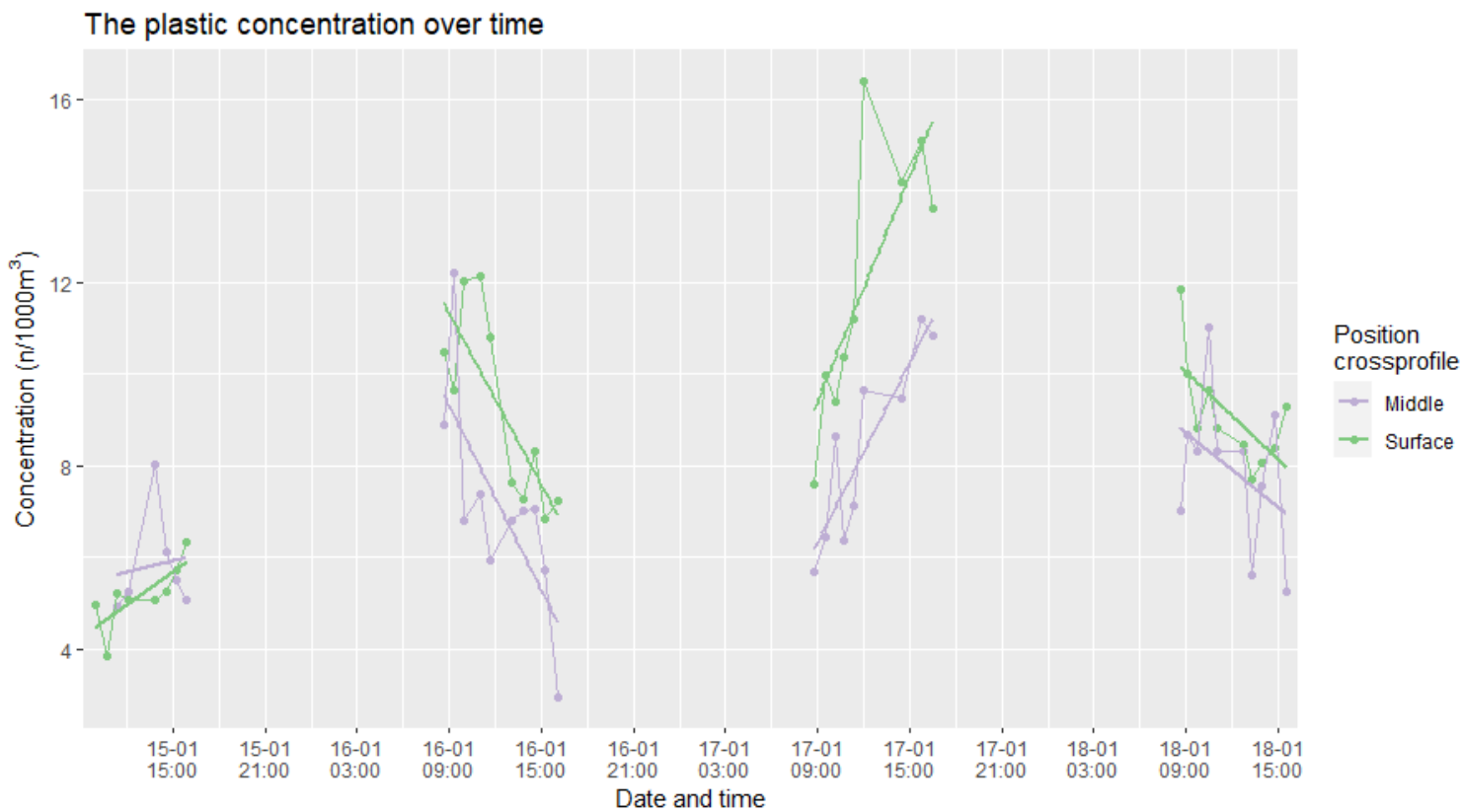


Figure 18 - The plastic concentration in  $n/1000m^3$  over time

#### 5.4.2 Hysteresis

A hysteresis analysis is normally done with data during an entire discharge peak. However, the data of this sampling period is only retrieved during a rise in discharge. Therefore a complete hysteresis analysis could not be performed and the results of this section are focused on the plastic concentration during a rising discharge.

Figure 19 shows the plastic concentration against the discharge, with the concentration of the plastic in the middle and the surface of the water column separated. This is done because they were found to be significantly different and to reduce complexity. At first glance no signs of hysteresis are seen, which would be recognized by a clockwise or anticlockwise loop. However, the sampling period only took place during the increase of the discharge, and therefore it can not be suspected that a loop is present. There are two peaks in concentration noted in the figure, one at lower discharges and one at higher discharges. The peak of plastic concentration at the higher discharges is occurring before the peak in discharge.

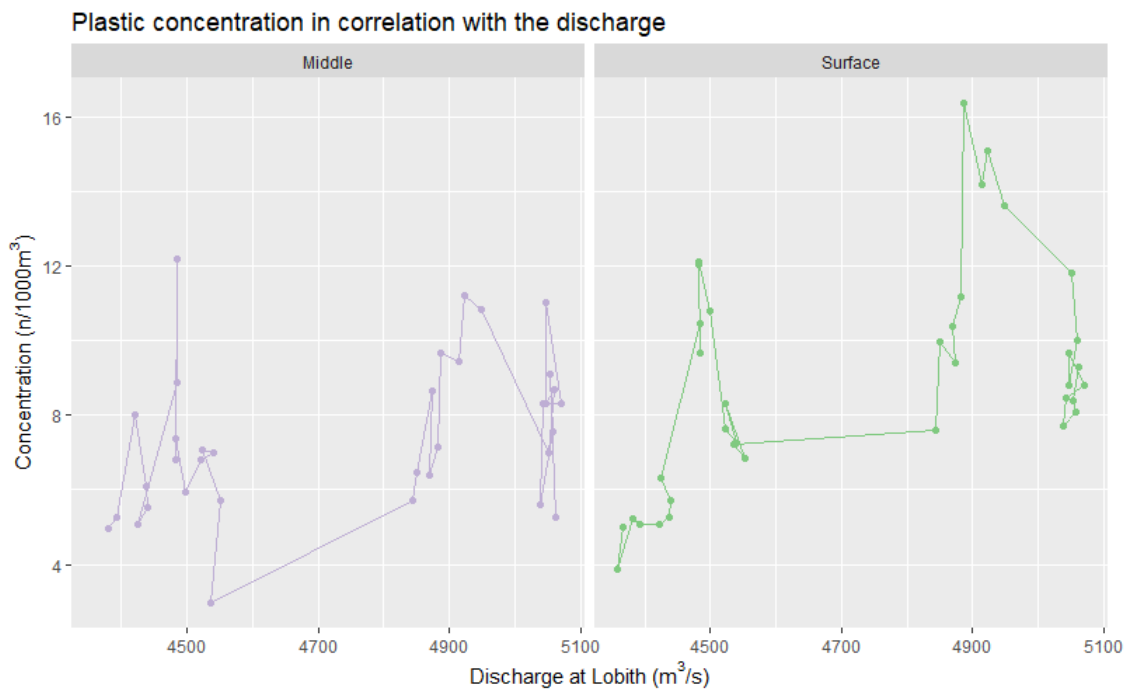


Figure 19 - The plastic concentration in correlation with the discharge at Lobith. Left: the plastic concentration of the middle of the water column, Right: the plastic concentration for the surface of the water column.

The most abundant individual categories ( $\geq 15$  pieces categorized) were also individually plotted against the discharge. The most striking categories are the unidentifiable hard plastics (meso and macro), unidentifiable styrofoam pieces (meso- and macro size) and plastic cotton swabs, which are mostly found at the surface of the water column. All these categories have a peak in concentration on the third day and at higher discharges. This rise concentration is occurring before the discharge peak and is falling when the discharge is still rising. The styrofoam pieces rarely occur at discharges below  $4800 \text{ m}^3/\text{s}$ , shown in figure 20 the cause for this is unknown. The categories that have this peak, are all categories of plastics that primarily float and are mainly found at the surface of the water column. A lot of categories have no clear pattern in their concentration, for example the strings and cord category, shown in figure 20. The string and cord category is found in both the middle and the surface of the water column, however there are no clear peaks or other patterns observed.

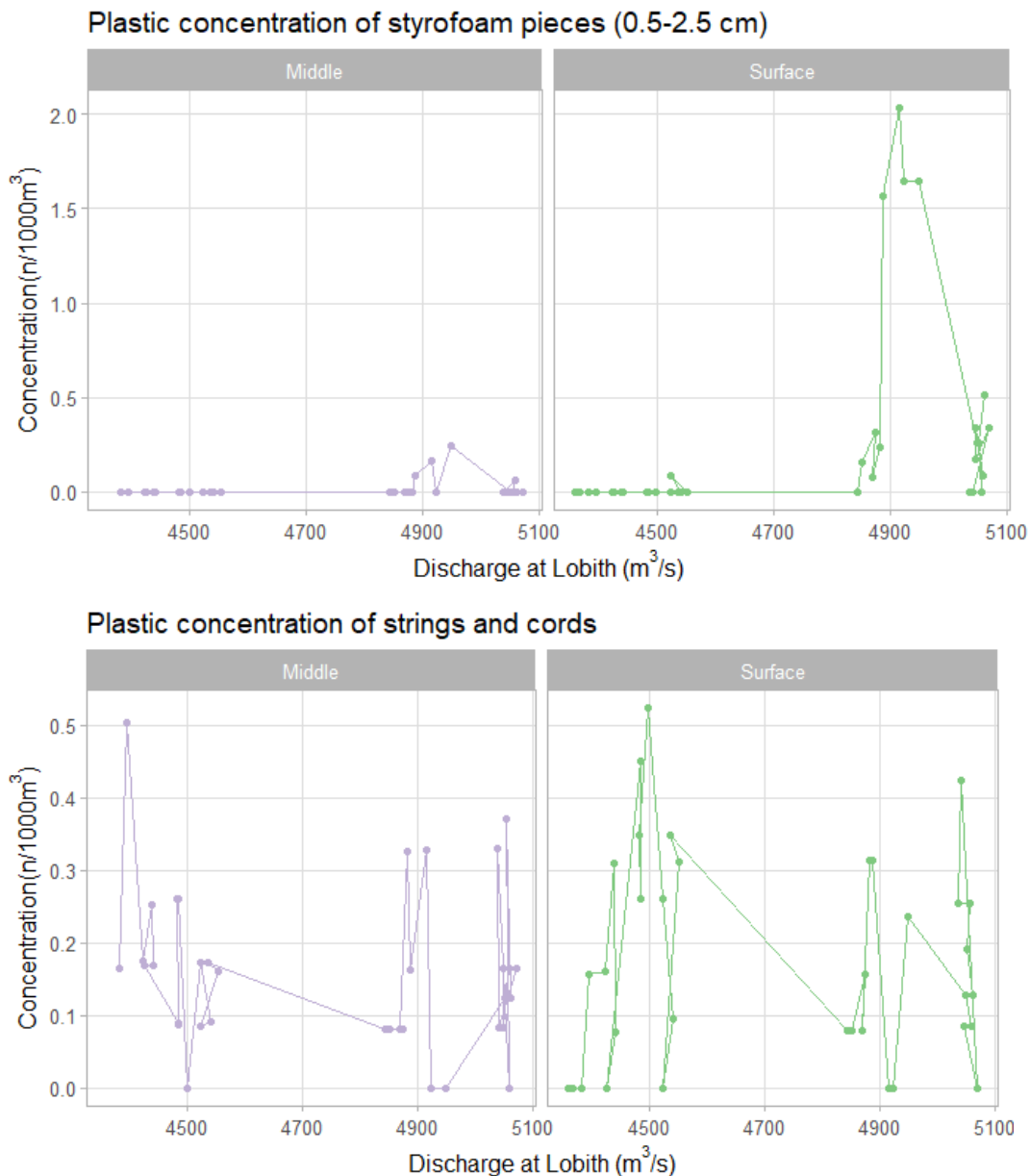


Figure 20 - Top: Plastic concentration of styrofoam pieces (0.5-2.5 cm) plotted against the discharge at Lobith. Bottom: The plastic concentration of strings and cords plotted against the discharge. The left sides are the plastics concentrations of the middle and the right side are the plastic concentrations of the surface of the water column.

#### 5.4.3 Plastic concentration compared to previous years

The total plastic concentration of this sampling year showed significant differences in concentration between the middle and surface of the water column. However during the sampling of 2021 and 2022, concentration did not differ significantly between the middle and surface of the water column. The table below shows the mean plastic concentration of the different years. The samples of 2023 had a higher plastic concentration than the samples of 2021 and 2022. When comparing the concentration of plastic in combination with the discharge (Appendix B), it is noted that with higher discharges in 2023, higher concentrations of the plastics are found. The difference in concentrations between 2021 and 2022 is however not significant.

Concentration of the right side of the river (items/1000m ³ )	2021	2022	2023
	Mean	Mean	Mean
<b>Middle</b>	2.19	5.77	7.4
<b>Surface</b>	1.80	7.56	9.0

Table 6 - The mean plastic concentration for the different sampling years.

In 2021, 12 samples were taken at the right side of the river in the middle and surface of the water column. These were taken on the first and last day of sampling, as shown in appendix C. The figure does not give any insights in a relationship between the plastic concentration over time. The discharge during the sampling period decreased from 1800 to 1600 m³/s as shown in Appendix D. Figure 21 shows the plastic concentration data plotted against the discharge. The few data points prevents from any patterns to arise.

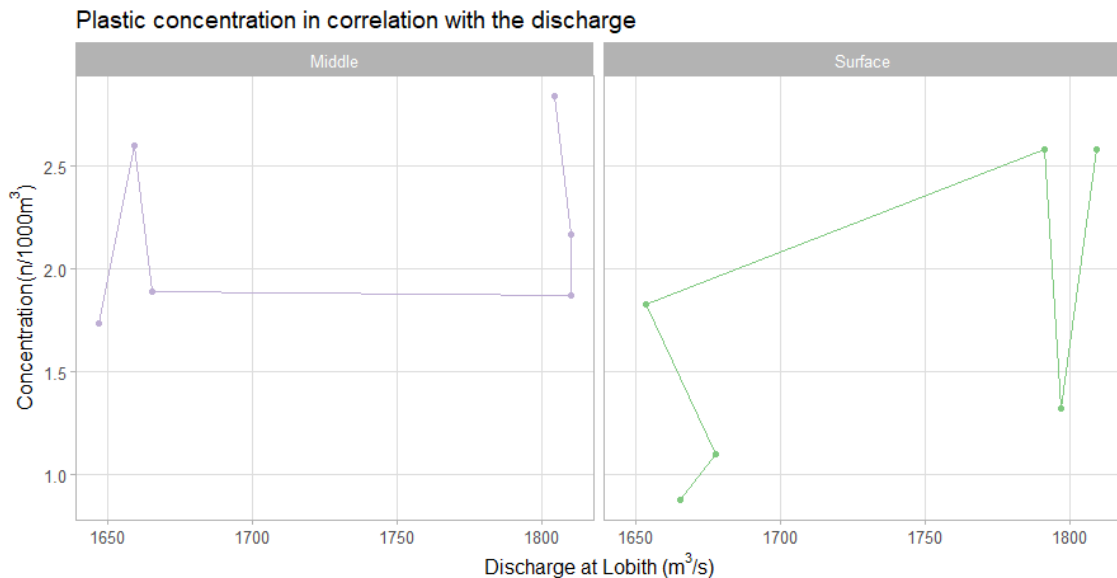


Figure 21 - The plastic concentration in correlation with the discharge at Lobith during the sampling of 2021. Left: the plastic concentration of the middle of the water column, Right: the plastic concentration for the surface of the water column.

In 2022, 15 samples were taken on the right side of the river in the middle and at the surface of the water column. These samples were taken throughout the sampling period, shown in appendix E. During this sampling period, a peak in discharge was observed, however no clear peak in plastic concentration was detected. Figure 22 shows the plastic concentration data plotted against the discharge. The concentration at the peak discharge is higher than the plastic concentration at the lower discharges. After the peak discharge, the plastic concentration at the surface stayed higher than before the peak discharge. Since no significant differences were found in the data of 2022 between the middle and the surface, figure 23 combines the two vertical positions.

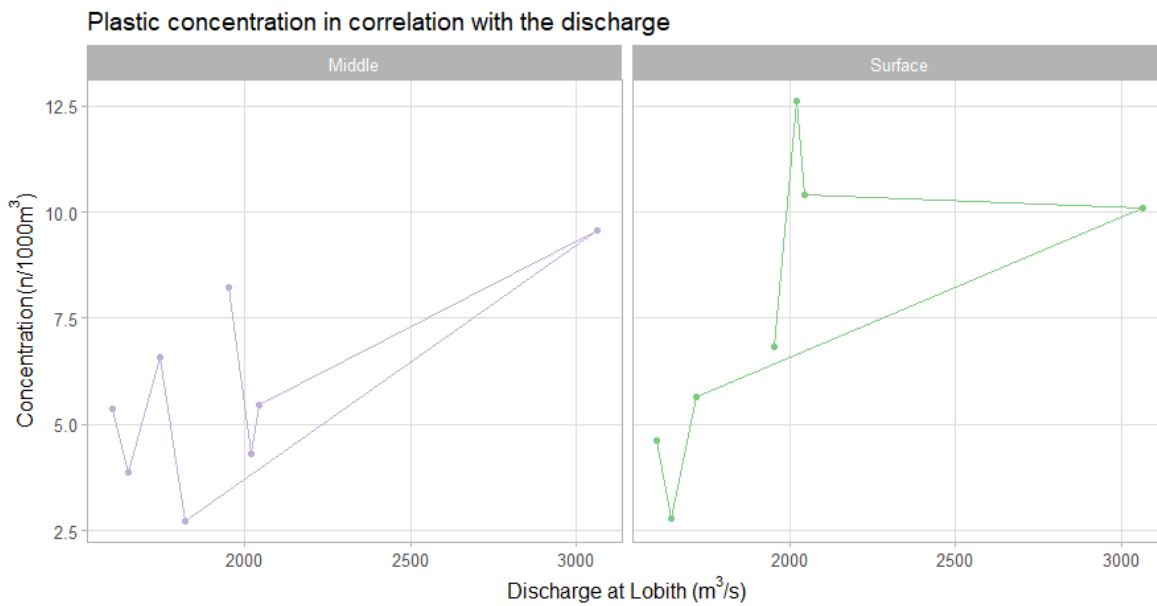


Figure 22 - The plastic concentration in correlation with the discharge at Lobith during the sampling of 2022. Left: the plastic concentration of the middle of the water column, Right: the plastic concentration for the surface of the water column.

When combining the surface and middle data of the plastic concentration, a pattern is formed. Starting with low concentrations at low discharges (between 1500-1800 m³/s), the plastic concentration rises significantly when the discharge rises around 1500 m³/s between the samples. After the peak discharge, the plastic concentration does not lower to the concentrations before the peak discharge. This pattern could be defined as an anti-clockwise hysteresis pattern.

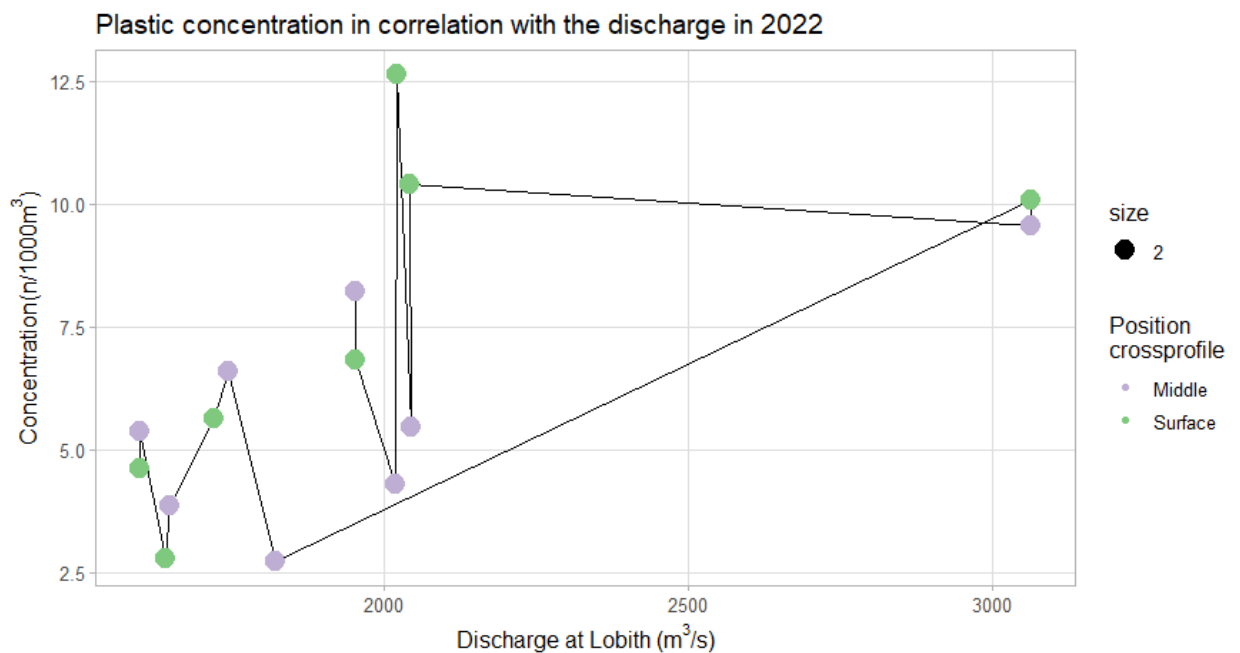


Figure 23 - The plastic concentration in correlation with the discharge at Lobith during the sampling of 2022. The line starts at the first sample to the last sample. The colors indicated the position of the sample in the water column.



## 6. Discussion

To find out the correlation between the plastic particles in the Rhine and the discharge, several objectives were answered. Different kinds of plastic found in the river were studied in combination with respect to their vertical positioning in the river. The distribution of the plastic categories found during high discharges were also compared to the categories found during low discharges. The concentration of the plastic was then calculated with the local flow velocity, from where spatial and temporal variation in plastic concentration was searched. This plastic concentration was again compared with concentration data of previous year.

### 6.1 Categories

The most dominant category according to the River OPAR-method was 'plastic/polystyrene pieces of soft plastic' which contributed to 83.5% of the total plastics. This was around 5% more than the years 2021 and 2022. The origins of these plastics are hard to define since they are smaller fragments from possibly plastic bags, (food) products or the clear plastic films that are around a lot of food products. These plastics are fragmentating due to navigation or weathering because they are heavily affected by turbulence (van Emmerik & Schwarz, 2020). Other abundant categories were unidentifiable hard plastics and food and snack packaging.

Remarkable 'rest plastic' finds were small stickers from a glass company, which are attached onto new glasses like wine glasses. These small stickers were found six times on different days in different samples. The factory of this German glass brand is situated in Bad Dribur, however it located at the edge of the drainage basin of the Rhine and flows into the drainage basin of the Weser ("Company - Leonardo," 2023). This suggest that these stickers do not come from the factory but from people who removed the stickers from their glasses and did not dispose them correctly or from spillage during transportation.

#### 6.1.1 Categories over vertical position

The distribution of the categories over the vertical position of the water column was not evenly divided. Some categories were mostly found at the surface of the water column. 100% Of cotton swabs, 95% of unidentified styrofoam pieces (meso and macro) and 90% of plastic/polystyrene pieces of hard plastic (2.5-50 cm) are found at the surface of the water column. Particles in surfaced transport are influenced by buoyancy, turbulence and surface tension, where surface tension becomes more important when the particle is small (Valero et al., 2022). The size of the plastics that are found in the Rhine are relatively small as a result of shipping, and are therefore more influenced by surface tension than larger particles. Where the buoyancy of a plastic is also influenced by their density, when it is fully submerged in water it will create a higher upward force. This results in the plastic particle reaching the surface, which will further enhance the high upward force. Particles which are more creased will also have a stronger surface tension since they have a larger interfacial contact line (Valero et al., 2022). Only turbulence can create a drag force, from which the plastic will move down again. This process will repeat itself, until the characteristics of the plastic particle change and a new balance in forces establishes. This process could have influenced the abundance of plastic particles at the surface of the water column during the sampling of 2023. However, data of the bottom of the water column is also necessary to fully establish the correct relationship between forces on a plastic particle. The ratio of the pieces at the surface and in the middle can be used to eventually extrapolate this data for the entire water column, however this is only applicable for the surface and the middle of the water column with this data. The ratio with the bottom of the water column is unknown and needs to be investigated in future sampling.

Some rare categories e.g. cigarette butts, straws, pieces of fireworks, band/tie-wraps) were predominantly located in the surface of the water column. The reason for this can be because of their density or it can be a coincidence. The lack of data on these categories prevents from further conclusions and more data is needed. 95 Percent of the unidentified styrofoam pieces, for both macro- and mesoplastic, are located at the surface of the water column, this is likely caused by their density, which is lower than water. Likewise 90 percent of hard plastic/polystyrene pieces (2,5-50 cm) are found in the surface of the water column and 74 percent of large hard plastic/polystyrene pieces (2,5-50cm) are also found at the surface. High surface tension and buoyancy are likely the cause for this. Performing laboratory tests to find the plastic flow characteristics per category can help to improve the knowledge about this. This should include drops tests in water and flow test to find the category specific characteristics of plastic transport, taking the average size of the plastic pieces in the Rhine into account. Valero (2022), mainly investigated the transport characteristics of large plastic pieces, like plastic cups and mouth masks, which are not found regularly in the Rhine.

#### *6.1.2 Categories compared to previous years*

There were substantially less sanitary wet wipes found in the samples of 2023 compared to 2021 and 2022. Only 0.13% of the plastic were sanitary wet wipes in 2023 (n=9), compared to 6.5% and 3.8% of sanitary wet wipes in 2021 and 2022 respectively. However, the data of the previous years also included other horizontal and vertical locations. When comparing the sanitary wet wipes concentrations of 2023 to the other years sanitary wet wipes concentration of the right side of the river and the surface and middle of the water column, no big differences are noted. Most of the sanitary wet wipes in the previous sampling years were found on the left side of the river at the bottom of the water column. The sanitary wet wipe concentration of 2023 is thus a good representation of the concentration at the sampled locations, however it is an underestimation of the total concentration of sanitary wet wipes in the cross-section of the river, since most are found on the left side of the river.

The strings and cords category (diameter <1cm) was found less than in the previous years. It was only found 2.2% percent compared to 6.5% and 6% in 2021 and 2022 respectively. Possible sources are geotextile or fishing gear (Collas et al., 2021). Oswald (2021), even found a contribution of strings and cord of 34.4% in the Upper-Rhine, it should be noted that this sampling was done with finer nets. Oswald noted that most strings and cord were found at the bottom of the water column, which could be a reason for the lower abundance in the 2023 samples (Collas et al., 2021). Another reason can be the shape of the strings and cords, since they are mostly long and thin, they can easily flow through the nets. This will cause an underestimation from the actual presence in the water column. A recommendation can be to do a net efficiency test, which can be performed for this category as well as for other categories to make a correction factor.

A total of 40 categories were identified during this sampling period. Which is less than the number of categories found in 2022 (42) but more than 2021 (33). Rus (2022), suggested that the difference in the total categories between 2021 and 2022 had to do with the discharge or the measuring duration. Since the discharge was much higher in 2023 compared to the previous years, this can probably be rejected. A reason for this difference can be the lack of different positions of sampling, since no bottom samples were made as well as no left side and middle of the river samples. Other reasons for the difference could be the different total duration time which was less in 2023 (2130 minutes) compared to 2022 (2947 minutes), the number of sampling days, different source areas or other storage densities.

## 6.2 Concentration

Significant differences were found between the middle and the surface of the water column on the right side of the river. Local differences in flow velocity caused by helical flow could explain these differences. In 2021 and 2022 no significant differences were found between the middle and the surface of the water column at the right side of the river. Hohenblum et al., (2015) found a homogenous distribution of plastics in the Danube river and attributed this to turbulence due to high discharges and flow velocity. At a lower discharge Hohenblum even found a more layered plastic distribution in the Danube river (Hohenblum, 2015). However, during this sampling period the discharges were relatively high (4000-5000 m³/s) compared to the average discharge (2300 m³/s). High turbulence and flow velocities could create a homogenous plastic concentration as it would with suspended sediment concentration (Charlton, 2007). A high abundance of floating plastics was found at the surface of the water column, these plastics have a high surface tension and buoyancy and will therefore keep floating (Valero et al., 2022). The turbulence did not play a large role in this for the particles found at the surface, or it did not play a large enough role for the particles. Thus the discharge of the river is not the only factor that contributes to the mixing of plastics vertically since surface tension and buoyancy also play a large role. Since this sampling period only took place at the right side of the river due to the high discharges and lack of time, extrapolating the concentrations from this side of the river to the whole horizontal cross section is not advised. It would probably overestimate the concentrations since the flow velocity is highest at the right side of the river because of the meander bend. Carrying out ADCP measurement across the whole cross section of the river will gain information about the flow velocity at the other points in the river. This can prove to be useful when trying to extrapolate the concentrations for the whole cross section and depth of the river. Caution is necessary however, since exact ratios of plastic concentrations are not yet known.

### 6.2.1 *The plastic concentration over time*

The results of the plastic concentration over time followed a striking pattern. It did not follow the same curve as the discharge and had two peaks in concentrations. On the first day, the plastic concentration rises at the surface and has a small peak in the middle of the water column. This small peak can be a result of high turbulence since the concentrations of the surface during that peak are smaller than the concentrations at the middle. However, this can not be stated with complete certainty and more factors can contribute to this peak. The concentration on the second day both descend at the surface and the middle of the water column, which is not in line with the rising discharge. When looking more closely at the discharge graph of this day, we see a small stagnation in the rise of discharge. The second rise in concentration is seen on the 3rd day which on the 4th day descends again. This peak of plastic concentration is occurring before the discharge peak which is noteworthy since it again does not follow the same curve. The cause between the two peaks in concentration and the rise in discharges is hard to pinpoint. For example, a lot of plastics could have been picked up from the riverbanks during the rising water level somewhere along the Rhine and the peak concentration of this happening far before the peak discharge. Since not all riverbanks are located at the same level above the water level, they could flow over at different moments in time, causing different periods of high plastic concentrations. Knowledge about the heights of individual riverbanks mainly in Germany is needed to validate this theory. Another reason for the peaks in discharge can be a result of a sewer spillage from which the plastic concentration rose. A suggestion is to get information about sewer outlet locations and moments they are in use.

### 6.3 Hysteresis

A hysteresis analysis with the available data could not be fully completed since there only was data of the plastic concentrations during a rising discharge. This results in only concentrations of plastics at 1 phase of a hydrograph and lacking the plastic concentrations during a decreasing discharge. However, even with only this data, a rising and descending concentration is observed during the rising of the discharge. This can be a precursor of a clockwise hysteresis pattern, which means that the plastic concentration is ahead of the discharge peak. A possible cause for a clockwise hysteresis loop is that the sediment supply areas, and in this case plastic supply areas, are close to the river (de Boer & Campbell, 1989). Plastics located on the riverbanks can be remobilized and enter the river again by higher water levels (van Emmerik, Strady, et al., 2019). There is a high probability that for this discharge event, plastics that were located on riverbanks were entrained by the rising water levels. The last time a event with a discharge higher than 4500 m³/s occurred, was in the beginning of January in 2022. Thus a lot of time has passed since this, which enhanced the amount of plastic debris that accumulated on the riverbanks. Along with the fact that not all riverbanks are the same height above the water level, there are different periods with higher concentrations of plastics when different riverbanks are flooded. The relation between the discharge and plastic concentration is still largely unknown and it is recommended that for further research, sampling should take place before, during and after a peak in discharge. Accomplishing this not an easy task, as the sampling period is determined long before the discharges of the Rhine are known. One solution for this could be measuring in a period when the discharge of the Rhine fluctuates the most due to storms and rainfall.

An individual concentration-discharge graph was made for styrofoam pieces, which showed a clear peak around the discharge 4900 m³/s. Before this peak, almost no styrofoam pieces were found which further supports the origin of the plastic pieces coming from the riverbanks. This peak is also seen in the hysteresis graphs of the unidentifiable pieces of hard plastic (both meso and macro) and styrofoam pieces larger than 2.5 cm. For all these categories, the peak in concentration is occurring before the peak in discharge which suggest a clockwise hysteresis loop will form. The plastic concentration of strings and cords showed no sign of hysteresis and seems to be completely random, so not all categories have an explainable relationship with the discharge and multiple factors seem to influence the plastic concentration.

The concentration-discharge data of this year was compared to the concentration data of 2021 and 2022. In order to do this, only the data from the middle and surface of the right side of the river was taken. This resulted in only a low number of samples for these years, which makes it challenging to get usable information. The data of 2021 was proven to be unworkable, as only 12 samples were taken at these positions. Together with only a small decrease in discharge no patterns could be observed. Data of 2022 has a bit more samples and an anti-clockwise hysteresis pattern could be seen. However, keep in mind that this is based on only a few samples (n=15). A higher concentration in plastic was observed after the peak discharge. The water level before the peak discharge was not high enough to inundate the groynes but the water level after the peak discharge did overflow the groynes. This emphasizes the findings in the concentration data of 2023 that suggest the most plastic originates from the riverbanks. Since the riverbanks did overflow during the sampling period in 2022 and a higher plastic concentration was observed after this situation.

### 6.4 Sampling method

The total number of plastic pieces that have been found during this study is an underestimation of the total plastic pieces in the river, as there are multiple steps in this methodology that can contribute to losing or missing plastics. The mesh size of the nets was

6 millimeter, the lower size limit of meso- plastic is 5 millimeter, which can cause an underestimation of mesoplastic particles. Additionally, elongated particles, for instance strings, cords and tie-wraps can flow through the nets which adds to the underestimation of meso- and macro plastics. During the cleaning of the nets on the boat, the trawls are shaken which can cause small plastic particles to fall through the net. Plastic particles that fell onto the boat during the shaking, were afterwards picked up by the fishermen to reduce this problem. During the sorting process on the boat, where the plastics were separated from the organic matter, it is always possible that some plastic particles were missed, mainly when a lot of organic material was present. To reduce this error, all samples were sorted out by multiple people and as precisely as possible.

Using this sampling method is recommended for future campaigns. Using a vessel is beneficial as it allows monitoring at various positions in the river in contrast to visually monitoring from bridges. Further investigation of the plastic distribution at high discharges is needed to be able to further analyze hysteresis patterns and eventually find the major source areas of the plastics.

## 7. Conclusion

This study provided new insights in the plastic categories and concentrations during high discharges with a return period of one year in the Rhine river. We measured significant differences between the middle and surface of the water column (on the right side of the river) which were contradicting the results of earlier years. However, these differences can possibly be linked to the high abundance of floating plastics during the monitoring period. These floating plastics are less affected by turbulence because of their low density and therefore will stay more at the surface of the water column. It is also noted that turbulence does not only influence the spatial distribution but buoyancy and surface tension also play an important role in the vertical distribution of plastic pieces.

The plastics were divided into different OSPAR-categories which provided a good insight into the different categories that are present in the water column of the Rhine. The most abundant found category of meso- and macroplastic was the plastic/polystyrene pieces of soft plastic, which origins are difficult to locate. It is highly likely that these are fragments of food packaging, plastic bags or other thin plastics that degrading due to their transport in the river. Extrapolating the plastic concentration data to the whole cross section of the river is not advised. For example, the sanitary wet wipe concentration data of 2023 was multiple percent lower than the concentration in 2021 and 2022. However, when comparing the sanitary wet wipe concentration data of 2023 to the data of the previous years of the same locations in the river, no big differences were observed. Thus, the sanitary wet wipe concentration data of 2023 was a good representative for the locations in the middle and surface of the water column of the right side of the river, however not for the entire water column since most sanitary wet wipes found in the previous years were on the left side of the river.

The influence discharge has on the plastic concentration is still complex. However, a precursor of a clockwise hysteresis pattern was detected in the total plastic concentration and in the plastic concentration of styrofoam and unidentifiable hard plastics. The peak of the plastic concentration is located before the peak of the discharge which shows that the plastics that were present in the water column originated from nearby sources, such as nearby riverbanks. These riverbanks were overflowed by the rising water level and the plastics located at those riverbanks were entrained by the water. This is further reinforced by the plastic concentration data of 2022. During this sampling period a peak in discharge was observed, as well as a peak in plastic concentration. However, a higher concentrations of plastic was found after the peak discharge, which could be caused by the riverbanks that were overflowed during the rise in discharge. However, the relationship between the discharge and the plastic concentration is very complex and is influenced by hydrological variables, anthropogenic factors and more. More sampling during high discharges is advised, which should be before, during and after a discharge peak to get the full scope of this relationship.

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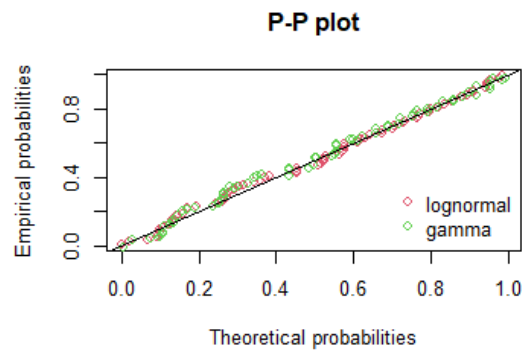
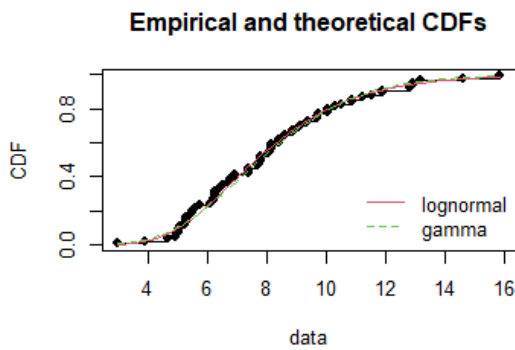
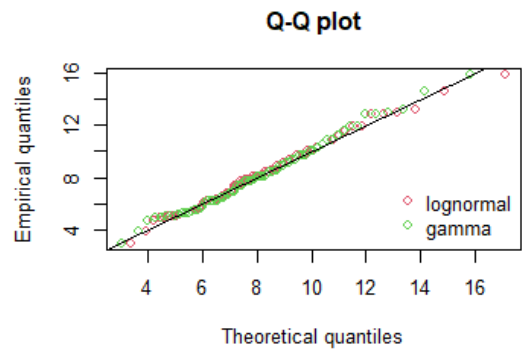
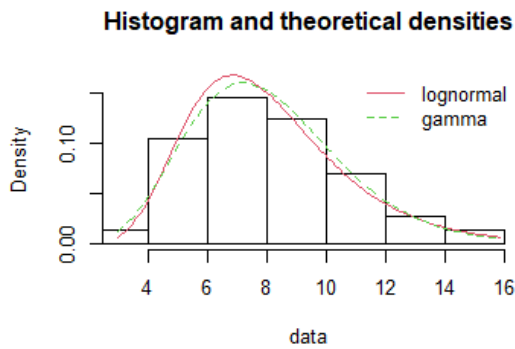
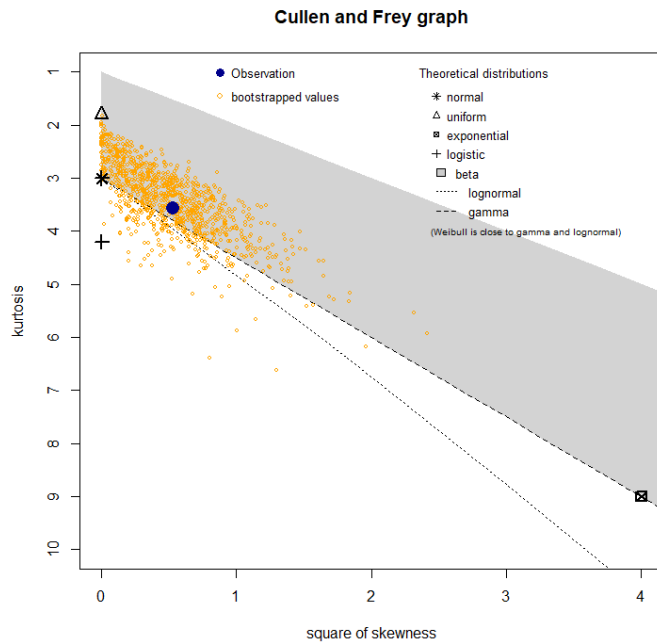
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# 9. Supplementary material

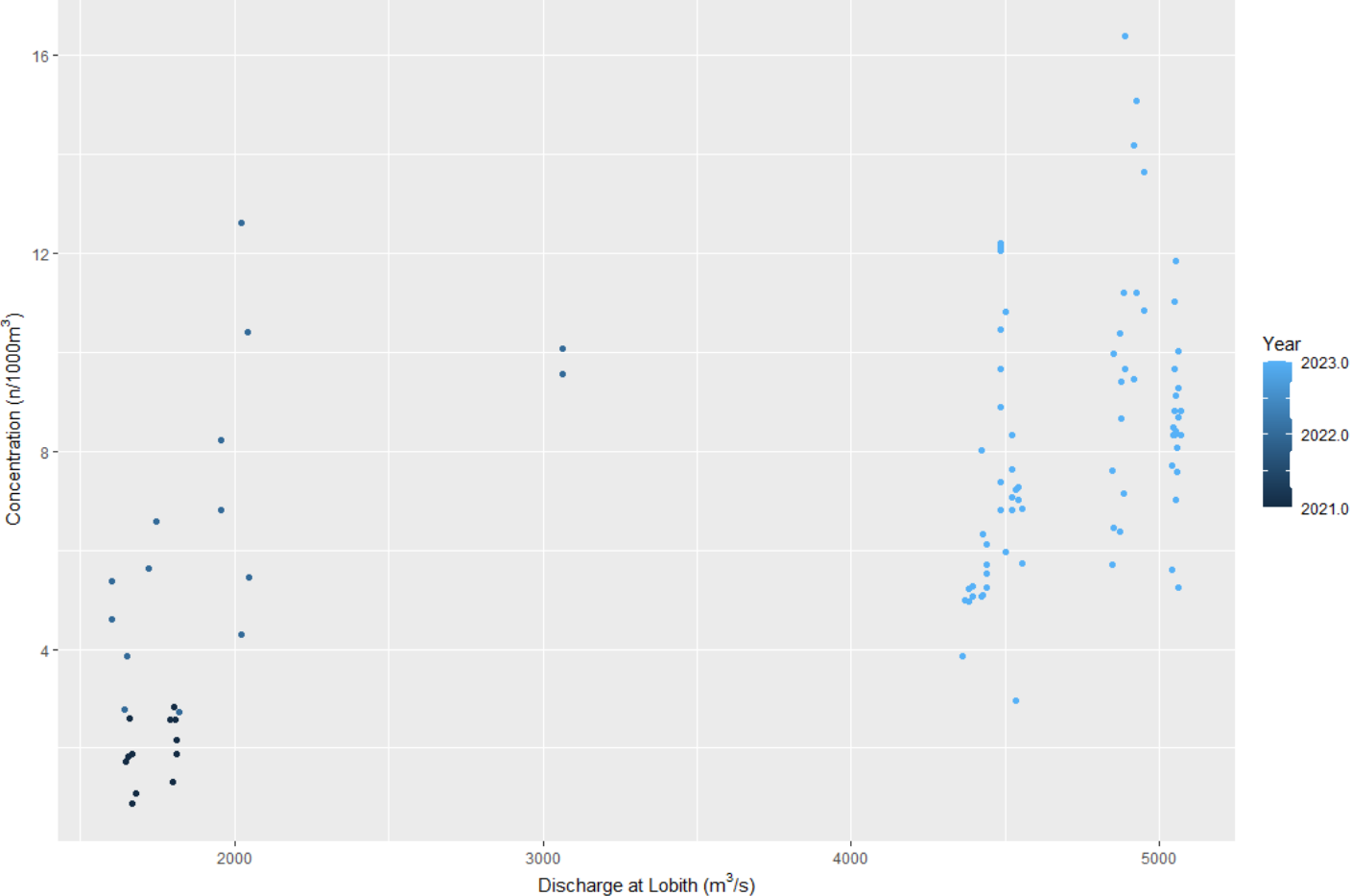
## Appendix A

Histogram, cumulative distribution and Cullen and Frey graph of the concentration data



# Appendix B

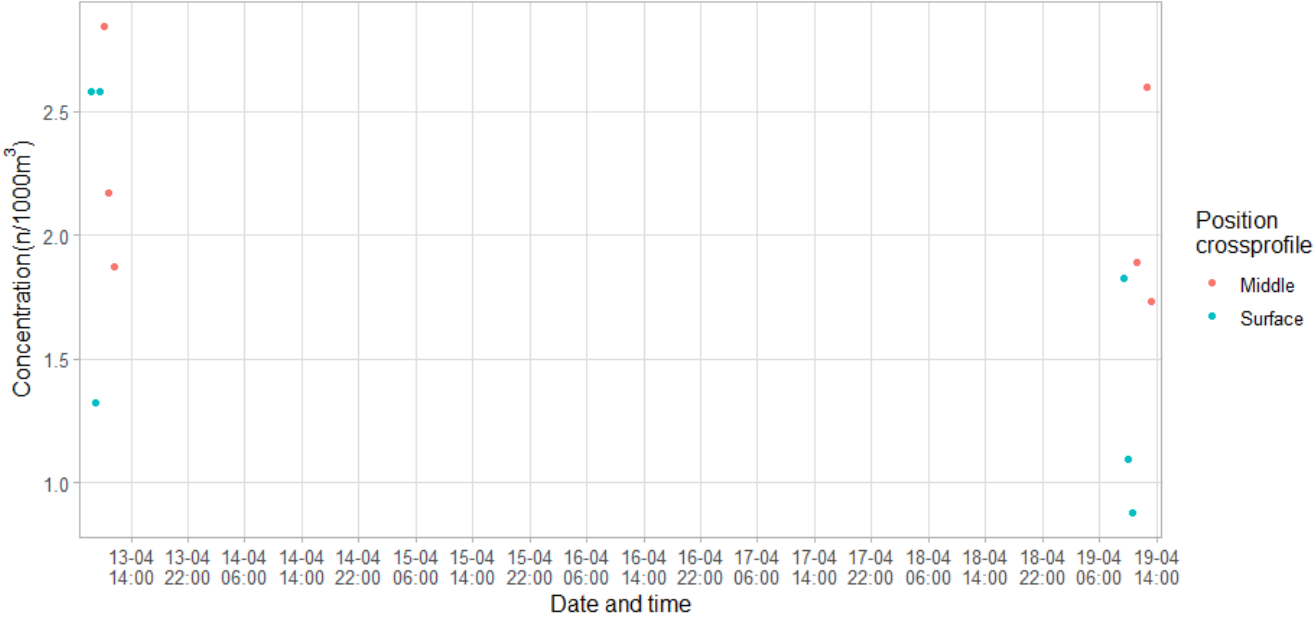
The concentration of the plastic particles in 2021, 2022 and 2023 plotted against the discharge at Lobith.



# Appendix C

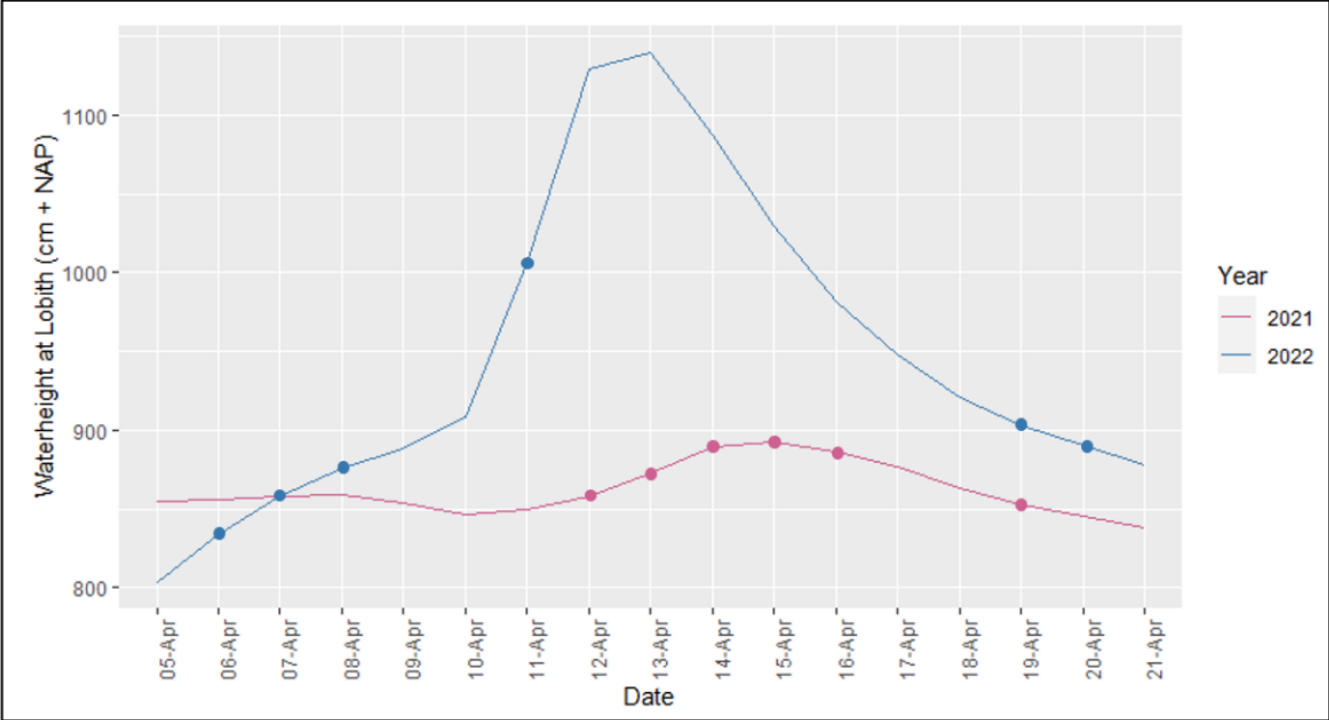
The plastic concentration of 2021 over time.

The plastic concentration over time (2021)



# Appendix D

The water level at Lobith over time during the monitoring periods of 2021 and 2022 (Rus, 2022)



# Appendix E

The plastic concentration of 2022 over time.

