Microplastic pollution in the Vaal River system

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DECLARATIONS

All samples were collected and analysed by me at the North-West University. Additional FT-IR analysis conducted by Aalborg University, Denmark.

I conducted the data analysis and interpretation and drafted all chapters. Co-authors were consulted for clarification only.

This document is my own work and has been checked for plagiarism.

Chapters two, three and four are published works and chapters five, six and seven are manuscripts ready for publication.

I have received permission from my supervisor and co-authors to submit all manuscripts as part of my dissertation for the completion of the degree, Doctor in Environmental Science with Environmental Science at the North-West University.

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Carina Verster

Preface

This thesis concerns microplastic pollution in South African freshwater systems. It focuses on the Vaal River and two of its tributaries, the Klip River and Vals River, but includes data from adjacent catchments. Surface water and riverine sediment were used as matrices. This thesis is presented in article format and consists of eight chapters:

Chapter 1:	An introduction to the topic of microplastics as a global issue which contextualises the knowledge gaps addressed in the chapters to follow.
Chapter 2:	An overview of the South African plastics industry and where microplastics fit into the country's socio-economic landscape.
Chapter 3:	This chapter forms part of a scientific review series on oceanic plastic around South Africa. It identifies the land-based sources of marine plastic and the importance of rivers as pathways and sinks of plastic waste.
Chapter 4:	An overview of airborne microplastics as possible source of this ubiquitous pollutant. It also highlights the possible adverse effects of microplastics.
Chapter 5:	The first scoping study for microplastics in Gauteng rivers, including the Vaal River, conducted in conjunction with the Water Research Commission. Gaps identified in this study lead to the experimental design of the study discussed in the next two chapters.
Chapter 6:	Spatial distribution of microplastics in the Vaal River catchment and the factors that affect it.
Chapter 7:	How microplastic concentrations vary over time – the effect of seasonality.
Chapter 8:	Discussion, conclusion and recommendations.

"*Much that is natural, to the will must yield*."

- Thom Gunn, On the move

This is line from a poem about man's restless pursuit of what remains just out of reach. It stuck with me since I had first read it in Mrs Lorraine's English class at high school. Each person gathers as much as possible from a fear of not having or being enough, and nature is the ill-defended victim of our greed. I believe that if you can alter the will of man, you can limit the measure to which nature must yield. But how do we do that?

The will of man is driven by the notion of self-preservation. If there is no evident benefit to an action, whether the benefit is physical, emotional, spiritual or social, it is very unlikely that the action will be done. The current societal structure does not allow for most individuals to realise the benefit and even dependency they have from and upon nature. A person therefore sees no plausible reason to sacrifice his/her comfort or profit in the short term, to ensure the preservation of the benefits nature offers in the long run.

You cannot blame a person for choosing what he thinks best if you do not understand the adverse effects on him/her and her/his family. We must get people back to nature to see, taste, smell and realise our dire dependency upon it. If you are one of the lucky few who have realised your dependency upon mother nature, make it your mission to help others see her benefits as well. Create opportunities for your neighbour to see the adverse consequences of their actions and that in hurting the environment, they are hurting themselves.

The study and thesis to follow is an attempt to do exactly this.

"*Unless someone like you cares a whole awful lot, nothing is going to get better. It's not*."

- Dr Seuss, The Lorax

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ABSTRACT

This thesis aimed to address the escalating issue of plastic pollution in South Africa's freshwater systems across several key objectives. Findings from the study highlighted the extensive presence of microplastics (MPs) throughout the Vaal River system, particularly in sediment samples, with concentrations in some areas comparable to highly polluted rivers in other regions. Notably, the heavily polluted Klip River exhibited the highest MP concentrations, including an unprecedented high presence of microbeads, indicating the severity of pollution in urbanized and densely populated areas where waste mismanagement is prevalent.

Identification of plastic pollution sources revealed a strong correlation between macroplastic presence and MP concentrations in water and sediment, emphasizing local environmental plastic pollution as a significant predictor for riverine MP loads. Urbanized catchments, characterized by dense populations and inadequate waste management, were found to host the highest MP concentrations. Furthermore, the temporal dynamics of MPs in South African freshwater systems demonstrated contrasting trends between heavily polluted and less impacted rivers during the rainy season, emphasizing the influence of runoff on MP concentrations.

Insights into the sinks of microplastics underscored sediments as temporary repositories for MPs, particularly during low-flow periods, with rapid settling observed, especially for microbeads. Additionally, wetlands were identified as effective filters, trapping MPs in sediment during low-flow periods, raising concerns regarding the accumulating MP concentrations in these sensitive ecosystems.

Addressing mitigating actions, recommendations emphasized the urgent need to improve waste management infrastructure, enhance functionality at wastewater treatment plants to minimize MP release into rivers, restrict microbead production, raise public awareness, conduct regular river cleanups, and remove macroplastics from the environment to prevent further generation of MPs. These findings collectively underscore the critical importance of strategic interventions and holistic measures to curtail and remediate plastic pollution in South Africa's freshwater systems.

Keywords: Microplastic, Vaal River, South Africa, sediment, waste management, seasonal, wetland

OPSOMMING

Die doel van die studie was om die toenemende plastiekbesoedeling in Suid-Afrika se varswatersisteme aan te spreek via verskeie sleuteldoelwitte. Bevindinge uit die studie het die uitgebreide teenwoordigheid van mikroplastiek (MP's) regdeur die Vaalrivierstelsel beklemtoon, veral in sedimentmonsters, met konsentrasies op sommige plekke vergelykbaar met hoogs besoedelde riviere in Europa en Asië. Die hoogs besoedelde Kliprivier het die hoogste MP-konsentrasies getoon, asook ongekende hoë teenwoordigheid van mikrokorrels, wat die erns van besoedeling in verstedelike en digbevolkte areas waar afvalbestuur gebrekkig is, aandui.

Identifikasie van plastiekbesoedelingbronne het 'n sterk korrelasie tussen die teenwoordigheid van makroplastiek en MP-konsentrasies in water en sediment aangetoon, wat die plaaslike omgewingsplastiekbesoedeling as 'n belangrike voorspeller vir MP in riviere beklemtoon. Stedelike opvanggebiede, gekenmerk deur digte bevolkings en ontoereikende afvalbestuur, het die hoogste MP-konsentrasies getoon. Verder het die verandering van MP oor tyd in Suid-Afrikaanse varswatersisteme teenstrydige tendense tussen sterk besoedelde en minder geaffekteerde riviere getoon tydens die reënseisoen. Besoedelingsvlakke van afloopwater is dus ŉ bepalende faktor.

MP en veral mikrokorrels gaan lê in riviersediment tydens droë tydperke, en kan weer uitgewas word tydens hoë vloei. Vleilande is geïdentifiseer as effektiewe filters wat MP's in sediment vasvang tydens tydperke van lae vloei, wat kommer wek oor die toenemende MP-konsentrasies in hierdie sensitiewe ekostelsels.

Met betrekking tot die voorgestelde plastiek bekampings maatreëls, is aanbevelings gemaak om die dringende behoefte te beklemtoon om afvalbestuursinfrastruktuur te verbeter, funksionaliteit by waterbehandelingsaanlegte te verbeter om MP-vrystelling in riviere te minimaliseer, produksie van mikrokorrels te beperk, openbare bewustheid te verhoog, gereelde rivierskoonmaakaksies uit te voer, en makroplastiek uit die omgewing te verwyder om verdere generasie van MP's te voorkom. Hierdie bevindinge beklemtoon die belang van strategiese ingrypings en holistiese maatreëls om plastiekbesoedeling in Suid-Afrika se varswatersisteme te verminder en te verlig.

Sleutelwoorde: Mikroplastiek, Vaal Rivier, Suid-Afrika, sediment, afvalbestuur, seisonaal, vleiland

TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

Chapter 1

Introduction: Microplastic pollution in the Vaal River system

1.1 Introduction

1.1.1 Plastic material

Plastic as a material is generally understood to include synthetic (fossil-fuel-derived) or natural organic (biomass-derived) polymers that can be formed into desired shapes and forms (Wagner & Lambert, 2018). It is a widely used, low-cost material used in packaging, medical applications, piping, construction, vehicle parts, toys, electronics, etc. (Karasik *et al*., 2023).

There are various plastic polymers and several ways to classify plastic such as chemical and crystalline structures, production process, hardness, design, density, capacity to absorb water, conductivity, and degradability (Barrick *et al*., 2021). In some cases, plastics are pure polymers made from their constituent monomers and contain no additives to change its properties—like polyethylene and polypropylene (Wiesinger *et al*., 2021). Others have additives like plasticizers and colourants added to give the material desired properties (Hahladakis *et al.*, 2018), but when released in the environment, can bioaccumulate or act as persistent organic pollutants (POPs) in organisms (Barrick *et al*., 2021). Globally polymers differ in demand—the most consumed polymers are polypropylene at 21%, followed by polyethylene (18%), polyvinyl chloride (17%), high-density polyethylene (15%), polystyrene (8%), and polyethylene terephthalate (7%) (Hahladakis *et al.*, 2018). These polymers can also be produced as mixtures.

1.1.2 Benefits of plastic

Plastics have many benefits, with various applications in packaging as it is relatively inert and some virtually impenetrable (Andrady *et al*., 2009). It protects foods and other products from getting spoiled, soiled, or contaminated. Packaging is also used as marketing and product recognition tools. Other benefits include plastics being light weight thereby reducing transport costs (Karasik *et al*., 2023) and plastic piping and storage containers that reduce the chances of potable water pollution (Hahladakis *et al.,* 2018). Besides many benefits of plastic, its characteristics, when not managed properly, can have adverse impacts on the environment (Karasik *et al*., 2023). Many plastic products, plastics especially single use items, have a much longer lifespan than its intended time of use, creating large volumes of plastic waste that end up in the natural environment (Wang *et al*., 2021).

1.2 Microplastic

1.2.1 What are microplastics?

Microplastic (MPs) is a group of plastics sized between macroplastics and nanoplastics. For the purposes of this thesis and consistent with much of literature, I defined the size range for MPs as between 1 - 5000 µm (5 mm) (Frias *et al*., 2019). Macroplastics are generally understood to be easily visible objects like bottles, bags, and food containers. MPs are smaller, less distinct particles usually needing a microscope to visualise (Figure 1.1).

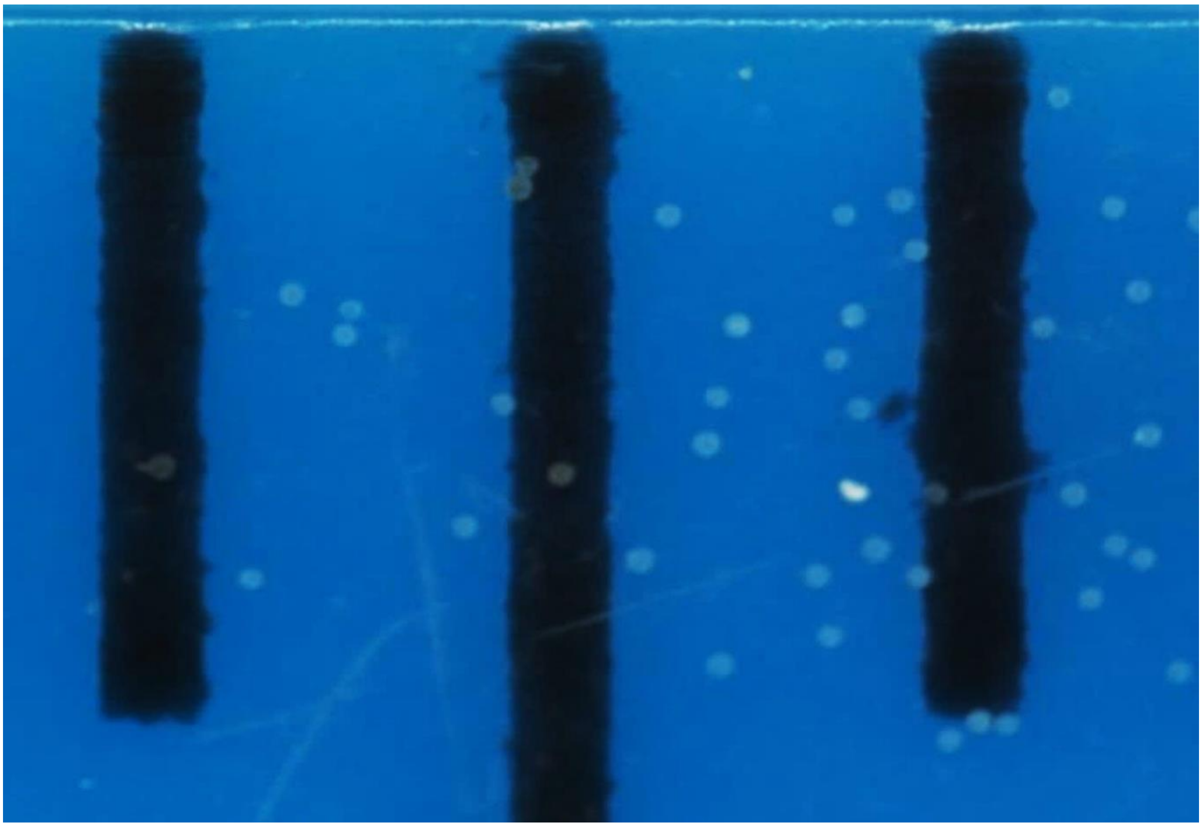

Figure 1.1: 60 µm microbead on a ruler with 1 mm increments. (Photo: C. Verster)

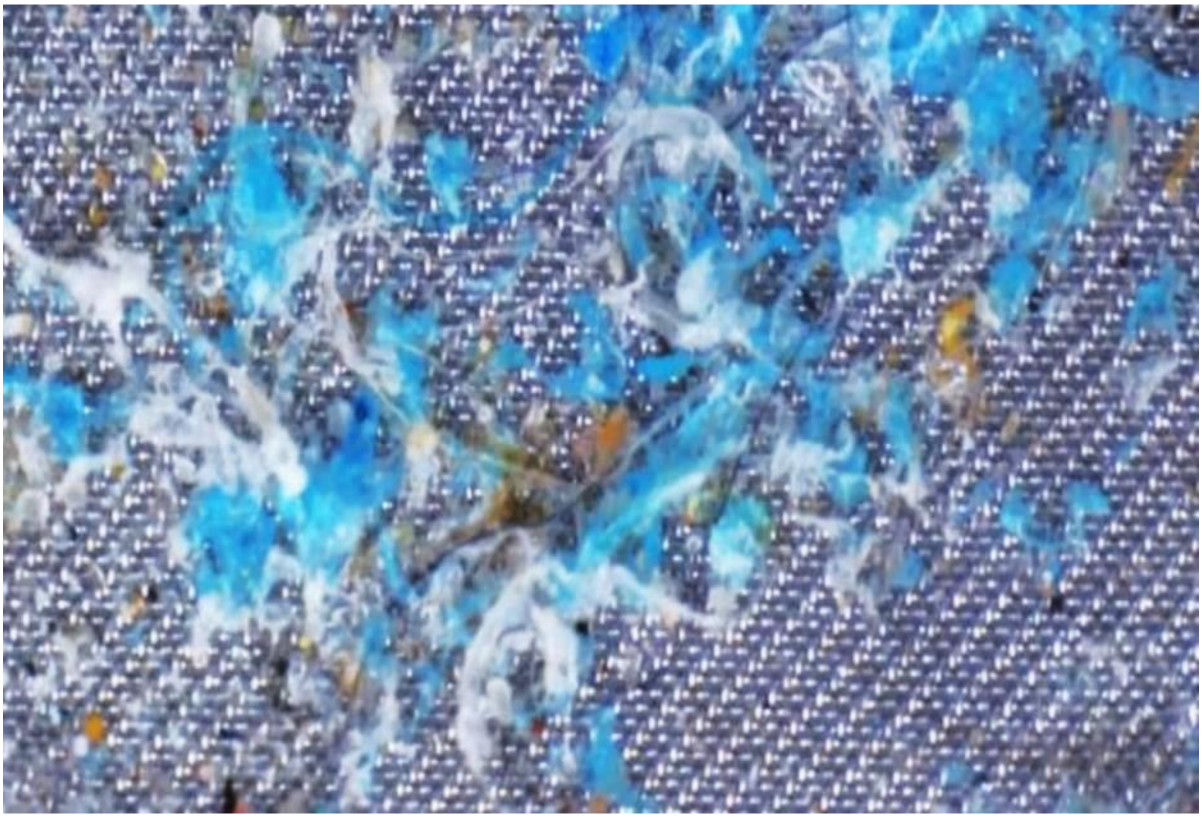

Figure 1.2: Fibre and fragments from Vaal River water on a stainless-steel sieve (Photo: C. Verster)

MPs can be described according to its source, size, shape, colour, and polymer composition (Wagner & Lambert, 2018). Fibres (Figure 1.2), pellets, fragments (Figure 1.2), beads (Figure 1.1), and films are some of the common MP morphotypes (Kooi *et al*., 2019). Some MPs are manufactured between 1 - 5000 µm and are called primary MPs (Cauwenberghe *et al.*, 2015). These include plastic pellets and nurdles used as raw plastic in industry, or cosmetic plastic particles used in cosmetics, toothpaste, exfoliators, and a host of other uses. They end up in water systems, often through municipal wastewater as a carrier (Arthur *et al.*, 2008).

1.2.2 Microplastic research

Since MPs were first detected in the ocean it has been associated with the degradation of larger macroplastics (Carpenter *et al*., 1972) and is used as proxy for general plastic pollution in the environment (van Emmerik, 2021). These MPs are classified as secondary MPs and include fibres (Dris *et al*., 2017), fragments (Thompson *et al*., 2004), and films (Claessens *et al*., 2011). Hydrodynamic interactions of MPs are different than that of its larger, often air-filled macro counterparts (Tsiaras *et al*., 2021) which must be kept in mind when using MPs as an indicator of general plastic pollution. Besides having varying sources (Osman *et al*., 2023), different MP morphotypes also have different hydrodynamic interactions due to differences in surface-area to volume ratios and densities (Dioses-Salinas *et al*., 2020). Fibres as a microplastic have become a recent field of interest in the microplastic community as it originates secondarily from wear and washing of textiles (Stanton *et al*., 2019). Fibres can be found in even the remotest of streams as it gets transported over long distances by air, and rural communities wash clothes in rivers or as washing effluent ends up in rivers (O'Brien *et al*., 2020).

Plastic is a ubiquitous contaminant in all environments (Loganathan *et al*., 2023). It is so common that it has been proposed to be used as the stratigraphic indicator for the Anthropocene (Zalasiewicz *et al.*, 2016), also called the Plastisphere (Pietrelli *et al.*, 2017). Although concentrations vary geographically, temporally, and in various matrices and trophic levels, MPs are found in almost every sample in which it is looked for—both biological and abiotic (Loganathan *et al*., 2023). They occur in aquatic, terrestrial, and atmospheric systems (Wagner & Lambert, 2018), and biota (Scherer *et al*., 2018). Oceans act as a sink for plastics as great volumes of terrestrial plastic make its way into aquatic systems and discharged into the oceans (Woodall *et al*., 2014). Microplastic particles have been found in surface water from all parts of the ocean (Andrady *et al*., 2011; Barnes *et al*., 2010; Him *et al*., 2015) as well as in deep-sea sediment, generally considered to be the major final sink for most marine microplastics (Van Cauwenberghe *et al*., 2015; Woodall *et al.*, 2014).

Microplastic as a research field is continuously developing. Its analysis is complex and resource intensive. The number of MP publications published has seen exponential growth since the start of the previous decade (Chaukura *et al*., 2021). Some assumptions and knowledge gaps regarding polymers, morphotypes, and sources may need to be acknowledged or assumed when conducting MP studies. Practically, in most studies the smallest size is determined by the size of the net or mesh sieve used for sampling (Blair *et al.*, 2019). As MPs continuously break down to smaller particles, smaller MPs are more prevalent in the environment, so the minimum detection size will greatly influence the results of a study (Lindeque *et al*., 2020). Another challenge faced when conducting MP research is contamination by atmospheric fallout (Dris *et al*., 2016). Fibres, and in some cases, fragments become airborne and contaminate samples if preventative or corrective measures are not taken (Klein *et al*., 2019; Dris *et al*., 2016).

1.2.3 Global overview

There is a notion that the ocean is the last great frontier of human exploration. The subsequent pollution of the ocean due to human activities is therefor considered as tainting the 'untouched'. It is most likely due to this sentiment that research on plastic and MP pollution has focused on the marine environment (Andrady *et al*., 2011; Cole *et al*., 2011; Lusher *et al*., 2015; Thompson *et al*., 2015). Peter Ryan from the University of Cape Town was the first to notice plastics in seabirds (Ryan, 1987) and marine waters (Ryan *et al*., 1990) of South Africa in the late 1980s. Although marine plastic pollution is of great concern, most people live further than 100 km from the ocean (Kummu *et al*., 2016) and rarely come in direct contact with marine products or environments. When the aim is to investigate plastics that have a direct impact on human livelihood and health, it is sensible to consider freshwater as well. It is estimated that about 90% of the global population lives within 10 km of a freshwater body (Kummu *et al*., 2011). In developing countries especially, much of the population is still directly reliant on these freshwater bodies as water source for drinking, cooking, and washing, and have direct, daily contact with it.

People that use raw and treated water, are susceptible to the effects of contaminants in the water. Microplastic is a contaminant of which the effects are only starting to be understood (Prata *et al*., 2020). Microplastics have several potential adverse environmental effects. Most plastics are very durable. Plastics may remain in the environment between hundreds and thousands of years (Barnes *et al.*, 2009). It poses an ever-increasing problem, now and probably for centauries to come, as it keeps on accumulating from current discard. Even though many plastics are naturally buoyant, particles and microorganisms (biofilm), can attach to the particles, increase the density, and cause these particles to settle in sediments (He *et al*., 2022). Environmental impacts have been tested with laboratory exposure studies and shown plastic can cause harm, inter alia, by means of ingestion (Cole *et al*., 2011; Wesch *et al*., 2016; Lusher *et al*., 2020), entanglement (O'Connor *et al*., 2020; Naidoo *et al*., 2020), reduced feeding, and reproductive impairment (Anbumani *et al*., 2018). Much of these studies have, however, been conducted with MP concentrations that are not environmentally realistic (Lenz *et al*., 2016; Cunningham *et al*., 2019).

Microplastic sizes falls within the size range of zooplankton (Lima *et al.*, 2014), filter feeders (Avio *et al.*, 2015), and even some smaller fish species (Luis *et al.*, 2015). The ingestion of plastic particles by filter and suspension feeders at the base of the food web raises toxicity concerns form pollutants (Anbumani *et al*., 2018). Microplastics accumulate hydrophobic pollutants present in the environment including persistent organic pollutants (POPs) (Rodrigues *et al*., 2019). The ingestion of these particles by organisms may also lead to accumulation in the digestive tract causing starvation due to a false sense of satiation, or even perforation of the gastro-intestinal tract (Naidoo *et al*., 2020). In addition, organisms that have taken up microplastics may now also pass this on to predators, including, say, from fish to humans (Farrel *et al*., 2013; Seltenrich, 2015; Sharma *et al*., 2017). This therefore potentially involves the accumulation of microplastics and their associated pollutants to higher trophic levels (Engler, 2012).

MPs likely affect human health, although little conclusive evidence has been found of direct impacts of MPs on human health (Blackburn *et al*., 2022). Most studies, however, agree that MPs are linked to immune and stress responses, developmental and reproductive toxicity (Blackburn *et al*., 2022) and possible carcinogenesis (Kumar *et al*., 2022). Plastic, in general, is considered a hazard due to toxicity of certain monomers, leaching of additives and plasticizers (Barrick *et al*., 2021), and physical obstruction (Heshmati *et al*., 2021). These apply to MPs as well, but with the added risk of being able to cross intestinal and cellular membranes (Stock *et al*., 2021). A more extensive discussion of the adverse effects of MPs on human health is included in Chapter 4.

1.2.3.1 Interventions – International

Efforts have been made by some countries to limit or restrict use of plastic in certain applications (Liu *et al*., 2021). Single-use plastics have been the object of most bans or reduction measures (Xanthos *et al*., 2017). The strongest interventions have been implemented in Europe as an EU-wide ban from July 2021 on trade of single use plastics for which there are no alternatives (European Commission, 2021). Microbeads in cosmetic products have also been banned in most of Europe, North America, and several Asian countries like China, India, South Korea, and Thailand (Walker *et al*., 2023). Talks have started about a microbead ban in South Africa (Molewa, 2018), but action has failed.

The United Nations Environmental Programme (UNEP) are leading efforts towards a global, legally binding plastic treaty by the end of 2024 (UNEP, 2023). Three sessions have been held in which South Africa was also represented, and the next is scheduled for April 2024 (UNEP, 2023). We were asked to give input on priority areas from a South African perspective and the current study was there for represented in the third meeting in Nairobi, Kenya. The goal of these negotiations is to create a legally binding instrument (a convention in other terms) which will comprehensively prevent, progressively reduce, and eliminate plastic pollution throughout its lifecycle (UNEP, 2023).

1.2.4 Africa

Africa is the second largest continent with the fastest growing population on earth (UNFPA, 2023). Africa and Asia are expected to remain a global hotspot of mismanaged plastic waste through the rest of this century (Lebreton *et al*., 2019). Both these continents have growing populations and developing economies that are not able to effectively manage the growing volumes of waste it produces. This waste enters the natural environment (Jambeck *et al*., 2018). It is expected that the population of sub-Sahara Africa will double by 2050 to reach around 2.5 billion people (Statista, 2023) with an increasing annual per capita plastic consumption that was at 16 kg per person in 2016 (Sadan *et al*., 2021).

1.2.5 South Africa

South Africa is a country rich is natural resources and biodiversity. Despite the abundance of potential, the country is battered by socio-economic, infrastructure and environmental degradation due to a lack of proper management of these resources, which is discussed in Chapter 2 (Verster *et al*., 2017). Each South African use an estimated 19 kg of plastic each year (Verster *et al.,* 2020). Half of all plastic converted into products in South Africa is used for packaging and 14% in the construction industry (Plastics SA, 2022). Polymers commonly used for single-use packaging include PE-LD, PP, PE-HD, and PET (Plastics SA). Poverty, rapid urbanisation, and inadequate waste management infrastructure is seen throughout the country. In addition, 39% of South African households do not have access to formal waste disposal (Plastics SA, 2022) and at least 29% of waste is informally disposed of (Verster *et al.,* 2020). Informal dumping of waste in streets, common areas, and unused lands like river green strips, is common practice in underserviced areas (Haywood *et al*., 2021).

1.2.5.1 Interventions – South Africa

A levy has been placed on single use shopping bags since 2003 but has not been very effective in reducing environmental plastic pollution (Bezerra *et al*., 2021). In an effort to promote recycling and reduce the amount of plastic entering the environment, legislation on Extended Producer Responsibility (EPR) has been in place since 2021 in Section 18 of the National Environmental Management Waste Act 29 of 2008. Importers and manufacturers pay an EPR levy per tonne and carry increased end-of-use responsibility for packaging (Arp, 2021). Specific collection and recycling targets have also been set for materials for the next five years (Arp, 2021). Investment in waste collection infrastructure and consumer awareness campaigns will also be advanced by EPR (Plastics SA). The South African Manufacturing and retail industries has also joined with the South African Plastic Pact and agreed to act towards a circular economy with set goals for 2025 (SA Plastic Pact, 2021). They aim to have 100% of plastic packaging reusable, recyclable or compostable, 70% of plastic packaging effectively recycled and 30% average recycled content across all plastic packaging by 2025 (SA Plastic Pact, 2021).

1.2.6 Vaal River

South Africa is a water-stressed country and has to protect its limited aquatic resources. The Vaal River is the life vein of Gauteng - the most economically active and densely populated region of the country (Tempelhoff *et al*., 2007). The Vaal River, especially, is a water resource worth protecting, as its catchment supports circa 60% of the county's economic activities (du Plessis, 2017). It serves as a reflection of the state of the country's natural resources and illustrates to a great extent the dire state of affairs. At the start of this study, no data was available regarding plastic pollution in the Vaal River system.

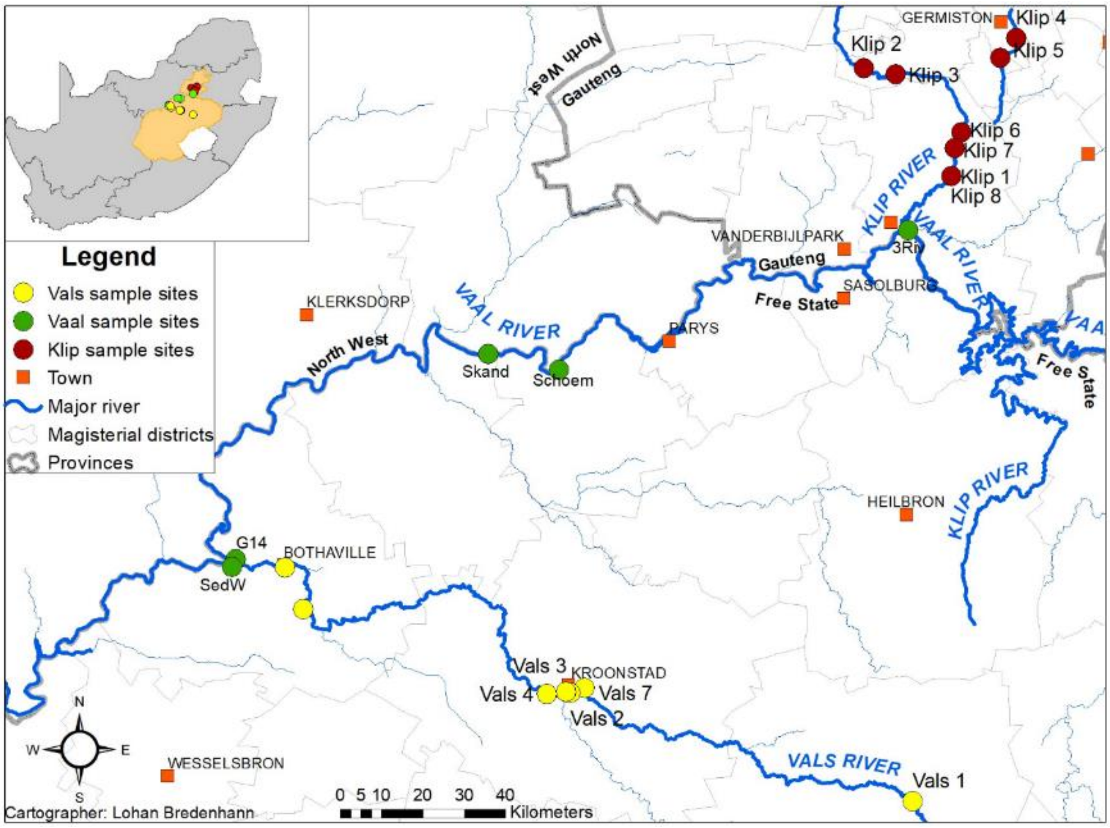

Figure 1.3: Map of the study area: Vaal River with the Klip and Vals rivers as tributaries .

1.2.6.1 Other studies

I conducted a scoping study with the Water Research Commission to establish baseline data for MPs in South African freshwater and determine research and intervention priorities. Findings from this study are summarised in Chapter 5. Findings highlighted the need for a deeper understanding of the spatial differences and temporal variation of MPs in the Vaal River and its tributaries, the Vals and Kilp rivers (Figure 1.3). The first samples represented in this thesis were taken in January 2017 and the last in March 2020. Since the onset of this study, a few studies have been published on inland river MPs in South Africa: Weideman *et al*. (2020) sampled 10 L of water along the Vaal and Orange rivers and found a mean of 1.7 ± 5.1 n/L which were mostly fibres (n= number). These results did however not correspond with findings from our initial scoping study, as the dominant morphotype in the scoping study were fragments, and further research had to be conducted. Dahms *et al*. (2020) found between 0.16 and 2.08 n/L in water from the Braamfontein spruit, also in the Gauteng area. Corresponding sediment samples showed concentrations of 4 – 1348 n/kg (Dahms *et al*., 2020). Ramaremisa *et al*. (2022) and Saad *et al*. (2022) detected $0.61 \times 10^{-3} \pm 0.57 \times 10^{-3}$ n/L in water with a mean of 460 ± 280 n/kg in sediment, respectively, in the Vaal River catchment. The present thesis focussed on the mainstream Vaal River and two of its tributaries - the Klip River and the Vals River.

1.2.6.2 The Klip River

Figure 1.4: Plastic pollution in the Klip River, Gauteng, South Africa (Photo: C. Verster)

The Klip River (Figure 1.4) drains urban Johannesburg and the historical mining region of the Witwatersrand (Tempelhoff *et al*., 2007). It is characterised by extensive wetlands in its upper reaches, improving the water quality from its catchment (McCarthy *et al*., 2007). Irrigation channels, wastewater treatment plants (WWTP) outlets, and developments in wetland areas have led to the decrease of wetland area and the formation of a single flow channel within wetlands, reducing the purification efficiency of the wetland system (McCarthy *et al*., 2007). Rapid urbanisation of southern Gauteng since the 1980s brought about a housing shortage in the area and have caused extensive informal settlements along the Klip River catchment (Tempelhoff *et al*., 2007). The growing population in the area also over-extended waste water treatment capacity.

1.2.7 Plastic dynamics

There are many factors that can affect the amount and morphotypes as well as the distribution between water and sediment of MPs at a given place and time (Stanton *et al*., 2020). MPs have varying sources (Mintenig *et al*., 2020), are differently affected by local hydrodynamics (Vianello *et al*., 2013), and display seasonal and temporal differences (Stanton *et al*., 2020). Chapter 4 is a scientific review on the known sources and pathways of plastics to the ocean in a South African context.

1.2.7.1 Waste management

The most prominent source of MPs is the breakdown of macrolitter (Stanton *et al*., 2020). Physical and chemical abrasion cause the fragmentation of larger plastics (Sipe *et al*., 2022). Plastic waste in the environment is a common site in South Africa. Delivery of waste disposal service ranges between 92% of households in the Western Cape to 20% in Limpopo (Verster *et al*., 2020). Waste management in informal settlement is a grave problem in South African waste management (Haywood *et al*., 2021)—most people in underserved areas in terms of waste have no option other than ‘illegal’ dumping (Figures 1.5 & 1.6) and informal incineration of waste (Adeniran *et al*., 2022).

Figure 1.5: Informal dumpsite in an informal settlement - Gauteng, South Africa (Photo: C. Verster)

1.2.7.2 Wastewater treatment plants (WWTPs)

Effluent from WWTPs contribute to the MP loads in rivers (Sun *et al*., 2020). The MP removal efficiency of WWTPs in different areas differ from 11% in the Netherlands (Leslie *et al*., 2017) to 99.9% in Sweden (Magnusson *et al*., 2014) and the United States (Carr *et al*., 2016). WWTPs in South Africa are notoriously inefficient due to overuse and inadequate maintenance (DWS, 2022). In the scientific assessment (Chapter 4) we report that 40% of South African waste waters enter the environment untreated due to faulty infrastructure (Verster *et al*., 2020). In the light of the DWS Green Drop report, this number is probably higher as WWTPs in South Africa are used beyond its capacity; 113 of the assessed 115 WWTPs (98 %) were in a ‘poor state’ or ‘critical state’ (DWS, 2022). Waste water and WWTP effluent are thus expected to be a direct contributor of MPs to South African rivers.

1.2.7.3 Informal settlements

Informal settlements in South Africa are areas with high population densities and narrow access ways. Most are underserviced in terms of waste removal (DEA, 2018). Some streets are too narrow for waste removal trucks to access (Verster *et al*., 2020). Due to this and a lack of public awareness of plastic pollution, waste is informally and/or illegally disposed of in informal dumpsites (Adeniran *et al*., 2022). These are typically in open areas and green strips (Haywood *et al*., 2021), especially along rivers edges. This causes broken down MPs and larger macrodebris to enter waterways by wind and runoff.

1.2.7.4 Wetlands

Wetlands improve water quality by removing pollutants from water (Verhoeven *et al*., 1990; Hammer *et al*., 2020). This is also the case for plastic and MPs (Dalvand *et al*., 2023). Wetlands have been shown to remove between 53 and 99% of MPs from water (Su *et al*., 2022; Ouyang *et al*., 2022). The protection of functional wetland ecosystems is vital in the fight for suitable quality water (McCarthy *et al*., 2007). Wetland sediment and biota, however, become sinks for pollution, and possible secondary sources when wetlands are degraded irreparably (Townsend *et al*., 2019; Davland *et al*., 2023).

Figure 1.6: Illegal dumping next to the Vals River in the Free State (Photo: C. Verster)

1.3 Aims, objectives and hypothesis

General aims for this these were to:

Aim 1: Quantify MP pollution in the Vaal River catchment.
Hypothesis 1: High MP abundances will be present in the Vaal River system.

Aim 2: Identify plastic pollution sources.
Hypothesis 2: Local environmental plastic pollution level is the major indicator for riverine MP load, meaning improper waste management is the main cause of MP pollution in South Africa
Hypothesis 3: Different MP morphotypes (fragments, fibres and beads) have different sources and pathways.

Aim 3: Describe sinks of MPs in South Africa.
Hypothesis 4: During low flow periods, sediments act as temporary sink for MP.
Hypothesis 5: Wetlands act as MP filters, trapping it in wetland sediment as sink.

Aim 4: Describe temporal patterns of MP for South African freshwater systems.
Hypothesis 6: Riverine MP load is affected by flow rate.

Aim 5: Propose mitigating actions to reduce freshwater plastic pollution in South Africa.

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Chapter 2

Marine and freshwater microplastic research in South Africa

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ABSTRACT

South Africa has a vibrant plastics manufacturing industry, but recycling is limited and insufficient with a notable proportion of the unmanaged waste entering the environment. South Africa is a developing country with microplastics research in its inception. Very little is known about freshwater microplastics, and studies on South African marine microplastics are limited but actively being pursued. In a water-scarce country, protection of freshwater resources remains a priority, but in the face of other socioeconomic issues (poverty, unemployment, and HIV/AIDS), it receives insufficiently effective attention. The full impact and risks of microplastics pollution in water is yet to be discovered. The risks may be enhanced in a developing country where many communities remain largely dependent on the land and natural waters. With South Africa being a water-scarce country, the quality of its aquatic resources is at an even greater risk with an assumed increasing background of microplastics, emphasizing the need for further research. A South African Water Research Commission–funded project is being undertaken to derive research priorities, but there is an immediate need for improved recycling and waste management.

Keywords: South Africa, Microplastics, Water-scarce environment, Plastic waste recycling, Aquatic pollution.

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Note 1: This article has been updated with an addendum following this text.
Note 2: This article was published in 2017 when microplastic research in South Africa was in its infancy. Its aim was to motivate action regarding plastic waste research and awareness, which have since taken hold, as witnessed by the many studies published since.
Note 3: This article has been cited 39 times (Google Scholar) by December 2023.
Note 4: The citation and reference style in the publication has been changed to be consistent with the style of this thesis.

2.1 Introduction

Plastic is used in every sector in the South African economy, with the plastic manufacturing industry contributing 1.6% to the gross domestic product and 14.2% to the manufacturing sector in 2014. About 60 000 people are employed (formally and informally) by about 1800 companies across the supply chain. Turnover was about $3.6 billion in 2014, with exports of $1.25 billion, and further imports of $2 billion (DTI, 2016). About 1 490 000 metric tons of virgin plastic and 310 600 metric tons of recyclate were used across a broad spectrum of South

African industries in 2015 (Plastics SA 2016; http://www.plasticsinfo.co.za/). Packaging uses about 55% of the plastics and, building and construction materials about 15%. Sectors using 6% or less each include electronics and electrics, automotive and transport, engineering, agriculture, and domestic products (Plastics SA, 2016).

The South African government has identified the plastics industry as a priority sector to promote economic growth through DTI the Industrial Policy Action Plan (DTI, 2016), stimulating aspects such as export, trade policy measures, innovation, and recycling. Quoting the Department of Trade and Industry IPAP document (DTI, 2016 - based on 2014 data), “At the moment, however, an overwhelming 72% of plastic packaging is not recovered at all: 40% is landfilled and 32% leaks out of the collection system—that is, either it is not collected at all, or it is collected but then illegally dumped.” It may be assumed that a similar low level of recycling and leakage is also applicable to some of the other sectors. From a different perspective, Jambeck *et al.* (2015) calculated 630 000 metric tons of mismanaged plastic waste for the coastal areas of South Africa, placing it eleventh in the world as a possible contributor to plastic marine debris.

Figure 2.1: Plastic debris in the Mooi River, Potchefstroom, South Africa. This is a small stream, typical of many such streams in South Africa. (Photo taken in September 2006 by H Bouwman; reprinted with permission).

Although legislation is in place to promote recycling and sustainable use of natural resources, the recycling of plastic-based materials, with particular reference to packaging materials, is implemented predominantly through corporate initiative (Nahman, 2010). In 2014, 315 600 metric tons of plastics were recycled by about 1800 convertors, which are mainly small businesses (PlasticsSA, 2016); these efforts are increasing. Inadequate waste disposal protocols and infrastructure are likely to cause much of this plastic to end up in aquatic systems (Figure 2.1), impacting upon the country's water quality.

Microplastics are derived from the breakdown of larger pieces of plastics, as well as fibers and manufactured microplastics. Of the latter category, not much information on manufacture, consumption, and release to the environment in South Africa is generally available, but its inclusion in many products can be safely assumed.

2.2 Issues faced in a South African context

South Africa is a developing country (United Nations, 2016) with a strong chemical manufacturing sector. The slow growth of the South African economy influences the prioritization and application of capital to stimulate development. Therefore, ecological issues such as the presence of microplastics, in the face of poverty, high unemployment, and HIV/AIDS, receive little attention. The South African plastics industry employs 60 000 people. With unemployment figures growing annually and the plastics industries employing more people every year (Plastics SA, 2016), it is unlikely that plastic use and production will decrease.

South Africa is a water-scarce country situated in the arid mid latitudes. The sustainability and protection of its freshwater resources is of critical socioeconomic and ecological importance. With microplastics being a relatively new field of study (GESAMP, 2015), very little is known about microplastics in South African aquatic systems. A scoping study funded by the South African Water Research Commission (WRC) is being conducted to determine what is known about aquatic microplastics pollution, and what future research is needed. A probable cause for the lack of knowledge is the prioritizing of research and resources to the more pressing socioeconomic issues. In many areas of rural South Africa, food and water security is largely attributed to subsistence farming and openly accessible natural waters (Baiphethi & Jacobs, 2009). With subsistence farming, consumers are therefore directly dependent on natural resources. When microplastics-contaminated water and soil are used for drinking and crop production, respectively, water and food security, as well as the well-being of the population, may be affected negatively.

This direct link between the human population and the environment, now (most likely) also with an increasing microplastic burden, is of concern. South Africa and many other African countries still use persistent organic pollutants (POPs), such as DDT for malaria control, especially in remote rural areas (Bouwman *et al.*, 2013). The links between pollutants in microplastics and potentially more vulnerable rural populations need to be examined. Although the full impact of microplastics on the environment and biota is not yet understood, the potential threats should not be taken lightly.

Another issue adding to the necessity of microplastics quantification and research in South African aquatic systems (both marine and freshwater) is the country's rich natural biodiversity. Studies are being conducted on the impact of microplastics on biota, but the full extent of this relationship is yet to be understood. Recent findings suggest that organisms at all trophic levels, with a variety of feeding strategies, have the potential to ingest microplastics (Eerkes-Medrano *et al.*, 2015). If the impact of microplastics on biota is as speculated (Galloway &

Lewis, 2016), a great deal of work must be done in South Africa and the rest of the continent to ensure progress in the field, and to reduce and/or eliminate sources of pollution.

2.3 Scope of studies conducted

Microplastics are plastic particles less than 5mm in diameter, about which little research has been conducted in South Africa. Most of the published work was on particles greater than 5mm (e.g. Eriksson *et al.*, 2013; Ryan *et al.*, 2014a; Ryan *et al.*, 2014b; Fazey & Ryan, 2016). Even less is known about microplastics in South African freshwater systems. As is the international trend, most of the research done on microplastics in this region has focused on the marine environment (e.g., Naidoo *et al.*, 2015), paying much attention to the ecological effects on seabirds (e.g., Ryan, 2008). A handful of studies have quantified microplastics along different parts of the South African coastline.

As expected, the highest densities of microplastics occur around major coastal metropolitan areas such as Durban and Cape Town (Nel & Froneman, 2015). Very little is known about the less-densely populated Atlantic Coast area. Some of the plastics studies conducted in the Antarctic region were South Africa–based (especially the work done by PG Ryan and colleagues), contributing to global knowledge of plastic distribution, accumulation, and trends. The knowledge gap regarding aquatic microplastics is a matter of concern because of the strong interaction between terrestrial biota (including the human population) and the aquatic environment (Eerkes-Medrano *et al.*, 2015).

Another area of focus in South Africa is the interaction of microplastics with POPs. Polyethylene pellets are used as indicators of POPs because of their ability to absorb POPs (Ryan *et al.*, 2012). These pollutants are known to have negative effects on humans and ecosystems, although the full extent thereof is yet to be understood. A number of South African universities are now conducting plastic debris and microplastics research. These include the University of Cape Town, North-West University, Rhodes University, University of Kwa-Zulu Natal, and the University of South Africa. In addition, Plastics SA are conducting campaigns on waste management and recycling, resource efficiency, information and communication, beach clean-ups, education and training, and supporting research (http://www.plasticsinfo.co.za/). The WRC scoping study referred to above will collate all research and, through consultation, will derive a list of priorities for future studies.

2.4 Conclusions

To find practicable solutions to address the microplastics issue in South Africa, the situation and conditions (including developing-country perspectives) must be fully understood, highlighting the need for further studies in the field. The uncertainty regarding susceptibility of humans and biota to harm by microplastics further emphasizes this need. While the impacts of microplastics are still poorly understood, sustainable solid waste disposal, better water purification, as well as recycling should receive higher priorities, even in the absence of more knowledge, so as to minimize potential future costs and impacts. At the same time, research capacity and funding must be strengthened to assess the issues we have identified, to keep abreast of international developments and knowledge, as well as to address yet unknown impacts and problems.

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Data availability. Data are available by contacting corresponding author Hindrik Bouwman (henk.bouwman@nwu.ac.za).

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2.7 Addendum

This article was published in 2017 when microplastic research in South Africa was in its infancy. Its aim was to motivate action regarding plastic waste research and awareness, which have since taken hold, as witnessed by the many studies published since. This article has been cited 39 times in Google Scholar by October 2023.

The following updates can be made regarding the South African plastics industry based on the latest figures released by PlasticsSA (2022):

- The plastic manufacturing industry has increased its contribution to the GDP from 1.6% to 2.3% and its contribution to the manufacturing GDP from 14.2% to 20% in 2022 (PlasticsSA, 2022).
- The South African plastic industry has an estimated value of R83.2 billion. A trade deficit, currently around 25% of total industry value, has been increasing over the last five years (PlasticsSA, 2022).
- In 2022, 1 904 924 metric tons of polymer have been converted into plastic of which which 21.7% was recycled locally (PlasticsSA, 2022). The use of plastic for packaging decreased from 55% to 50%, and building material from 60% to 15%. There has been a slight increase in the proportion of plastic converted in other sectors like agriculture, transport, and electrical applications.
- The reported mass of recycled plastic converted into raw material in South Africa in 2014 was 315 600 tons. This number climbed to 352 500 tons in 2019 after which the industry took a knock during the Covid-19 pandemic, with additional obstacles like loadshedding hindering recycling. It has since recovered to a total tonnage of 344 527 recycled in 2021 (PlasticsSA, 2022).
- There has also been a significant increase in the number of publications on environmental plastic in South Africa, whether in be macroscopic waste or microplastic. Details of these publications are discussed in Chapter 1.

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Chapter 3

Land-based sources and pathways of marine plastics in a South African context

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ABSTRACT

We review and evaluate the major land-based sources and pathways of plastic waste that leads to marine pollution in a South African context. Many of the formal solid waste and wastewater management facilities in South Africa are not fully functional, contributing towards plastic releases to the environment. Much plastic also enters the environment directly by informal and illegal dumping. Once in the environment, plastic is transported and distributed by air, inland waterways, and human activity, with complex dynamics that are not fully understood. Depending on the size and type of plastic and environmental factors like wind action and runoff, plastic can be deposited into sinks such as soil, river sediments, and vegetation, or carried to the ocean. Contrary to an initial assumption that South Africa is the 11th worst contributor to marine plastic pollution, we estimate from more accurate and recent data that between 15 000 to 40 000 tonnes per year is carried to the oceans. This six-fold less than a previous estimate. Despite many data and information gaps that require urgent attention through research and monitoring, it is clear that the status quo will lead to a worsening of already severe plastic pollution of all environments. South Africa needs to reduce plastic entering the environment by reducing illegal and informal dumping, effectively implementing and improving waste management infrastructure, and intensify long-term awareness campaigns. Most importantly however, immediate and effective mitigation is required.

Significance

- More accurate and recent data show that between 15 000 and 40 000 tonnes of plastic is carried to the oceans from South Africa per year – six-fold less than the widely used previous estimate.
- Riverine sediments are potentially major sinks for plastic en route to the ocean.
- Management of treated waste-water sludge, as well as the state of waste-water treatment plants (WWTPs) are key concerns. WWTPs are reported to remove most plastic from the water content. The state of South African WWTPs have deteriorated to such an extent that up to 40% of the country's waste water is untreated and data and management practices of sludge are unavailable.
- There are major data gaps in the South African waste sector, which lead to miscalculations and uncertainties about the country's contribution to marine plastic debris.

Keywords: South Africa, Microplastics, Water-scarce environment, Plastic waste recycling, Aquatic pollution.

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Note 1: This article has been cited 61 times as of December 2023.
Note 2: The citation and reference style in the publication has been changed to be consistent with the style of this thesis.

3.1 Introduction

Marine plastic debris is a global concern that needs urgent attention and mitigation (Kühn, 2015). Although numerical estimates differ (Ryan, 2020), the majority of plastic reaching the marine environment comes from land-based sources. Li *et al.* (2016) estimates that up to 80% of marine plastic debris is from land-based sources (Li *et al.*, 2016), but this is largely based on data from the Caribbean islands and the proportion of land-based to sea-based sources show great regional variation (Ryan *et al.*, 2019). Land-based plastic debris enters the marine environment mainly from formal, informal, and illegal debris, carried about by rivers, waste- and storm- water outlets, and being blown directly into the oceans via wind (Boucher). Recently, microplastic has also been found in air (Dris *et al.*, 2016) (Dris *et al.*, 2017) expanding our knowledge on plastic mobility and long-range distribution. Although most literature on plastic pollution remains marine based, more attention is being given to riverine research as rivers act as a major transport pathway of plastics to the oceans (Eriksen, 2014; Leslie *et al.*, 2013; Gasprei *et al.*, 2014; Castaneda *et al.*, 2014; Zhang *et al.*, 2015; Lebreton *et al.*, 2012). Rivers play a role in the transformation of plastic into smaller pieces due to abrasion, chemical, biological or UV degradation (Andrady, 2012). Freshwater sediments also act as sinks for plastic that may become secondary sources during floods or high-flow conditions.

Generally, one can distinguish three major categories of plastics found the environment. Large plastic items, arbitrarily termed macro plastics (>5 mm in longest dimension) are items such as packaging, foams, plastic bags, and ear bud stems. Large debris breaks down through a myriad of processes into smaller pieces called microplastics (<5 mm in longest dimension). Fibres released from fabrics (often from washing of clothes) are also considered microplastics due to their size. Not only do macro plastics cause direct harm to larger animals through ingestion, suffocation, and entanglement (GESAMP, 2015), but microplastics cause similar problems to smaller animals.

Many plastics are manufactured as complex mixtures of chemicals. Plastics can also take up additional chemicals from the environment such as persistent organic pollutants (POPs) and metals such as mercury. The incorporated and accumulated chemicals could be transferred to terrestrial, freshwater, and marine organisms that have taken them up through ingestion or assimilation, posing a threat to human, biotic, and ecosystem health (Van Cauwenberghe *et al.*, 2015; Woodall *et al.*, 2015).

Here, in a South African context, we consider the land-based sources of macro- and microplastics. We will discuss the sources of plastic that can become marine plastic, its distribution mechanisms, and how plastics eventually reach the oceans. An understanding of the underpinning factors and knowledge gaps is necessary to inform effective and integrated land-based remediation and intervention options and policies.

3.1.1 Plastics are complex

There are many types of polymers with many ways to characterise their properties, such as chemical and crystalline structures, production processes, design, density, hardness, capacity to absorb water, electrical conductivity, and degradability (Elberg *et al.*, 1995). Table 3.1 provides a summary of common polymers, some common uses, as well as their typical densities. The densities of various polymers become important as it relates to buoyancy in fresh and marine water that is pertinent to the current series of articles. It should be noted that densities given here are approximate.

Table 3.1: Types and some uses of selected polymers arranged according to their typical densities, as well as the densities of different waters (adapted - Bouwman *et al.*, 2018, Schwarz, 1995).

Type of polymer	Density (g/cm^3)	Common uses
Natural rubber	0.016 - 0.36	Cool boxes, floats, cups
Polyethylene - low density	0.91 - 0.93	Plastic bags, outdoor furniture
Polyethylene - high density	0.94 - 0.97	Bottles, pipes
Polypropylene	0.85 - 0.94	Rope, bottle caps, gear, strapping
Polystyrene - expanded	0.016 – 0.36	Cool boxes, floats, cups
Polystyrene	0.96 - 1.05	Utensils, containers, microbeads
Polystyrene - high impact	1.04	Shelves, printed graphics
Polyamide (‘– Nylon’)	1.12 - 1.14	Fishing nets, rope
Polycarbonate (bisphenol-A)	1.2	CDs, glass alternative, lenses
Polyurethane	1.2	Rubbers, sealants, paints
Metacrylate (acrylic)	1.19	Alternative for plate glass
Cellulose acetate	1.28	Cigarette filters, fabric fibre
Cellulose nitrate	1.35	Printing inks, nail polish, foil
Polyvinyl chloride	1.38	Film, pipe, containers
Polylactic acid (biodegradable)	1.21 - 1.43	Packaging, cups
Polyethylene terephtelate (21)	1.34 - 1.39	Bottles, strapping bands
Melamine	1.57	Flooring, dinnerware, dry boards
Polytetrafluoroethylene	2.15 - 2.20	Bearings, lining of pipes, non-stick cookware
Distilled water	1.00	
Brackish water	1.005 - 1.012	
Sea water	1.025 - 1.027	

Although many plastic items consist of only one monomer such as ethylene or propylene, there are plastic products consisting of multiple monomers called co-polymers (GESAMP, 2015; Hahladakis *et al.*, 2018) to address existing or specific needs. Depending on polymerisation efficiency, monomers trapped in the polymer matrix may leach or desorb to the environment, or into organisms that have ingested them. Bisphenol-A is one such monomer that is known to leach and has endocrine disruptive properties (GESAMP, 2015; Hahladakis *et al.*, 2018).

Many kinds of additives are incorporated into plastics to attain desired properties, some listed in Table 3.1 (Bouwman *et al.*, 2018). Some of these additives (up to 70% of the mass) may be released from the article to the environment and to organisms that have ingested them. There are many known toxicological implications associated with both the monomers and additives (GESAMP, 2015).

In addition to the chemicals incorporated during manufacture, synthetic polymers that are mostly made up of non-water-soluble organic materials, act as organisms do by absorbing or adsorbing pollutants such as metals and persistent organic pollutants from the environment concentrating pollutants from land, refuse dumps, water, and perhaps even from air (Wagner & Lambert, 2018; Eriksson *et al.*, 2013; Graca *et al.*, 2013). Mercury and DDT for instance, have been detected at higher concentrations in plastics than in water, supporting a concentration effect akin to bio-concentration. Plastics, suspended matter, and biota passively concentrates hydrophobic molecules from water through adsorption (therefore remaining in solution in the plastic matrix), absorption (such as ionic, steric, or covalent binding), or a combination thereof depending on matrix volume, polymer characteristics, and ambient concentrations (Hartmann). Plastics that thus had their chemical compositions altered in freshwater and reaching the marine environment via rivers and outflows (such as industrial and sewage outflows) should therefore be considered as transport facilitators of concentrated chemicals to the oceans. The incorporated and accumulated chemicals could be transferred to terrestrial, freshwater, and marine organisms that take them up through ingestion or assimilation, posing a threat to human, biotic, and ecosystem health (Van Cauwenberghe *et al.*, 2015; Woodall *et al.*, 2015).

Table 3.2: Examples of additive type, function, and chemical name that can be found in manufactured plastics (Bouwman *et al*., 2018).

Additive	Function	Example
Accelerants	Speeds up curing	Ethylene thiourea
Antidegradents	Reduces degradation	N,N'-bis(1,4-Dimethylpentyl)-p-phenylenediamine
Antioxidants	Slow down oxidation	2-2-Hydroxy-5-tert-octyphenyl-benzotriazole
Antizonants	Slows degradation by ozone	Nickel dibutyldithiocarbamate
Cross-linking additives	Links polymer chains	2-Mercaptobenzothiazole
Flame retardants	Reduces flammability	Tetradecachloro-p-terphenyl
Photosensitizers	Absorbs radiation	Benzophenones
Plasticisers	Making the material more pliable	Bis(2-Ethylhexyl)terephthalate
Surfactants	Modifies surface properties	Polysiloxanes
UV stabilizers	Protects against UV damage	2-(2-Hydroxy-5-methylphenyl)benzotriazole

3.2 Sources of plastic in the environment

3.2.1 Waste in South Africa

Jambeck *et al.* (2015) ranked South Africa eleventh in a list of countries contributing an estimated 90 000 to 250 000 tonnes to marine plastic in 2010, based on an estimate of 56% mismanaged waste with little actual data to support this. Key reasons considered for the loss of plastics to the environment were lack of waste removal infrastructure, logistical challenges in informal settlements and out-lying communities, poorly managed waste, and littering (PlasticsSA, 2019). There are however, concerns that some of the quantitative assumptions used in various assessments may not be accurate and results in an over-estimation of the actual amounts that enters the South African marine environment (Ryan *et al.*, 2020).

Solid waste removal is primarily a function of local government (Statistics South Africa (Stats SA, 2016). According to the 2018 South African State of Waste Report (SoWR) (DEA, 2018), total non-mining waste generated in South Africa for 2017 was 54.2 million tonnes, which is 1.0 ton per capita of 56.5 million people. South Africa generated 1.1 million tonnes of plastic waste in 2017 (DEA, 2018) equating to 19 kg plastic per capita per year, or 53 g per person per day. Jambeck *et al.* (2015) used 2 kg per day of all waste (not only plastic waste) and estimated that 12.9 million people living within 50 km of the coast of South Africa, amounting to 505 000 tonnes of plastic waste per year in the coastal areas (assuming equal distribution between inland and coastal plastic waste generation figures).

SoWR (DEA, 2018) calculates that 43.7% of plastic waste is recovered and/or recycled, with the rest being disposed of (618 880 tonnes). Assuming that 29% of the 12.7 million tonnes of household waste does not enter the formal waste management stream, 3.67 million tonnes of waste mismanaged plastic in South Africa (Rodseth *et al.*, 2020) (Figure 3.1). Of the domestic waste handled (GW01, GW50, GW51, GW52, GW54), 11% per mass is plastic and tyre waste (DEA, 2018). Assuming a similar proportion of unmanaged waste is plastic and tyre waste, South Africa releases 440 000 tonnes of unmanaged plastic waste into the environment. The 12.9 million coastal inhabitants living within 50 km from the coast release 100 000 tonnes of plastic waste into coastal environment. Jambeck *et al.* (2015) assumed that 15 to 40 percent of the mismanaged plastic waste would enter the oceans. For South Africa’s coastal population, we calculate 15 000 to 40 000 tonnes of plastic could reach the oceans (Figure 3.1), more than six-fold less than the Jambeck *et al*.’s (2015) estimate of 90 000 to 250 000 tonnes coastal plastic waste. Although this does not include formally managed waste that also enters the environment via secondary pathways and other factors, such as burning of portions of formally unmanaged waste, we highlight that the estimate contribution of South Africa’s plastic input to the ocean is significantly less than previously claimed.

An important data uncertainty remains however – illegal and informal waste dumping. Illegal waste was recognised in the SoWR (DEA, 2018) but no estimates were provided. However, we do not believe that the difference between estimates can be made up by illegal waste dumping. For higher resolution and more accurate numbers, more data should be collected locally and used to improve estimates.

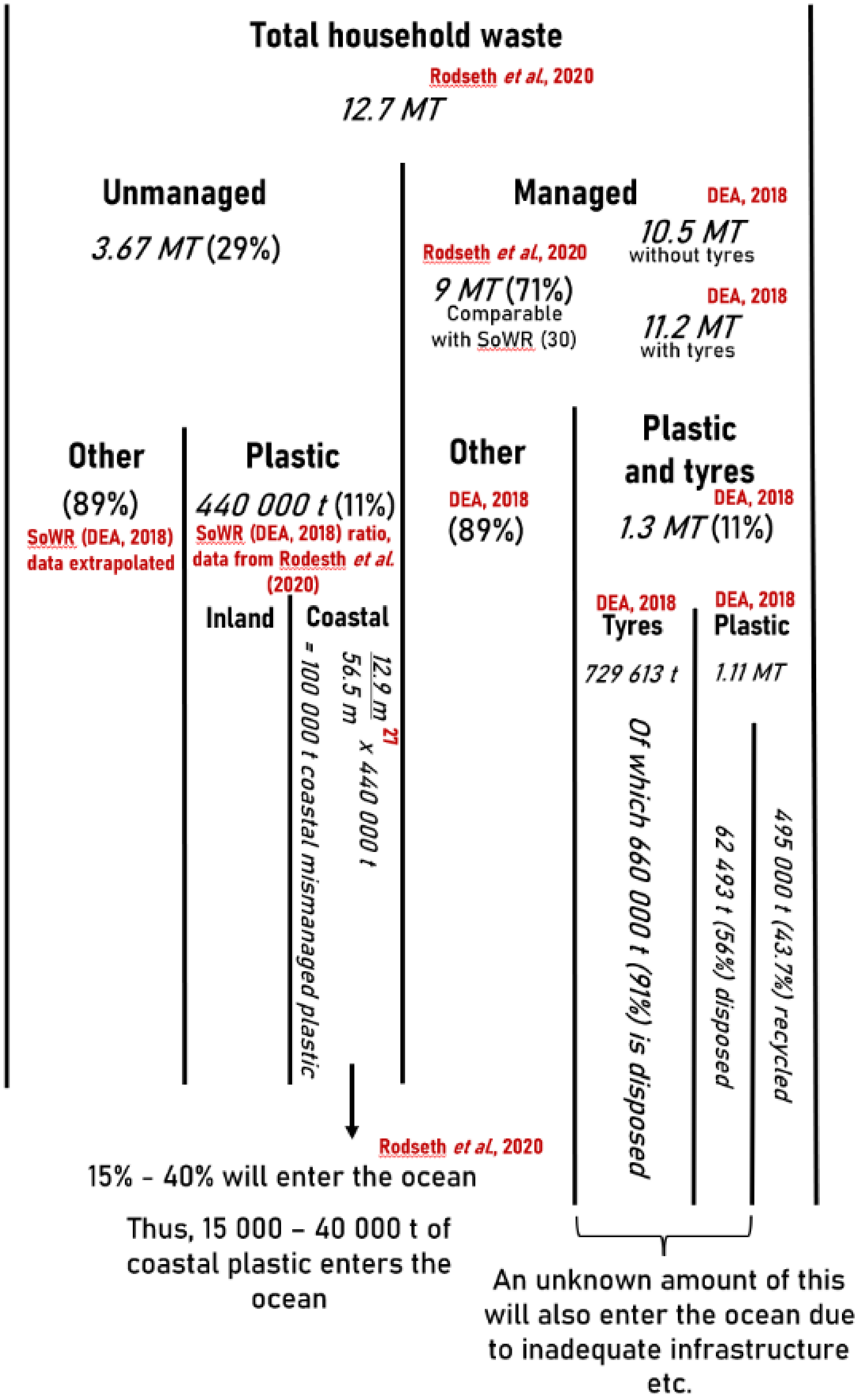

Figure 3.1: Breakdown of available data on household and plastic waste in South Africa (data source in red).

3.2.2 Socioeconomics and mismanagement of waste

Major drivers associated with plastic debris in the environment of an area are economic challenges and disadvantaged communities (DEA, 2018). Ninety-one percent of South African households are low-income households (Stats SA, 2016). In urban municipalities, 82.7% of households have weekly solid waste removal services, while only 4.9% use own dumpsites (Stats SA, 2016). In rural municipalities, only 1% of households have formal waste collection at least once a week; while 75.1% make use of own refuse dumps (Stats SA, 2016). Poverty, combined with rapid urbanisation and insufficient waste management results in logistical challenges in waste collection (DEA, 2018). Roads in informal settlements are often too narrow to be accessed by garbage trucks. Weak waste management by municipalities leaves many individuals, households, and communities with the responsibility to get rid of their own waste. Waste that is not formally collected is disposed of on communal dumps (DEA, 2018). Without proper infrastructure, plastic and other waste is lost to the environment by wind and water runoff (DEA, 2018). Vandalism of fencing at waste management sites also allows the leakage of plastic through wind (Pers. Obs. CV and HB).

Table 3.3: Breakdown of waste collection services in each province for 2016 (DEA, 2018).

Province	Formal waste removal	Communal/own refuse dump	Communal container/central collection point	Other
Western Cape*	92%	4%	4%	1%
Eastern Cape*	39%	53%	1%	8%
Northern Cape*	68%	25%	1%	6%
Free State	74%	21%	1%	5%
KwaZulu-Natal*	43%	49%	2%	6%
North West	58%	37%	1%	4%
Gauteng	88%	7%	2%	3%
Mpumalanga	40%	52%	1%	8%
Limpopo	20%	72%	0%	7%

Waste removal includes removal by local authorities, private companies, or community members (Table 3). It ranges from 92% in the Western Cape to 20% in the Limpopo province. The Western Cape and Gauteng have the most efficient formal waste collection systems, while Limpopo and the Eastern Cape have the lowest formal waste collection availability and inevitably the highest portions of informal or communal refuse dumps (DEA, 2018).

Excluded from the SoWR and data used for national waste estimates is the portion of mismanaged waste (Rodseth *et al.*, 2020). Of total domestic waste generated in South Africa, 29% (3.67 million tonnes per annum) is not collected or treated via formal waste management processes (Rodseth *et al.*, 2020). Because of inadequate waste management and a lack of consumer awareness and education, waste that is not collected is littered or illegally dumped (DEA, 2018; Rodseth *et al.*, 2020) (Figure 3.2). Rural communities may be largely ignorant of the adverse effects of plastics in the environment, resulting in a lack of motivation to keep the area clean (Wiseman *et al.*, 2012). We highlight the need for education about proper waste disposal practices and the provision of formal waste management services, especially in rural communities, as both income and settlement type largely determine the efficiency of waste management (Rodseth *et al.*, 2020).

Coastal cities report large debris loads deposited into the ocean directly via storm water drainage systems (Marais *et al.*, 2004b; Marais *et al.*, 2004a; Armitage *et al.*, 2000a; Armitage *et al.*, 2000b). Between 2000 and 2002, some 3 000 to 4 000 tons of debris were estimated to be deposited into the ocean by the City of Cape Town each year, most of which originated from informal settlements on the banks of canals (Marais *et al.*, 2004a). Data from beach clean-ups and debris booms in Cape Town suggest an increase in the plastic load during rainy seasons (Armitage *et al.*, 2000a). Recent beach clean-up data from Cape Town shows 9 of the 10 most frequently found items are associated with fast food containers, with the 10th being earbud sticks (Chitaka *et al.*, 2019).

Access to running water for households is related to microplastic concentrations in rivers – particularly to that of fibres (de Villiers *et al.*, 2019). If access to running water and proper wastewater treatment is limited, as is the case of many rural communities in South Africa, wastewater is discharged directly from households into river systems and clothes are often washed directly in rivers. Since mechanical (Bouwman *et al.*, 2018) and hand-washing of fabrics in water releases fibres, washing may contribute significant amounts of fibres to rivers. An average mechanical wash load of 6 kg of clothes can release more than 700 000 fibres per wash (Napper *et al.*, 2016). However, we could find no useful data on laundry activities in South Africa.

Waste management in South Africa is mainly not compliant with applicable regulations (DEA, 2018). Some issues that were identified at disposal sites were lack of access control, daily covering, auditing, and monitoring. To tackle this problem, infrastructure is needed, and waste removal and treatment services delivered to all communities. Education and awareness raising will lay the groundwork to reduce littering and burning. Education campaigns in schools and local authorities have been implemented in Gauteng, North West, Western Cape, and the Free State.

The informal waste sector is an integral part of the South African waste removal and recycling system, with more than 25 000 trolley pickers at kerbside and 36 000 landfill waste pickers in 2014 (DEA, 2018). Waste pickers tend to select high value products and often leave the rest, which can then enter the environment.

Figure 3.2: Illegal dumpsite next to a river in the Free State (Photo: C. Verster)

3.2.3 Transport sector

Global estimates conclude that automotive tyre wear or ‘rubber dust’ contributes up to 0.81 kg/year/person to the environmental microplastic load (Kole *et al.*, 2017). Road transport is the dominant mode of transport in South Africa. It will continue to be so in the foreseeable future as 71% of the national transport infrastructure budget in 2018 went to road infrastructure improvement. Although no data are available on tyre wear in South Africa, it is likely to be a source contributing to the environmental microplastic load that will also reach the oceans.

3.2.4 Industry

The plastic manufacturing and packaging contribute to the load of environmental plastic debris, but the amount of leakage is badly understood. Much of the leakage is in the form of primary pellets, recyclate flakes, and powders released to the environment during manufacturing or transport. During the 2015 coastal clean-up campaign, 53.9% of the number of micro-plastics found on beaches were industrial pellets (Plastics SA, 2015). Microscopic plastic particles are mixed with silica and other materials as abrasives and in sandblasting that is likely to leak to the environment if not properly contained (Bouwman *et al.*, 2018).

Operation Clean Sweep was initiated in the USA and globally launched in 2011 to contain primary plastic and recyclates within the manufacturing process, which is endorsed by Plastics SA to combat the release of plastics into the environment during production and recycling (Plastics SA, 2019).

3.2.5 Microplastics

3.2.5.1 Rivers and wastewater

Microplastics in aquatic ecosystems come from sources such as wastewater treatment plant (WWTP) effluent, sewer overflows, discharge, and runoff from sludge used in agricultural applications and industries (Mani *et al.*, 2015). In South Africa, urban runoff and informal settlements are other possible sources due to littering and inadequate waste management.

Microplastics may enter an aquatic system in two different forms. They can enter the system as primary microplastics (Mintenig *et al.*, 2017) or as secondary microplastics that form as breakdown products of larger items. When using cosmetic products like facial scrubs, between 4 600 and 94 500 microbeads, which are primary microplastics, can be released (Napper *et al.*, 2015) but little data is available on their retention by WWTPs (Mintenig *et al.*, 2017). Microbeads are also used in other applications such as sandblasting, soaps, and washing powder. Although microbeads have not been banned in South Africa as in Canada, the United States, United Kingdom, France, Sweden, Taiwan, South Korea and New Zealand, the South African cosmetics industry has taken some initiatives to replace microbeads with other materials. Where WWTP outflows are directly to the sea, any microplastics that remain in the effluent will be also be directly released to the sea.

When released into the environment untreated, wastewater can add large amounts of microplastics, especially microbeads, to riverine loads. Even though international results show that WWTPs can remove 97% - 99% of microplastics, treated wastewater still release large numbers due to high volume (Mani *et al.*, 2015; Mintenig *et al.*, 2017). Many of the WWTPs in South Africa are no longer fully functional. Of 68 audited WWTPs, only 8.2% were compliant with effluent quality (DEA, 2018). In 2014, about 30% of the country's sewage treatment plants were considered to be in a 'critical state' (needing urgent intervention), and another 25% in a 'high risk' state (DWS, 2014). This leaves up to 40% of the country's wastewater untreated (Donnenfeld *et al.*, 2018), increasing the likelihood of increased microplastic release to receiving marine and freshwaters. This plastic then becomes trapped in sludge, which is then often deposited on agricultural land (Nizzetto *et al.*, 2016). Runoff by water and pickup of microplastics by wind from agricultural land should therefore be considered a possible source of microplastic to rivers and oceans.

Only a hand full of studies have looked at microplastics in South African freshwater systems (Nel *et al.*, 2018; Bouwman *et al.*, 2018). High concentrations of microplastic fragments were found in sections of the Vaal River associated with more turbulent flow (Bouwman *et al.*, 2018) (Figure 3.3). Urban rivers like the Crocodile and Klip Rivers had microplastic levels up to 4.5 n/L (Figure 3.3). Levels of microplastic in sediments of the Bloukrans River ranged between 6 and 160 particles per kg dry sediment in summer (high flow) and winter (low flow), respectively (Nel *et al.*, 2018).

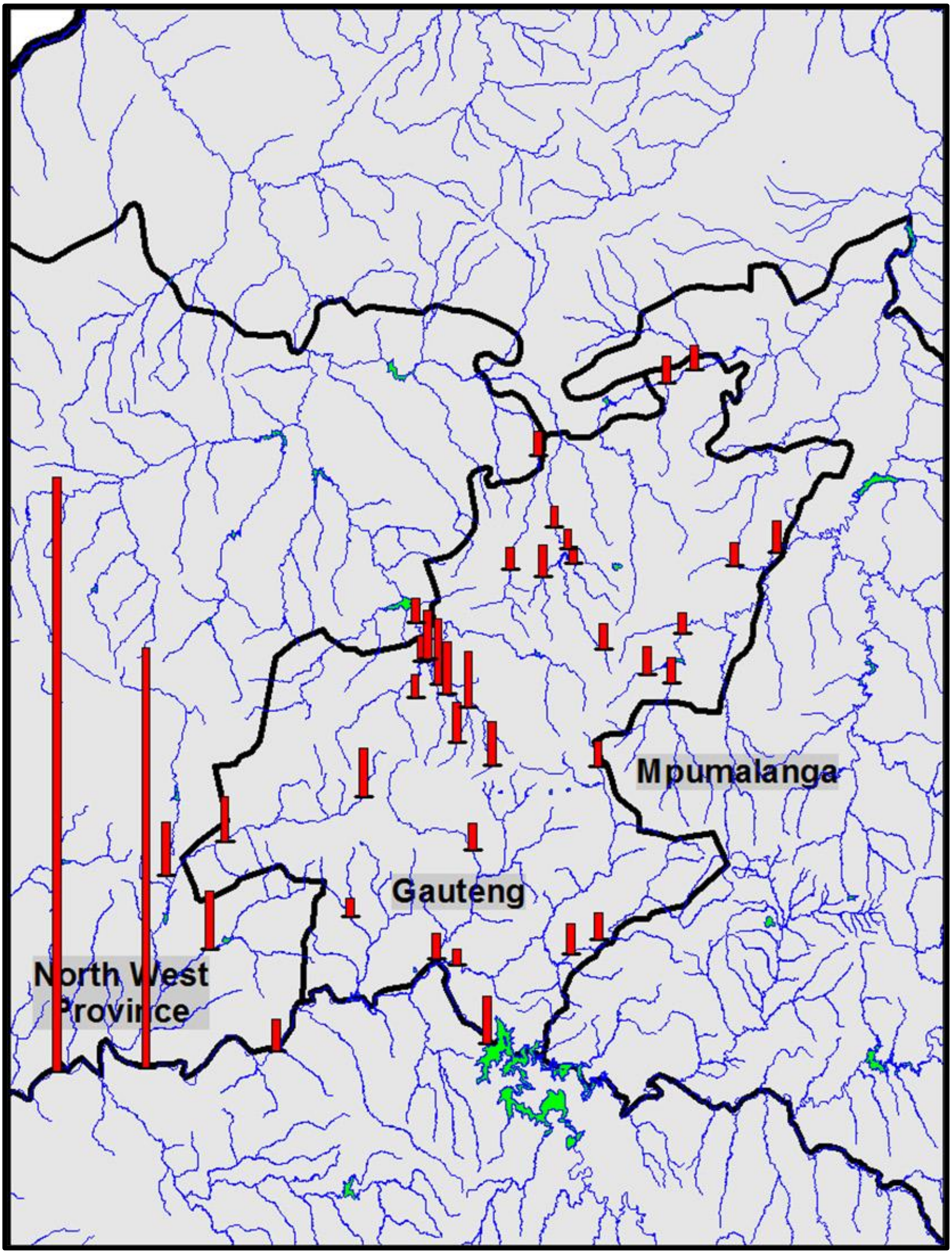

Figure 3.3: Distributions of total particles (fragments and fibres) per litre. The tallest bar represents 56 n/L (Bouwman *et al*., 2018).

3.3 Pathways

Size differences in plastic items influence environmental transport after release. Small microplastics (< 200 µm), even heavier-than-water polymers like PET (Table 1), tend to be retained in the water column, while larger particles precipitate faster (Nizzetto *et al.*, 2016). Larger, less buoyant items like bottles with air trapped inside, foams, and low-density polymer items, are found in surface water and riparian zones.

Plastic in the water column becomes covered by layers of biofilm through biofouling (Fazey, 2016). The more biotic material attaches to the plastic particle, the heavier it becomes, and sinks. This happens quicker for smaller particles. This process affects the movement and distribution of plastic particles and debris in fresh water (Fazey *et al.*, 2016), and probably its transportation potential to the marine environment.

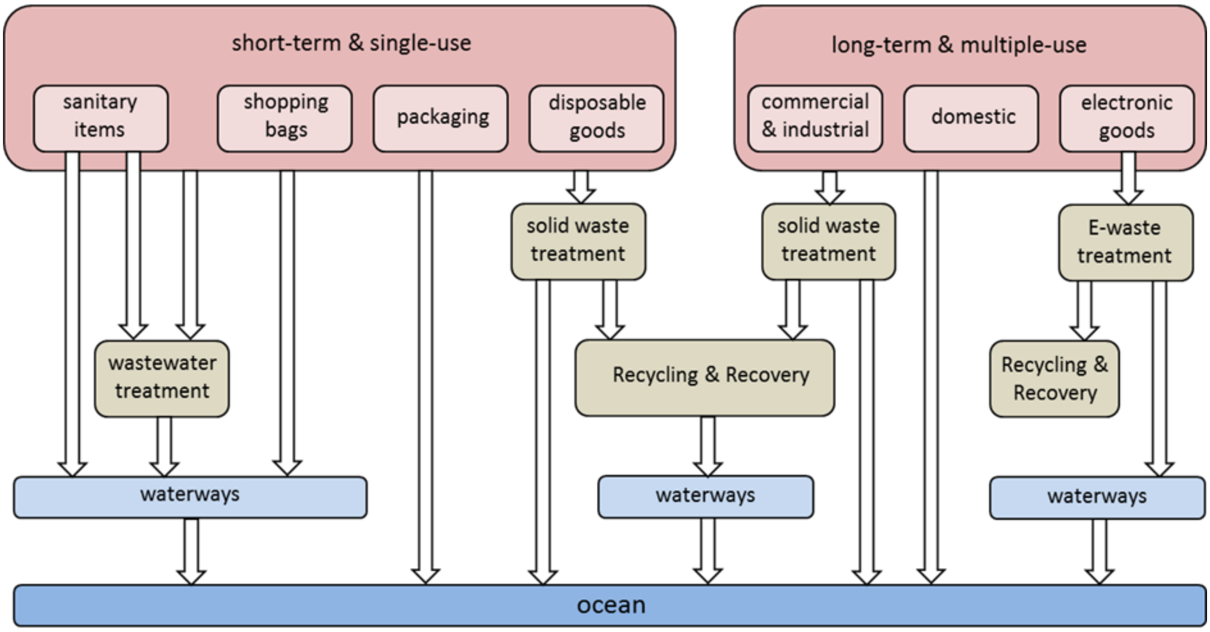

Figure 3.4: Schematic representation of generic land-based pathways of representative macroplastics reaching the ocean. (Permission: Peter Kershaw (GESAMP, 2015), and adapted to South African conditions).

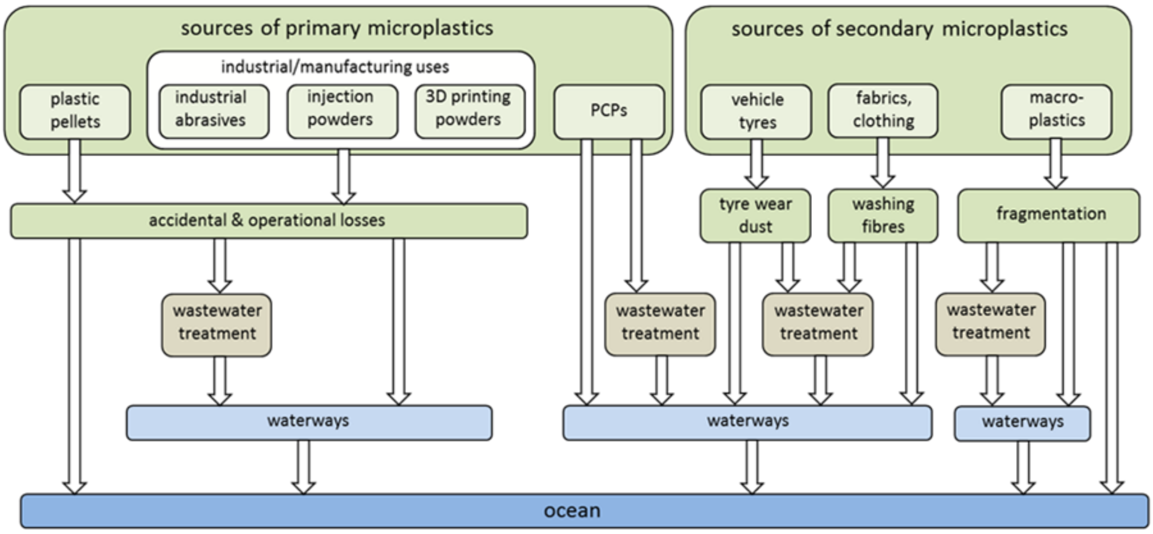

Figure 3.5: Schematic representation of generic land-based pathways of representative primary and secondary microplastics reaching the ocean. PCPs is personal care products. (Permission: Peter Kershaw (GESAMP, 2015), and adapted to South African conditions)

Figures 3.4 and 3.5 illustrate the major sources and pathways of examples of macro- and microplastics reaching the marine environment. The reality is, however, far more complex and nuanced. Plastic in the environment is subject to many factors that influence its movement, distribution, shape, and toxicity. Rivers act as the main conduits for marine plastic (Figure 3.4). Rivers also play a role in the transformation of plastic. As plastic can sink, especially in less dense freshwater, riverbeds can act as temporary sinks for plastic that can get resuspended and carried further downstream during high flow events. Hydrodynamics and the effect of impoundments play a critical role in the movement and distribution of plastics in any freshwater system (Nel *et al.*, 2018). These movements and interactions are quite well documented for marine systems (Sherman *et al.*, 2016, Hardesty *et al.*, 2017), but such understanding for riverine systems is lacking for South Africa. A scoping study on microplastic for riverine surface water found microplastic concentrations (both fibres and fragments at near equal proportions) ranging between 0.32 n/L in the Suikerbosrand River to 56 n/L in the Vaal River after heavy rains (Bouwman *et al.*, 2018). Preliminary results for South African groundwater indicate the presence of predominantly fibres at 0.17 n/L (Bouwman *et al.*, 2018).

3.3.1 Airborne

Microplastic fibres have been found all over the globe in the remotest of environments (Prata, 2018). It is assumed that these fibres are deposited via air (Figures 3.4 and 3.5). Although the study of microplastic pollution in air is in its infancy, significant numbers of plastic, especially fibres, have been found in settled dust and atmospheric fallout (Prata, 2018). It is estimated that between 1 600 and 11 000 fibres m^2/day can be deposited in urban areas (Dris *et al.*, 2017). Most are natural fibres like cellulose and an estimated 29% are petrochemical-based synthetic fibres (Dris *et al.*, 2016). There is a strong correlation between anthropogenic activity in an area and the number of fibres found in the air (Dris *et al.*, 2016). Although a novel field of enquiry, microplastics have been shown to travel more than 95 km from point sources (Bank *et al.*, 2019). An estimated 7% of the number of ocean plastic may be deposited through atmospheric fallout (Boucher *et al.*, 2017). Although no data have been published for airborne plastic settling in South Africa, preliminary results indicate the presence of fibres in remote arid areas in the country suggesting deposition of plastic much further than the 95 km suggested by Bank *et al.* (2019). Plastic fibres were found in dry runoff sediments (up to 315 particles per m^2, unpublished data, J Louw and H Bouwman pers. obs.) in the Nama Karoo near Brandvlei where it has not rained for many years.

Lightweight macro- and microplastic is also transported about by winds. Distances travelled might not be as far as that of smaller particles and fibres, but Jambeck *et al.* (2018) suggests that areas downwind from sources act as plastic sinks. Especially in rural areas without proper waste disposal infrastructure, plastic debris can spread quickly outside the bounds of informal dumps, contaminating large areas of rural land. Plastic debris can thus be directly transported to the ocean, carried about by winds or blown into rivers that carry debris to the ocean (Figures 3.4 and 3.5).

3.3.2 Sinks

Riverine sediments can act as a sink for plastics released into the environment, containing 40 times more microplastic than in surface waters (Kawecki *et al.*, 2019). Sediments in weirs had increased levels of plastic because particles settle out in these slower flowing parts of rivers (Mani *et al.*, 2016). Some 16–38% of microplastic denser than water settles out into sediments (Fazey *et al.*, 2016). Particles larger than 200 µm are also retained in riverine sediment with possible resuspension during high flow periods (Nizzetto *et al.*, 2016). From 0 to 567 fibres·dm-3 were found in sediments of lower reaches of water catchments along the South African coast (de Villiers *et al.*, 2019). Although no data is available, this is likely to be the case for macro debris as well. Microfibre content in river sediments of Kwa-Zulu Natal and the Eastern Cape also show a very strong association with socio-economic development indicators like access to water (de Villiers *et al.*, 2019).

Elevated levels of micro litter are found in rivers associated with densely populated areas (Mani *et al.*, 2016). There are some conflicting findings in literature as to how far plastic will flow down a river before it becomes stuck in sediment or vegetation. Mani *et al.* (Mani *et al.*, 2016) claims that plastic loads increase immediately downstream of sources, while Jambeck *et al.* (2018) states that downstream areas of high plastic input in rivers act as plastic sinks. Bouwman *et al.* (2018) suggests that in the Gauteng study area, microplastics show little patterns in terms of population or downstream accumulation. Larger fragments were slightly more common upstream closer to the Vaal Dam, while smaller particles dominated downstream sites of presumed sources, which suggest that larger particles do not stay suspended in the water column as long, and sites downstream of sources most likely act as sinks for larger plastic pieces. This is in accordance with findings by Nel *et al.* (2018) in the Bloukrans River system in the Eastern Cape where low-flow winter periods yielded higher sediment microplastic concentrations (160 particles per kilogram dry mass) when compared with high-flow periods (6.3 particles per kilogram dry mass). There are indications of very high microplastic loadings in sediment from rivers flowing through the Kruger National Park (P Shikwambana pers. comm.). Although not conclusive, flow rate seems to be an important hydrodynamic factor with the greatest effect on the plastic load in rivers of South Africa due to settling out in low-flow areas and seasons.

3.3.3 Soil

Although images of land-based environmental macro-debris are common, scant data are available in a South African context on amounts and distribution. The largest datasets available in this regard report amounts and composition of plastic on beaches (Plastics SA, 2015) which reported a recent increase in disposable nappies on beaches close to informal settlements. Interaction with biota on land is also less reported on, but examples include reports of cattle eating plastic in grazing areas (Wiseman *et al.*, 2012).

3.4 Current uncertainties

Compared with marine plastic debris research, information and data on inland sources and pathways in South Africa are scarce. To some extent data, findings and models can be extrapolated from research done elsewhere. However, as pointed out by Jambeck *et al.* (2018), South Africa faces distinctive socio-economic challenges and unique environmental and ecological dynamics affecting the load and movement of land-based plastic. Wrong assumptions may lead to wrong conclusions that may adversely affect policy and interventions. Here we discuss some of these uncertainties in terms of difficulties to extrapolate global findings to a South African context.

- Although visibly an issue, volumes and hotspots of illegal dumping and informal dumps are still unclear and needs to be quantified in order to motivate mitigation.

- Considering the unique socio-economic issues faced by South Africa when compared with countries with more complete datasets for sources and pathways of plastic, plastic management and regulations implemented in other parts of the world might not be as effective here or have unintended consequences (Verster *et al.*, 2017). In order to tailor a plastic policy for South Africa, more spatial and temporal data are needed for freshwater bodies to determine areas in need of protection, areas of highest threat, and processes that may be targeted for intervention.

- The deposition of plastic in riverine sediment as a possible plastic sink (Nel *et al.*, 2018) correlates with global findings. The effect of deposition or transport of plastic by rivers in these different regions need to be better understood and might be part of the answer to the missing plastic problem (Ryan *et al.*, 2020). If rivers do act as a temporary sink for plastic, more emphasis will have to be placed on knowledge of the amounts and impacts of plastic in freshwater systems.

- Freshwater and estuarine sediments may act as a long-term secondary source of plastics to the oceans, possibly long after effective mitigation on plastic releases have been achieved.

- Preliminary results show low microbead counts in South African rivers compared with developed countries. Although surface water microplastic concentrations in Gauteng and North West province rivers ranged between 0.33 and 56 n/L, microbeads were only found at two of the sites, and in very low concentrations (<0.01 particle per litre). Microbead data from South Africa's freshwater sediments are yet to be reported but can be expected to be higher than that of surface water – international data range up to 103 beads per litre of sediment (Castaneda *et al.*, 2014). Global estimates show microbeads originating from cosmetics only make up 2% of the marine plastic load by number (Boucher *et al.*, 2017). It would be beneficial to consider import, production, application and distribution of plastic microbeads since it attracts much international attention. South Africa needs to determine whether banning microbeads is a realistic and achievable national priority, and an easy first action to reduce the release of manufactured microplastics.

- A lack of data about polymer and pollutant composition of plastic debris in the environment is another area of study that will help refine, identify, and mitigate the greatest threats.

- Recently it has been suggested that anti-microbial resistance genes are associated with microplastic biofilms. These microplastic particles act as vectors for these genes, especially in plastics released by WWTPs (Eckert *et al.*, 2018). This will possibly translate to agricultural sludge applications as media in which anti-microbial resistance genes spread through the environment. How the movement of anti-microbial genes from land-based sources to the sea is actual and a threat needs further investigation.

3.5 Evidence gaps

- Plastic debris from land-based sources reach the ocean largely by means of rivers and rivers could act as sinks for plastic. When considering that many out-lying communities in South Africa source water, often untreated, directly from these systems and the country has limited freshwater resources, several concerns arise. Knowledge gaps in this regard include the volume of plastic trapped in freshwater systems and the retention time of plastic in freshwater sediment acting as a temporary sink and possible secondary source of plastic debris.

- Due to the diverse marine and freshwater aquatic biodiversity of South Africa, very little is known about specific ecosystem health risks of plastic debris in South Africa. To our knowledge, no published toxicity tests or ecological risk assessments have been conducted on freshwater organisms. Since it is evident that plastic is present in South African aquatic systems, we need to know the effect on freshwater ecosystems.

- Factors affecting breakdown of plastic in terrestrial and freshwater ecosystems are inadequately quantified in South African conditions. There are many physical and biological factors that play a role – the effects on eventual micro- and nanoplastics (<100 nm in longest dimension) formation remains unknown.

- Global estimates show that WWTPs remove more than 99% of microplastic from wastewater (UKWIR, 2019). Sludge from the wastewater treatment process is often applied as fertilizer to agricultural soils, transferring microplastics to agricultural soil (Nizzetto *et al.*, 2016). However, the retention rate of South Africa's wastewater treatment plants has not been tested, and the extent of sludge addition is not well documented. It is thus necessary to determine the amount of plastic in sludge. Sludge is also a secondary source via wind and runoff. Therefore, more information is needed on how sludge is managed in South Africa, to determine whether intervention is needed.

- Vehicle tyre wear could be a significant source of microplastics in developed countries. The South African transportation system relies heavily on road transport. One can therefore expect notable additions to the freshwater and marine environments. This topic has not yet been considered in South Africa.

- Preliminary results indicate the cosmopolitan distribution of microplastic fibres (Gasprei *et al.*, 2015). The extent to which this is true in South Africa is worth examining. Certain aspects of dust models are available for South Africa and may be adapted, but this will require additional information on the plastic content of dust in air. Long-range transport of plastic is an issue of concern as it can contaminate remote environments, including marine ecosystems.

3.6 Implication and actions

Municipalities should prioritise improvements in waste removal and management – especially in informal settlements, for hygienic and environmental reasons. Systems must be designed and/or implemented to the needs and conditions of communities (Boucher *et al.*, 2017) to improve recycle supply chains, and loose less plastic to environment.

We encourage the development of a standardised solid waste monitoring programme to monitor high risk areas (Ryan *et al.*, 2009). Issues such as illegal dumping needs to be monitored and enforced.

Further public and private sector incentives, awareness raising, and civil society pressures are needed to improve the situation to reduce land-based sources to both freshwater and marine environments. Risk communication and education efforts about the environmental and possible health effects of plastic are of great importance if public participation is to be expected (Bouwman *et al.*, 2018). Public realisation of the value of plastic as economic resource could motivate public participation in recycling and clean-up efforts (Plastics SA, 2019). Public sector assistance in extended producer responsibility programmes will assist industry mediators e.g. PETCO (SST, 2017) to encourage and administrate producer responsibility and contribute to the circular economy concept.

In moving towards a circular economy, research and development resources must be applied to develop alternatives for difficult-to-recycle plastics e.g. PS (Plastics SA, 2019). As certain polymers and polymer compositions are less economically rewarding to recycle, much of these are sent to landfill. However, it should be noted that landfill space is limited and so diversion from landfill is ideal (DEA, 2018).

However, it is clear that maintaining the status quo in the face of increasing population, industry, consumerism, and wealth, will increase the land-based plastic loadings to the sea. Urgent interventions, awareness, voluntary actions, and regulations are needed to stem the flow of plastics to our oceans. An understanding of the underpinning factors and knowledge gaps is necessary to inform effective and integrated land-based remediation and intervention options and policies.

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Authors' contributions

C.V.: Writing – the initial draft; writing – revisions; project management.

H.B.: Writing – revisions; student supervision; funding acquisition; project leadership.

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Chapter 4

Contextualization of airborne microplastic pollution in the South African environment

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ABSTRACT

Plastic has been found in all compartments of the environment, with specific research focus on the aquatic environment. An emerging body of literature highlights the presence of synthetic polymers suspended in the air column and the deposition thereof. The presence of plastic in the air that we breathe poses several potential health risks which are being investigated. Atmospheric microplastic might be the form of this pollutant that humans are most exposed to, as small particles are able to pass through the respiratory tract and come in contact with the respiratory surface of lungs. The presence of synthetic polymers in human lungs have been associated with cancer formation in lung tissue. Due to the uncertainty of the health risks associated with atmospheric microplastics and the presence of microplastic found elsewhere in the South African environment, it is recommended that further research is conducted on the scope of microplastics in the air of industrialised, residential, and natural regions of Southern Africa.

Keywords: Microplastics, atmospheric fallout, inhalation, ecotoxicology, environmental health, anthropogenic activity, health risk

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Note 1: This article was presented at the 2018 Conference of the National Association for Clean Air, South Africa and received the prize for best student paper at the conference.

Note 2: This article has been cited 3 times (Google Scholar) by December 2023.

Note 3: The citation and reference style in the publication has been changed to be consistent with the style of this thesis.

4.1 Introduction

4.1.1 Microplastics

Useful and highly variable material characteristics have made plastic the material of choice for an array of industries. The use of plastic outside of military applications began after World War II, seeing the plastics industry grow faster than that of any other man-made material (Geyer *et al.* 2017). The plastic industry in South Africa is not exempt from the international growth trend, growing at a faster rate than the local economy in recent years (Verster *et al.*, 2017). A total of 1.518 million tons of virgin polymer was converted into products in South Africa during 2016 (Plastics SA, 2017).

Microplastics are synthetic polymer particles smaller than 5 mm (Arthur *et al.* 2008) and can be categorised into primary and secondary microplastics (Cole *et al.*, 2011). Primary microplastics are particles produced to be microscopic in size, such as primary production pellets or microbeads used in the cosmetic, pharmaceutical and air blasting industries. Secondary microplastics are the result of breakdown of larger plastic items due to UV and mechanical weathering. This can be subdivided into fragments and fibres. Fibres are of importance as 90% of plastic particles in atmospheric fallout are fibrous (Dris *et al.*, 2015). These fibres are released due to the weathering of textiles products, of which clothing contributes the greatest fraction to atmospheric fibres (Dris *et al.*, 2015). A single wash from a laundry washing machine can release up to 1900 fibres into wastewater discharge (Browne *et al.*, 2011).

4.1.2 Brief history of microplastic research

Microplastic is a contaminant of emerging concern (STAP, 2012) which has received increasing attention in the past decade (Wagner & Lambert, 2018). The first studies identifying the presence of small plastic particles in the environment were conducted in the early 1970s (Carpenter *et al.,* 1972). Renewed interest in this topic began in the 2000s, increasing steadily to generate a substantial amount of literature, with specific focus on microplastics in the ocean (Magnusson *et al.*, 2016). The UNEP Yearbook 2014 proposed microplastics as one of ten emerging issues and identifies it as an important factor in the loss of biodiversity globally (UNEP, 2014). Plastic is found to be ubiquitous in the marine environment and the need to investigate freshwater pathways as source of oceanic plastic arose (Wagner & Lambert, 2018). This saw a gradual increase of studies on microplastics in freshwater, mainly on economically important rivers and lakes in North America and Europe. Isolated cases of such studies in developing countries such as India (Ramasamy, 2016), Mongolia (Free *et al.*, 2014), China (Fok & Cheung, 2015) and the Lake Victoria region in Africa (Biginagwa *et al.*, 2016) have been conducted. The first such study in South Africa, and the first riverine microplastic study in Africa recently determined the scope of microplastic pollution in fresh water in Gauteng and North West (Bouwman *et al.,* 2018). This highlighted the need for further investigation of major South African rivers such as the Vaal River.

More researchers are trying to track the sources and sinks of microplastic pollution in all water bodies, as well as quantifying how much of it is due to atmospheric fallout. Plastic particles in soil is also an area of emerging concern, as soil shares certain characteristics with the aquatic environment (Rillig, 2012). There are however no such known studies done in Africa to scope the impact of soil or airborne plastic particles.

4.1.3 The ‘Plastic Cycle’

Land-based plastic products are broken down and released into air and water, moving between different environmental compartments. Research has focused mostly on aquatic environments, but a new body of literature is highlighting the importance of terrestrial microplastics and the movement of plastic between these compartments.

Microplastics originate from the breakdown of larger plastics, whether it be fibrous or solid articles. This breakdown can happen on land or in water. Fibres are more prone to become airborne than fragments due to higher wind resistance, hence most synthetic particles found in the air are fibres (Dris *et al.*, 2016). These particles move around in the air and get deposited on land or in water. An estimated 7% of oceanic microplastics are deposited directly into the ocean by wind transfer (Boucher & Friot, 2017).

Wastewater treatment plants remove up to 97% of microplastics (Mintenig *et al.*, 2017). This plastic then gets sedimented out and deposited into agricultural land as wastewater sludge (Nizzetto *et al.*, 2016). Rivers carry plastics to the ocean, where most of it will remain and get broken down. Some of these particles can however get redeposited in air by sea spray (Prata, 2018). When plastic gets washed up on land and airborne it closes the so called 'plastic cycle' (Prata, 2018).

4.2 Airborne plastic

Although the study of microplastic pollution in air is in its infancy, significant amounts of plastic, especially fibres, have been found in settled dust and atmospheric fallout (Prata, 2018). Different air compartments are associated with different levels of airborne plastic, but it is estimated that between 1 600 and 11 130 fibres/m^2/day can be deposited (Dris *et al.*, 2017). Of this an estimated 29% are petrochemical based synthetic fibres (Dris *et al.*, 2016).

4.2.1 Indoors

The indoor environment contains much more synthetic fibres than outdoor air (Gasprei *et al.*, 2018). This is mainly due to the fact that the sources for microplastic fibres – clothes – are more abundant indoors. It has also been found that the dominant polymer fibre in the air correlates with the seasons fashion trends, reinforcing the idea that clothes are the main source of atmospheric fibres (Dris *et al.,* 2016). Approximately 33% of indoor fibres are petrochemically based (Gasprei *et al.*, 2018). Larger fibres settle faster and implicitly smaller fibres can stay airborne for longer. Workers in the textile and plastics industries run a great risk of pneumonic diseases due to chronic exposure to high concentrations of airborne microplastics in their occupational environment (Prata, 2018).

4.2.2 Outdoors

As textile materials are more frequently used and more concentrated indoors, outdoor concentrations of atmospheric fibres are comparably lower. Dris *et al.* (2017) found that fibres in outdoor air are significantly shorter than those found indoors. The concentration of outdoor fibres varied between 0.3 and 1.5 fibres /m^3, compared to the range of 1 to 60 fibres /m^3 indoors. The presence of microplastics in the outdoor air column can mostly be ascribed to mixing with indoor air and in the process also diffusing microplastics into the outdoor environment (Prata, 2018). Weather seems to play a role in the deposition of atmospheric plastics. Atmospheric fallout increased by a magnitude of 10 on rainy days (Dris *et al.*, 2016).

Microplastics suspended in air is carried about by weather (Prata, 2018). Plastic found in a remote mountain lake in China (Zhang *et al.*, 2016), where anthropogenic contact is very limited, is likely deposited by atmospheric fallout. The effect of plastic on the total environment is thus augmented by its distribution through air.

4.3 Possible effects

4.3.1 Biota

The adverse effects of plastic on biota have been an emerging field of study over the past decade. The bulk of work is done in the aquatic environment. As airborne microplastic contamination is associated with anthropogenic activities (Dris *et al.,* 2017), and less animals naturally live in densely populated industrial and residential areas compared to rural areas, the effect on airborne plastics and animals would not be that much of a concern. Plants in densely populated areas are however of concern, as synthetic fibres have been found in flowering plants (Prata, 2018).

4.3.2 Effects on humans

There are several health problems associated with inhaling microplastics. The method of microplastic quantification limits the minimum size of fibres. Most studies are only able to detect fibres up to 50 µm or even 100 µm (Gasprei *et al.*, 2018). Particles this large – when compared to nano plastics – are much easier filtered out by respiratory tract defence mechanisms before entering the lung (Prata, 2018). Although the hydrophobic qualities of polymers make it harder for mucus and villi to remove the particles, it is unlikely that particles reach the respiratory surface of the lung (ibid.). We can therefore not accurately predict the number of plastic particles able to make it to the lung surface, as these size particles cannot be detected by standard microplastic analysis. Studies also show that the smaller the particles get, the higher the abundance, as larger fibres break down into smaller pieces (Gasprei *et al.*, 2018; Wagner & Lambert, 2018). These smaller particles also have a higher surface to area ratio and can adsorb more pollutants than their larger counterparts. These pollutants can be released once the particle has entered the body (Naidoo *et al.*, 2015). There is therefore a need to improve methods of microplastic sampling, quantification, and characterisation in air, and to detect smaller sized microplastics.

Particles not removed by natural defence mechanisms become trapped in the respiratory tract can have several effects. Because of the small particle size, many plastic particles are able to cross the cellular membrane (Prata, 2018). When inside the cell they can release adsorbed toxins, plasticisers and other pollutants that have been shown to associate with microplastics in the environment. In this way they act as a vector for chemicals and pollutants to get released intracellularly (Bouwman *et al.*, 2018). The presence of these xenobiotic particles is also known to stimulate the production of reactive oxygen species (ROS) in cells (Prata, 2018). The carcinogenic effects of ROS are well known. The plastic particle is also able to directly cause mechanical damage to genetic material that can lead to cancer formation (Prata, 2018). Also, as much as 97% of malignant pneumonic tissue specimens contain synthetic fibres (Pauly *et al.*, 1998).

As this research is in its infancy, very little is known about the extent of health risks regarding inhaled plastic. Dust is a likely source of microplastic exposure when ingested and pose threats specifically to small children (Prata, 2018). Trace amounts of Al, Na, Ca, Mg and Si have been detected on the surface of microplastics found in settled dust (Dehghani *et al.*, 2017).

4.4 Microplastics in South African air

Microplastic research in South Africa has been lagging (Verster *et al.*, 2017). Plastic was found in all compartments of the South African freshwater environment including surface water from rivers and ground water (Bouwman *et al.*, 2018). Considering the movement of plastic across different compartments of the environment as depicted by Prata (2018), there is reason to believe that the Gauteng area in South Africa would contain significant levels of microplastic pollution. There is a definite correlation between anthropogenic activity in an area and the amount of fibres found in the air (Dris *et al.*, 2016) and the Gauteng province is the industrial hub of South Africa.

In the report commissioned by the Water Research Commission (Bouwman *et al.*, 2018) elevated levels of fibres were found in the Jukskei and other rivers surrounding the metropolitan area. A possible source for these fibres is atmospheric fallout, as not all the riverine areas highly contaminated with fibres are directly in contact with heavy anthropogenic activities. Due to the lack of knowledge on the health risks pertaining to inhaled microplastics and the levels of synthetic fibre pollution found in the rivers in and around Gauteng, we suggest that the state of atmospheric microplastics in South Africa specifically, and Southern Africa in general should be investigated. A better understanding of aerial plastics may eventually assist in mitigation of plastic contamination of the entire environment and alleviate pressure on human and ecosystem health.

4.5 Acknowledgments

We kindly thank the Water Research Commission for funding a foundational microplastic project in South African fresh waters and availing the data.

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Chapter 5

A scoping study on microplastics in water environments

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Abstract

As the first study to quantify freshwater microplastics in South Africa, we report baseline data of environmental microplastic concentrations for the country. All major rivers and streams in the Gauteng province, as well as river and groundwater from the North West provinces were sampled—all contained microplastics. Potable tap water from residential areas in Gauteng also contained microplastic, although in lower concentrations then that of rivers. The mean concertation in river water was 4.0 n/L, while that of tap water and groundwater were 1,3 n/L and 0.17 n/L, respectively. Two adjacent sites in the Vaal River inexplicably had MP concentrations an order of magnitude greater than the mean, warranting further investigation. Microplastic concentrations in river water showed great spatial variation, ranging from 0.33 to 56 n/L. The most prominent questions that arose were regarding sources and temporal variation of MPs in South African freshwater.

Keywords: Microplastics, South Africa, rivers, fragments, fibres, groundwater, drinking water.

Note 1: This article is adapted from the Water Research Committee report 'Microplastic in freshwater environments – A scoping study' (Bouwman *et al*., 2018). This report has been cited 43 times as of December 2023.

Note 2: Presented here is a shortened version intended for eventual submission.

Note 3: Figures and tables from the original report are included in the article.

Note 4: Table 5.6 has been updated.

Note 5: Section 5.4.3 has been updated with a recent publication I did with Rand Water on microplastics in tap water (Swanepoel *et al*., 2023).

Note 6: Gaps identified in this chapter informed the experimental design of the studies discussed in chapter 6 and 7.

5.1 Introduction

Plastic as environmental pollutant has been increasing and its effects are becoming more obvious. It is detrimental to environmental health (Prata *et al*., 2021). Microplastics (MPs) from land-based sources enter rivers (Horton *et al*., 2018) where it either gets deposited and interacts with freshwater ecosystems and water users, or gets transported to the ocean (Jambeck *et al*., 2015). It is clear that concerted action needs to be taken regarding plastic and waste management, but a lack of consensus on how the problem should be approached without doing major socio-economic damage has been an obstacle (Verster *et al*., 2017). The collection and interpretation of environmental data is critical to identify the most appropriate interventions and assess its implementation efficiency. At the onset of this study, no data on South African MPs had been available.

Microplastics are plastic particles larger than 1 µm and smaller than 5 mm (Frias *et al*., 2019). It can be produced to be this small, in which case it is referred to as primary microplastics (Boucher *et al*., 2017). These include microbeads, used as abrasive in various processes, and pellets or nurdles (Boucher *et al*., 2017). The latter are the virgin material or recyclate that will be used to manufacture plastic products. Macroplastic can also break down to the microplastic range by physical and chemical abrasion (Lv *et al*., 2022). This process often happens in natural environments where plastics are released and left to the forces of the elements. Microplastic detected in the environment can therefor either be due to the release of primary microplastics of the breakdown of larger plastic pieces that are already there. Fibres are due to the breakdown of textiles and can either be released due to washing of textiles (*Napper et al*.., 2016) and WWTP discharge (Chan *et al*., 2021), or be deposited by atmospheric fallout (Dris *et al*., 2016)

This study arose from a need to gather baseline data as a scoping study on MP concentrations in the water of the Gauteng and North West provinces and compare it on a global scale. Gauteng is South Africa's smallest but most urbanised and densely populated province (Parker *et al*., 2022). Results would then be used to do an initial assessment on the MP concentrations in South Africa and identify and priority areas for further research.

5.2 Materials and methods

The aim of this study was to conduct a limited scoping survey of microplastics in surface waters, ground water, and tap water to test robust sampling and counting methods.

5.2.1 Site selection for water sampling

We conducted a survey of surface water, tap water, and ground water from Gauteng and North West Province. Site selection was done according to location and accessibility of rivers. Most sites were selected along bridges or riverbanks where deeper parts of the river would be easily accessible. Three major river catchments were covered in this study: The Vaal to the south, Olifants to the north-east and Limpopo to the north-west of the study area. We selected 43 freshwater sampling sites across Gauteng and the North West Province to sample water (Figure 5.1). We also analysed tap water samples from Johannesburg and Pretoria. Ground water was collected from four boreholes in Potchefstroom. Tap water was collected from residences in Tshwane and City of Johannesburg municipalities.

5.2.2 Experimental procedures

To develop and test a robust method to sample and quantify microplastics in South African freshwaters, we selected the NOAA standard microplastic protocol (Masura *et al*., 2015), and adapted some of the procedures for South African conditions.

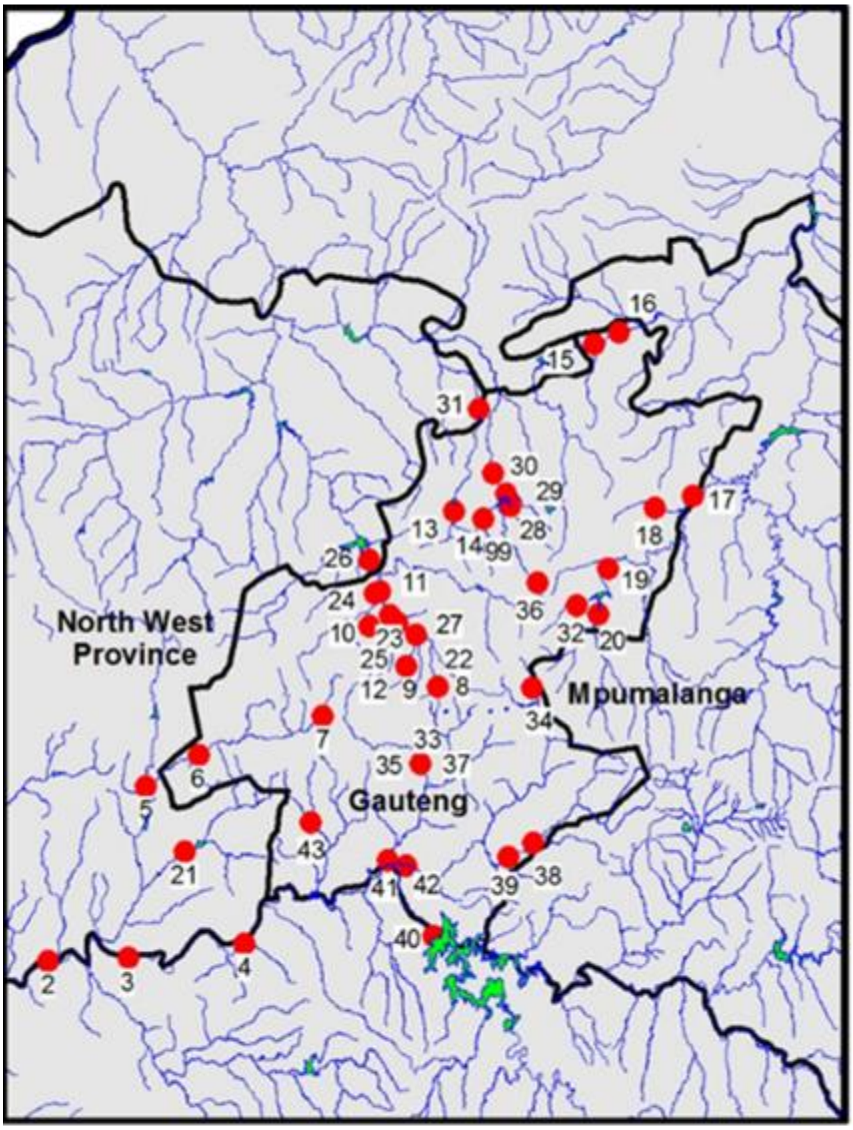


Figure 5.1: Sampling sites in Gauteng and North West Province. Large water bodies are indicated in green.

5.2.2.1 Sampling

From literature, we ascertained that to sample adequate volumes of water, we would need 90 L. This is of course too much to be easily transported back to the laboratory. We devised a method where we sampled six replicates of water with a 15 L metal bucket and filtered the required volume of 90 L through a 25 µm sieve. The sieve was pre- and post-cleaned with filtered double-distilled water to minimise contamination. The content of the sieve after filtration was rinsed and stored in pre-cleaned HDPE bottles and transported to the laboratory.

5.2.2.2 Sample clean-up and analysis

Samples were decanted into glass beakers and the bottle rinsed a minimum of three times to remove all remaining visible debris. The glassware was then covered with tin foil or a watch glass and placed in a drying oven at 90°C until all the water had evaporated.

Dried material was subjected to wet peroxide as described in Masura *et al.* (2015) to digest biological material. Density separation was then done similar to the method described in

Masura *et al.* (2015). Sodium iodide was used instead of sodium chloride, as a sodium chloride solution does not allow PVC to be floated in a density separation, and a sodium iodide solution of the same molar concentration does so. After samples were left overnight in the density separators (Figure 5.2), and clear settling of sediments could be observed, the sedimented layer was removed and retained in a petri dish. This was subsequently inspected for microplastic using a dissection microscope. If any plastics were found it was removed using a pair of forceps and added to the rest of the collection for that sample.

The remainder of the liquid containing the less dense material such as plastic was filtered through a custom-made 25 µm stainless steel filter (Figure 5.3) that is spring clamped in the place of filter paper, membranes, or sintered discs in a normal 47 mm glass vacuum filtration system. The disc is made entirely of metal and has black rubber O-rings to ensure a seal when clamped. Filtered and dried samples were carefully stored in petri dishes until counting.

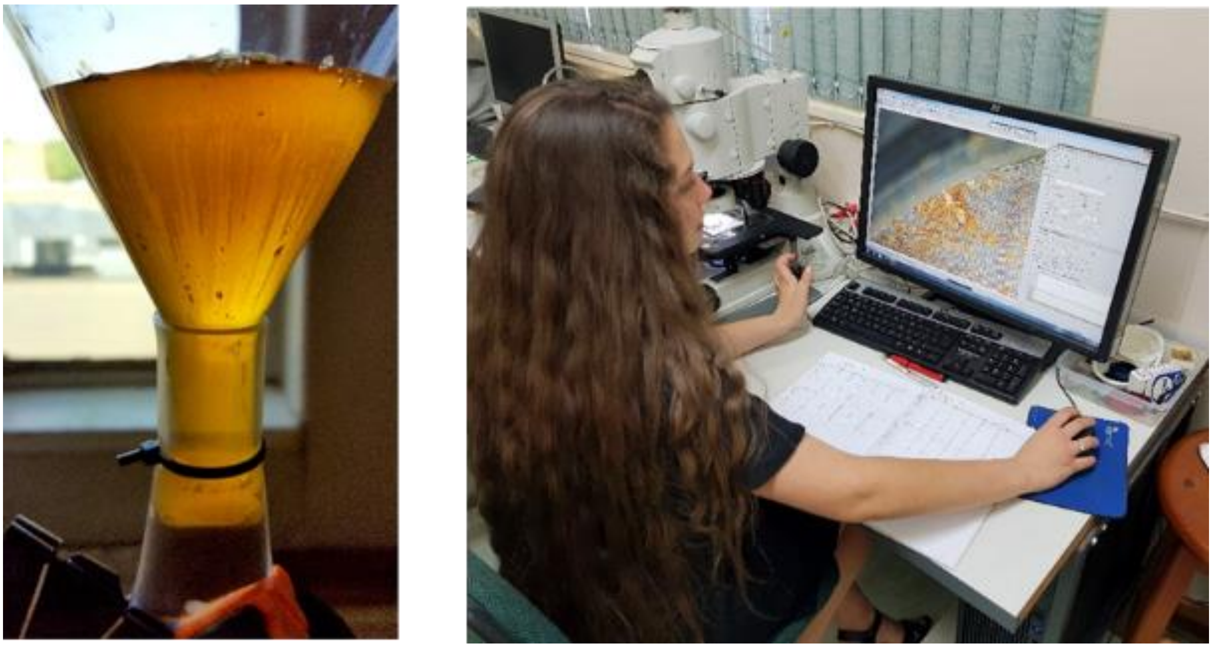


Figure 5.2: Sample preparation and analysis

Each concentration disc was inspected using a Nikon EZ 100 multizoom compound binocular microscope (Figure 2.2). We measured the longest dimension of each particle. MPS were divided into two morphotypes. Fragments and fibres were counted plotted on counting matrices according to colour and longest dimension measured. A subsample of larger plastic fragments was then manually removed and analysed on an ATR-FTIR for polymer identification using Agilent ResPro (e.g. Figure 5.4).

Figure 5.3: Custom-made stainless-steel filter, with microplastics filtered from 90 L of surface water.

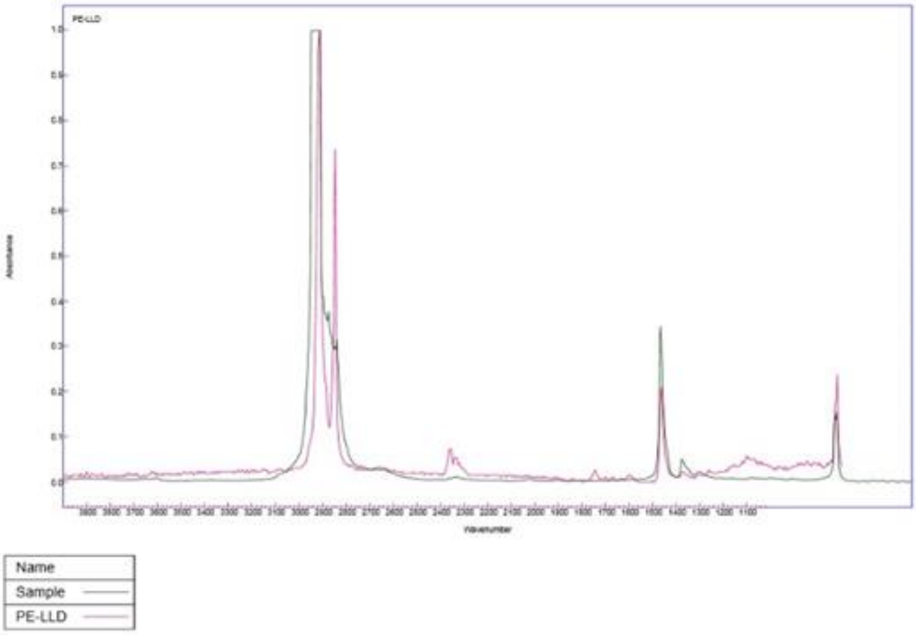


Figure 5.4: ART-FTIR spectra of sample (green) and library for LD-PE (red)

5.3 Results

5.3.1 Surface water results

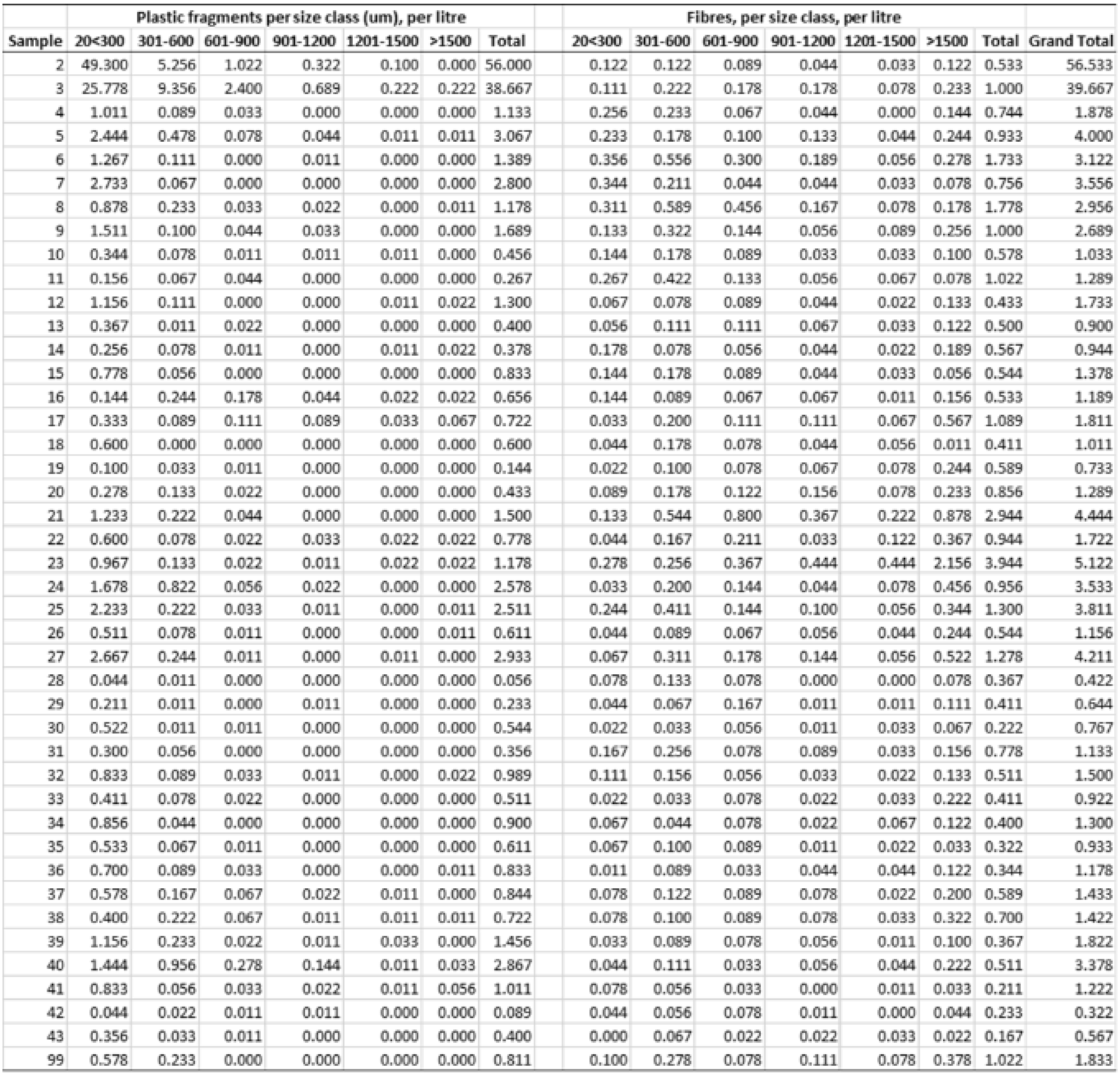
5.3.1.1 Descriptive statistics

We counted a total of 15 526 fragments and fibres from 43 freshwater samples. The quantitative results are displayed in Table 5.2 as fragments or fibres (n/L), respectively. Two samples (2 and 3), both from the Vaal River, immediately stands out because of the order of magnitude higher particle counts. In most cases, these two samples were not used for statistics or graphs, because of distortions.

Excluding the samples from sites 2 and 3, there were 3 963 plastic fragments, and 3 281 fibres. This translates to about 55% of the detected were fragments. The mean concertation in river water was 4.0 n/L, while that of tap water and groundwater were 1,3 n/L and 0.17 n/L, respectively.

The various size classes for fragments and fibres were plotted and presented in Figure 5.5. The log-transformed plots of the data are also shown, as the distributions per size class were not Gaussian. Please note that samples 2 and 3 are not included in these results as the were removed as outliers.

Table 5.1: Counts of fragments and fibres (n/L) of water from 43 sites according to size classes.



	Plastic fragments per size class (um), per litre							Fibres, per size class, per litre							
Sample	20<300	301-600	601-900	901-1200	1201-1500	>1500	Total	20<300	301-600	601-900	901-1200	1201-1500	>1500	Total	Grand Total
2	49.300	5.256	1.022	0.322	0.100	0.000	56.000	0.122	0.122	0.089	0.044	0.033	0.122	0.533	56.533
3	25.778	9.356	2.400	0.689	0.222	0.222	38.667	0.111	0.222	0.178	0.178	0.078	0.233	1.000	39.667
4	1.011	0.089	0.033	0.000	0.000	0.000	1.133	0.256	0.233	0.067	0.044	0.000	0.144	0.744	1.878
5	2.444	0.478	0.078	0.044	0.011	0.011	3.067	0.233	0.178	0.100	0.133	0.044	0.244	0.933	4.000
6	1.267	0.111	0.000	0.011	0.000	0.000	1.389	0.356	0.556	0.300	0.189	0.056	0.278	1.733	3.122
7	2.733	0.067	0.000	0.000	0.000	0.000	2.800	0.344	0.211	0.044	0.044	0.033	0.078	0.756	3.556
8	0.878	0.233	0.033	0.022	0.000	0.011	1.178	0.311	0.589	0.456	0.167	0.078	0.178	1.778	2.956
9	1.511	0.100	0.044	0.033	0.000	0.000	1.689	0.133	0.322	0.144	0.056	0.089	0.256	1.000	2.689
10	0.344	0.078	0.011	0.011	0.011	0.000	0.456	0.144	0.178	0.089	0.033	0.033	0.100	0.578	1.033
11	0.156	0.067	0.044	0.000	0.000	0.000	0.267	0.267	0.422	0.133	0.056	0.067	0.078	1.022	1.289
12	1.156	0.111	0.000	0.000	0.011	0.022	1.300	0.067	0.078	0.089	0.044	0.022	0.133	0.433	1.733
13	0.367	0.011	0.022	0.000	0.000	0.000	0.400	0.056	0.111	0.111	0.067	0.033	0.122	0.500	0.900
14	0.256	0.078	0.011	0.000	0.011	0.022	0.378	0.178	0.078	0.056	0.044	0.022	0.189	0.567	0.944
15	0.778	0.056	0.000	0.000	0.000	0.000	0.833	0.144	0.178	0.089	0.044	0.033	0.056	0.544	1.378
16	0.144	0.244	0.178	0.044	0.022	0.022	0.656	0.144	0.089	0.067	0.067	0.011	0.156	0.533	1.189
17	0.333	0.089	0.111	0.089	0.033	0.067	0.722	0.033	0.200	0.111	0.111	0.067	0.567	1.089	1.811
18	0.600	0.000	0.000	0.000	0.000	0.000	0.600	0.044	0.178	0.078	0.044	0.056	0.011	0.411	1.011
19	0.100	0.033	0.011	0.000	0.000	0.000	0.144	0.022	0.100	0.078	0.067	0.078	0.244	0.589	0.733
20	0.278	0.133	0.022	0.000	0.000	0.000	0.433	0.089	0.178	0.122	0.156	0.078	0.233	0.856	1.289
21	1.233	0.222	0.044	0.000	0.000	0.000	1.500	0.133	0.544	0.800	0.367	0.222	0.878	2.944	4.444
22	0.600	0.078	0.022	0.033	0.022	0.022	0.778	0.044	0.167	0.211	0.033	0.122	0.367	0.944	1.722
23	0.967	0.133	0.022	0.011	0.022	0.022	1.178	0.278	0.256	0.367	0.444	0.444	2.156	3.944	5.122
24	1.678	0.822	0.056	0.022	0.000	0.000	2.578	0.033	0.200	0.144	0.044	0.078	0.456	0.956	3.533
25	2.233	0.222	0.033	0.011	0.000	0.011	2.511	0.244	0.411	0.144	0.100	0.056	0.344	1.300	3.811
26	0.511	0.078	0.011	0.000	0.000	0.011	0.611	0.044	0.089	0.067	0.056	0.044	0.244	0.544	1.156
27	2.667	0.244	0.011	0.000	0.011	0.000	2.933	0.067	0.311	0.178	0.144	0.056	0.522	1.278	4.211
28	0.044	0.011	0.000	0.000	0.000	0.000	0.056	0.078	0.133	0.078	0.000	0.000	0.078	0.367	0.422
29	0.211	0.011	0.000	0.011	0.000	0.000	0.233	0.044	0.067	0.167	0.011	0.011	0.111	0.411	0.644
30	0.522	0.011	0.011	0.000	0.000	0.000	0.544	0.022	0.033	0.056	0.011	0.033	0.067	0.222	0.767
31	0.300	0.056	0.000	0.000	0.000	0.000	0.356	0.167	0.256	0.078	0.089	0.033	0.156	0.778	1.133
32	0.833	0.089	0.033	0.011	0.000	0.022	0.989	0.111	0.156	0.056	0.033	0.022	0.133	0.511	1.500
33	0.411	0.078	0.022	0.000	0.000	0.000	0.511	0.022	0.033	0.078	0.022	0.033	0.222	0.411	0.922
34	0.856	0.044	0.000	0.000	0.000	0.000	0.900	0.067	0.044	0.078	0.022	0.067	0.122	0.400	1.300
35	0.533	0.067	0.011	0.000	0.000	0.000	0.611	0.067	0.100	0.089	0.011	0.022	0.033	0.322	0.933
36	0.700	0.089	0.033	0.000	0.000	0.011	0.833	0.011	0.089	0.033	0.044	0.044	0.122	0.344	1.178
37	0.578	0.167	0.067	0.022	0.011	0.000	0.844	0.078	0.122	0.089	0.078	0.022	0.200	0.589	1.433
38	0.400	0.222	0.067	0.011	0.011	0.011	0.722	0.078	0.100	0.089	0.078	0.033	0.322	0.700	1.422
39	1.156	0.233	0.022	0.011	0.033	0.000	1.456	0.033	0.089	0.078	0.056	0.011	0.100	0.367	1.822
40	1.444	0.956	0.278	0.144	0.011	0.033	2.867	0.044	0.111	0.033	0.056	0.044	0.222	0.511	3.378
41	0.833	0.056	0.033	0.022	0.011	0.056	1.011	0.078	0.056	0.033	0.000	0.011	0.033	0.211	1.222
42	0.044	0.022	0.011	0.011	0.000	0.000	0.089	0.044	0.056	0.078	0.011	0.000	0.044	0.233	0.322
43	0.356	0.033	0.011	0.000	0.000	0.000	0.400	0.000	0.067	0.022	0.022	0.033	0.022	0.167	0.567
99	0.578	0.233	0.000	0.000	0.000	0.000	0.811	0.100	0.278	0.078	0.111	0.078	0.378	1.022	1.833

5.3.1.2 Particle sizes

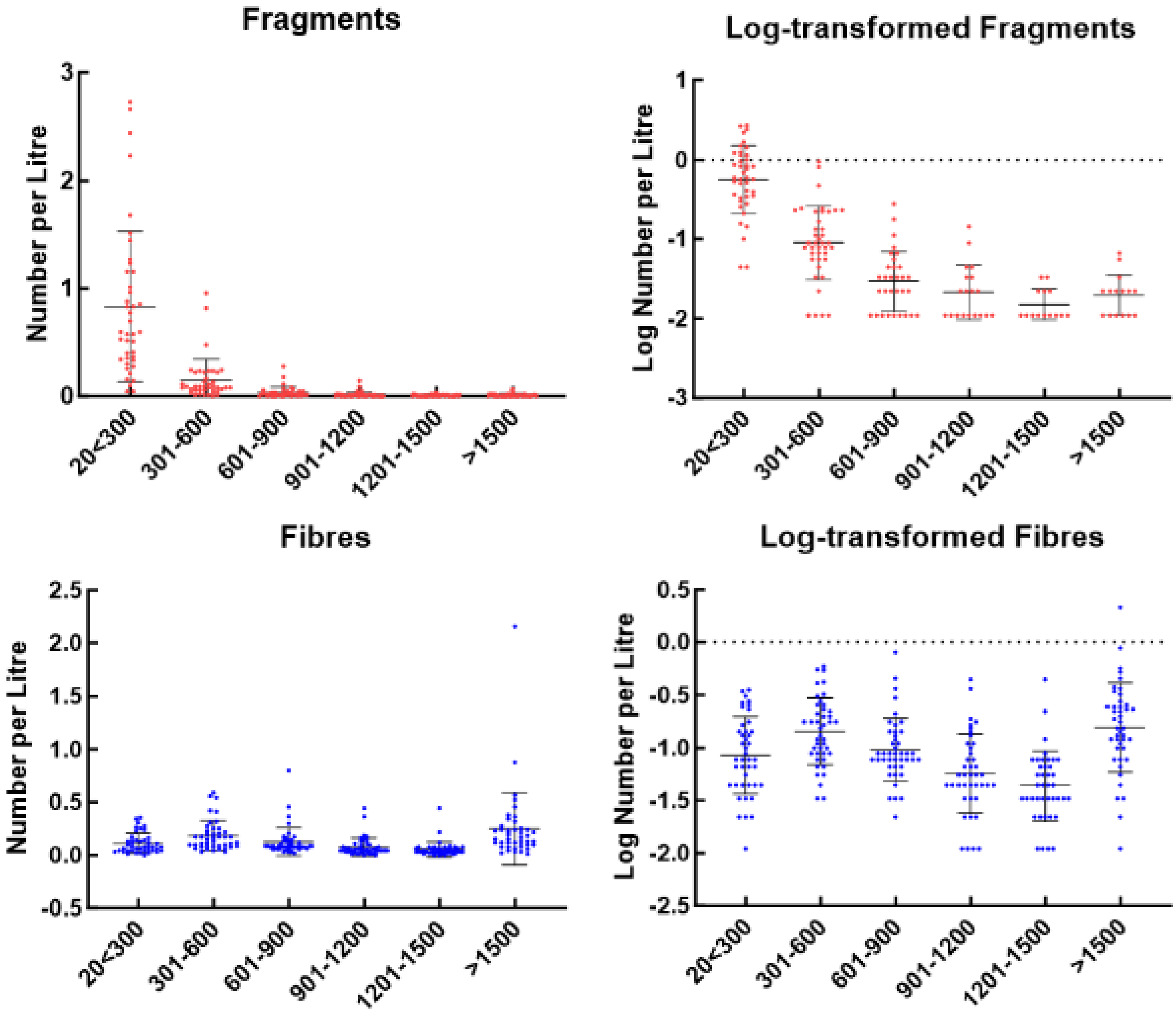


Figure 5.5: Scatterplots of untransformed and log-transformed size-class data for fragments (a and b) and fibres (c and d). Mean and standard deviations are shown. Two samples (2 and 3) were not included in any plot.

For fragments, there was a clear skewing towards the smaller sizes (Figure 5.5a and b). The Kruskal-Wallis tests for log-transformed data (Figure 5.5b) showed a highly significant difference between particle size classes ($p < 0.0001$). Subsequent Dunn's multiple comparisons tests showed significant differences between the 20 < 300 and 301 - 600 µm, and the 301 - 600 and 601 - 900 µm size classes (log-transformed) only. A post-test for linear trend between log-transformed size classes was also highly significant ($p < 0.0001$).

There was no discernible pattern for fibre size-classes. The Kruskal-Wallis test for log-transformed fibres was also highly significant ($p < 0.0001$) (Fig 5.5d). Dunn's multiple comparisons showed significant differences between 20 - 300 µm and 301 - 600 µm ($p = 0.03$), and between 301 - 600 µm and 601 - 900 µm ($p < 0.0001$) size classes. The summary statistics for fragments and fibres separate are presented in Table 5.3.

	Fragments						
	20<300	301-600	601-900	901-1200	1201-150	>1500	Total
Minimum	0.04	0.00	0.00	0.00	0.00	0.00	0.06
Maximum	2.73	0.96	0.28	0.14	0.03	0.07	3.07
Median	0.60	0.09	0.02	0.00	0.00	0.00	0.58
Mean	0.83	0.15	0.03	0.01	0.01	0.01	0.81
Standard deviation	0.70	0.19	0.05	0.03	0.01	0.02	0.83
	Fibres						
Minimum	0.00	0.03	0.02	0.00	0.00	0.01	0.17
Maximum	0.36	0.59	0.80	0.44	0.44	2.16	3.94
Median	0.08	0.16	0.09	0.06	0.03	0.16	0.58
Mean	0.12	0.19	0.13	0.08	0.06	0.25	0.82
Standard deviation	0.09	0.14	0.14	0.09	0.07	0.34	0.72

Table 5.2: Summary statistics for the 41 freshwater sites (n/L). Samples 2 and 3 are not included.

The summary statistics for fragments and fibres combined are shown in Table 5.3.

Table 5.3: Summary statistics for fibres and fragments combined (n/L). Column titled ‘All’ includes the data for sites 2 and 3. The column titled ‘Depleted’ have the data for sites 2 and 3 omitted.

	All	Depleted
Minimum	0.32	0.32
Maximum	56.53	5.12
Median	1.42	1.38
Mean	4.01	1.86
Standard deviation	10.10	1.24

The log-transformed data, that includes the data for Sites 2 and 3, are plotted in Figure 5.6.

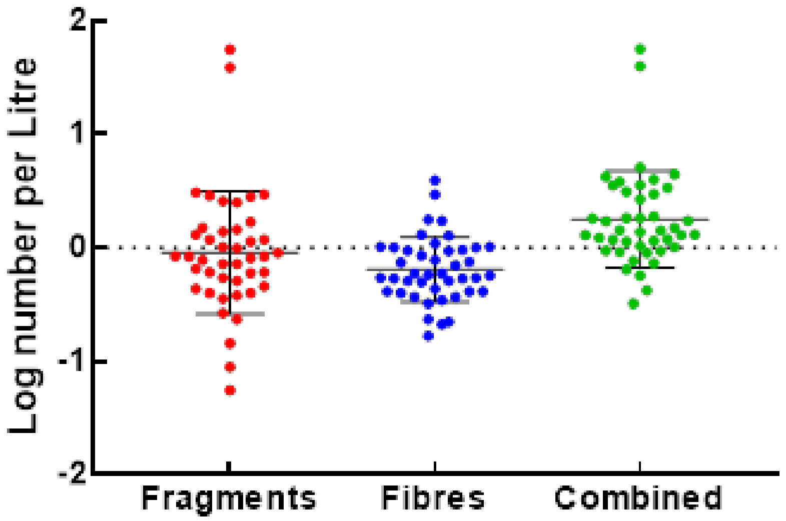


Figure 5.6: Log transformed data of fragments, fibres, and combined data, that includes data from Sites 2 and 3, are presented in this scatterplot. Means and standard deviations are shown.

A two-tailed Wilcoxon matched-pairs signed rank test of log-transformed fibre and fragment data (excluding data from Sites 2 and 3), produced a p-value of 0.13. The pairing though, was significant ($p = 0.003$) for a Spearman correlation r^2 of 0.42, showing some relationships between numbers of fibres and numbers of fragments in the same samples.

A linear regression between fibres and fragments (again excluding Sites 2 and 3), showed a poor fit of the data to the regression line ($r^2 = 0.077$) and a non-significant regression at $p = 0.08$ (Figure 5.7a). For log-transformed data, however, there was a significant regression ($p = 0.005$) (Figure 5.7b), but the fit of the points to the regression line remained poor ($r^2 = 0.18$).

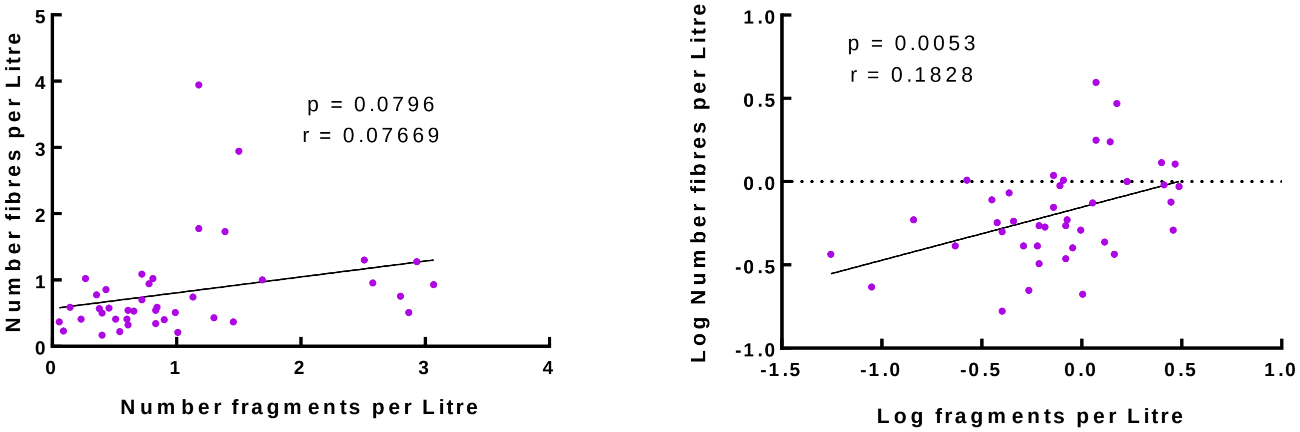


Figure 5.7: Linear regression of untransformed (a) and log-transformed (b) numbers of fragments and fibres (n/L).

5.3.1.3 Geographic distribution

Knowledge on the distributions of plastic particles in water allows a determination of where risks may be expected, and where interventions will be most effective. Figure 5.8 shows the distribution of the combined fibre and fragment numbers per L in 41 freshwater samples.

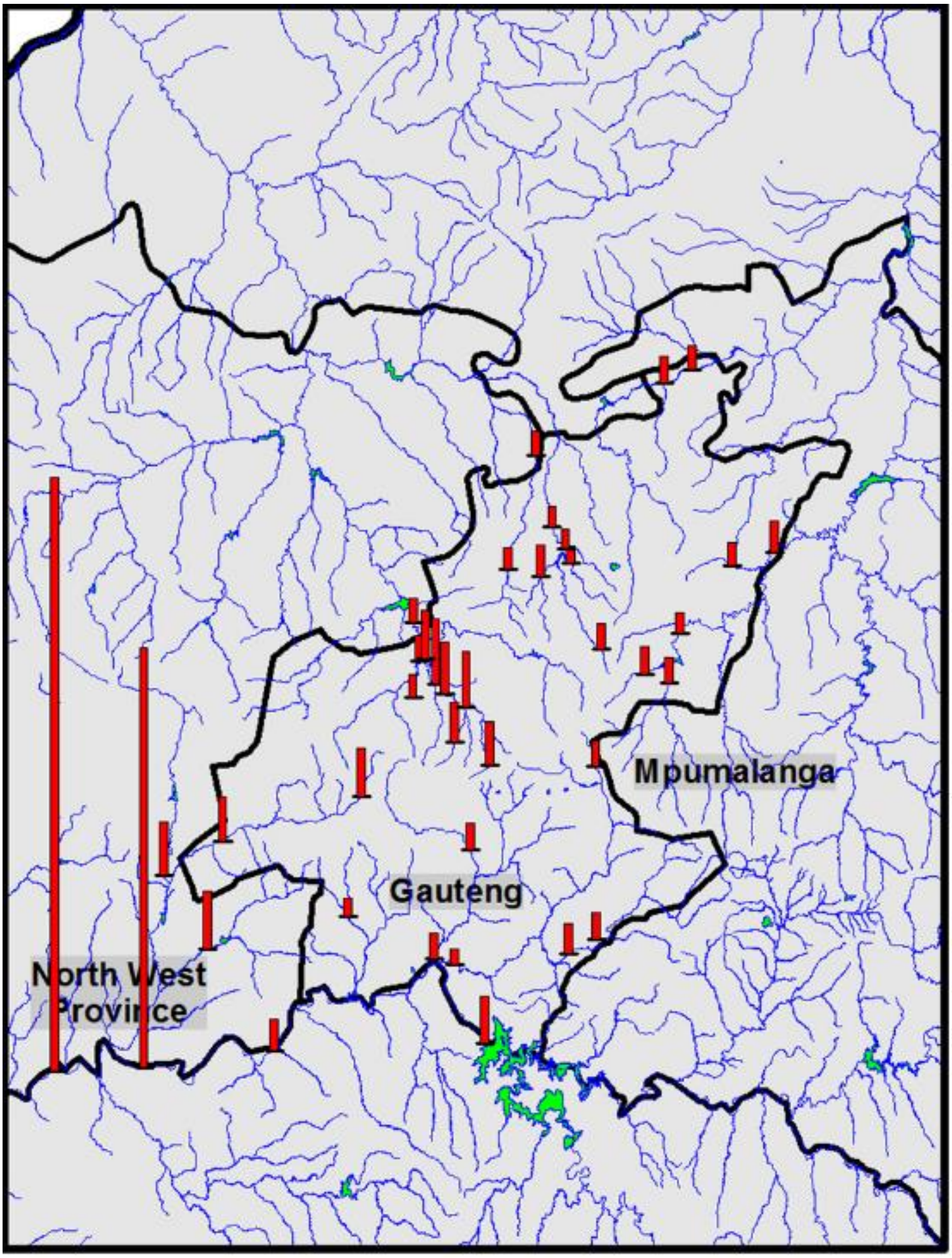


Figure 5.8: Distributions of total microplastic (fragments and fibres) concentrations in freshwater at all sampling sites. The tallest bar represents 56 n/L.

The relatively high numbers for the two Vaal River sites 2 and 3 skewed the results and made comparisons with other sites difficult. The data from these sites were excluded form some of the statistics and maps.

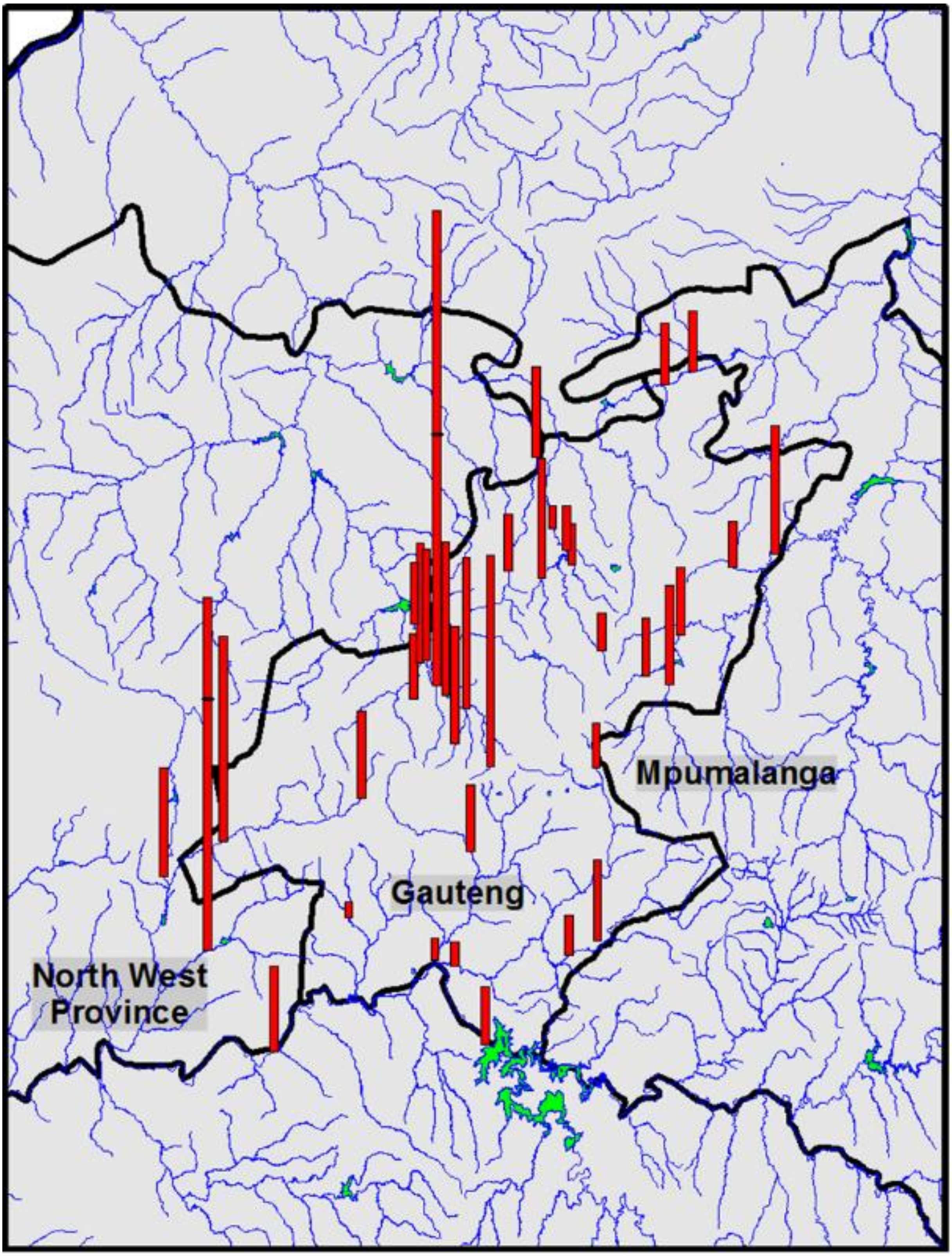


Figure 5.9: Distribution of total particles (fragments plus fibres) per L of water at all sampling sites. Sites 2 and 3 are excluded. The tallest bar represents 5.12 n/L.

Besides high concentrations in the Vaal River (Figure 5.8), the Jukskei River draining central Gauteng, and the Mooi River in the North West Province had high MP concentrations (Figure 5.8).

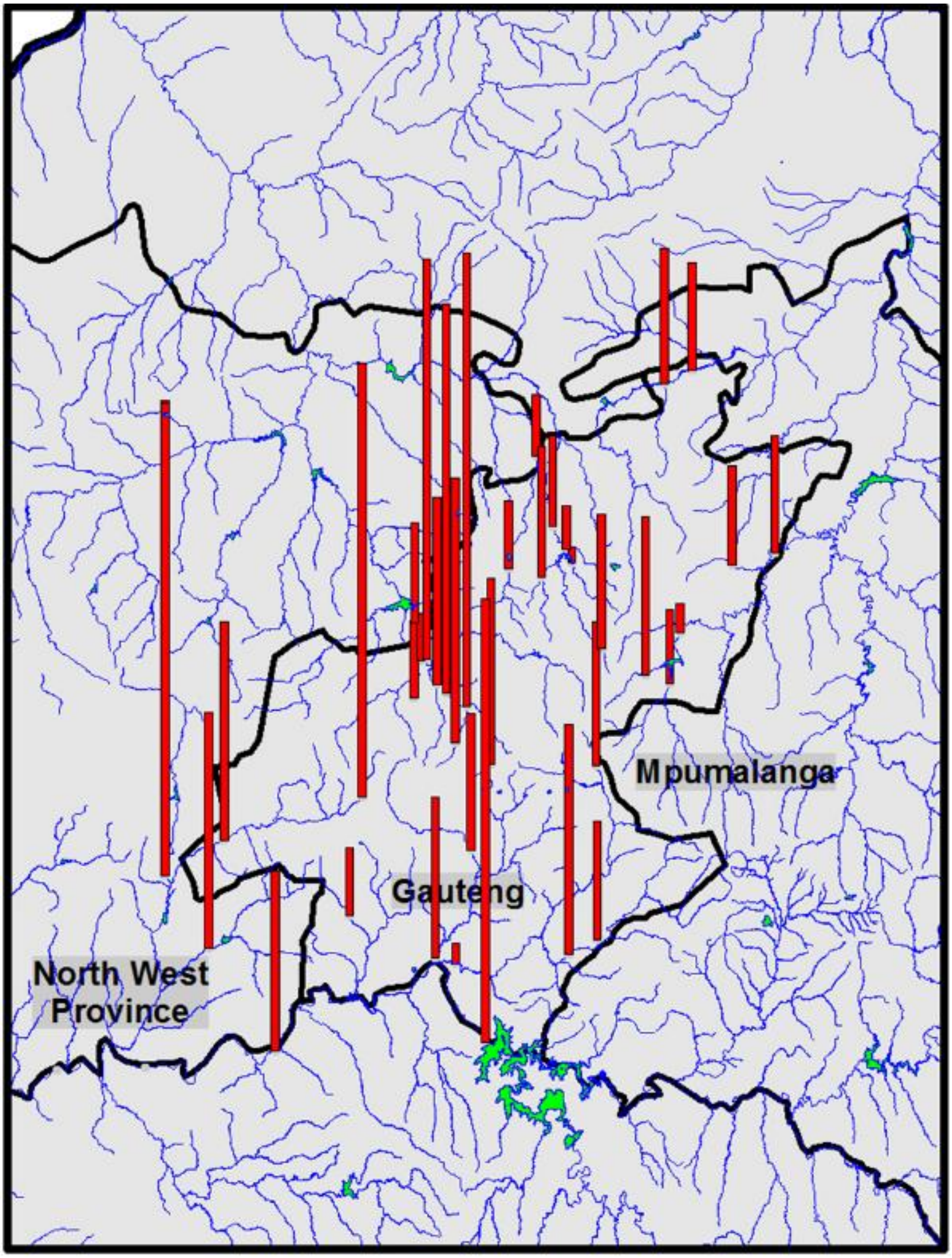


Figure 5.10: Fibres per L of water, per collection site, excluding Sites 2 and 3. The tallest bar represents 4 n/L.

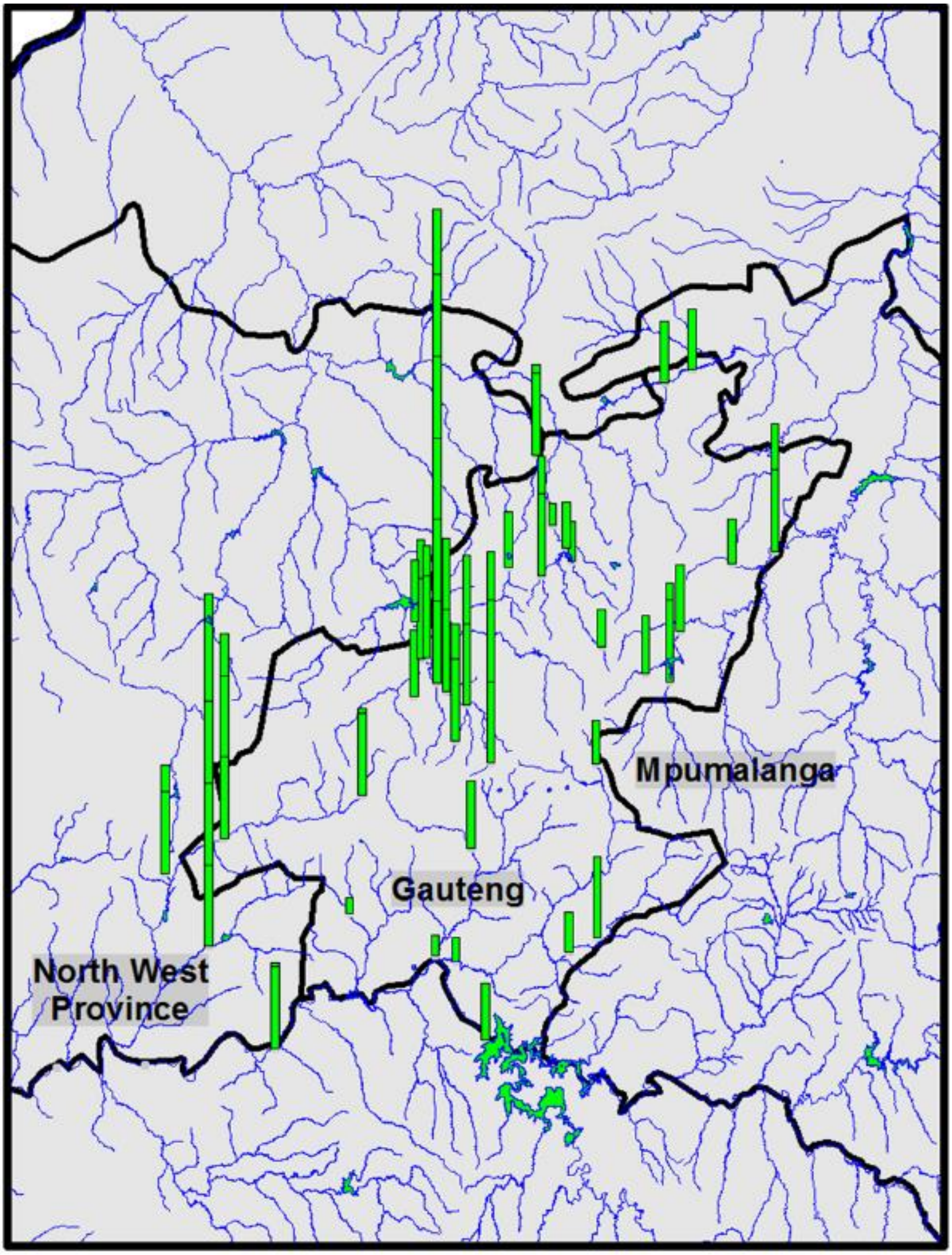


Figure 5.11: Fragments per L of water, per collection site, excluding Sites 2 and 3. The tallest bar represents 3.9 n/L.

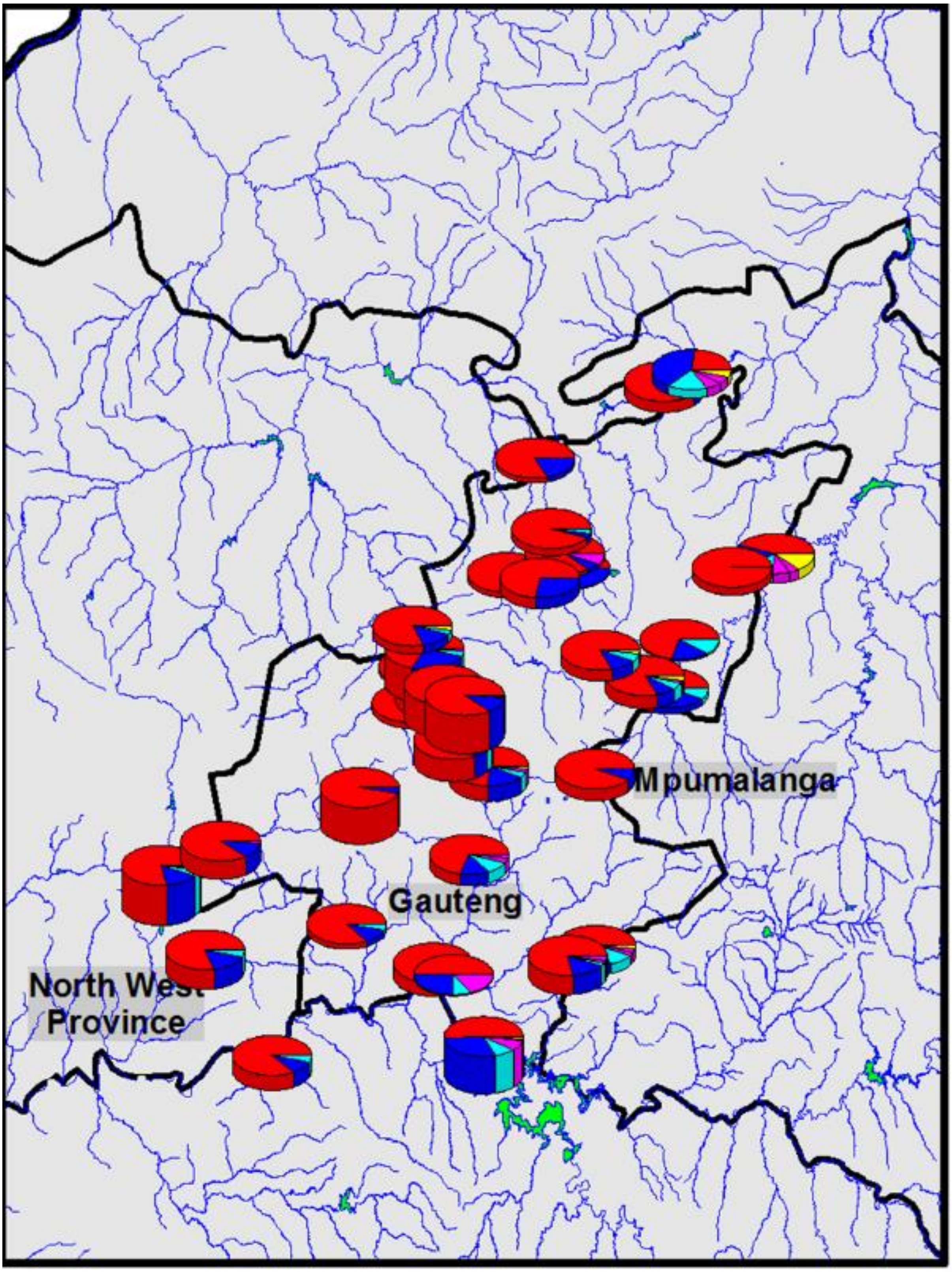


Figure 5.12: Pie charts of the size (µm) composition profiles of fragments. The height of each pie represents the number of fragment particles. Red = 20<300; Purple = 3001-600; Dark blue = 601-900; Light blue = 901-1200; Green = 1201-1500; Yellow = >1500.

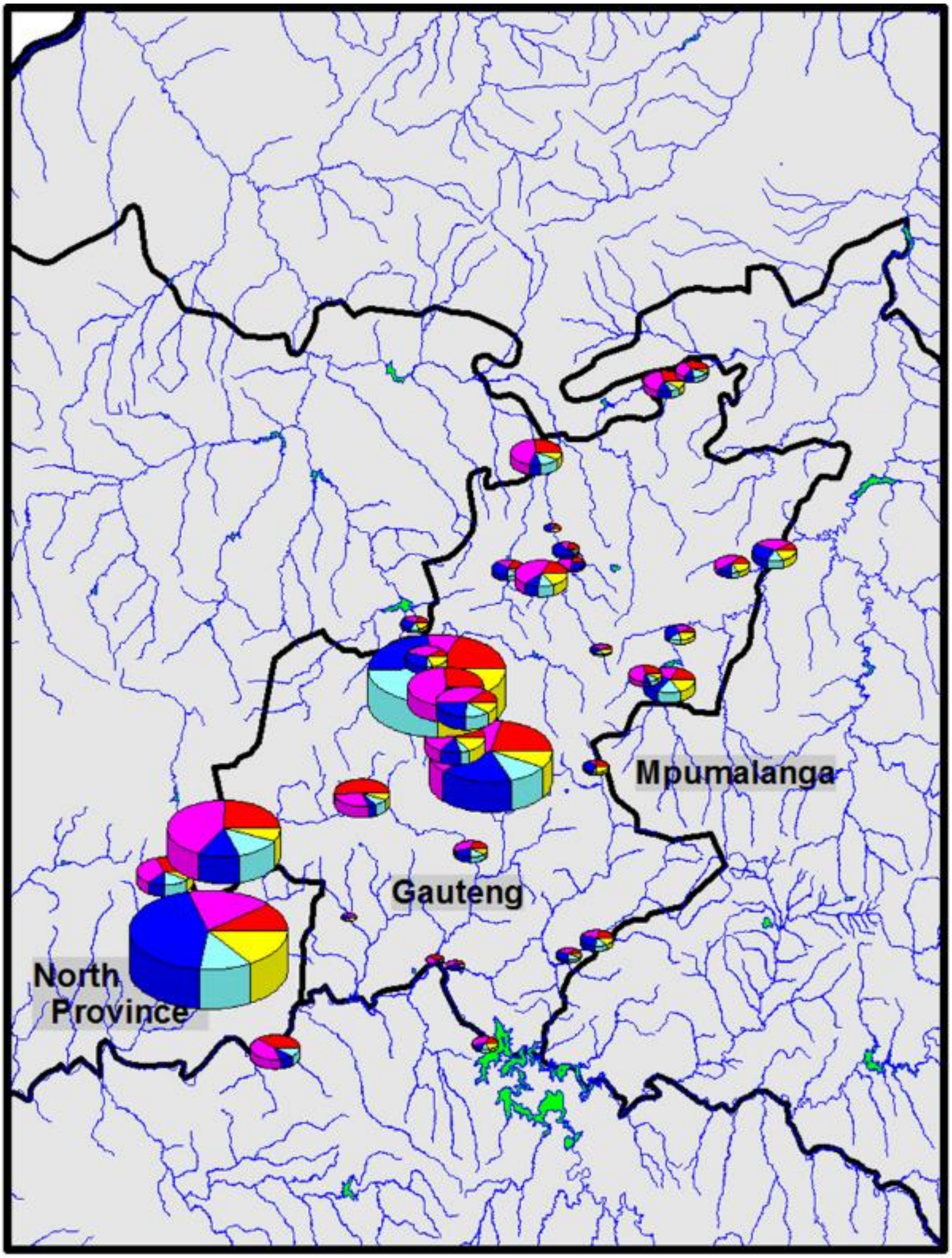


Figure 5.13: Pie charts of the size (µm) composition profiles of fibres. The height of each pie represents the number of fibres. Red = 20<300; Purple = 3001-600; Dark blue = 601-900; Light blue = 901-1200; Green = 1201-1500; Yellow = >1500.

5.3.2 Ground water results

Groundwater from four boreholes in Potchefstroom had very low MP concentrations (Table 5.5) when compared with surface water concentrations (Table 5.2). In surface waters, fragments and fibres had the same mean of~0.8 n/L. Particles in ground water was an order of magnitude lower that in surface water, but fibre concentrations were similar.

Table 5.4: Counts of fragments and fibres (n/L) of groundwater from four boreholes in Potchefstroom, according to size classes (µm).

	Plastic fragments per size class (um), per litre								Fibres, per size class, per litre							
Sample	20<300	301-600	601-900	901-1200	1201-1500	>1500	Total		20<300	301-600	601-900	901-1200	1201-1500	>1500	Total	Grand Total
52	0.044	0.022	0	0	0	0	0.067		0	0.022	0.011	0	0.011	0.022	0.067	0.133
54	0.011	0	0	0	0	0	0.011		0	0	0.033	0.011	0	0.067	0.11	0.122
55	0	0	0	0	0	0	0		0.011	0.022	0	0.022	0.033	0.033	0.12	0.122
56	0.078	0.011	0	0	0	0	0.089		0	0.022	0.044	0.022	0.044	0.067	0.2	0.289
Mean	0.033	0.008	0	0	0	0	0.042		0.0028	0.017	0.022	0.014	0.022	0.047	0.125	0.167

5.3.3 Comparison of size distributions between ground and surface water

Proportional composition between size classes of fibres and fragments (Fig. 5.14) shows that fragment profiles were distinctly skewed towards the smallest size class in both ground and surface waters, while fibres showed an opposite trend with the greatest relative proportion in the largest size class.

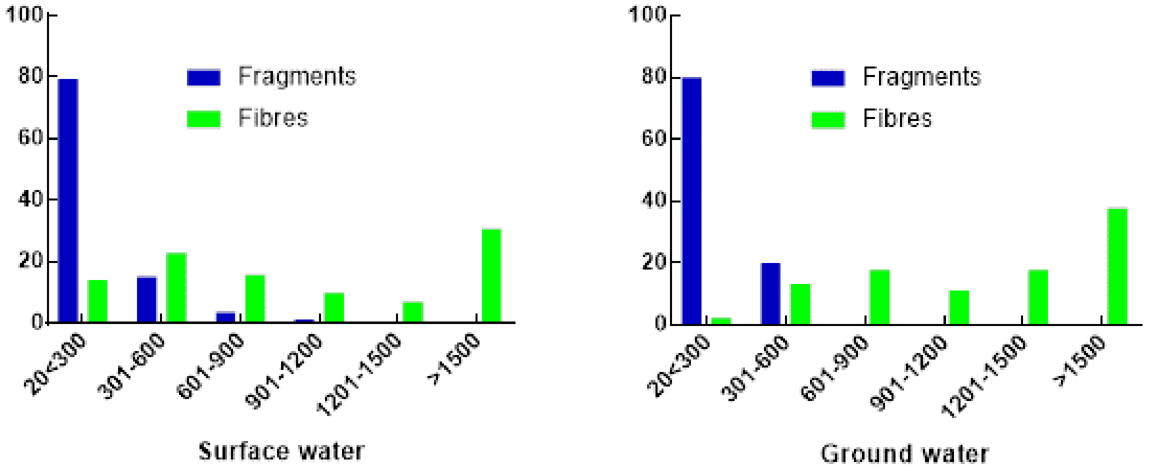


Figure 5.14: Comparisons of relative distributions (percentages) of size class composition of fragments and fibres in surface and ground water samples.

5.3.4 Tap water results

Two water samples were taken to determine a baseline for MP concentrations in South African potable water. Johannesburg water had a higher MP concentration than Pretoria, and both samples contained more fragments than fibres (Table 5.5).

Table 5.5: Counts of fragments and fibres (n/L) of tap water from Johannesburg and Pretoria, according to size classes (µm).

	Plastic fragments per size class (um), per litre								Fibres, per size class, per litre							
Sample	20<300	301-600	601-900	901-1200	1201-1500	>1500	Total		20<300	301-600	601-900	901-1200	1201-1500	>1500	Total	Grand Total
Pretoria	0.17	0.022	0	0	0	0	0.19		0.13	0.28	0.12	0.089	0.078	0.17	0.87	0.87
Johannesburg	0.13	0.28	0.12	0.089	0.078	0.167	0.87		0.10	0.36	0.28	0.26	0.11	0.70	1.8	1.8

5.4 Discussion

5.4.1 Surface water

South African surface waters contain microplastics between 0.33 and 56 n/L. Two sites in the Vaal River had very high concentrations of MPs; 56 and 39 n/L, respectively. We excluded these from most analyses, as the rest of the samples had a mean of 1.86 n/L (Tables 5.2, 5.3 and 5.4). These two sites were adjacent to one another in the Vaal River and are probably the result of some direct form of MP contamination. Other sites in the Vaal River had up to two orders of magnitude less MP in its water (Site 42 – Table 5.1). Most MPs at site 2 and 3 samples were blue and white fragments of more or less the same size range and the same breakage pattern. As this is a scoping study, the focus was not to identifying sources, but identifying a baseline and aspects for further research. We recommend that further research should be done to investigate sources of MP in this area.

Fragments had a very clear size class pattern (Figure 5.5 a). The smallest size class (20-300 µm) had four times more fragments than the other fragments size classes combined. This trend was significant (Figure 5.5b) and correspond with other findings (Bai *et al*., 2022). It is likely due to to the continuous breakdown of environmental MPs to smaller sizes (Lv *et al*., 2022). It could also be due to a significant release of small manufactured fragments in excess of larger particles, but findings from literature and the macroplastic pollution level of the South African environment concur with the former. Most MPs found, were either blue or white (including transparent), which are common colours found in plastic used for packaging.

For fibres, there was no size-class pattern discernible (Figure 3.5 c and d), although the Kruskal-Wallis analyses suggested some size-class differences. Longer fibres, conceivably, would break down into smaller particles the same as for fragments particles. The largest size class (>1500 µm) covered a greater size range than the other size classes (e.g. 301-600 µm). The distribution of fibres across size classes were therefore proportional to the range of the respective class, indicating a relatively homogenous size distribution of fragments.

Fragments and fibres might behave and distribute differently in the same aquatic medium, due to possible differences in water resistance, reaction to turbulence, specific mass, or density, and surface to volume ratios. They should also have very different sources e.g. fragments resulting from the breakdown of larger plastic pieces, and fragments deposited by atmospheric fallout. Heavier fragments might also settle out of water faster into sediments while fibres of the same mass might be prone to remain in suspension for longer. Smaller particles would be less prone to settle, dominating the smaller size-classes in water. These factors would need more investigation, including the analyses of fragments and fibres in sediments. The effect of water flow velocity at each site should also be considered in future studies. Size classes might affect impact, as smaller sizes could cross membranes more easily (Stock *et al*., 2021).

The geographic variation is also insightful. The heavily-used Crocodile River that drains most parts of Johannesburg dominated the total particle, fragment, and fibre concentrations (Figures 5.8 – 5.11). Fragments and fibres were also prominent to the west near Potchefstroom, while northern and eastern parts had noticeably lower MP concentrations. Detailed analyses should in future focus on individual streams and possible sources to inform location-specific interventions.

As has already been shown, small particles dominated at all sites (Figure 5.5). However, at Vaal Dam and towards the north, larger particles made up greater proportions. This study was also only conducted in only one sampling period. It is likely that MP concentrations at the same site would vary over time and seasons (Talbot *et al*., 2022) warranting further investigation.

At the time of publication of the report in 2018, MP research in freshwaters was in its infancy with only a few studies that had methods and units comparable with our results. As our study quantified MPs per volume of surface water and not per surface area sampled, the results of contemporary studies applying a similar sampling method are shown in Table 5.6.

Table 5.6: Comparable contemporary results from other studies (n/L).

Location	Microplastics in water (n/L)	Reference
Austrian Danube, Austria	Mean: 0.32 x10^{-3} Maximum: 5.0 x10^{-3}	Lechner *et al*., 2014
Goiana Estuary: Brazil	Maximum: 0.15 x10^{-3}	Lima *et al*., 2014
WWTP effluent: Paris, France.	Untreated waste water: 0.26 - 0.32 Effluent: 14 x 10^{-3} - 50 x 10^{-3}	Dris *et al*., 2015
Italy: Lake Bolsena and Lake Chiusi	27 x 10^{-3} 34 x 10^{-3}	Fischer *et al*., 2016
Netherlands: Amsterdam canals	Mean: 100 Max: 187	Leslie *et al.,* 2017
USA general	WWTP effluent: 0.5 ± 0.02	Mason *et al*., 2016
North America: 29 Great Lakes tributaries	Mean: 4.2 x 10^{-3} Maximum: 32 x 10^{-3}	Baldwin *et al.,* 2016
China: Lake Taihu (developed area)	3.4–26	Su *et al*., 2016
China: Three Gorges Dam	Mean: 4.1 Maximum: 12.6	Di & Wang, 2017
Yangtze Estuary	Mean: 4.1 Maximum: 10.2	Zhao *et al*., 2014
China: Lakes, Wuhan	8.9	Wang *et al.,* 2017
China: Manas River Basin	21 ± 3 – 49 ± 3	Wang *et al*., 2020
China: Jinze Reservoir Huangpu River Suzhou Creek	28.3 ± 4.1 26.2 ± 9.6 14.4 ± 5.1	Chen *et al*., 2020
Nigeria Ghana	410 – 1556 90	Chaukura *et al*., 2021
South Africa: Vaal River	1.7 ± 5.1	Weideman *et al*., 2020
South Africa: Braamfontein Spruit	0.16 – 2.08	Dahms *et al*., 2020
South Africa: Vaal River	0.61 x 10^{-3} ± 0.57 x 10^{-3}	Ramaremisa *et al*., 2022
Gauteng and North West Province	**Mean: 3.37 ± 4.42** **0.3 - 56.5**	**This study**

5.4.2 Ground water

Microplastic fragments leach from agricultural soils due to sludge applications (Kumar *et al*., 2020). Fibres and particles can be expected to behave differently in soil water then in open surface waters (Nizzetto *et al.*, 2016; Rillig *et al.*, 2017). Fibres can also remain in soil-applied sludges for 15 years and may travel horizontally and laterally along flow paths and via earthworms (Rillig *et al.*, 2017). As far as we are aware, this study was the first that quantified fibres and fragments into size classes.

The size class profiles seem similar between groundwater and surface water, albeit with much higher concentrations in the latter. The fragment proportion is also much lower in ground water compared to surface water (Tables 2.2 & 2.5). Only fragments in the two lowest size classes were found in soil water, but fibres occurred in all size classes (Fig. 3.13). The reasons for this difference between ground and surface water are not known. However, in many places, people get their prime household consumption water from groundwater. Therefore, more studies would be needed to determine the factors involved, as well as the possible health implications it may have.

5.4.3 Tap water

Concentrations of MPs found in drinking water are generally much lower than that found in environmental samples (Sharma *et al*., 2022) as water treatments plant have been shown to remove a significant portion of MPs from source water (Swanepoel *et al*., 2023). We analysed tap water from two metros with one sample each, to get an appreciation of the presence of microplastics in potable water in the area (Table 5.6). Pretoria tap water seemed to have fewer fragments (0.19 n/L) and fibres (0.87 n/L) compared with Johannesburg (0.87 and 1.8 n/L, respectively). In Johannesburg tap water, particles seem to be distributed homogenously between different size classes, while Pretoria tap water only had particles in the two smallest size classes. Although the presence of MPs in Gauteng drinking water has been confirmed, one sample in each metropolitan area is not enough to draw conclusive results from, and more research should be done in this regard.

5.5 Conclusions and recommendations

We showed the presence of substantial MP concentrations in South African freshwaters. All samples of surface water, potable tap water, and ground water contained MPs with concentration differences of up to three orders of magnitude. River water contained the highest concentrations of MP, and ground water the lowest. The developing science on the toxicological impact hereof precludes risk statements, but it allows for the first time a baseline to be set for South African inland waters.

The aim of the study was to cover a wide variety of water sources and sites to identify further research priorities. The following priorities have been identified:

1. Include sediment samples at sites to investigate the water-sediment MP distribution.
2. Sample sites over a longer periods to investigate temporal variance in MP concentrations and the impacts of seasonality.
3. Determine major sources of plastic and MP in an African and South African context.
4. Define first step interventions to curb plastic pollution.
5. Identify the factors that play a major role in the distribution of MPs in South African water.
6. Determine the contribution of longer inland rivers to the oceanic MP load.
7. The efficiency of water treatment plants and wastewater treatment plants in South Africa, as well as the fate and MP distribution of resulting sludge.

8. Expand tap- and groundwater studies to determine more realistic means and identify sources.
9. Health impact of MPs on river users and freshwater biota.

The geographical patterns seen, the differences between sites, and the differences between size classes according to fibres and fragments, show that the technique developed is robust. However, the following needs to be done to validate fully the technique.

1. Expand the use of field blanks by taking clean water and filtering on site, to determine possible procedural contamination.
2. Check for repeatability, by taking more than one sample per site, alternating the filtering of sub-samples between different containers.
3. Use standards in the extraction process to determine any possible losses and extraction efficiencies.

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Chapter 6

Spatial distributions of microplastics in the Vaal River and two of its tributaries: South Africa

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ABSTRACT

The Vaal River is the second largest but economically most important river in South Africa. High concentrations of microplastics (MPs) were found throughout the system with the highest concentrations in the heavily industrialised and densely populated Klip River tributary. Sediment per kilogram had three orders of magnitude higher MP concentrations than water per litre. Sediment in the Klip River wetlands had microplastic concentrations an order of magnitude higher than the most heavily polluted wetlands in Asia. To our knowledge, samples from the Klip and Vals rivers contain the highest concentrations of microbeads in water and river sediment, respectively, ever measured. Different MP morphotypes have different sources and hydrodynamics, with microbeads found mostly localised to point sources such as untreated wastewater. MPs do not get transported far from point sources except likely during flooding events. The ubiquity of fragments as most abundant MP morphotype attests to insufficient and inadequate waste disposal practices and infrastructure in developed areas in South Africa. MPs were mostly polypropylene. MPs in sediment were circa 22 times heavier than in water. Due to microplastic retention in sediments, the Vaal River and its tributaries may therefore not a be major contributor to marine MPs but leaves the most utilised water source in Southern Africa in a poor and probably worsening state regarding MPs, requiring interventions.

6.1 Introduction

It is well established that microplastics (MPs) are ubiquitous in the natural environment. While most MP research is done in the marine environment (Blettler *et al*., 2017), recent years have seen an increased interest in freshwaters as up to 80% of marine MPs can trace its origins to land-based sources (Li *et al*., 2016) with rivers as the main vector (Lebreton *et al*., 2019; Jambeck *et al*., 2015; Meijer *et al*., 2021).

6.1.1 Global perspective

Globally, circa 39% of people live within 100 km from the sea (Kummu *et al*., 2016), meaning most people will not come into direct contact with marine plastics except for occasional seafood ingestion. Humans are dependent upon freshwater, and although accessibility thereof has been simplified by modern urban infrastructure, 90% of the world population lives within 10 km of a freshwater body (Kummu *et al*., 2011). Almost everything humans come in contact with, including consumed goods, utilises freshwater for production and functioning. Freshwater is used to grow crops, feed livestock, supply industries, and for washing and recreational activities, to name but a few. Most plastic deposited in inland rivers do not make it to the ocean (van Emmerik *et al*., 2022) and is an important origin of plastic exposure for humans and freshwater biota, warranting greater research interest. While the effects of MPs on aquatic ecosystems are being elucidated, its effects on human health remains uncertain (Blackburn *et al*., 2022). In humans, MP exposures are *inter alia* associated with DNA damage, oxidative stress, immune responses, and metabolic disorders (Li *et al*., 2023b).

6.1.2 Africa

Africa remains the continent with the greatest proportion of mismanaged waste and might be the second largest contributor to marine plastic pollution (Lebreton *et al*., 2019). Although 70–80 % of plastic waste on the continent is recyclable, only 4% of the total is recycled (Minakshi *et al*., 2023). Africa faces many socio-economic and health problems, that, in the eyes of the population and many governments, outweigh environmental challenges (Verster *et al*., 2017). In South Africa, only 1% of rural communities have access to formal waste collection and three quarters of these households make use of their own refuse dumps (Statistics SA, 2016). Neglect of waste disposal, water treatment infrastructure, and consumer behaviour have led to waste mismanagement in South Africa on a grand scale. Annually, an estimated 440 000 metric tonnes of plastic waste are released into the South African environment due to mismanaged waste (Verster & Bouwman, 2020) placing the problem in numeric context.

The aim of this study was to better understand the sources and distributions of MPs in South African freshwaters using the Vaal River System as case study. The Vaal River is part of the greater Orange-Vaal River catchment which drains half of the country's surface water (Swanevelder, 1981). It is the life stream of the South Africa's economic hub that provides potable water, drives major industries like mining, electricity generation, agriculture and livestock production, and is used for traditional activities and recreation. The estimated economic value of the Vaal River system was about ZAR 13.3 billion at the start of the century (Mirrilees *et al*., 2003). Increasing anthropogenic activities and river use subjects this system to increasing pollution levels (Gyedu-Ababio *et al*., 2004). The first MP data from the Vaal River found concentrations comparable with that of major European rivers (Bouwman *et al*., 2018). Puzzlingly high MP concentrations with high variability between sites informed this more detailed investigation into the possible sources and distribution trends of MPs in the Vaal River system.

6.2 Methods

6.2.1 Study sites

The Vaal River Catchment drains the most economically active and densely populated region of South Africa (Figure 6.1). Twenty sampling sites were selected based on accessibility, safety, and position above and below possible point sources in the Klip River (n = 8) and Vals River (n = 7) - both tributaries of the Vaal River, as well as the mainstream Vaal River (n = 5) (Figure 6.1).

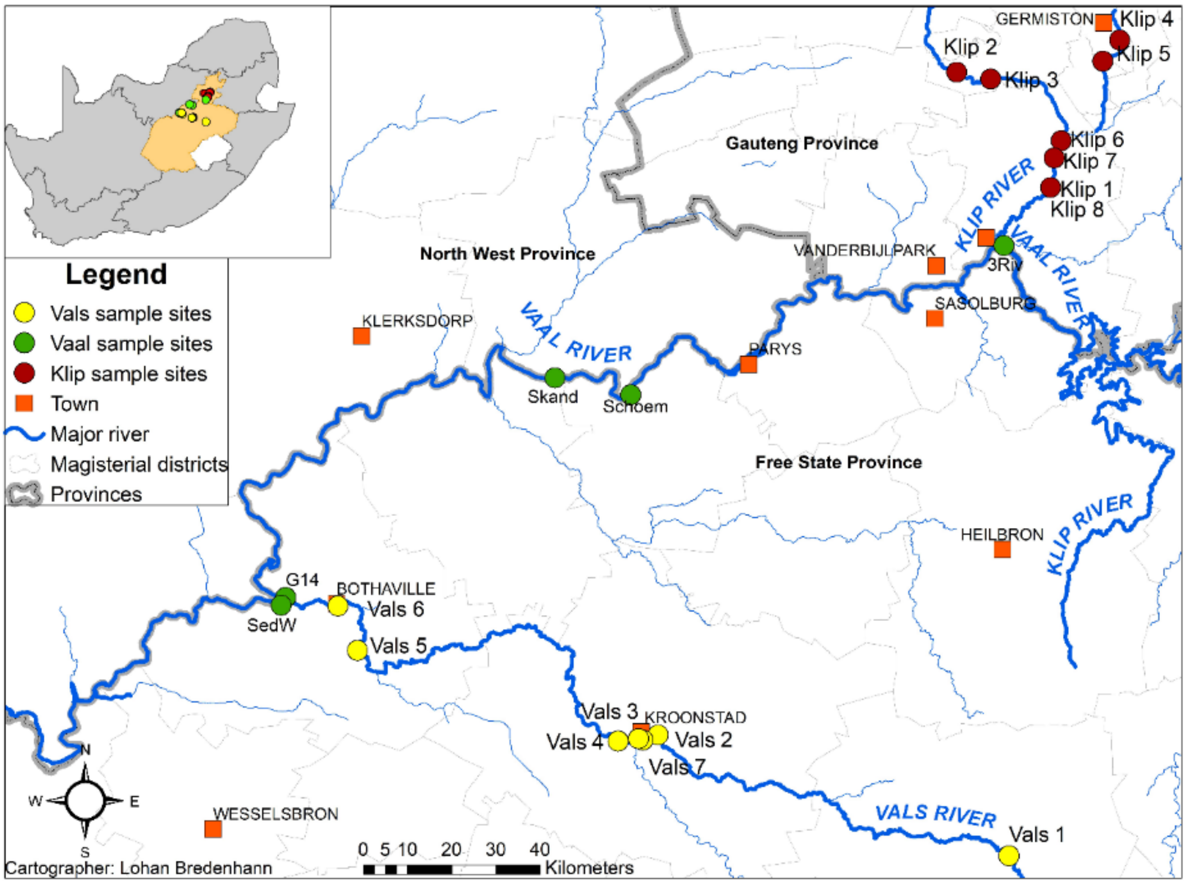


Figure 6.1: Location of sample sites in Klip River (red), Vals River (yellow) and mainstream Vaal River (green)

Sampling took place from May 2019 to March 2020 at two-month intervals. Sampling campaigns include May 2019, July 2019, October 2019, November 2019 (during first rains of the season), January 2020, and March 2020. Not all sites were sampled during each sampling episode due to logistical and COVID reasons, but data for sampling months with fewer samples will still be included in findings.

6.2.2 Sampling

Surface water samples were taken from the main flow channel of each river by wading where possible, or from infrastructure extending into or over the main flow channel. Sediment samples were taken at the water's edge. Bulk water sampling was done with a stainless-steel bucket to filter 90 litres (six replicates of 15 litres) of surface water through a 25 µm mesh stainless steel sieve, on site. Residue was rinsed into a Schott bottle pre-cleaned with pre-filtered water. The opening was covered with aluminium foil before closing the bottle cap. Sediment was removed from water's edge using a steel shovel and covered with rinsed aluminium foil. Basic water quality parameters (temperature, total dissolved solids (TDS), dissolved oxygen (DO), pH, and in-stream flowrate was measured in situ. A count of all visible plastic items 50 m upstream and downstream from the sampling site on both sides of the riverbank was made and noted. Sixty-seven samples in all were collected and analysed.

6.2.3 Sample processing

Filtered water residue and sediment was taken to the lab and dried at 50°C while lightly covered with foil to prevent contamination from atmospheric fallout. A 50 g dry mass subsample of sediment was taken. Fifty grams was the best sample size we determined in a prior method-optimising scoping study for the available equipment with optimum results in removing MPs (Bouwman *et al*., 2018). Dried water and sediment samples were separately subjected to temperature-regulated Fenton digestion (Masura *et al*., 2015). Density separation was done with a 0.5 M sodium iodide solution (1.05 g/ml). After separation, settled material was visually inspected under a light microscope for possible residual MPs as PVC and PET have higher densities than the solution and some samples with fine material trapped obvious MPs in settled material. These MPs were manually removed and placed on the sieve. If deemed necessary due to incomplete separation, another round of density separation was carried out and floating plastic was filtered onto the same sieve. Stainless steel sieves used for in situ filtration as well as filtering floated material during density separation had a 25 µm mesh size, making 25 µm the lower size limit for this study, although smaller particles might be trapped on top of larger.

All equipment and materials in contact with samples during sampling and lab processing were triple rinsed with pre-filtered, double distilled water. Samples were covered with rinsed aluminium foil or glass watch glasses throughout processing to avoid contamination by atmospheric fallout. Procedural blanks were also run alongside each batch of samples being processed.

6.2.4 Quantification, characterisation, and measurement

Samples were dried and visually inspected on a Nikon AZ100M microscope using 300x magnification. MPs were counted and the longest dimension measured with Nikon AZ100 imaging software/NIS Elements-D imaging software. All MPs were noted in a matrix according to morphotype (fragment, fibre, and bead), size groups (25 – 300 µm, 301 – 600 µm, 601 – 900 µm, 901- 1200 µm, 1201 – 1500 µm and > 1500 µm), and colour (blue, black, red, green, yellow/brown, white/transparent, and purple).

The identification of microbeads was relatively simple as very few natural objects have a distinct solid, spherical appearance; most plastic beads having standard sizes e.g., 50 µm or 250 µm. Inorganic fibres are also relatively easy to identify as most natural fibres have cellular structures or started showing signs of breakdown due to aggressive digestion during sample clean-up. Fragments identification was the most complex. In this study, fragments were classified as irregularly shaped MPs caused by the breakdown of larger plastics and include films. Visual criteria for fragments excluded those particles with cellular or organic structure or metallic lustre which may be clay particles. Colour, breakage patterns and crystalline structure was also considered.

Procedural blank mean counts were subtracted as background per batch from fragments and fibres of each sample respectively, to account for method contamination. Blanks had no beads. Counts were converted to n/L in water samples and n/kg dry mass in sediment samples. For certain purposes, where visual comparisons were needed, the order-differences between water (n/L) and sediment n/kg dry weight) we chose to convert the sediment concentrations to n/g. This conversion did not affect proportional analyses.

6.2.4.1 Polymer determination

A selection of samples was sent for focal plane array-micro-Fourier transform infrared (FPA-μFTIR) analysis to Aalborg University, Denmark, for polymer determination according to Maurizi *et al*. (2023). Samples were resuspended in a known volume of pure ethanol and an aliquot of the sample was deposited on a zinc selenide (ZnSe) transmissive window. The analyses were done in transmission mode using an FPA–μFTIR (Cary 620 FTIR microscope, integrated with a Cary 670 IR spectroscope; Agilent Technologies, USA). A background scan was done beforehand for background correction. The result was a chemical image of the sample area where each pixel held a corrected IR spectrum. Images were analysed using the siMPle software v. 1.3.1β (Primpke *et al*., 2020). Dimensions and mass of MPs were generated. Due to sample leakage during transport, FPA-μFTIR results were used only to for relative (%) polymer composition, and not for concentration determination.

6.2.5 Statistical analyses

Statistical analysis was done using GraphPad Prism 10 software (www.grahpad.com). Descriptive statistics, linear regressions of log transformed concentrations with meta data, and Spearman correlations of concentrations were conducted for various data groupings (per month, per river, wet and dry season). One Way ANOVAs were conducted for mean MP concentration between sites in water and sediment, respectively, in each river to elucidate variance between sites in a river. Chi-square tests were used to determine proportional differences between morphotypes in the three rivers, as well as proportional distribution of MPs between matrices.

PCORD 7 was used to perform non-metric multidimensional scaling (NMS) analysis on relativised data of various groupings according to matrix (water or sediment), site, month, river (Klip, Vals and Vaal), flow (laminar or turbulent), surrounding land use, proximity to formal residential areas, informal settlements, industries, agricultural activities, WWTPs and dams or weirs. Non-metric Multidimensional Scaling (NMS) was conducted using data relativised per sample for logical data groupings using PCORD 7.08 software. This was done to identify major relationships between variables and data. Gower-ignore-zero was used as distance measure, starting from random coordinates. Five initial dimensions was allowed, with 250 runs of real and randomised data. NMS plots indicating strong associations between variables were retained and used to guide further interpretation.

6.3 Results

6.3.1 General results

All sediment and water samples at every site during all seasons (n = 134) contained MPs. The mean total MP concentration in water for this study (n = 67) was 3.4 ± 4.4 n/L. The river with the highest mean MP concentration in water was the Klip River. The Vals River contained almost half of the overall mean, and the mainstream Vaal River had the lowest mean MP concentration of the three rivers (Table 6.1).

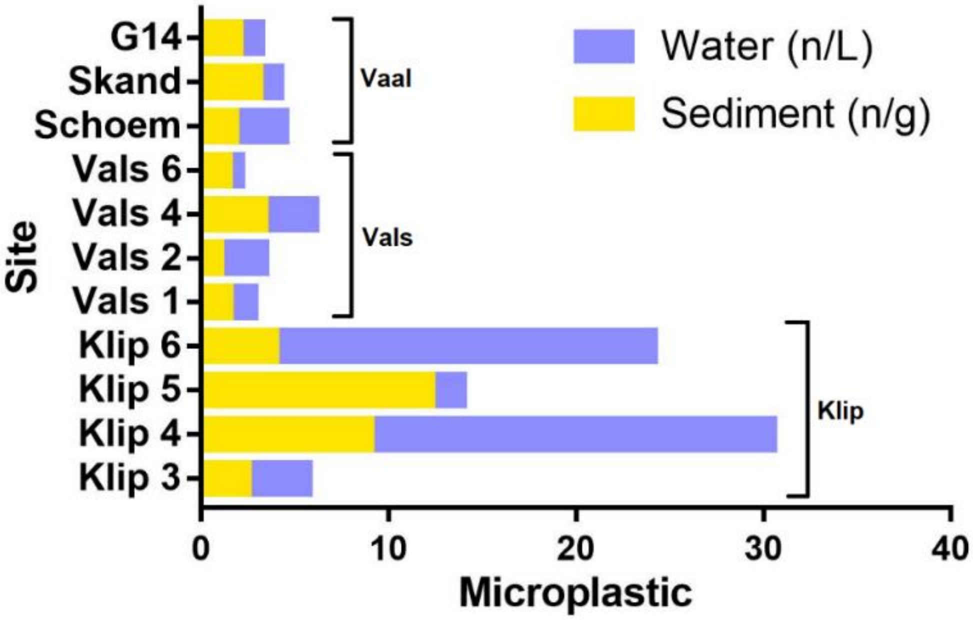
Table 6.1: Summary mean microplastic concentration, standard deviations and percentage coefficient of variations in water and sediment for all 67 sites and %CV overall and per river.

		Water (n/L)		Sediment (n/kg dry mass)	
River	**Sample size**	**Mean & SD**	**%CV**	**Mean**	**%CV**
All rivers	67	3.4 ± 4.4	146	3600 ± 4500	89
Klip River	24	6.1 ± 5.8	112	6200 ± 6400	70
Vals River	24	1.9 ± 2.8	42	1900 ± 2400	76
Vaal River	17	1.6 ± 1.6	55	2600 ± 1600	26

The MP concentrations in sediment were three orders of magnitude greater per kilogram dry mass than per litre of water (Table 6.1). Mean MP concentrations in sediment samples of the three rivers followed a similar trend per kilogram dry mass: the Klip River had the highest mean sediment concentration. The Vals River and mainstream Vaal River sediment samples contained about a quarter to a third of the MPs found in Klip River sediment. The greatest coefficient of variation (%CV) was in sediment samples from the Vals River (357%) and the least variation was in mainstream Vaal River sediment (63%).

The lowest concentration in water was seen during July in the mainstream Vaal River, ranging between 0.02 – 0.7 n/L. Vals 2 consistently had the lowest MP concentration in sediment, dropping to only 40 n/kg during March, at the end of the rainy season.

Figure 6.2: Mean MP concentrations per matrix across all months for sites with sufficient (n>2) water and sediment data (sediment data converted from n/kg to n/g for visual comparison with water).



One-way ANOVA tests for MP concentrations in water and sediment, respectively, in each river were conducted. There was no statistically significant difference between sites in the mainstream Vaal River (p = 0.42), and Vals River sites had no significant differences in terms of water MP concentrations (p = 0.61). The greatest variation was seen between water sample means in the Klip River (p = 0.15).

Figure 6.2 accounts for plastic distribution between water and sediment per site, expressed in particles per gram of dry mass sediment (n/g dry weight), while water data was expressed as particles per litre (n/L). A comparative volume of sediment generally had three orders of magnitude higher MP concentration than the corresponding litre volume so only the relative compositions should be noted. Again, the greatest proportional difference was seen in the Klip River, especially Klip 4 and Klip 5 (Figure 6.2). This reflects the results of the ANOVA analyses above.

6.3.2 Spatial distribution

The Klip River had the greatest differences in MP concentrations between sites and no consistent pattern of spatial distribution (Figure 6.3). The highest concentrations of MP in water were in the upper reaches of the eastern branch of the river (Klip 4) and after the confluence of the two rivers (Klip 6). The highest mean MP concentration in sediment was in the wetland site (Klip 5) below the heavily polluted Klip 4.

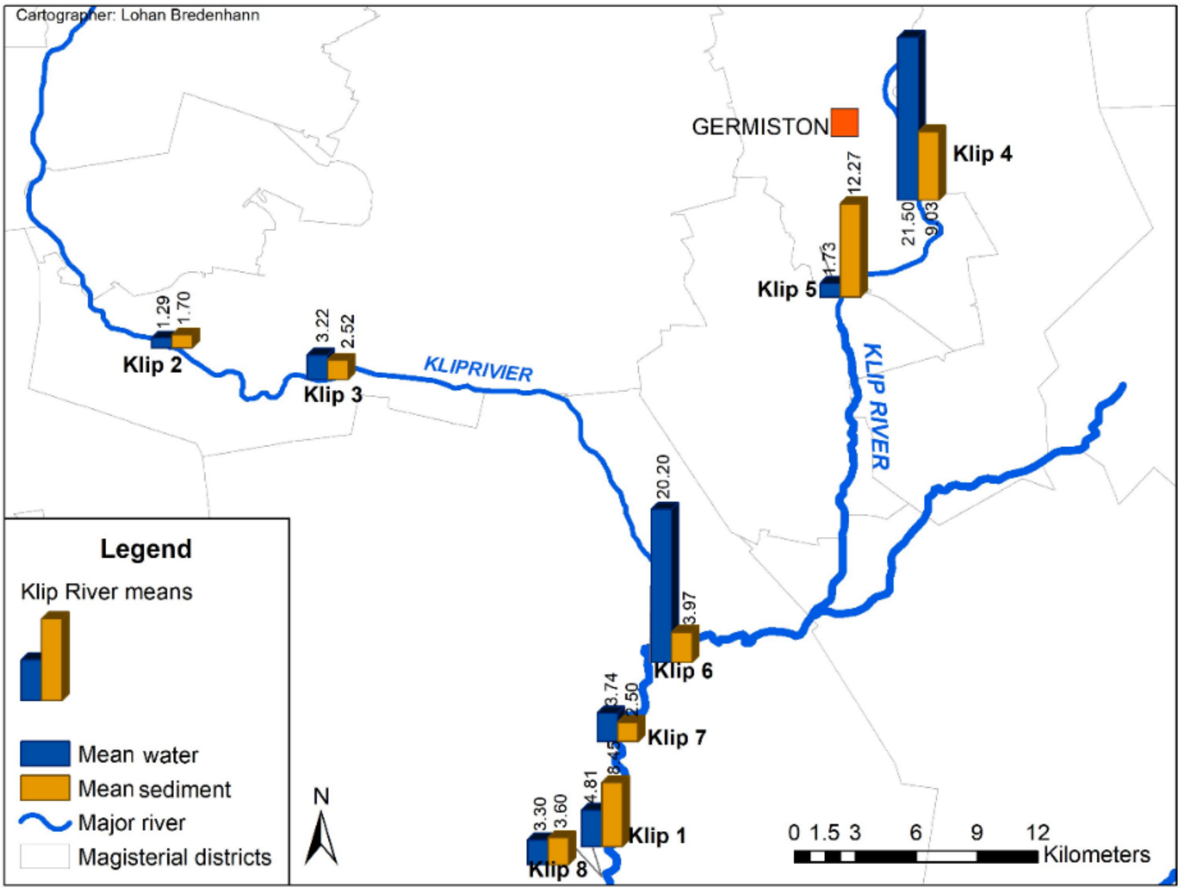


Figure 6.3: Mean microplastic concentrations in surface water (n/L) and sediment (n/g) of the Klip River.

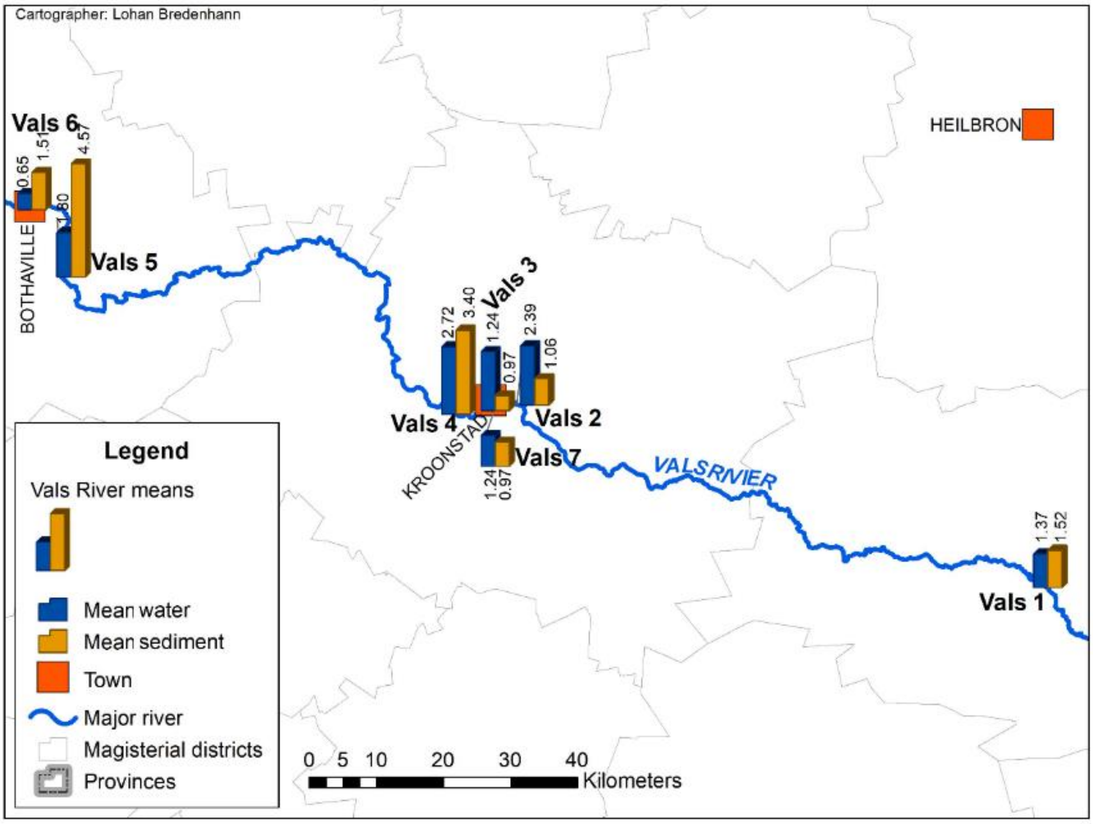


Figure 6.4: Mean microplastic concentrations in surface water (n/L) and sediment (n/g) of the Vals River.

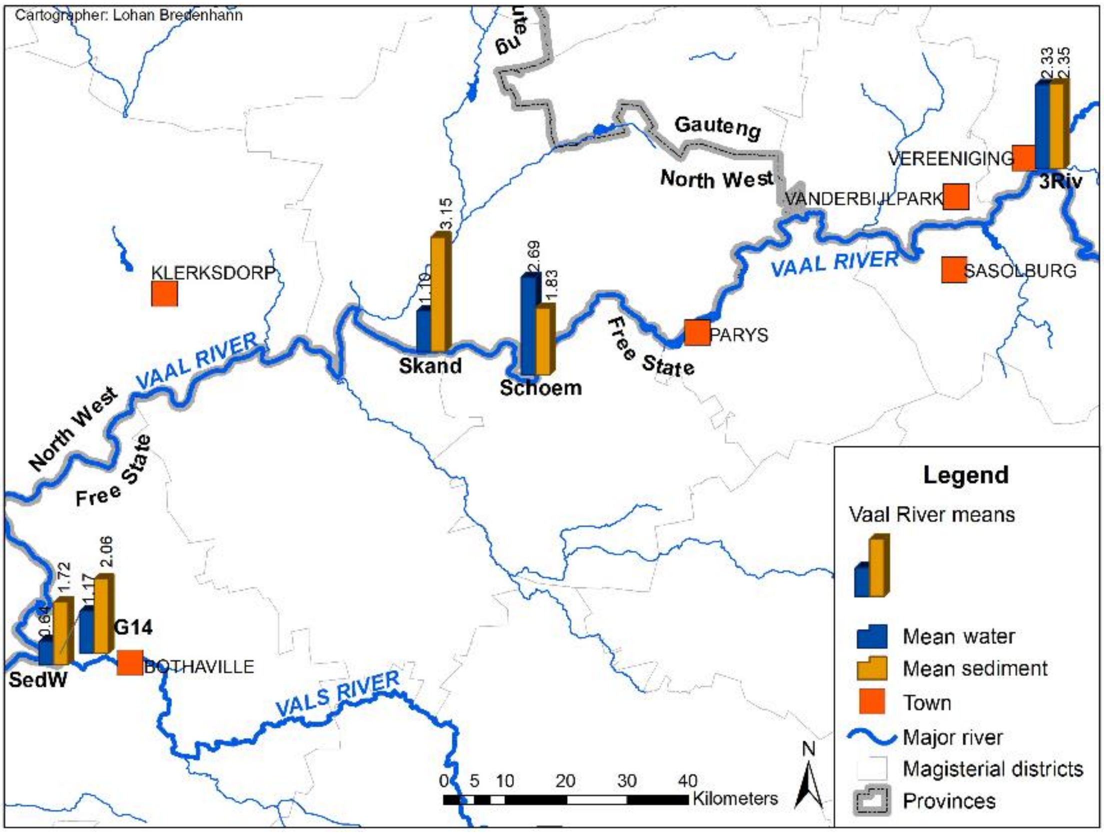


Figure 6.5: Mean microplastic concentrations in surface water (n/L) and sediment (n/g) of the Vaal River.

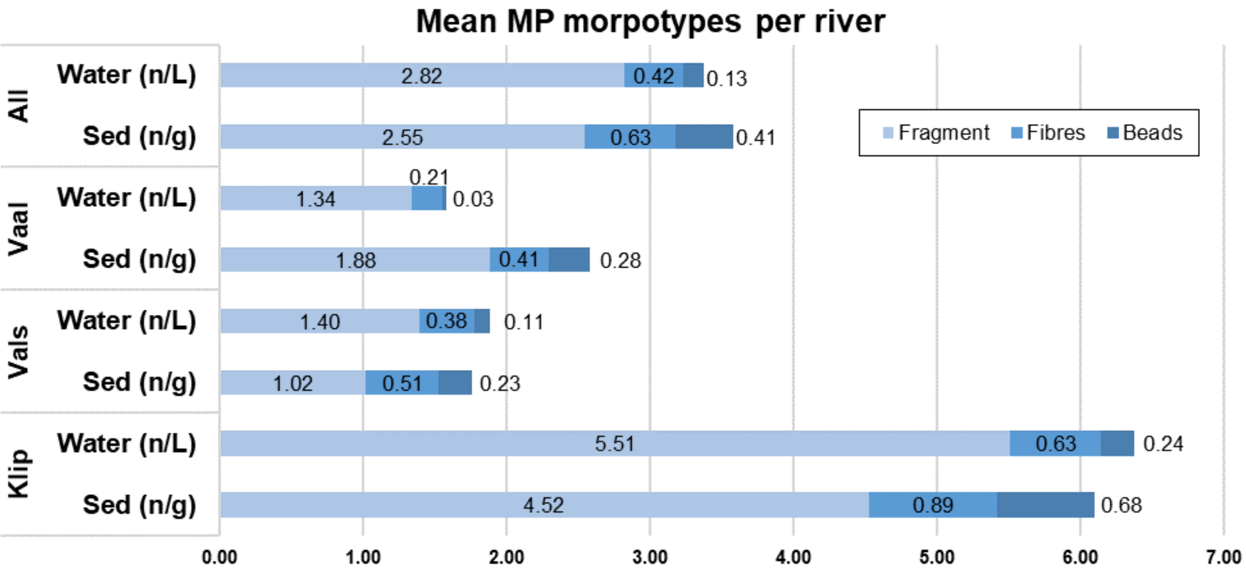
The Vals River is much less modified than the Klip River, with agriculture as primary land use, with and only two substantial towns—Kroonstad and Bothaville—along its course in the studied stretch of the river (Figure 6.4). Samples were mostly taken near the towns with the exception of sites Vals 1 and 5. MP concentrations in water from the Vals River showed less variation (%CV = 42%) between sites than corresponding sediment samples (%CV = 76%) (Table 6.1).

MP concentrations in the Vaal River were more consistent between sites compared with the tributaries (Figure 6.5). Sediment MP concentrations from the mainstream Vaal River were relatively consistent along the course of the river, and sediment concentrations were comparatively higher than its corresponding water samples when compared with the other two rivers (Figures 6.2 - 6.5).

6.3.3 Morphotype composition

Fragments were the most abundant morphotype, followed by fibres and then microbeads (Figure 6.6). Beads made up a notably larger proportion of MP in sediment (11%) than it did in surface water samples (3.9%). No nurdles or other forms of primary MPs were found. A complete results table is attached in supplementary data.

Figure 0.6: Mean microplastic morphotype compositions per river (sediment data converted from n/kg to n/g for visual comparison with water).



All Chi-square analyses between the Klip, Vals, and Vaal Rivers of proportions of the three morphotypes (fibres, fragments, and microbeads), as well as between sample matrices per river, yielded significant results (p<0.0001). Therefore, the proportional compositions of morphotypes differed between water and sediment within each river, and between waters and sediments between each river. Each river, therefore, had a unique morphotype composition in waters and sediments.

6.3.4 Factors that affect MP distribution

Various metadata parameters were used during sample analysis to detect factors that might affect plastic loadings at a site. Linear regressions with metadata as independent variable and log-transformed MP concentrations as dependent variable were done for various data groupings – all data, each sampling episode (month), and each river (Klip, Vals and Vaal River) respectively. Parameters considered in linear regression analysis were pH, water temperature, TDS, DO, and sediment particle size results (expressed as % silt + clay). Water temperature showed no significant relationships with plastic concentrations. TDS, pH, and % silt + clay yielded sporadic significant relationships; 7 out of 218 (3.4%) regressions run had a $P < 0.05$. It should be noted that with so many regressions, 5% (11 out of 218) might be spurious at $p<0.05$).

	Water Frag	Water Fib	Water Bead	Water Total	Sed Frag	Sed Fib	Sed Bead	Sed Total
All months	-0.48 p = 0.0007	none	none	-0.28 p = 0.0008	2.87 p = 0.018	2.05 p = 0.0003	2.02 p = 0.01	2.94 p = 0.0003
July	none	none	none	none	none	none	1.80 p = 0.0282	none
January	none	none	none	none	0.37 p = 0.03	0.59 p = 0.002	0.46 p = 0.03	0.42 p = 0.006
May	-0.79 p = 0.008	none	none	-0.57 p = 0.06	none	none	none	none
October	none	none	none	none	none	none	none	none
Klip	none	none	none	none	none	1.53 p = 0.0009	1.94 p = 0.02	3.17 p = 0.02
Vals	none	none	none	none	none	none	none	none
Vaal	none	none	none	none	none	none	none	none

Table 6.2 Significant linear regressions of various concentrations with macroplastics observed per site expressed as slope and p-value. Positive slopes are green and negative in orange.

Despite this qualification, DO and Klip River data for January (high-flow) did show some significant associations with plastic concentrations. DO yielded 4/8 (50%) significant relationships for January fragment data ($p < 0.001$; slope = -2.43), fibres ($p = 0.01$; slope = -1.96), and total surface water concentrations ($p = 0.001$; slope = -2.04), as well as fibres in sediment ($p = 0.03$; slope = -1.92). In Klip River samples, fibres in water ($p = 0.03$; slope = -1.51) and sediment data ($p = 0.049$; slope = -1.92) respectively had a significant negative relationship with DO. Flow rate at a site also yielded significant positive associations with concentrations but this will be discussed in Chapter 7 as part of temporal changes.

The amount of macroplastics observed at site surrounding was the parameter that had the most significant association with MP concentrations in water and sediment which is shown in Table 6.2. More significant regressions were found between macroplastics and sediment MP concentrations than that of water MP concentrations (Table 6.2). Klip River sediment MPs was most consistent with the number of macroplastics surrounding the site (Table 6.2)

Spearman correlations were carried out between various MP concentrations (fibres, fragments, beads, and total in water and sediments, respectively). October was the only month in which there was a significant correlation between water and sediment concentrations which were correlations between beads in water and total sediment MPs ($r^2 = 0.66$; $p = 0.04$). The only other significant correlation between water and sediment data was seen in data from the mainstream Vaal River between water and sediment microbead concentrations (r^2 = 0.99; p = 0.008).

6.3.5 Polymer composition

MPs in a water sample (n=666), and 265 MPs from sediment were analysed by FPA–μFTIR (Table 6.3) for polymer composition, size range, and mass (Table 6.3). Polyethylene made up the bulk of both samples, with heavier polypropylene and polystyrene present in sediment but not water. The MP size range were much smaller in water than in sediment, and mean mass of MPs in sediment were 22 times greater.

Table 6.3: Polymer composition and parameters of microplastics in water and sediment

Polymer	% in water (n = **666**)	% in sediment (n = **265**)
Polyethylene	98	68
Polyester	1	4
Polyurethane	1	0
Polypropylene	0	17
Polystyrene	0	11
Size range % (μm)	(>25=9) (25-50=69) (50-100=22)	(>25=11) (25-50=25) (50-100=28) (100-300=28) (300-600=8)
Total/mean mass (ng)	3866 / 5.8	34489 / 130

6.3.6 Water purification by a wetland

We saw a significant increase in MP concentrations in water from Klip 4 to Klip 5, and a corresponding increase in sediment MPs between the two sites. Figure 6.7a shows how MP concentrations between the matrices at these two sites changed over the course of the sampling period. Klip 4 drains the most heavily polluted site of the study judged by visible macroplastics during sampling, and Klip 5 is in the wetland directly downstream from Klip 4. Figure 6.7 b shows the percentage of MPs in water from the upstream site, Klip 4, being retained in wetland sediment of Klip 5. In January, almost all MPs from the inflowing water was retained by the sediment.

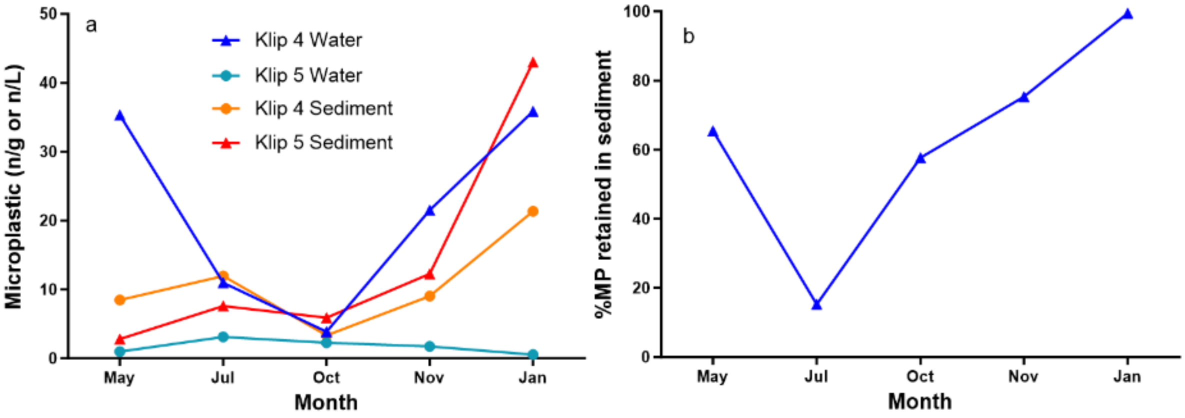


Figure 6.7: Microplastic concentrations in water and sediment at Klip 4 (above wetland) and Klip 5 (inside wetland). A - temporal variation. B - Proportion of microplastics retained in sediment at Klip 5 from water at Klip 4.

6.4 Discussion

6.4.1 MPs as indicator of river condition

The environmental MP concentrations at a site in this study were largely determined by the amount of plastic pollution that directly surrounded the site (Table 6.2). Beter general water quality (as DO) was also associated with lower MP concentrations. High MP concentrations were therefore an indication of poor river health at a site. The less polluted catchment of the Vals River (McCarthy *et al*., 2007) correspondingly had lower mean MP concentrations in water and sediment than the heavily industrialised and populated Klip River catchment (Table 6.1; Figure 6.2). Catchment use and anthropogenic activity affect the MP concentrations in a river (Townsend *et al*., 2019; Chen *et al*., 2020).

6.4.2 Sources

Polymer composition of plastics generally correspond to household polymer use. The most common polymer in samples were PE (Table 6.3). µFTIR does not distinguish between HDPE and LDPE and groups them together as PE, as infrared spectra of these two polymers are virtually identical. HDPE and LDPE (PE) are commonly found in hard and flexible single-use packaging, accounting for 35% of the plastic used in South Africa (Plastics SA, 2022). Microplastic composition of samples analysed lacked the widely used polymers PET and PP. This could be ascribed to leakage during transport or to the success of increased recycling efforts of these polymers in recent years (Plastics SA, 2019).

There are two factors known to affect the distribution of MPs within a river—hydrodynamics and surrounding land use—the latter known to be more indicative of MP pollution (Li *et al*., 2023a; Vianello *et al*., 2013). The three morphotypes in this study portrays vastly different behaviour indicating different sources and hydrodynamic interactions. Deviating vectors in NMS plots (supplementary data) and variability between nearby sites reinforce this finding. Literature suggests that there is variation in the dominant microplastic morphotype found in different studies. Many studies suggest fibres to be the dominant morphotype, this being especially true for water samples (Weideman *et al*., 2020; Bai *et al*., 2022), while other studies found fragments (resulting from breakdown of larger plastics) as the dominant morphotype, especially in sediment samples (Vianello *et al*., 2013; Nel *et al*., 2015; Naidoo *et al*., 2019; Badylak *et al*., 2021). In this study, for all samples, fragments were the most prevalent morphotype. Fibres were second and beads made up the smallest proportion of total MP at most sites (Figure 6.6).

6.4.2.1 Fragments – the breakdown of macrodebris

Fragments had the highest abundances most likely because of its abundant source in the study area—larger plastic near the rivers that decay to fragments. Macroplastics were visible along riverbanks at all sites during all sampling periods. The number of macroplastics observed at a site during sampling varied greatly between rivers but corresponded with MP concentrations in water and sediment (linear reg $p_{water} = 0.0008$, $r^2 = 0.17$; $p_{sed} = 0.0003$, $r^2 = 0.2$). Mean observed macroplastic was highest along the heavily polluted Klip River (mean 146 ± 188 items per site), then the Vals River (mean 34 ± 51 items per site) and lowest along the mainstream Vaal River (16 ± 22 items per site). These plastics get broken down in situ and can account for high prevalence of fragments at the sites.

The high prevalence of illegal dumping in the country is ascribed to inadequate formal waste management and lacking consumer awareness and education (Verster *et al.,* 2020). Informal and illegal dumping of household waste was observed along the rivers at many sites during sampling – Klip 3, 4, 6, and 7. Vals 3, 4, 5, 6, and 7 all had illegal dumping sites within 100 m from the riverbank. An estimated 1.4 million tonnes of plastic waste are produced by South Africans annually (Rodseth *et al*., 2020). Conservative estimates indicate that 440 000 tonnes of this ends up in the South African natural environment each year (Verster *et al.,* 2020). This is likely to end up in natural soils and waterways, as less than 10% of this will reach the ocean (Verster *et al.,* 2020)

Of the surface water MP morphotypes, only fragments showed a significant positive linear association ($P = 0.0007$; slope = 0.39 ± 0.11) with observed macroplastics. Fibres ($P = 0.057$) and beads ($P = 0.115$) in the water likely have different sources than the breakdown of these debris.

6.4.2.2 Beads – Untreated wastewater

Although all samples had MPs, the three morphotypes were not always detected in all samples. Sediment samples had proportionately more microbeads than in corresponding water samples. This is likely due to the fast settling of beads compared to other morphotypes confirming that riverine sediment act as an accumulating medium for primary MPs. NMS results (supplementary data) suggests a strong association downstream from WWTP effluent input with microbeads. Various studies indicate varying removal efficiencies of microbeads at WWTPs (Gao *et al*., 2023). South African WWTPs are in a notoriously poor state with 98% of WWTPs in ‘poor’ of ‘critical’ state (DWS, 2022). There are also no restrictions on the use of microbeads in South African cosmetic products, which corroborates the high prevalence of beads where wastewater enters a river. The Klip River had the most beads in surface water, but unlike with fragments, was followed by the Vals river and then the mainstream Vaal River (Figure 6.6).

There were only two significant Spearman correlations between water and sediment concentrations (Table 6.2) i.e., water and sediment bead concentrations only in the mainstream Vaal River, and beads in water correlating with total sediment concentrations only in October. In both cases, it was the microbead component of water MPs correlating with sediment data. This reinforces the notion that microbeads settle out of the water column first and was only found in the water column close to direct sources like untreated wastewater or WWTP effluent inputs.

Previous sampling of the Vaal River and lower order rivers in the Gauteng area yielded no microbeads in water sampled with the same methods (Bouwman *et al*., 2018) (Chapter 5). This indicated either an increase in the amount of microbeads released into South African waterways in the three years between the two sampling campaigns, or the fact that prior samples were taken in flood conditions that had beads already flushed away and deposited in sediment. No sediment samples were taken during 2017 sampling (Bouwman *et al*., 2018).

6.4.3 How consistent are MP concentrations in a river?

Samples from both the mainstream Vaal River and the Vals River catchments had relatively consistent concentrations compared to the great differences in MP concentrations between sites of the Klip River (Figure 6.2, and compare the %CVs in Table 6.1). In the Vaal and Vals Rivers, the proportion of MPs in water to that in sediment at a site were more consistent, but the Klip River had much more variance between proportions of MP in water and sediment (ANOVA results; Figure 6.2).

All data groupings displayed significant correlations (Spearman correlations, $p < 0.05$) between the various morphotype concentrations in water and sediment, respectively. This suggests that, even though the three morphotypes had different sources (NMS figures – Supp. data), they occurred in constant relative proportions to one another in the respective matrices. These relative proportions of morphotypes between the three rivers were, however, different (Chi-square results). Findings from one river therefore cannot be extrapolated to another. This emphasises that each river must be assessed individually for interventions.

6.4.4 Downstream transport and local hydrodynamics

The MP load at a site was related to the proximity to sources such as untreated industrial and household effluent, nearby settlements due to waste mismanagement, and wastewater treatment plant effluents. Where water MP concentrations gave an indication of the immediate conditions of MP contamination at a site, the corresponding sediment sample most likely provides a longer-term picture of the cumulative MP load of the area – probably since the last flooding event (Hurley *et al*., 2018; Bai *et al*., 2022). Observed macroplastics is also an indication of cumulative pollution at the site, explaining the strong positive linear association between macroplastics and sediment fragments ($p = 0.019$; slope = 0.21), sediment fibres ($p = 0.0003$; slope = 0.37), and sediment microbeads ($p = 0.010$; slope = 0.26).

6.4.4.1 Microplastic dynamics in the Klip River

The presence of wetlands and local hydrodynamics also affect whether MPs will flow downstream or be deposited. For example, Klip 2 lies within a wetland and is one of the cleanest sites in the entire study area although located in the heart of a heavily industrialised and highly populated area (Figure 6.3). Klip 3, downstream of Klip 2, however, is situated directly below both the Olifantsvlei and the overloaded Bushkoppies WWTP discharge points, illustrating the impact of WWTP effluents on the riverine MP load (Figure 6.3).

Sites Klip 4 and Klip 5 (Figure 6.3) was a case study showing the water purification effect of wetlands in terms of plastic (Figure 6.7). Wetlands have long been known to remove organic and inorganic pollutants from water, thereby improving water quality (McCarthy *et al*., 2007). The wetland complex of the upper Klip River receives acid mine water, urban runoff, and industrial wastewater, and plays a critical role in the purification of water entering the Vaal River (McCarthy *et al*., 2007). Urban runoff has been shown to contain more than 8000 n/L MPs, which will significantly increase riverine MP concentrations (Wang *et al*., 2022).

The quality of water received in this section of the Klip River was reflected in elevated MP concentrations in surface water at Klip 4 and 6 (Figures 6.2 & 6.3). The largest component of plastic at Klip 4 and 6 was seen in water, while Klip 5, in-between these two sites, had the majority of its MP settled in sediment (Figure 6.2). Klip 4 in the upper reaches, draining the heavily industrialised Germiston, had the highest mean MP concentration (Figure 6.2 & 6.3). It had an especially high MP concentration during the high flow season – 39 n/L (mean = 22)—likely due to increased runoff. This site also accounted for the highest concentration of beads in surface water during May with 11 beads/L. River flow at this site was turbulent with a mean flow rate of 0.55 m/s. Heavier plastics settle out into sediment at the beginning of the dry season (Jul) but most stayed in suspension (Figure 6.7a).

Klip 5 in the wetland between polluted Klip 4 and 6, experienced at least an order of magnitude decrease in water MP concentrations. Here, up to 100% of MPs were trapped in wetland sediment (Figure 6.7b). Klip 5 had the highest total MP concentration in sediment in the entire study (42 984 n/kg) during January. The concentration of microbeads in sediment only (6120 beads/kg) in July was very high, second only to the wastewater-fed Vals 4 site with 6760 beads/kg.

The heavily polluted Klip 4 had the highest prevalence of microbeads in the study area To the best of our knowledge, we have detected the highest concentrations of microbeads ever found in both water (11 beads/L) and sediment (6760 beads/kg), meaning microbeads accounted for almost 0.2% of the dry mass of river sediment by our conservative estimates (Table 6.4).

Table 6.4: Comparison of maximum microbead concentrations in different studies.

Study	Catchment	Water (beads/L)	Sediment (beads/kg)
Mani *et al*. (2015)	Rhine	0.008	no data
Castenada *et al*. (2014)	St Louis	no data	3980
This study	Vaal	11	6760

The phenomenon of wetland deposition of MPs in the Klip River varied seasonally as the MP influx due to runoff was increased during the high flow season. Water inside the wetland (Klip 5) consistently had low MP concentrations throughout the sampling period (Figure 6.3). The trend of sediment MP in the wetland (Klip 5) corresponded with that of the received water (Klip 4) (Figure 6.7, because of conditions conducive to settling inside the wetland, except for May at the end of the rainy season (Figure 6.7). This is likely due to a gradual flushing of MPs further downstream through the wetland during periods of increased flow, as was found by others (Hurley *et al*., 2018; Treilles *et al*., 2022).

6.4.4.2 Microplastic dynamics in the Vals River

At Vals 1, in the upper reaches of the Vals River, the river flows through land used mostly for agriculture (Nealer, 2014). Vals 1 was relatively clean (Figure 6.2). Where the river then meanders through the town of Kroonstad, most water sites were also relatively clean while sediment plastic load slightly increased with laminar flow (Figure 6.4). A major spike in sediment MPs was seen at Vals 4 due to the combination of low river flow and raw sewage input (Observations: C Verster) (Figure 6.4). High MP concentrations in sediment, especially during July (Supp. data), at Vals 4 was likely a result of untreated wastewater discharged into the river with virtually no other flow, effectively creating a settling pond for plastic. The proximity of an informal settlement could also have contributed to MP concentrations here, but raw sewage seemed to be the major culprit. After leaving the town of Kroonstad, the Vals River meanders through farmland for about 100 km. No other major point sources were observed, except for possible runoff from agricultural land (Schell *et al*., 2022). The next site, Vals 5, was located just upstream from the town of Bothaville. It was expected to be relatively clean as major inputs from agricultural land use was not observed anywhere else in the study site. However, informal dumping in the vicinity of Vals 5 could explain relatively high MP concentrations in sediment observed during June (7720 n/kg) (Figure 6.4). Most MPs here were fragments (>99%), reinforcing this finding.

6.4.4.3 Microplastic dynamics in the Vaal River

Differences in water and sediment concentrations of the mainstream Vaal River from its tributaries (Klip and Vals Rivers) (Table 6.1) were presumably due to rapid settling of most MPs in the Vaal Rivers' tributaries and minimal MP transport from the tributaries to the main stream. Microplastic concentrations in sediment in the Vaal River were therefore more consistent (%CV = 26 – Table 6.1).

In the mainstream Vaal River, MP concentrations further upstream (3Riv), but also closer to the densely populated and highly industrialised Gauteng, were higher than downstream sites (G14 and SedW) (Figures 6.3, 6.4 & 6.5). Little downstream transport of MPs was noted in the mainstream Vaal River (Figure 6.5). As in the tributaries, hydrodynamics played a role in MP distribution - although Schoemans drift (Schoem) and Skandinawiëe drift (Skand) are relatively close to each other, MPs in water was higher in the corresponding sediment with turbulent flow at Skand, with this pattern reversed at the laminar flowing Skand (Figure 6.5).

6.4.5 Comparison of results

Microplastic concentrations found in water in this study were higher compared with those reported by most other researchers studying various parts of the Vaal River system (Table 6.5). This difference is likely due to difference in sampling methods (sample volume and sieve mesh size) and certain sampling sites in this study being close to point sources. Weideman *et al*. (2020) detected similar concentrations of MP in surface water of the mainstream Vaal River, but with fibres as the dominant morphotype. Sample sites from Weideman *et al*. (2020) were however mostly far form point sources and anthropogenic activity, with atmospheric deposition of fibres as the main MP source (Bai *et al*., 2022). This reinforces the finding that fragments and beads were deposited in sediment close to sources. Sediment data was comparable to Ramremisa *et al*. (2022), reporting between 1100 - 29000 n/kg dry weight.

Table 6.5: Comparison of results with other studies conducted in the study area.

			Water				Sediment			
Study	Sampling period	Minimum mesh size (µm)	Sampling method	Sample size	Digestion method	MP (n/L)	Sample size	Digestion method	MP (n/kg)	Major morphotype
This study	May 2019 to Mar 2020	25	Bucket bulk water sampling	90 L	Aggressive, temperature-controlled Fenton digestion.	Main stream Vaal: 1.58 ± 1.64 Vaal catchment: 3.37 ± 4.42	50 g dry weight	Aggressive, temperature-controlled Fenton digestion.	Main stream Vaal: 2878 ± 1631 Vaal catchment: 3642 ± 4533	>70% Fragments
Bouwman *et al*. (2018)	January 2017	25	Bucket bulk water sampling	90 L	Aggressive, temperature-controlled Fenton digestion.	Main stream Vaal: 45.75 ± 38.14 Vaal catchment: 9.65 ± 21.90	N.A.	N.A.	No sed. data	>90% Fragments
Weideman *et al*. (2020)	Apr to Nov 2018	25	Bucket bulk water sampling and Newston net	10 L	No digestion	1.7 ± 5.1	N.A.	N.A.	No sed. data	>99% Fibres
Dahms *et al*. (2020)	June 2019	53	Bucket bulk water sampling	100 L	Alkali digestion (10% KOH at room temp; 18h)	Braamfontein spruit min = 0.16; max = 2.08; mean = 0.7	500 g dry weight	None	Braamfontein spruit max = 1347.5; min = 4; mean = 166.8	Water: Fibres Sed: Fragments
Ramaremisa *et al*., (2022)	June 2021	55	Plankton net	Bulk	Fenton digestion over night at room temperature	$0.61 \times 10^{-3} \pm 0.57 \times 10^{-3}$	Not specified	Alkali digestion (10% KOH at 40°C; 24h)	460 ± 280	Fibres and fragments
Saad, *et al*. (2022)	Not specified	1.6	N,A,	N.A.	N.A.	No water data	30 g – 370 g dry weight	Alkali digestion (10% KOH at 40°C; 24h)	463.28 ± 284.08	Fragments

N.A = Not analysed

6.5 Conclusions

High concentrations of MPs were detected at various sites across the sampling area, indicating that the Vaal River system is heavily impacted by plastic pollution due to anthropogenic activities. Two sites – Klip 4 and Vals 4 – contained unprecedented levels of microbeads in soil, and Klip 4 in water as well. The majority of MPs detected – fragments and fibres – are caused by the breakdown of macroplastic waste. The issues contributing to high pollution levels are improper waste disposal infrastructure and consumer behaviour (Verster *et al*., 2020). It is not necessarily due to a lack of available resources, but negligence to maintain existing infrastructure and insufficient municipal service delivery. Poor condition of WWTPs and landfill sites as well as lacking formal waste disposal in informal settlements, combined with consumer ignorance and socio-economic issues, leaves South-Africa’s major commercial water resource—the Vaal River—in a poor condition. To address the issue, all stakeholders will have to buy into mitigation interventions which should include policy updates, proper resource and service delivery management, and consumer education and cooperation (Verster *et al.,* 2020).

MP concentrations did not increase as river flows downstream. It was considerably higher in the upper reaches and decreased downstream. This shows that the MP concentration at a site is determined by the directly surrounding environment and that MPs are not continually transported. Although great mobility in macroplastics have been shown before (Newbould *et* *al*., 2021), microplastics settle our fasted due to lower buoyancy than most macroplastics that are more easily transported downstream (van Wijnen *et al*., 2019).

Different morphotypes had different sources also influenced by different effects of hydrodynamics on morphotypes. The presence of microbeads, especially in sediment infers the proximity of a MP point source like wastewater. High fragment concentrations indicate the direct input of untreated wastewater or runoff but is also the trademark of poor solid waste disposal infrastructure and practices in the surrounding area. Fibres had a relatively even distribution due to its ability to be aerially transported over long distances before deposition (Tatsii *et al*., 2023).

Factors that greatly affect MP loads are poor waste management practices and illegal dumping, degraded state of WWTP infrastructure and functioning, and hydrodynamics. Wetlands are very effective at purifying the water column from MP contamination as we saw, but wetland sediments may become saturated by plastic pollution, reducing the long-term efficiency of wetlands and degrading wetland health.

Based on our data, the Vaal River system feeding into the Orange River may not be a major source of plastic pollution to the Atlantic Ocean. Although this is good news for oceanic plastic load, it poses a new, possibly more threatening question: What are the effects of MP in rivers, as the Vaal River system is the major water source of South Africa’s economically most important region. This could also shed a new perspective on the ‘missing plastics problem’ (Ryan, 2020) as river and wetland sediments are likely long-term sinks for broken down and primary MPs. It would also imply that only rivers with major pollution sources close to the river mouth will contribute to the oceanic plastic load. As the major pollution sources in the Vaal-Orange system is situated more than 1000 km inland, it leaves most of the plastic-associated threats to freshwater users.

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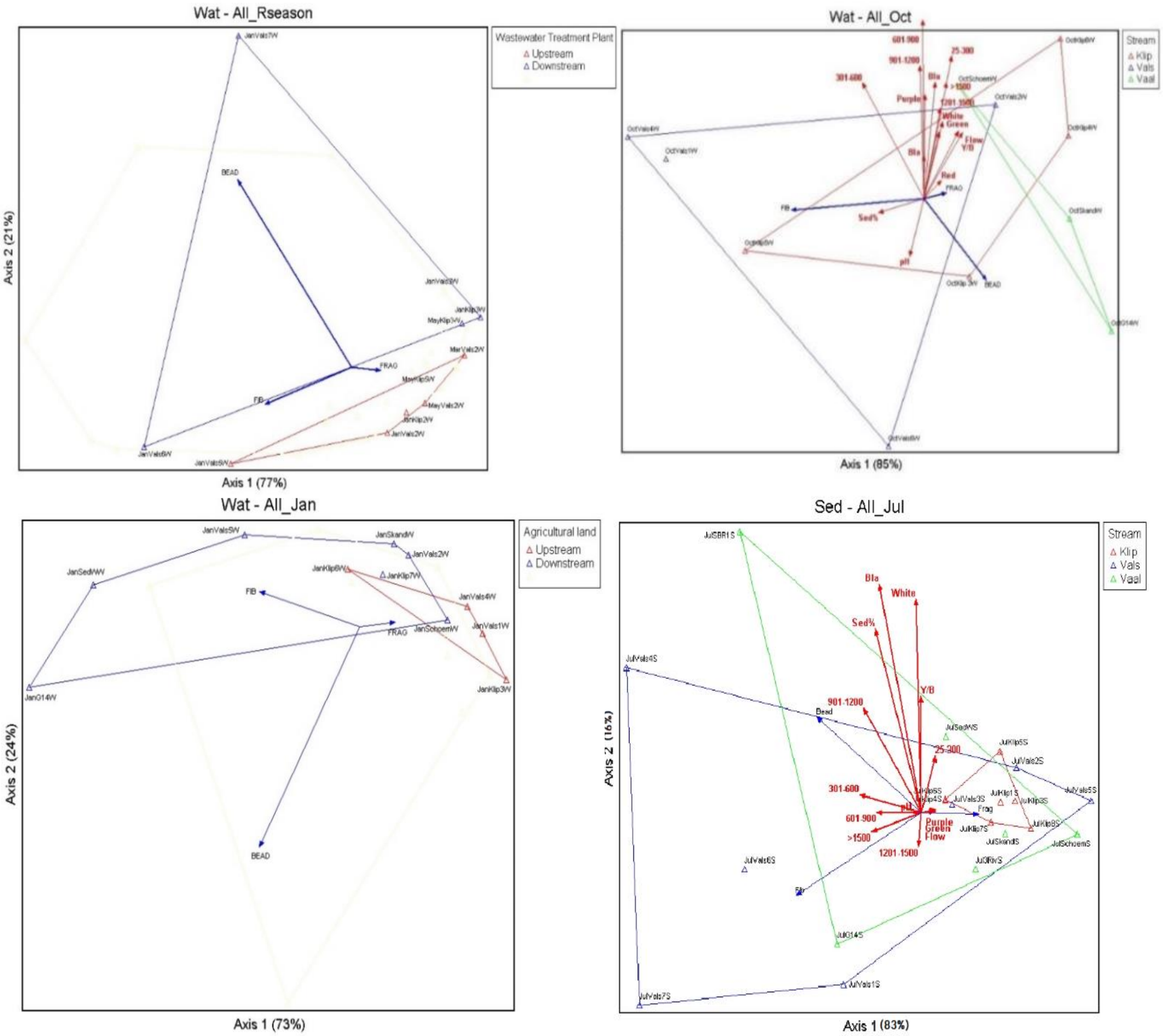
Chapter 6 Supplementary data

Supp Table 6.6: Means per stream

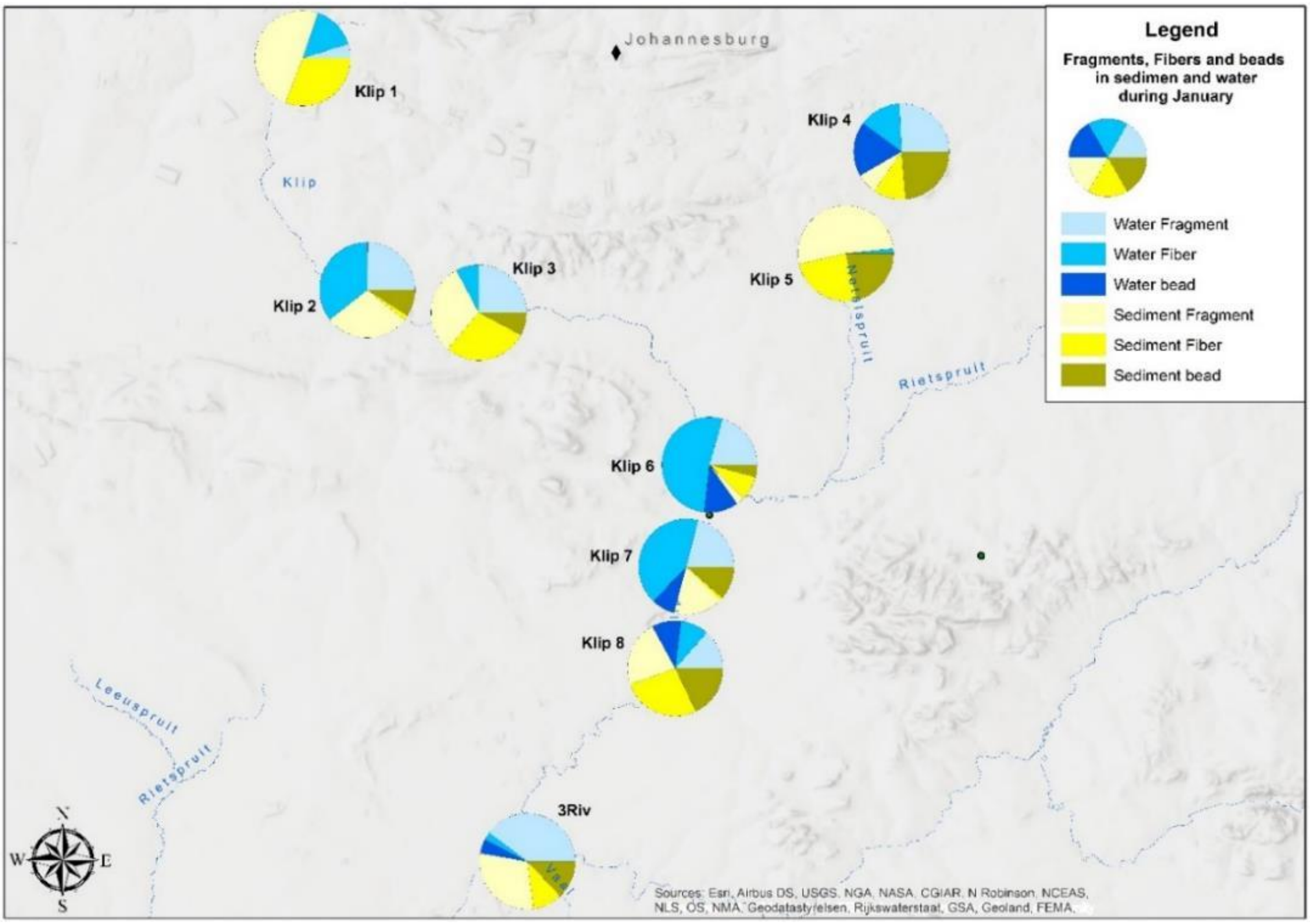
	Klip	Vals	Vaal	All
Water Frag	5.51	1.40	1.34	**2.82**
Water Fib	0.63	0.38	0.21	**0.42**
Water Bead	0.24	0.11	0.03	**0.13**
Water Total	6.11	1.88	1.58	**3.27**
Sed Frag	4522	1019	1882	**2546**
Sed Fib	893	509	411	**629**
Sed Bead	681	226	285	**407**
Sed Total	6150	1869	2578	**3642**
Sed/SSW	4693	3358	22135	**10226**
Macroplast's	146.20	33.95	15.94	**68.13**
Flow (m/s)	0.72	0.14	0.22	**0.36**
pH	6.92	7.19	7.74	**7.25**
DO	16.49	19.39	20.47	**18.72**
TDS (200 ppm)	5030	2399	4693	**3893**
Sed % silt + clay	8.08	8.96	10.70	**9.41**

Supp Table 6.7: Means per site

Site	Sed (n/kg)	Water (n/L)
Klip 3	2516	3.219
Klip 4	9032	21.51
Klip 5	12275	1.734
Klip 6	3972	20.2
Vals 1	1515	1.369
Vals 2	1060	2.391
Vals 4	3395	2.725
Vals 6	1507	0.648
Schoem	1830	2.686
Skand	3152	1.104
G14	2055	1.17



Supp Figure 6.2: NMS plots of relativised MP counts with spatial factors



Supp Figure 6.3: Morphotype composition in water and sediment in the Klip River during January

Chapter 7

Dry and rainy seasons affect fluxes of microplastics in rivers but are moderated by surrounding waste management conditions: Vaal River, South Africa

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ABSTRACT
Catchment characteristics govern the temporal distribution of microplastics (MPs) in rivers. This is the first study to show temporal trends of MPs in the Vaal River system – South Africa's most used river. We investigated three different rivers in the Vaal system. River sediments act as a MP sink, with a measure of resuspension happening in the rainy season. Opposing effects of increased stream flow on MP concentrations were observed in two rivers within the same larger catchment. A diluting affect was found during high-flow periods in a less polluted tributary catchment – the Vals River. In contrast, areas with poor waste management like the catchment of the Klip River tributary, polluted runoff following rains increased the riverine MP load. Variance of two orders of magnitude is present at certain sites over the course of a year. The highest concentration of MPs in water was measured directly after the first rains of the new rainy season - 8.12 n/L. The highest mean concentration in sediment was measured in July during the dry season - 4339 n/kg dry mass. At one site, beads composed 0.2% of dry sediment. Local hydrodynamics can affect the distribution of MP between the water and sediment with turbulent flow favouring MP in water. Fibres are more prevalent in sediment where flow is laminar. These sites also have higher proportions of MPs in its sediment. Interventions should be tailored to local catchment waste profiles, and a national intervention on beads is strongly advised.

7.1 Introduction

Microplastics (MPs) are plastic particles larger than 1 µm and smaller 5 mm (Zhang *et al*., 2020). One can expect to find MPs in virtually all environments due to the cosmopolitan use of plastics and wide array of release and transport pathways (Rede *et al*., 2023). Rivers are important pathways of MPs to the ocean but has also recently been shown to be an MP reservoir or sink (van Emmerik *et al*., 2023; He *et al*., 2021). When MP concentrations are observed over time, variations become apparent (Talbot *et al*., 2022). MPs in rivers can show temporal variation of up to eight orders of magnitude (Stanton *et al*., 2020). Though variation is expected, particular patterns are difficult to predict because of the multiple factors than can affect MP concentrations at a given moment within a landscape (Talbot *et al*., 2022).

Two factors have major effects om the MP profile at a given point in a river - pollution sources and local hydrodynamics (Vianello *et al.,* 2013). Both these factors are multi-faceted, and the interactions of these factors may differ vastly over time (Bai *et al*., 2022).

7.1.1 Pollution sources

Africa and Asia are expected to remain the global hotspots of mismanaged plastic waste through the rest of the century (Lebreton *et al*., 2019). Each South African uses at least 19 kg of plastic each year (Verster *et al.*, 2020). Conservative estimates show that 39% of South African households do not have access to formal waste disposal (Plastics SA, 2022). Severe waste mismanagement is reinforced by poverty, rapid urbanisation, and inadequate waste management infrastructure causing huge piles of rubbish often near water (Verster *et al.*, 2020).

The South African natural environment is visibly polluted with macrolitter. Land near underserviced areas in terms of waste collection are prone to extensive littering and illegal dumping in streets, vacant land, and public spaces like river fringes (Haywood *et al*., 2021). In South African townships, waste is often discarded informally, without polluters being aware of the consequences of environmental plastic pollution (Adeniran *et al.,* 2022). The presence of secondary MPs like fragments – the breakdown products of larger plastic debris – can be expected in elevated concentrations in these areas (Chapter 6).

Untreated wastewater is another major source of MPs in South African freshwater (Saad *et al.*, 2022). Of the smaller towns in the study area, none have more than 10% wastewater treatment plant (WWTP) functionality (Department of Water and Sanitation (DWS), 2022), so most wastewater enter the rivers untreated. In the Vaal River and Gauteng area, WWTPs are generally in better condition, but often operated at over-capacity (DWS, 2022).

7.1.2 Hydrodynamics

Microplastics and macroplastics behave differently in rivers. Larger, more buoyant pieces of macrolitter travel long distances in rivers (Newbould *et al*., 2021). Microplastics, on the other hand, tend to become deposited in sediment relatively quickly after entering the river, with longer range transport during high-flow events (van Emmerik *et al.,* 2023). Higher flows could increase the influx and mobilisation of MPs in rivers (He *et al*., 2022), but may in some cases also have a diluting effect (De Arbeloa *et al.,* 2023). Local hydrodynamics – whether flow is laminar or turbulent - will affect the reposition and suspension fluxes between MPs in water and sediment (Vianello *et al.,* 2013).

This study will focus on the dominant factors that affect MPs in the Vaal River and two of its tributaries – the Klip and Vals rivers, to give more refined insight into the temporal variation of MPs in South African freshwaters, characterised by high- and low-flows over rainy and dry seasons, and the effect of proximity of the sampling points to waste sites.

7.2 Methods

7.2.1 Sampling

The area of study consists of a section of the Mid-Vaal catchment which reaches across the Free State, North West, and Gauteng provinces of South Africa (Figure 7.1). It lies within a summer rainfall region with the mean annual precipitation (MAP) ranging between 500–900 mm (Mnguni, 2020). Study sites were selected on basis of accessibility, safety, distribution, places where both sediment and water can be sampled, and proximity to possible point sources. Eight sites were selected in the Klip River tributary, seven in the Vals River tributary, and five sites along the mainstream Vaal River, for a total of twenty study sites (Figure 7.1). An additional site in the Suikerbosrand Nature Reserve was also sampled initially as a reference, but disregarded because the river was dry for most of the sampling period.

Sites were sampled at circa two-month intervals between May 2019 and March 2020. Sampling episodes are described as months throughout this paper namely May, July, October, November during the rainy season, and January and March as the dry season, respectively. Due to logistical reasons, not all sites were sampled during each sampling campaign. The most complete sampling episodes are January, during the rainy season, and July, in the dry season.

Sampling at each site consisted of obtaining surface water and sediment. Surface water samples were collected using a 15 L stainless steel bucket to make a composite 90 litre sample which is filtered on-site through a 25 µm mesh stainless steel sieve. Sieve content was then retained in a Schott bottle and taken to the lab. Sediment samples were taken with a steel shovel at the water's edge and stored in rinsed tinfoil for transport to the lab. Metadata parameters were also measured at each site - instream river flow velocity, local hydrodynamics (laminar or turbulent), dissolved oxygen (DO), pH, total dissolved solids (TDS), and the number of visible macroplastic pieces within the immediate surroundings at water level.

Each batch of samples that was processed was run in conjunction with a procedural blank of which background counts were subtracted from sample counts after analysis. All surfaces that would come in contact with samples were triple-rinsed with double-distilled water that was re-filtered through a 25 µm mesh stainless steel. Samples were covered with tin foil and watch glasses as far as possible to avoid atmospheric fallout contamination.

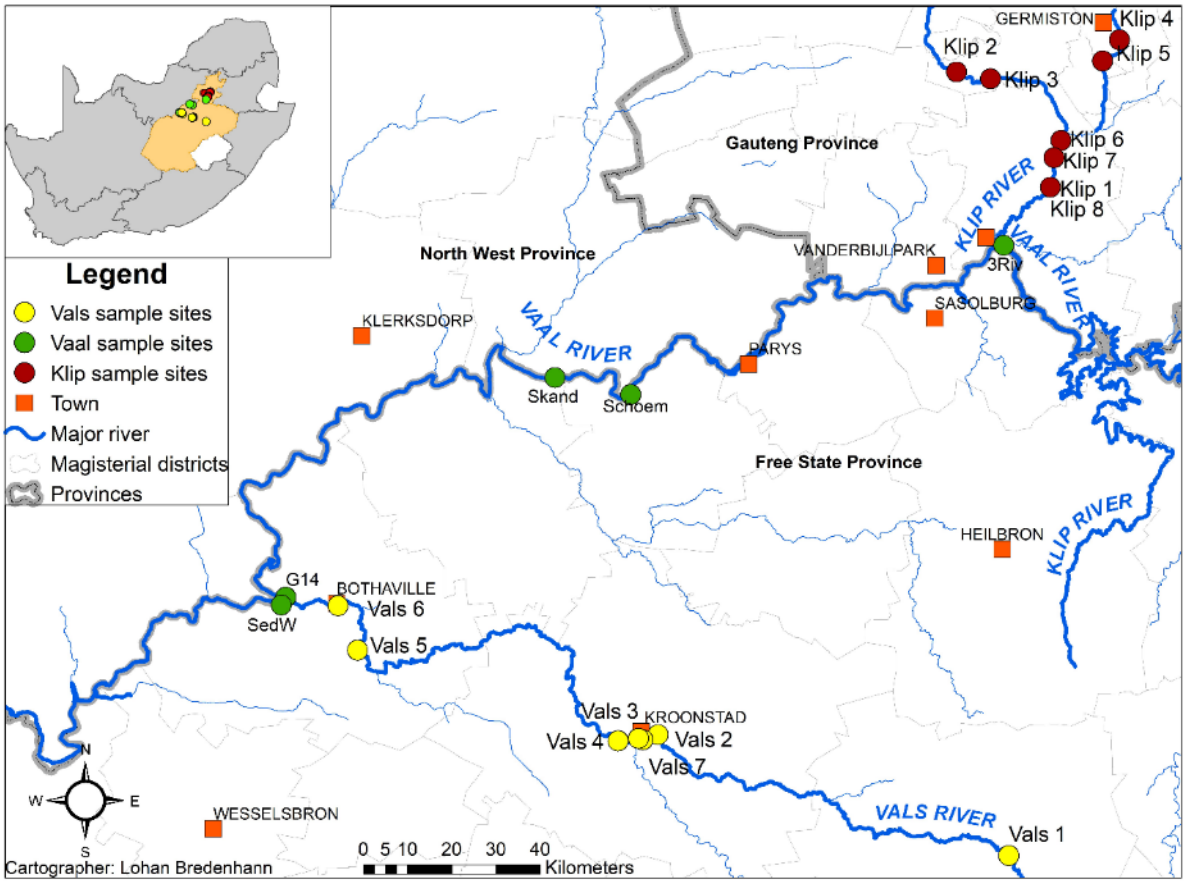

Figure 7.1: Map of study area with sampling sites in the Vals River (yellow), Klip River (red) and main stream Vaal River (green).

7.2.2 Sample processing

Water and sediment samples were dried at 50°C. A temperature regulated H_2O_2 digestion with Fenton's reagent (Savino *et al*., 2022) was then carried out on whole water samples, and on 50 g dry mass subsamples of sediment, respectively. Particle size distribution analysis of sediment samples was also done on separate sub-samples. An overnight, post-digestion density separation was then done with each sample in a 0.5 M sodium iodide solution (1.05 g/ml). Floated material was deposited on custom made 25 µm mesh stainless steel sieves. Settled material was visually inspected under a light microscope. Putative MPs were manually removed with forceps and added to the sieve. If separation was deemed insufficient, another round of density separation was carried out and floated material added to the same sieve.

7.2.3 Analysis

Samples on custom stainless-steel sieves were dried. Sieves were placed on a 1 X 1 cm counting square. Microplastics in each square was counted under a Nikon AZ100M microscope using 300 X magnification. Each MP particle was visually identified as follows: Microbeads were easily identified as distinct spherical shapes with smooth surfaces and standard diameters are not common in nature (Figure 7.2). Inorganic fibres were identified, as natural fibres have distinct cellular structures or started showing signs of degradation due to the aggressive digestion methods samples were subjected to. Fragments—irregularly shaped MPs caused by breakdown of larger plastic pieces—were identified based on colour, breakage patterns, and crystalline structure. Particles were excluded if it had metallic lustre, cellular or organic structures, and mineral crystalline structures.

The longest dimension of each particle was measured with Nikon AZ100 imaging software/NIS Elements-D imaging software and noted (Figure 7.2). The colour and morphotype of each particle were also recorded. Background subtractions were done per sample batch and raw counts were converted to counts per litre of water and per kilogram dry mass of sediment. Where sediment and water data were compared, sediment samples were converted to counts per gram dry mass because counts per kilogram are generally three orders of magnitude greater than corresponding water counts per litre.

Figure 7.2: Sample containing microbeads from Klip River water showing beads, fragments, and fibres. The large yellow fibre was considered as natural.

A subset of samples was sent polymer composition determination. Dried MPs were suspended in pure alcohol and a subsample was deposited on a zinc selenide (ZnSe) transmissive window. Polymer analyses was done using FTIR according to Maurizi *et al*. (2023) at the Department of the Built Environment, Aalborg University, Denmark. The analyses were executed using a Focal Plane Array– μFTIR. The Cary 620 FTIR microscope, integrated with a Cary 670 IR spectroscope (Agilent Technologies, USA), was used for scanning the ZnSe transmission windows' active area to a 128 × 128 MCT FPA detector. A separate background tile was collected before each sample and was made up of 120 co-added scans. Scanning was performed in transmission mode, covering a spectral range of 3750–850 cm−1 at 8 cm−1 resolution, with 30 co-added scans. The result was a chemical image of the sample area, where each pixel held a corrected IR spectrum. These images were analysed using the siMPle software Primpke *et al* v. 1.3.1β, which detects particles, quantifies their morphology, and estimates their volume and mass automatically (Simon *et al*., 2018).

Descriptive statistics and linear regressions were computed using GraphPad Prism software (Ver 10; www.grahpad.com) for various data groupings (each stream, each month etc.). Descriptive statistics and correlations were done on concentration data, and linear regressions were performed to investigate relationships between concentrations and various metadata parameters, including flow rate and macroplastic observed at a site. Non-metric Multidimensional Scaling (NMS) was carried out on relativised data per sample for various data groupings using PCORD 7.08 software to identify major relationships between data and variables. Gower-ignore-zero was used as distance measure from random starting coordinates. Five initial dimensions was selected, with 250 runs of real and randomised data.

7.3 Results

7.3.1 Monthly mean concentrations per morphotype

The highest mean concentration of MPs in water was recorded during November, directly after the first rainfall of the season (8.12 ± 10.3 n/L) (Table 7.1). The same sampling period had the lowest mean MP concentration in its corresponding sediment (1440 ± 817 n/kg). The cleanest sampling period for both water (0.27 ± 0.1 n/L) and sediment (1400 ± 1178 n/kg) MPs was during March, at the end of the rainy season. Only three of the 20 sites could however be sampled during March due to the onset of the Covid-19 pandemic. A general increase in MP concentrations in water can be seen throughout the dry season from May to November. The reverse took place during the rainy season when MP concentrations decrease from November to March. Mean MP concentrations across all sites in sediment did not display an obvious trend.

Table 7.1: Means and standard deviations of microplastic morphotypes (fragments, fibres, and microbeads) concentrations per month of all sites combined

		May	***Jul***	***Oct***	***Nov***	***Jan***	***Mar***	***All***
Water	**Fragments**	3.3 ± 5.2	2.6 ± 2.9	3.32 ± 3.2	7.8 ± 10	2.21 ± 4	0.20 ± 0.1	**2.82 ± 4.1**
	Fibres	0.3 ± 0.4	0.32 ± 0.4	0.74 ± 1.3	0.78 ± 0.6	0.42 ± 0.7	0.07 ± 0.04	**0.42 ± 0.7**
	Beads	0.2 ± 0.5	0.12 ± 0.2	0.11 ± 0.2	0.25 ± 0.4	0.12 ± 0.3	0.0	**0.13 ± 0.3**
	Total	3.5 ± 5.6	3.04 ± 3.2	4.21 ± 4.2	8.12 ± 10.3	2.58 ± 4	0.27 ± 0.1	**3.27 ± 4.4**
Sediment	**Fragments**	1680 ± 1108	3192 ± 3329	2233 ± 2686	1107 ± 763	2980 ± 4734	813.3 ± 888	**2546 ± 3390**
	Fibres	402 ± 816	513.5 ± 677	930.9 ± 467	186.7 ± 257	784.6 ± 1106	460 ± 762	**629.3 ± 820**
	Beads	520 ± 693	494.8 ± 937	145.5 ± 222	146.7 ± 254	486.8 ± 954	126.7 ± 155	**406 ± 784**
	Total	2602 ± 2341	4339 ± 3975	3309 ± 3178	1440 ± 817	4315 ± 6654	1400 ± 1178	**3642 ± 4533**

7.3.2 Monthly means per river

The Klip River had the highest mean macroplastic count, followed by maximum MP concentrations for total water MPs reached a maximum during the first rains of the season and a minimum in July and the greatest difference between sites in January (Table 7.2a). Sediment in the Klip River was at its peak in January and reached the lowest concentration in November (Table 7.2a), although the most polluted sediment site – Klip 5 – could not be sampled during November which could have affected the results.

The Vals River generally has lower macroplastic counts and MP concentrations when compared to the Klip River (Table 7.2b). Its water reaches a maximum MP concentration during October, and a minimum in March. Sediment is at its highest in July and at its lowest in May (Table 7.2b).

The mainstream Vaal River (Table 7.2c) had the lowest levels of surrounding pollution and MP concentrations and the most consistent MP concentrations in sediment. Water in the Vaal River reached a maximum difference during May and the highest concentration during October. Vaal River sediment was at a minimum MP concentration during November and a maximum in May.

Table 7.2: Monthly mean macroplastic and descriptive statistics for water and sediment for the Klip (a), Vals (b) and Vaal (c) rivers.

a) Klip River

		May	July	October	November	January	March	ALL
Macroplastics (Mean)		164	106	284	43	117		**238**
Water	**N**	4	7	4	1	8	0	**23**
	Mean (n/L)	11.78	6.38	11.69	41.56	6.46		**8.27**
	Standard dev.	15.97	2.61	18.37	0	12.12		**11.67**
	%CV	136	41	157	0	188		**141**
Sediment	**N**	4	4	4	1	4	0	**15**
	Mean (n/kg)	3425	5863	6025	2360	17 791		**8361**
	Standard dev.	3464	4857	4143	0	18 849		**11 122**
	%CV	101	83	69	0	106		**133**

b) Vals River

		May	July	October	November	January	March	ALL
Macroplastics (Mean)		8	26	68		53	3	**32**
Water	**N**	3	7	4	0	7	3	**25**
	Mean (n/L)	0.61	2.09	5.30		0.97	0.27	**1.87**
	Standard dev.	0.31	1.69	5.49		0.79	0.08	**2.72**
	%CV	51	81	104		81	29	**145**
Sediment	**N**	3	4	4	0	4	3	**14**
	Mean (n/kg)	906.7	3055	2040		1325	1400	**521**
	Standard dev.	427	4892	806		1011	1178	**1797**
	%CV	47	160	40		76	84	**345**

c) Vaal River

		May	July	October	November	January	March	ALL
Macroplastics (Mean)		5	19	7	16	23		**17**
Water	**N**	3	6	3	2	5	0	**17**
	Mean (n/L)	1.27	0.26	2.29	2.18	2.19		**3.93**
	Standard dev.	1.71	0.27	2.57	0.77	1.69		**9.84**
	%CV	135	103	112	35	77		**250**
Sediment	**N**	2	3	3	2	3		**10**
	Mean (n/kg)	3450	3280	1380	980	2820		**3082**
	Standard dev.	410	1680	453	255	2652		**1778**
	%CV	12	51	33	26	94		**58**

7.3.3 Temporal changes per site

Different sites and matrices (water and sediment) displayed different MP concertation fluctuations (Figure 3.7). When considering Klip River water, Klip 6 saw an increase in MPs in October and reached a maximum MP concentration during the first rains of the season in

November (Figure 7.3a). Klip River sediment saw increased MP concentrations as the rainy season progressed (Figure 7.3b). The Vals River had its highest concentrations during the dry months of July and October (Figures 7.3c & d). The site with the most polluted surroundings in the Klip River – Klip 4 – had its peak MP concentration in water during the rainy season (January to May) (Figure 7.3a; Supp. Figure 7.2), while the most polluted site in the Vals River – Vals 4 – peaked in the dry season (July) (Figure 7.3c).

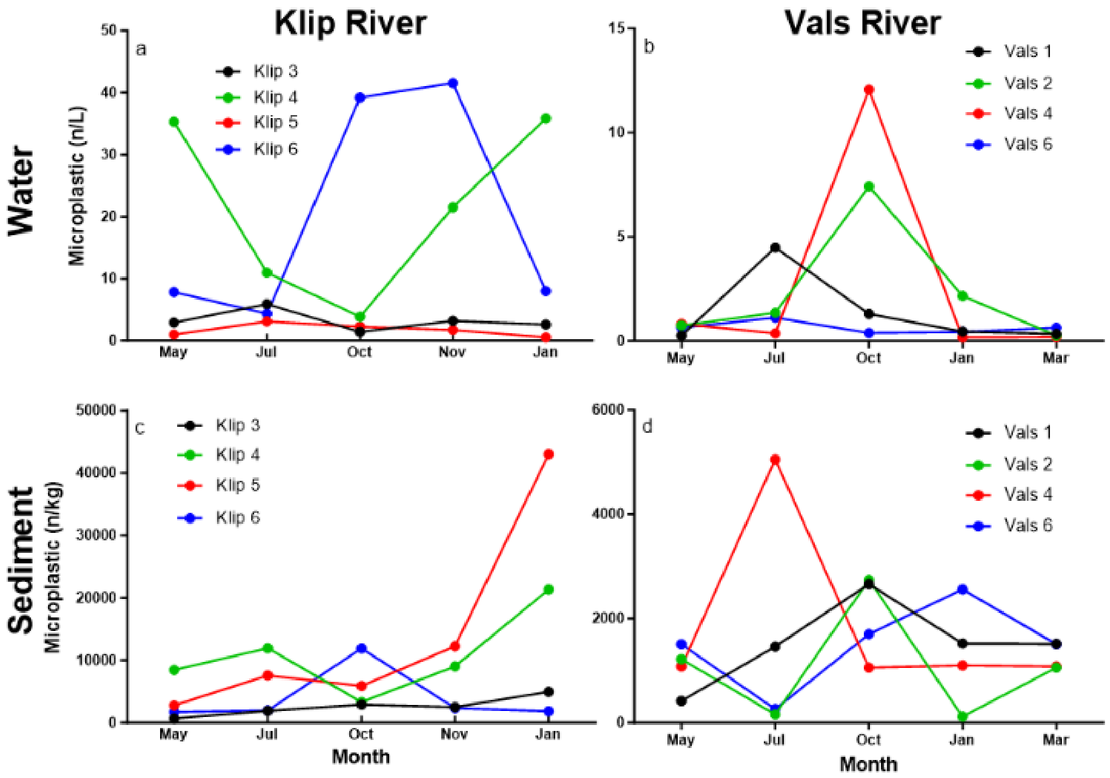

Figure 7.3: Temporal changes of microplastic concentrations in water and sediment of frequently sampled sites along the Klip (a & b) and Vals (c & d) rivers.

7.3.4 Seasonal maps

When considering the changes in MP concentrations over time at sites during low-flow (July) and high-flow (January) sampling periods, each river will be assessed separately due to differences in source and environmental variables (Figure 7.4).

The Klip River had high variability between MP concentrations throughout the wet season, while the dry season had a more consistent MP load throughout the river over time. This is the case for sediment (Figure 7.4a) and water samples (Figure 7.4b). Microplastic concentrations in Klip River sediment during July had lower pollution concentrations, with less variation than seen in corresponding high flow samples, but more variation when compared with its water MP concentrations. The highest plastic pollution in surface water in the study was taken at Klip 6 during November with 37 n/L. Klip 6 did not have the highest mean surface water MP load with a mean count of 14 ± 6, but a major spike in surface water MP was observed at this site directly after the first rains.

The Vals River displayed an obverse pattern of the high flow vs. low flow compared with the Klip River. Here, greater variability was seen in water and sediment for low-flow samples, and rainy season samples had more consistent and lower MP concentrations for surface water and sediment (Figures 7.4c & d).

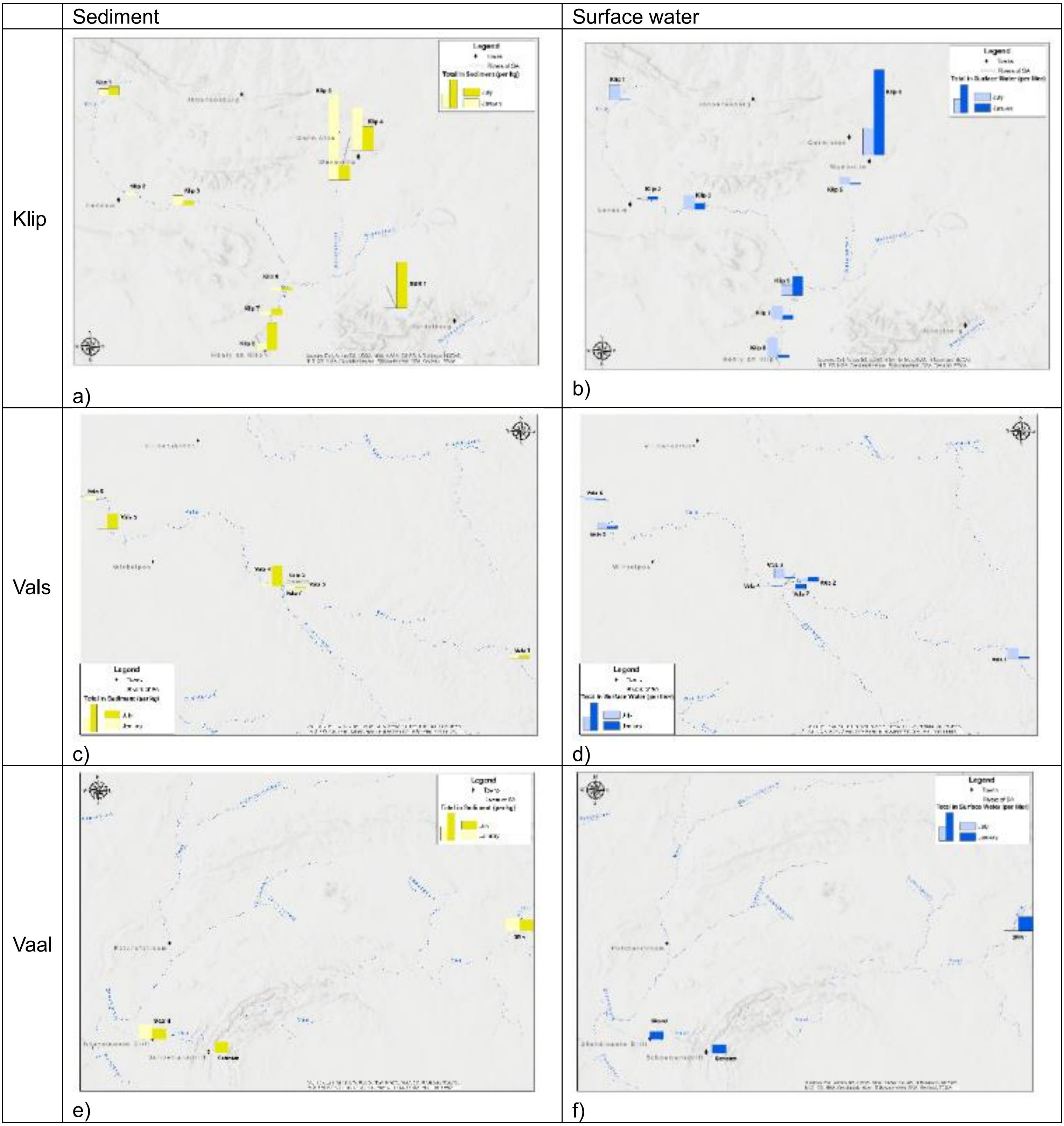

Figure 7.4: Comparisons of January (rainy season) microplastic concentrations as dark blue and light yellow, and July (dry season) microplastic totals as light blue and dark yellow at a site per river in sediment and surface water respectively.

Microplastic concentrations in the mainstream Vaal River were relatively consistent over time when compared with its two tributaries (Figures 7.4e & f). High-flow samples from the Vaal River showed consistently higher MP concentrations in water and sediment compared with the low-flow period. The only exception was at Schoemans drift where high flow sediment samples was virtually free from MPs (0.02 n/L).

7.3.5 Flow rates vs concentration regressions

Table 7.3: P-values and slopes of linear regressions of plastic counts (log-transformed) with flow rate. Significant p-values are in red cells, positive slopes in green cells and negative slopes in blue.

Stream	**Value**	**Water**			**Sediment**		
		Fragments	**Fibres**	**Beads**	**Fragments**	**Fibres**	**Beads**
Klip	P	0.041	0.289	0.023	0.032	0.280	0.031
	Slope	0.466		0.606	-0.403		-0.487
Vals	P	0.014	0.253	0.534	0.157	0.428	0.035
	Slope	-0.765					0.832
Vaal	P	0.457	0.358	0.250	0.061	0.546	0.038
	Slope						-0.723

Linear regressions of log-transformed MP concentrations with the flow rates measured at each site was done for the Klip, Vals and Vaal Rivers separately. The Klip River had more significant regressions than the other two (Table 7.3). The concentrations of fragments and beads in water had significant positive associations with river flow. Both fragments and beads in Klip River sediment had significant negative relationships with stream flow velocity at a site, meaning more beads and fragments are in sediment at slower stream flow velocity (Table 7.3).

The trend for significant linear regressions of MP concentrations with flow rate in the Klip River was reversed in the Vals River (Table 7.3). In the Vals River, only the number of fragments in the water had a significant negative association with flow rate (Table 7.3). In the less polluted Vals River, the higher the flow rate corresponded with lower fragment plastic concentrations. Only beads in the Vals and Vaal River sediment had a significant association with water flow velocity. Unlike the Vaal or Klip River, the stronger the stream flow, the more beads there were in Vals River sediment. Neither fibres in water nor sediment showed any significant association with stream flow (Table 7.3).

7.3.6 Surrounding pollution

We saw a significant positive regression between the number of macroplastics surrounding a site and log-transformed MP concentrations in its associated water (Figure 7.5). The most polluted catchment in our study was the Klip River with (mean macroplastics per site = 146 ± 189), then the Vals River (mean macroplastics per site = 34 ± 51). The cleanest catchment in terms of plastic and waste management practices was the Vaal River (mean macroplastics per site = 16 ± 21).

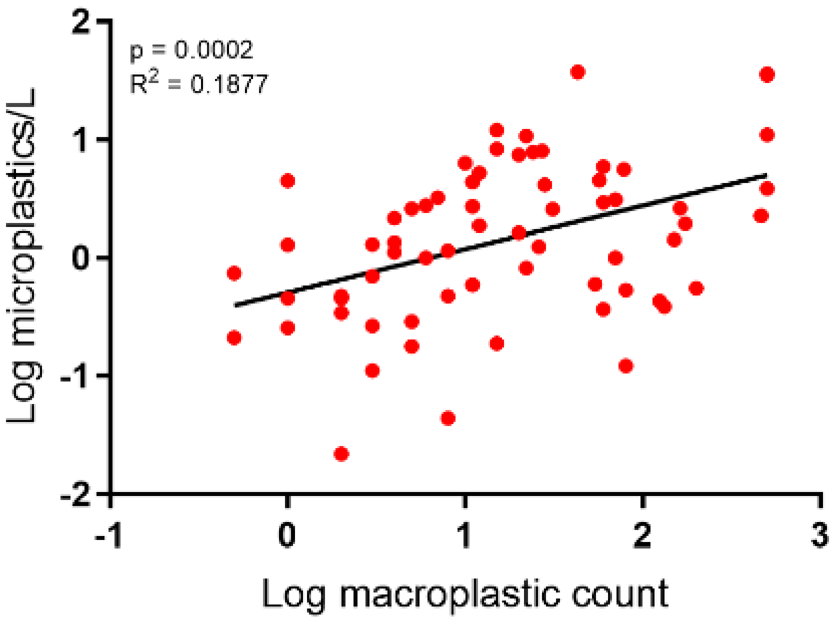

Figure 7.5: Linear regression of log transformed macroplastic scores with microplastic concentrations in water.

7.3.7 Hydrodynamics affects morphotype composition in water and sediment.

Vectors (blue arrows) of the three different morphotypes diverged in almost all NMS analyses conducted (Figure 7.6) showing that morphotypes originates independently of each other. The fragment vectors associated strongly with turbulent flow, while fibres showed less distinct patters in water. The bead vectors diverged (180°) from turbulent flow to laminar flow polygons, especially in sediment (Figure 7.6a). In both NMS examples, a sample from Vals 7 ordinated distinct from the others (Figure 7.6). Vals 7 is a site in Kroonstad where water flow was minimal. Its sediment sample during July (low-flow) was also distinct because of a high prevalence of fragments (600 n/kg). Its January (high-flow) water sample was distinct due to a relatively high concentration of beads (1.04 n/L).

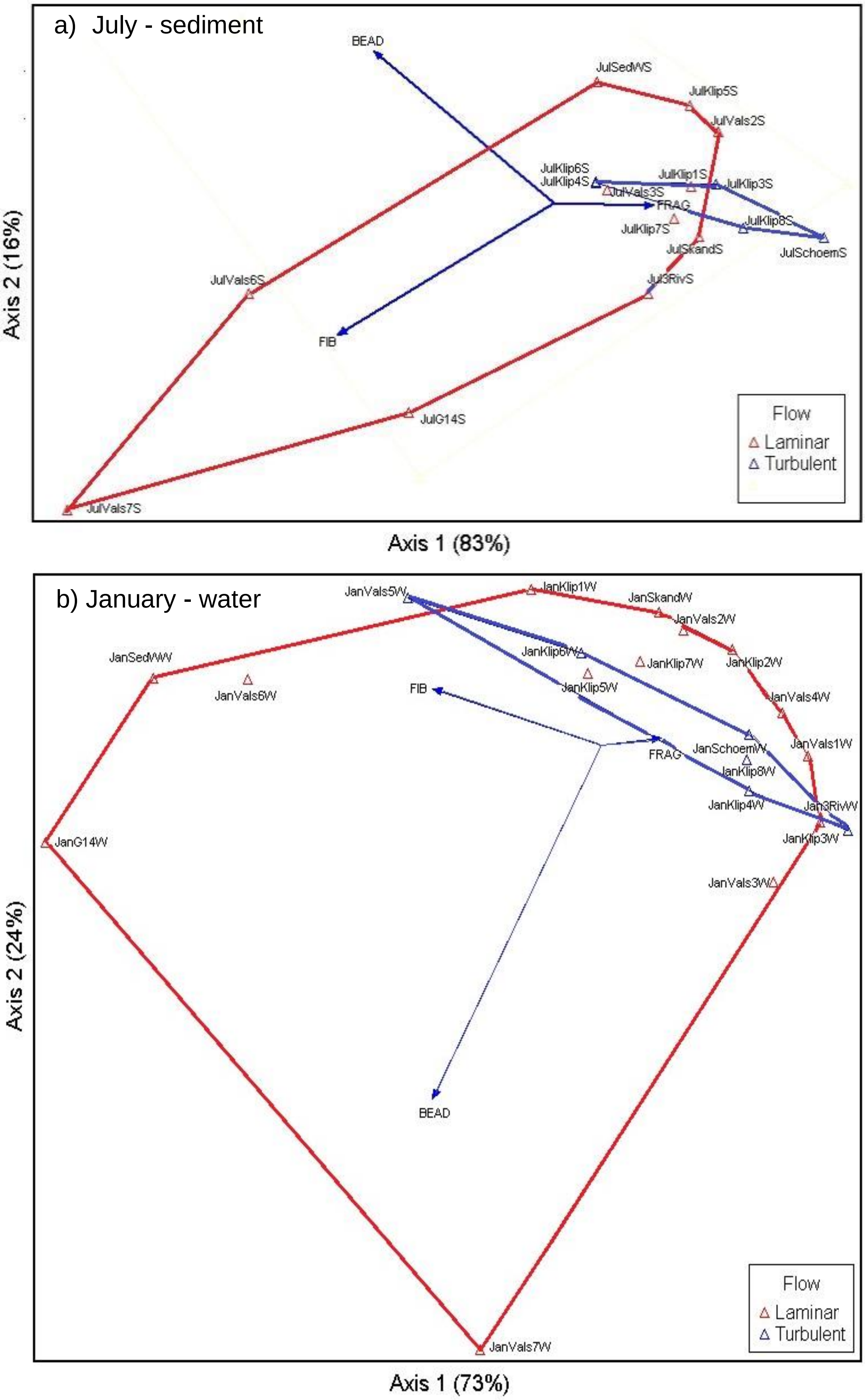

Figure 7.6: Nonmetric multidimensional-scaled ordinations of sediment data for July (a) and water data for January (b).

Schoemans Drift (Schoem) is located in an area of the mainstream Vaal River where it flows through rocky rapids. At Skandinawië Drift (Skand), about 20 km downstream from Schoemans Drift, the river settled into a steady laminar flow profile. The proportional distribution of fragments, fibres and beads in sediment and water was examined during January (rainy season) and July (dry season) to highlight the 1) variation between MP proportions of high and low-flow situations and 2) laminar and turbulent river dynamics (Figure 7.7).

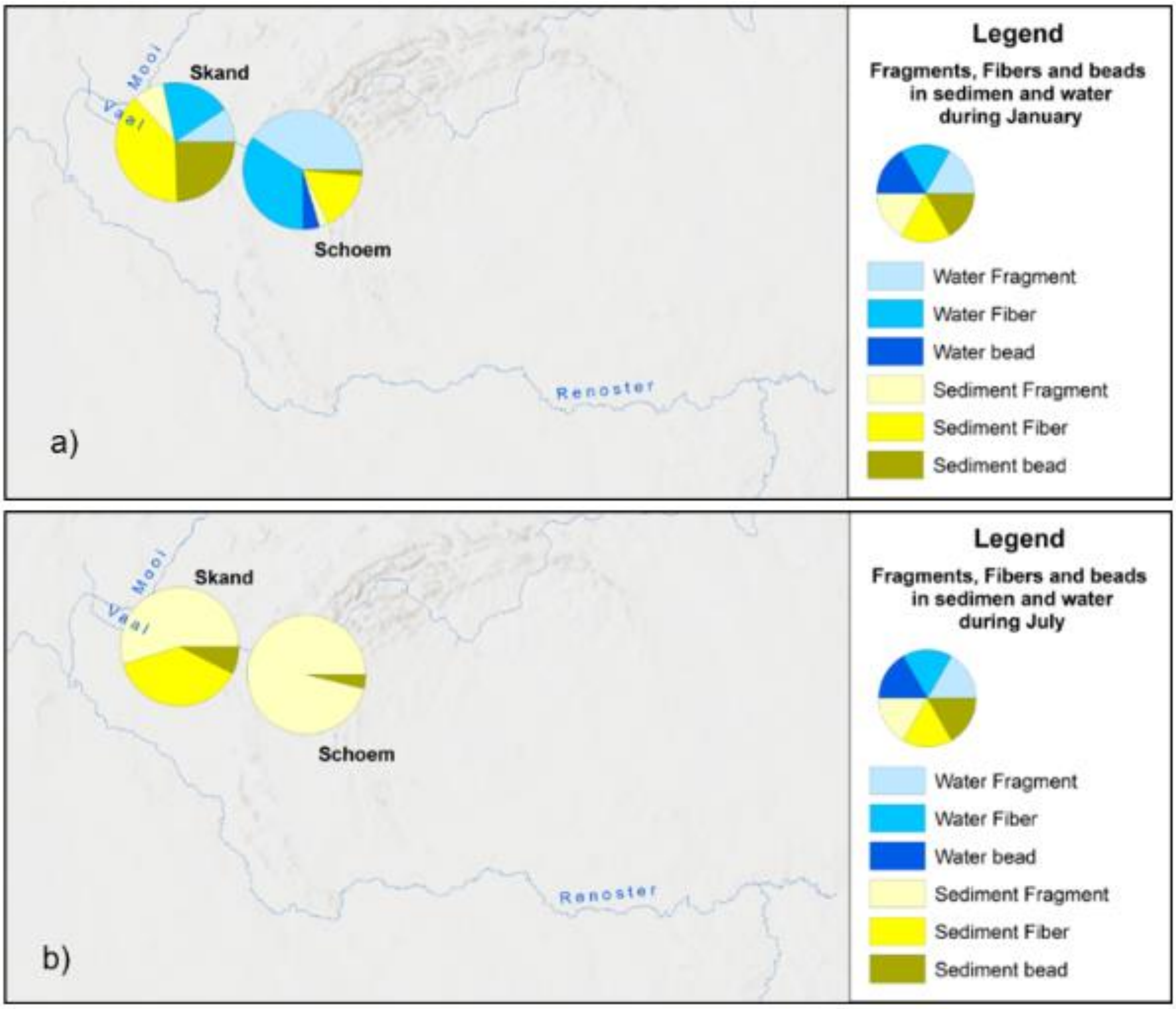

Figure 7.7: Comparisons of morphotype distributions in water and sediment at Schoemans Drift (turbulent) and Skandinawië Drift (laminar flow).

In a high-flow scenario (January) (Figure 7.5a) a much greater proportion of MP is suspended in water when compared to a low flow situation (July) (Figure 5b). When one considers the variation between the two sites, one turbulent and one laminar, there is also a clear difference in proportions of morphotypes, even though no direct point sources are present between the two sites. During a high-flow, the more turbulent site (Schoem) has most of its MP suspended in water with sediment virtually free from MPs, while sediment MP dominate in laminar flow (Figure 7a). Fragments at the turbulent site are also more prevalent than in laminar flow. In both laminar flow scenarios, beads make up a greater proportion of sediment MPs.

7.3.8 Polymer composition

Four samples – one water, two sediment and one background – was sent for µ-FTIR analysis. All samples were not analysed for polymer composition due to time and budget constraints. The dominant polymers found in the three environmental samples were high- and low-density polyethylene (HD-PE and LD-PE), which combined have a prevalence of 89% of analysed samples. Polypropylene (PP) composed 5.7% of MPs, polystyrene (PS) 3.8% and polyester 1.7 %. A single polyurethane particle was detected in the mainstream Vaal River sample. The control sample contained 10 polyethylene particles, all smaller than 100 µm.

Table 7.4: Polymer composition and parameters of microplastics in one water and two sediment samples.

Polymer	% in water (n = 666)	% in sediment (n = 265)	% in sediment (n = 1660)
Polyethylene	98	68	99.7
Polyester	1	4	0.30
Polyurethane	1	0	0
Polypropylene	0	17	0
Polystyrene	0	11	0
Size range % (µm)	>25=9 25-50=69 50-100=22	>25=11 25-50=25 50-100=28 100-300=28 300-600=8	>25=4.1 25-50=56 50-100=27 100-300=11 300-600=0
Total/mean mass (ng)	3866 / 5.8	34489 / 130	37320 / 22.5

7.4 Discussion

The microplastic profile the the three streams investigated, are different from one another, which confirms that catchment characteristics affects a rivers MP profile (Talbot *et al.*, 2022). Local macroplastics surrounding a site had a significant correlation with MP concentrations in river water (Figure 7.5).

The study area lies within a summer rainfall area with the rainy season during the sampling period ranging from November to March. The area sampled along the Klip river has a mean annual precipitation (MAP) of 700-800 mm per annum (Cropsmonitor, 2023). The Vals river is in a slightly more arid area of the country with a MAP of 500-600 mm per annum. The area sampled along the Vaal River has an MAP of 600-700 mm per annum. Flow rate at a site is a general reflection of the amount of water the river is receiving. Flow rates are higher in rainy months. MP concentrations in water generally increased throughput the dry season from May to November, reaching a maximum during the first rains of the season in November (Table 7.1). Thereafter, it decreased as the rainy season progressed from January to March.

7.4.1 Comparison between Klip and Vals Rivers

In the Klip River, increased runoff in the rainy season generally increases riverine MPs, as opposed to the Vals River where increased runoff had a diluting effect.

The Klip River drains the densely populated City of Johannesburg, containing areas of high industrial activity. Increased flow in the Klip river is mostly due to runoff, resulting in an influx of MP from polluted surroundings. Urban areas are known to be associated with higher concentrations of MPs in rivers (Talbot *et al.*, 2022). Surface water MP concentrations in the

Klip River increased as flow rate increased, while sediment MP concentrations decreased (Table 7.3) due to rapid settling of incoming fragments and beads (Table 7.1). Likely for the same reason, there was greater variation among sites in water and sediment MP concentrations during January, than during July in the Klip River (CV% - Table 2a; Figure 3a & 4a & b). Polymer composition of samples (except for the absence of PET, likely lost during density separation in sample cleanup) correspond with household plastic use (Plastics SA, 2022). In the Klip River, summer months (November to March) also had more riparian vegetation able to trap MPs and increase deposition in sediments.

In contrast with the Klip River, surface water MP concentrations from the Vals River decreased, and sediment concentrations increased with an increase in streamflow (Table 7.3). Agriculture is the main economic activity in the Vals River catchment, and is, is much less densely populated the the Klip River catchment. In the Vals River, greater volumes of water diluted plastic from point sources during January (Figure 7.3b; Table 7.2b), but the impacts of isolated point sources were observed in the July samples (Figure 7.3b, 7.4c & d). Unlike the Klip River's sediment MP peak of mean 17 791 n/kg in January (Table 7.2a) in the rainy season, the month with the highest mean MP concentration in Vals River sediment was July (3055 n/kg) in the dry season (Table 7.2b).

A study by Dahms *et al*. (2020), conducted in the Braamfontein Spruit close to the Klip River catchment showed results similar to that found in the Klip River. An increase in filaments (comparable to fragments in this study) associated strongly with higher flow rates. The deeper a river sediment sample was taken, the more fragment type particles were present (Dahms *et al*., 2020). Nel *et al*. (2015) investigated temporal variation in sediment from the Bloukrans River in the Eastern Cape of South Africa and also found increased MP concentrations during low-flow periods. Most data available for South African river plastic data was taken in shorter, lower order rivers in the eastern part of the country and display this trend.

Sporadic dilution was however also seen in the Klip River: Klip 3 downstream of Klip 2 is situated directly below the Olifantsvlei and Bushkoppies WWTPs discharge points and showed the impact of WWTP effluent on the riverine MP load (Figure 7.1a & b). During July, there was a higher concentration of MPs in surface water at this point, likely because the amount of plastic being discharged by the WWTP remained relatively constant, but the amount of water it got diluted in instream was decreased during low-flow periods (Figure 4a &b). Dilution of MPs due to the effluent from large WWTPs was also observed by Schmidt *et al*., (2020), while smaller WWTPs had less of a diluting effect on riverine MPs. The decreased MP concentration in Klip 3 water during January (Figure 4a &b) was thus the result of dilution.

The two tributaries therefore showed contrasting responses in MP concentrations during the wet season. This difference is likely due to the significant difference in the macroplastic waste counted on the shorelines (Table 7.2), emphasising the importance of applying interventions tailored to local situations.

Other studies have found that rainy seasons affect MP concentrations by means of runoff and the resuspension of deposited particles (Talbot *et al*., 2022). Both these were found to be true in our results. Some studies found an increase in MPs during the wet season (Campanale *et al*., 2020; Chen *et al.,* 2020; Eo *et al*., 2019) as was seen in the Klip River, and others found decreases (Weideman *et al*., 2020; Wang *et al*., 2021; de Carvalho *et al.,* 2021) consistent with the trend seen in the Vals River. Some studies have found no association between MP concentrations and rainfall (Mani *et al*., 2020; Mintenig *et al*., 2020).

7.4.2 Flushing MPs downstream

Waste that enters the natural environment can stay deposited in soil or be washed into waterways and broken down by physical and chemical mechanisms (Corcoran, 2022). Little signs of significant continuous downstream carriage were seen in any of the study areas, as was the case elsewhere (Matjašič *et al*., 2023; Horton *et al*., 2018; Sigfried *et al*., 2017). The main-stream Vaal River had more consistent MP concentrations in both water and sediment throughout the sampling period (Table 7.2, Figure 7.4e & f) which suggests that only a small portion of MPs released in the tributaries make it to the mainstream Vaal River and get carried downstream. This is consistent with recent findings that only flooding event cause major transport of MPs in rivers (van Emmerik *et al*., 2023). Mean concentrations in sediment (2575 ± 1631 n/kg) and water (1.58 ± 1.64 n/L) were comparable to that of the Vals River, but with much less variation (Table 7.2). Higher MP concentrations in the Vaal River in both water and sediment during the rainy season (Table 7.2; Figure 7.4e & f) suggest that more MPs were carried to the Vaal River from its tributaries during this period. In dry months, MPs mostly remained trapped in the sediment of tributaries close to point sources.

An increase in stream flow velocity in the Klip River will likely resuspend deposited fragments and beads causing a ‘flush’. An increase in streamflow was related to a decrease in sediment plastic load (Table 7.3) showing that energy from the water effectively flushes MPs from sediment in the Klip River. Microplastics imported by polluted runoff and wastewater will likely get transported further downstream in the Klip River than in the other two rivers in this study.

Little ‘flushing’ was seen in sediment data of the Vals River during periods of increased flow, except for MP concentration in Vals 4 sediment that decreased by a factor of ten from July (10340 n/kg) to January (1100 n/kg) (Figure 7.3b). Water samples at this site saw two orders of magnitude decrease in MP concentrations from October (end of the dry season) to January (Figure 7.3).

7.4.3 Microbeads

Microbeads have the lowest surface-area to volume ratio and settles out from the water column quickly (Kumar *et al*., 2021). It was only beads in Vals River sediment that showed a significant positive association with stream flow velocity (Supp. Table 7.2). Beads, throughout this study were only found close to pollution point sources like an influx of untreated wastewater in the Vals River (Supp Figure 7.1) and Klip River (Table 7.3). An increase in sediment beads was therefore associated with increased stream flow because more beads can be deposited close to the source of the water. A similar trend of beads in sediment was noted in the mainstream Vaal River (Supp. Table 7.1). Untreated waste water thus strongly associated with an increase the sediment load of microbeads.

Microbead concentrations in sediment in the Vals River showed great variation with a %CV of 190% (Table 7.1). The number of beads found in sediment during low-flow (July) (Supp. Fig 7.1) was closely related to the amount of waste water influx in close proximity to the site. The highest concentration of beads in sediment detected throughout the study, and to the best of our knowledge, globally, was at site Vals 4 in July, when river was virtually stagnant with only the influx of raw wastewater causing slight flow. The concentration equated to 0.2% of dry sediment. The site effectively formed a wastewater settling pond with great numbers of microbeads from wastewater trapped in riverine sediment (Figure 7.6). Microbead concentrations in the environment is a function of policy and waste water management practices. Mentions of policy regulating the use of microbeads in South Africa faded around 2019 and currently no restrictions are in place for the distribution and use of microbeads in South Africa.

7.4.4 Impact of local hydrodynamics

Besides seasonal fluctuations in water flow, local hydrodynamics also greatly impacted the MP distribution between water and sediment, as well as morphotypes present, clearly made visible by relatively consistent Vaal River MP concentrations (Figure 7.7). In both water and sediment, during rainy and dry seasons, fragments showed a strong association with turbulent flow (Figure 7.6). Fibres were more prevalent in sediment samples of sites with laminar flow (Figure 7.6), giving easily suspended fibres opportunity to settle out of the water column. Our results concur with a laboratory study determining the deposition rates and transport lengths of fibres and fragments (Hoellein *et al*., 2019). They found that fibres have slower deposition rates and longer transport distances than fragments. This is due to differences in densities and surface area to volume ratios of different morphotypes (Kumar *et al*., 2021).

Other than this, the fibre concentration in rivers showed no significant relation to stream flow of any kind, possibly indicating a completely different source for fibres Table 7,1; Supp Table 7.1). The most likely other source of fibres was atmospheric deposition (Dris *et al*., 2016; Zhang *et al*., 2020). Wastewater, treated or untreated, did not seem to be a source of fibres in the Vaal River system, as was initially expected.

7.5 Conclusions

The initial driver behind plastic and MP research in rivers was the investigation of the function of rivers as plastic carriers to the ocean (Jambeck *et al*., 2015). With an increase in data coming out on riverine MP research, rivers are also starting to be seen as important reservoirs for plastic, and especially MPs (van Emmerik *et al*., 2023). It is therefore critical to begin to understand the dynamics of MPs in rivers as plastic interface with humans and biota.

Temporal variation of microplastics in smaller tributaries are governed by source inputs and local pollution, rather than hydrodynamics. Different streams draining differing land uses and population zones displayed vastly different temporal variations in MP concentrations. Catchment characteristics are key to understanding MP dynamics. Water inputs to polluted streams like the Klip River showed runoff is a major source of pollution in this area of high anthropogenic activity. Runoff in less populated, rural areas like the Vals River catchment tends to dilute MP concentrations in water. Untreated wastewater also adds to the riverine MP load and is strongly associated with the concentrations of microbeads in sediment.

Lower flow during the dry season sees an increase in sediment MP concentrations acting as a temporary sink for MPs. Sediment can be a long-term sink for less buoyant morphotypes like microbeads and fragments but flushing of sediment MPs were seen to a certain extent. This is especially true for the highly polluted and rapid flowing Klip River. With no new inputs from water, turbulent flow can scour the top layers of riverine sediment of most of its MPs, as seen at Skandinawië Drif.

Although little evidence was found for downstream transport of MPs, the mainstream Vaal River had slightly elevated MP concentrations during the rainy season due to increased inputs from tributaries. The effect of local hydrodynamics was much clearer in the mainstream Vaal River. Turbulent flow caused the resuspension of sediment MPs like fragments. Fibres were more prevalent in sediment of sites with laminar flow. A much greater proportion of the MPs at a site is deposited if flow is mainly laminar.

When conducting future studies on temporal variations in South African freshwaters, it is important that catchment characteristics, land use, population, waste, and hydrodynamics be taken into account. It is also advisable to include water and sediment samples to extrapolate more realistic plastic concentrations in rivers. Relatively high concentrations of MPs were found at many sites, and no sample was free of MPs. The highest concentration of microbeads detected as yet, was found in the sediment of the Vals River in Kroonstad, strongly suggesting that national regulatory interventions are advised. Interventions to reduce the MP concentrations in the Vaal River system, improved waste management practices, and more responsible consumer behaviour are the most important issues to tackle. Improving WWTP infrastructure will also reduce the number of MPs that make it to our rivers.

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Chapter 7 Supplementary data

(a) Water means

Klip River

	May	July	October	January	March
Fragments	8.44	5.63	11.04	5.60	
Fibres	0.49	0.61	0.44	0.72	
Beads	2.84	0.56	0.22	0.14	
Total	**11.78**	**6.79**	**11.69**	**6.46**	

Vals River

	May	July	October	January	March
Fragments	0.47	1.68	3.81	0.65	0.20
Fibres	0.13	0.22	1.44	0.16	0.07
Beads	0.00	0.20	0.04	0.16	0.00
Total	**0.61**	**2.09**	**5.29**	**0.97**	**0.27**

Vaal River

	May	July	October	January	March
Fragments		0.17	2.03	1.85	
Fibres		0.08	0.22	0.31	
Beads		0.02	0.05	0.02	
Total		**0.27**	**2.29**	**2.19**	

(b) Sediment means

Klip River

	May	July	October	January	March
Fragments	1885	5281	4370	7705	
Fibres	800	435.7	1355	1212	
Beads	740	662.3	300	1180	
Total	**3425**	**6379**	**6025**	**10097**	

Vals River

	May	July	October	January	March
Fragments	760	1397	1200	737	813
Fibres	73	637	770	440	460
Beads	73	1000	70	43	127
Total	**907**	**3034**	**2040**	**1220**	**1400**

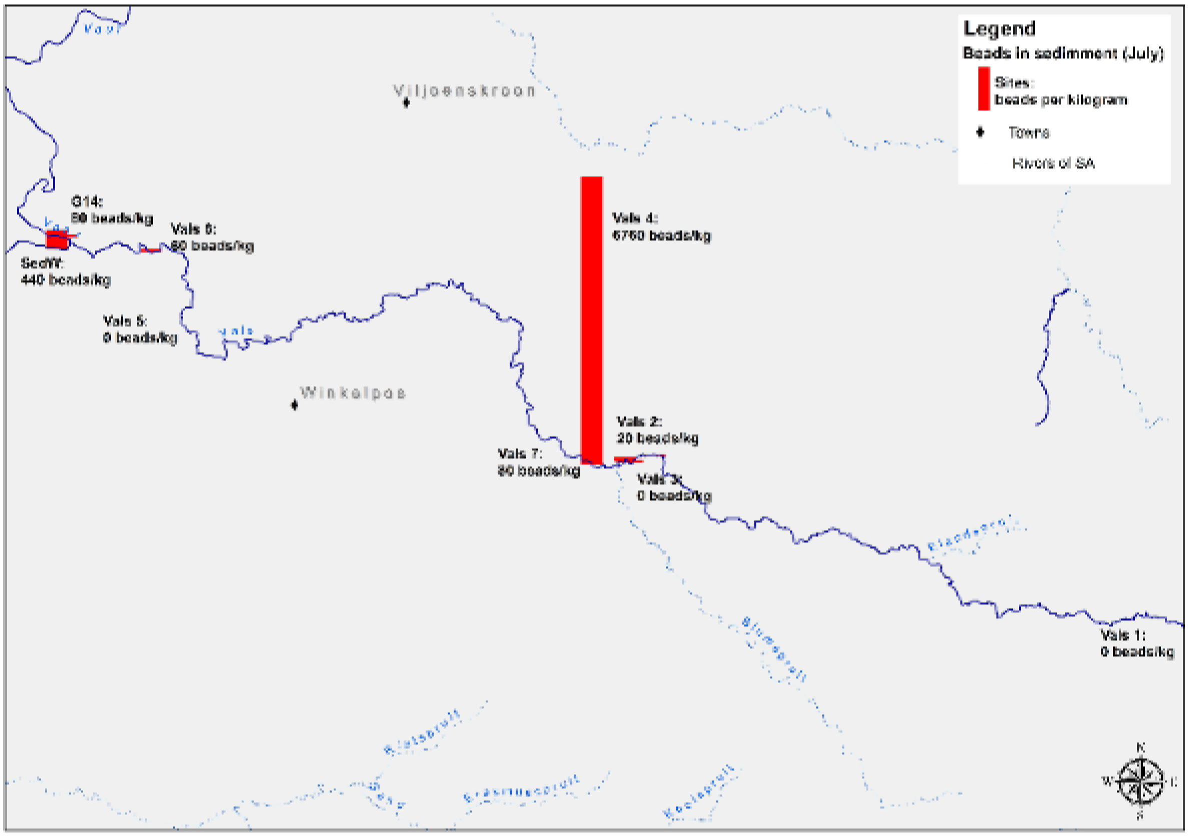
Vaal River

	May	July	October	January	March
Fragments		2604	760	1948	
Fibres		360	580	584	
Beads		204	40	464	
Total		**3168**	**1380**	**2996**	

Supp table 7.1: Mean fragment, fibre and microbead concentrations in each river in water (a) and sediment (b).

Supp. Table 7.2: Mean + Standard deviation of plastic concentrations and metadata parameters per month.

		May	**Jul**	**Oct**	**Nov**	**Jan**	**Mar**	**All**
Water	**Fragments**	3.3 ± 5.2	2.6 ± 2.9	3.32 ± 3.2	7.8 ± 10	2.21 ± 4	0.20 ± 0.1	**2.82 ± 4.1**
	Fibres	0.3 ± 0.4	0.32 ± 0.4	0.74 ± 1.3	0.78 ± 0.6	0.42 ± 0.7	0.07 ± 0.04	**0.42 ± 0.7**
	Beads	0.2 ± 0.5	0.12 ± 0.2	0.11 ± 0.2	0.25 ± 0.4	0.12 ± 0.3	0.0	**0.13 ± 0.3**
	Total	3.5 ± 5.6	3.04 ± 3.2	4.21 ± 4.2	8.12 ± 10.3	2.58 ± 4	0.27 ± 0.1	**3.27 ± 4.4**
Sediment	**Fragments**	1680 ± 1108	3192 ± 3329	2233 ± 2686	1107 ± 763	2980 ± 4734	813.3 ± 888	**2546 ± 3390**
	Fibres	402 ± 816	513.5 ± 677	930.9 ± 467	186.7 ± 257	784.6 ± 1106	460 ± 762	**629.3 ± 820**
	Beads	520 ± 693	494.8 ± 937	145.5 ± 222	146.7 ± 254	486.8 ± 954	126.7 ± 155	**406 ± 784**
	Total	2602 ± 2341	4339 ± 3975	3309 ± 3178	1440 ± 817	4315 ± 6654	1400 ± 1178	**3642 ± 4533**
Macroplastics		69 ± 154	51.05 ± 110	119 ± 187	24.67 ± 16	70.7 ± 119	6.667 ± 7.2	**68.1 ± 132**
Flow (m/s)		0.3 ± 0.3	0.2375 ± 0.3	0.4045 ± 0.7	1.183 ± 1.2	0.385 ± 0.4	0.2833 ± 0.2	**0.3619 ± 0.5**
pH		7.2 ± 0.79	7.187 ± 0.6	7.317 ± 0.6	7.72 ± 0.6	7.298 ± 0.5	6.71 ± 0.2	**7.247 ± 0.6**
DO			18.66 ± 4	20.49 ± 4.1	21 ± 1.6	18.69 ± 5.5	11.37 ± 8.1	**18.72 ± 5**
TDS (200 ppm)		6020 ± 6831	3452 ± 1963	4641 ± 2051	5565 ± 176	3202 ± 1359	201.3 ± 114	**3893 ± 3239**
Sed % silt + clay		10.9 ± 6.5	8.655 ± 6.5	8.117 ± 5.3	11.48 ± 7.6	10.3 ± 6.3	6.388 ± 2.2	**9.412 ± 6.1**



Supp. Figure 7.1: Microbeads in the Vals River during July (dry season)

Chapter 8

Summary and conclusions

8.1 Overview

Plastic is a persistent organic substance, long outliving its usage time, and if not recycled or incinerated, will end up in landfill or the natural environment. South Africa, as part of the African continent, has a growing population and a decreasing capacity to manage the increasing waste that is generated. Waste management and wastewater treatment infrastructure in South Africa is in a poor state. Urgent interventions need to be implemented to curb the adverse effects of plastic and subsequent microplastic (MP) pollution. The main objective of this thesis was to create data and knowledge to advise interventions (nationally and internationally), and identify the most important sources of environmental plastics in the South African context.

Chapter 1 gave a background from literature outlining the history of plastic, issues regarding plastic in the environment and need for data. It also outlined available data on South African MPs and described factors that affect the MP flux, both spatially and temporally.

Chapter 2 is a commentary article (Verster *et al*., 2017 – with updated data) on the socio-economic issues contributing to the environmental plastic load in Africa and South Africa. It also touches on how these issues must be addressed in order to improve South African waste management and reduce the environmental plastic load.

Figure 8.1: Plastic pollution in the Mooi River, North West, South Africa.

Chapter 3 is a scientific review and data analyses on the sources and pathways of South African marine plastic litter (Verster *et al*., 2020). It identified rivers as major pathways to the ocean, but also possible sinks for environmental plastic. It also addressed waste management issues and outlined plastic use and discard statistics in a South African context (Figure 8.2). It identified knowledge gaps and burning issues that need to be addressed. Crucially, the issue of the 'missing plastic' was addressed, finding that initial calculations that South Africa was the 11th worst contributor to marine plastic pollution globally was overstated by a factor of six. Where does the 'missing plastic' go? This was addressed in the forthcoming chapters.

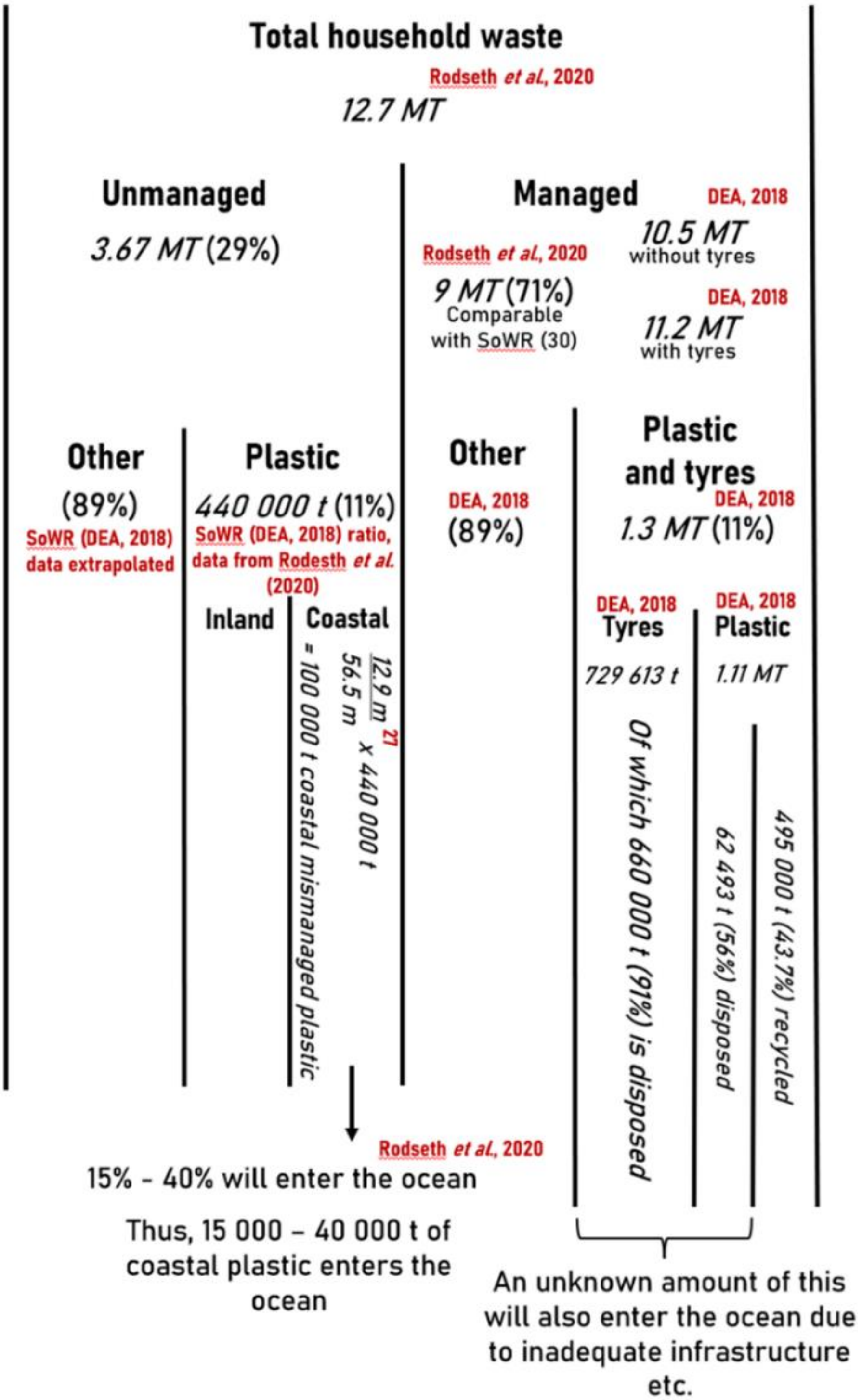


Figure 8.2: Breakdown of available data on household and plastic waste in South Africa (data source in red).

Chapter 4 is a review paper and assessment of airborne microplastics and how MPs could harm human health (Bouwman *et al*., 2018). It speaks to the effects of MP pollution in general and has relevance to this thesis as it shows that not all plastics are on land or water, and that MPs can settle out of the atmosphere, as referenced in Chapters 6 and 7.

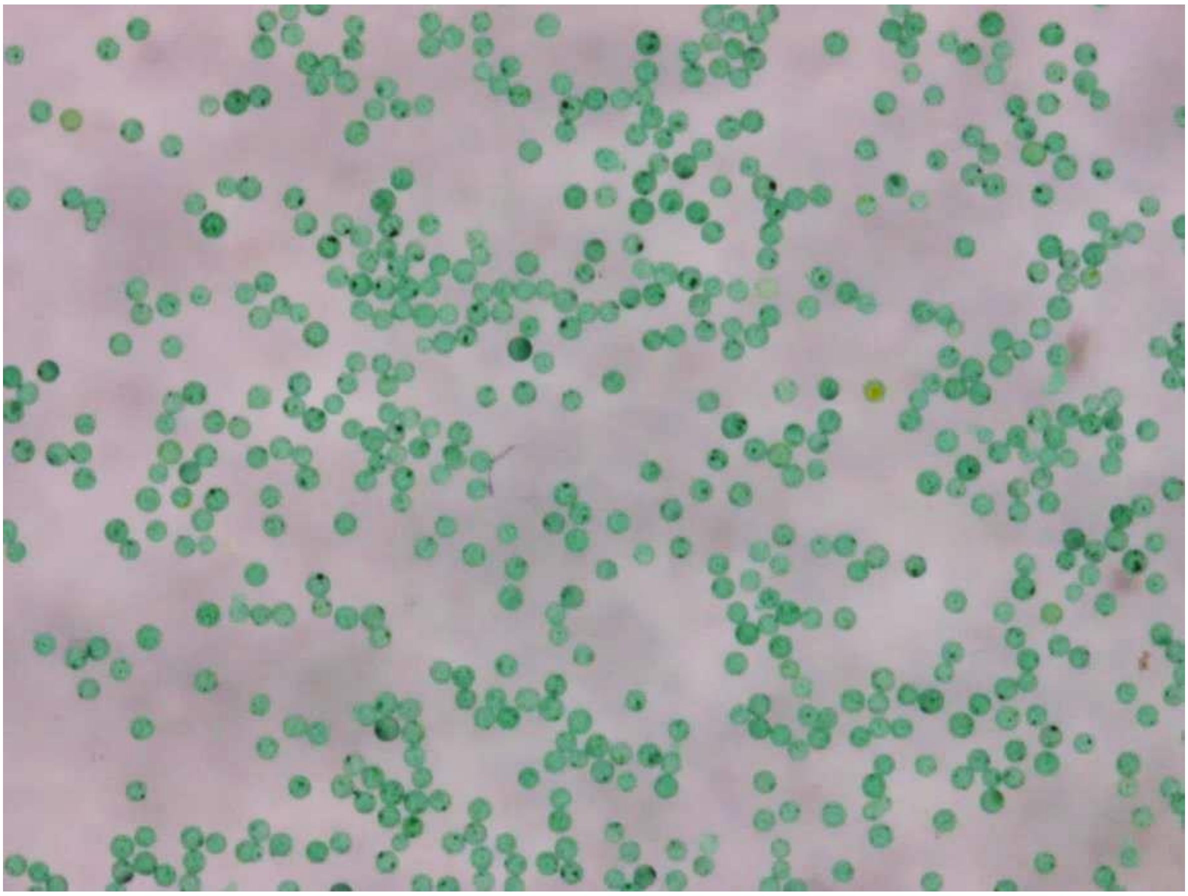


Figure 8.3: Synthetic plastic microbeads

Chapter 5 is a synopsis of the first study conducted in South African inland freshwater MPs (Bouwman *et al*., 2018), rewritten by me as a manuscript. It is a scoping study of surface water, tap water and groundwater from the Gauteng and the North West provinces, to determine baseline levels of MPs in the South African environment. Here, the aim was to identify future research needs and hotspots. From this study, questions regarding inexplicably high MP concentrations in the Vaal River arose. A detailed study of MPs in the Vaal River system was consequently planned, evidenced in the following two chapters.

Chapter 6 describes results in terms of spatial differences of the detailed Vaal River study that followed from the scoping study of Chapter 5. It describes the difference in sources and MP morphotype compositions between three streams—the mainstream Vaal River and two of its tributaries - the Klip and Vals rivers. It concluded that the pollution level of the surrounding environment had the greatest impact on riverine MP concentrations. It also identified different sources for fragments, microbeads, and fibres, and recorded the highest concentration of microbeads in water to the best of knowledge.

Figure 8.4: Microplastic from 90 L of Vaal River water on a custom-made 25 µm stainless-steel sieve

Chapter 7 outlines MP dynamics in water and sediments in a temporal perspective. It compares rainy and dry seasons in the same and different rivers, and describes the effect of stream flow velocity and local hydrodynamics on MP concentrations in both water and sediment. Microplastic concentration changes ranged up to three orders of magnitude over time, highlighting the importance of long-term monitoring.

8.2 Outcomes

Aim 1: Quantify MP pollution in the Vaal River catchment.

Hypothesis 1: High MP abundances will be present in the Vaal River system.

Microplastic concentrations comparable to the most polluted rivers in Asia were detected throughout the Vaal River system. Concentrations per litre of river water generally had three orders of magnitude lower MP concentrations than it corresponding sediment sample per kilogram dry weight (Table 8.1). The heavily polluted Klip River contained the highest MP concentrations. The highest concentration of microbeads to our knowledge in water (11 n/L) and sediment (6760 n/kg, at 0.2% dry weight) were also reported.

Table 8.1: Summary mean microplastic concentration, standard deviations, and percentage coefficient of variations in water and sediment for all 67 sites and %CV overall and per river.

		Water (n/L)		Sediment (n/kg dry mass)	
River	**Sample size**	**Mean & SD**	**%CV**	**Mean**	**%CV**
All rivers	67	3.4 ± 4.4	146	3600 ± 4500	89
Klip River	24	6.1 ± 5.8	112	6200 ± 6400	70
Vals River	24	1.9 ± 2.8	42	1900 ± 2400	76
Vaal River	17	1.6 ± 1.6	55	2600 ± 1600	26

c

Aim 2: Identify plastic pollution sources.

Hypothesis 2: Local environmental plastic pollution is the major predictor for riverine MP load, meaning improper waste management is the main cause of MP pollution in South Africa

Microplastic in water and sediment showed a strong correlation with the number of macroplastics surrounding the river at the sampling points. The most densely populated and urbanised river catchment also contained the highest MP concentrations in both water and sediment. Waste mismanagement was also prevalent in this area, and MP concentrations associated the strongest with mismanaged waste.

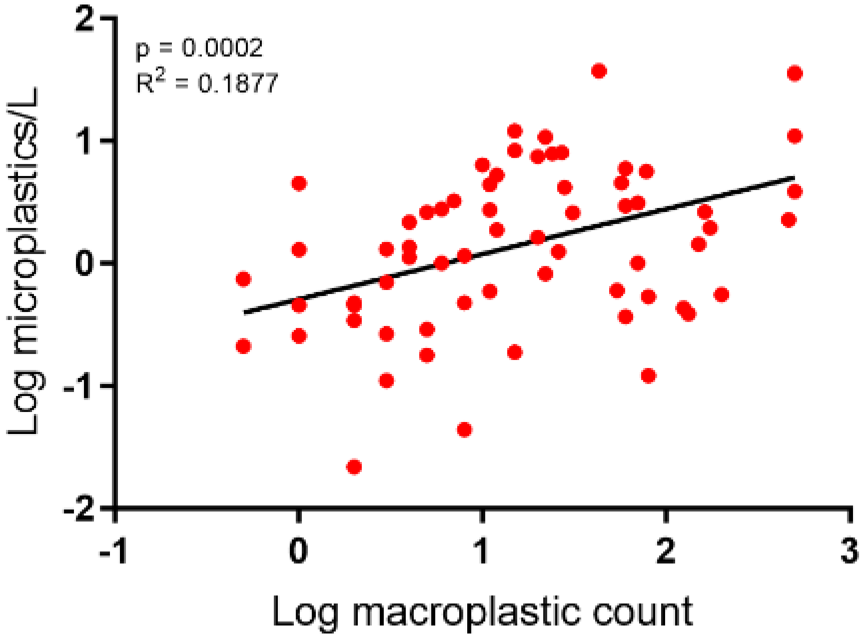


Figure 8.5: Linear regression of log transformed macroplastic scores with microplastic concentrations in water.

Hypothesis 3: Different microplastic morphotypes (fragments, fibres, and beads) have different sources and pathways.

The three morphotypes identified in this study were fragments, fibres, and microbeads. Fragments were the most prevalent morphotype throughout the study and showed the strongest association with environmental macroplastic pollution. Fragments are thus the breakdown products of larger plastic pieces and tend to settle out into sediment soon after entering a river. Fibres had relatively constant, low concentrations throughout the study period and area. Atmospheric deposition was the most likely source of of fibres, as it showed little increase close to other possible sources. Microbeads were localised close to point sources like untreated and insufficiently treated wastewater, where it rapidly settled into sediment. The three morphotypes therefore had different sources and dynamics, reflected in their compositions in both water and sediments.

Aim 3: Describe sinks of MPs in South Africa.

Hypothesis 4: During low flow periods, sediments act as temporary sink for MP.

MPs in sediment reached a maximum during July in the dry winter season, even though MP concentrations in water was not at its highest (Table 8.2). Lower flow periods with less in-stream energy causes rapid settling of MPs, especially microbeads. Measures of resuspension was noted again during higher flow periods, or at sites with more turbulent flow.

Table 8.2: Means and standard deviations of microplastic morphotypes (fragments, fibres, and microbeads) concentrations per month of all sites.

		May	*Jul*	*Oct*	*Nov*	*Jan*	*Mar*	*All*
Water	Fragments	3.3 ± 5.2	2.6 ± 2.9	3.2 ± 3.2	7.8 ± 10	2.2 ± 4	0.2 ± 0.1	2.8 ± 4.1
	Fibres	0.3 ± 0.4	0.3 ± 0.4	0.7 ± 1.3	0.8 ± 0.6	0.4 ± 0.7	0.1 ± 0.04	0.4 ± 0.7
	Beads	0.2 ± 0.5	0.1 ± 0.2	0.1 ± 0.2	0.3 ± 0.4	0.1 ± 0.3	0.0	0.1 ± 0.3
	Total	3.5 ± 5.6	3.0 ± 3.2	4.2 ± 4.2	8.1 ± 10.3	2.6 ± 4.0	0.3 ± 0.1	3.3 ± 4.4
Sediment	Fragments	1680 ± 1108	3192 ± 3329	2233 ± 2686	1107 ± 763	2980 ± 4734	813 ± 888	2546 ± 3390
	Fibres	402 ± 816	513.5 ± 677	931 ± 467	186.7 ± 257	785 ± 1106	460 ± 762	629.3 ± 820
	Beads	520 ± 693	494.8 ± 937	145.5 ± 222	146.7 ± 254	487 ± 954	127 ± 155	406 ± 784
	Total	2602 ± 2341	4339 ± 3975	3309 ± 3178	1440 ± 817	4315 ± 6654	1400 ± 1178	3642 ± 4533

Hypothesis 5: Wetlands act as MP filters, trapping it in wetland sediment as sink.

Two sites in the Klip River highlighted the effect of wetlands on the riverine MP load. Near 100% of MPs present in water upstream of the wetland were trapped in wetland sediments during low flow periods. Wetlands are thus effective in removing MPs from rivers, and wetland sediment is likely a major sink for plastics in rivers. Given the importance and sensitivity of wetlands, this increasing accumulation is of great concern.

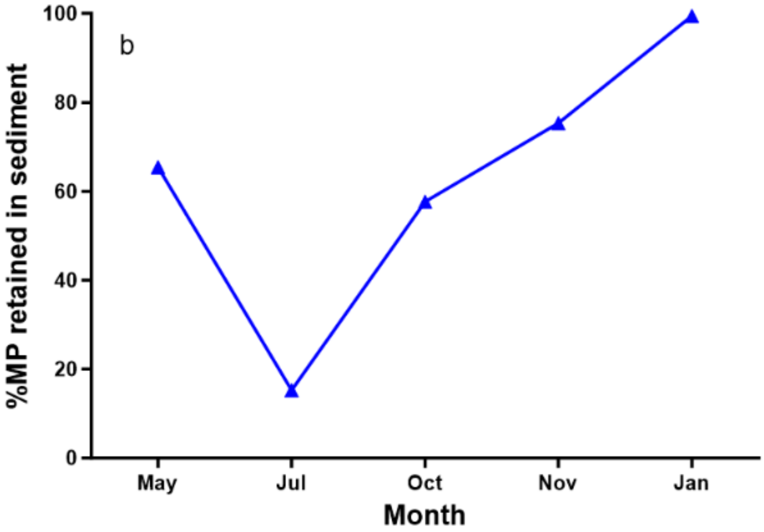


Figure 8.6: Proportion of microplastics from water at Klip 4 retained in wetland sediment at Klip 5.

Aim 4: Describe temporal patterns of MPs for South African freshwater systems.

Hypothesis 6: Riverine MP load is affected by flow rate.

The two tributaries of the Vaal River investigated in this study—the heavily polluted Klip River and the cleaner, less impacted Vals River—showed opposing trend during the rainy season. The Klip River water saw an increase in water and sediment MP concentrations, while the

Vals River showed dilution by runoff during the rainy season. Klip River runoff is more polluted than that of the Vals River and shows that results from one catchment cannot necessarily be extrapolated to another.

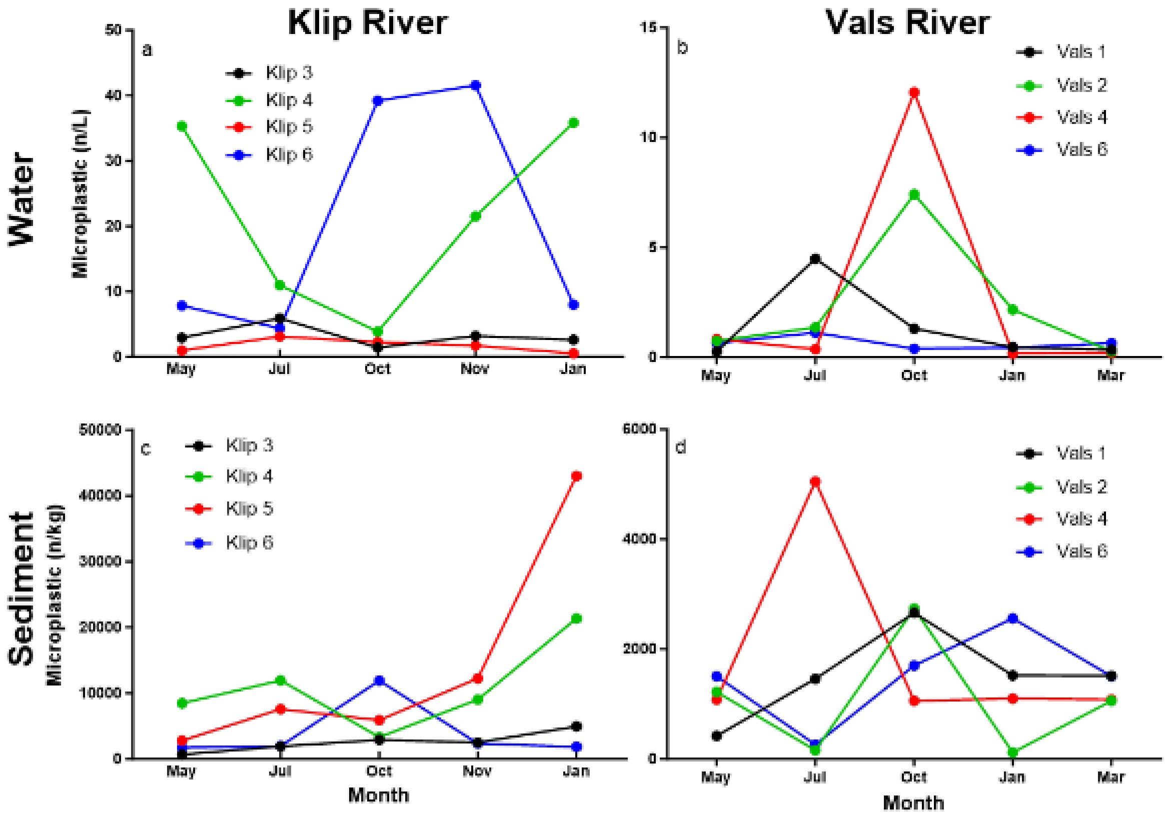


Figure 8.7: Temporal changes of microplastic (MP) concentrations in water and sediment of frequently sampled sites along the Klip (a & b) and Vals (c & d) rivers

<u>Aim 5: Propose mitigating actions to reduce freshwater plastic pollution in South Africa.</u>

- Maintaining and improving waste management infrastructure and services is the most important intervention to curb plastic pollution in Africa and South Africa.
- Increasing capacity and functionality of WWTPs will improve plastic removal by WWTPs and decrease MPs released into rivers.
- Banning or severely restricting the production and use of microbeads will help to mitigate their environmental impacts. Thise that are already trapped in sediment may already have severe consequences.
- Public awareness and environmental education must be increased. Long-term action will only be sustainable if all members of society, government, and industry take responsibility for their environment.
- Conduct regular river cleanups and install litter booms. Most MPs originate from the breakdown of larger plastic pieces and will most likely not be removed once it has broken down. Removing macroplastics from the environment is therefore crucial.

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