MICROPLASTICS IN THE NZ WATER ENVIRONMENT – SHOULD WE BE WORRIED?

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ABSTRACT

One of the most topical emerging contaminants in the water environment is microplastics. There are many sources of these microconstituents in our wastewater streams due to the ubiquity of plastics used in modern society, thereby becoming prevalent in all municipal and industrial waste streams, and in the water environment. The quantity and composition of microplastics entering wastewater treatment plants and ending up either captured or released into the environment are the subject of much international research effort. If captured in the liquid stream processes, there is also great interest in which stage microplastics ultimately sequester and what impact they have on the quality and end-uses of biosolids and reuse water streams, as well as how macro-plastics break down, over time, into micro- and nano-plastics.

It appears from university research and monitoring at USA and European WWTPs (*Microplastics in the Water Environment: Should We Be Worried and Why?,* Knowledge Development Forum: WEFTEC 2020, New Orleans) that conventional secondary and tertiary treatment processes are very successful at removing a range of microplastics, however given the large effluent flow volumes, even small concentrations of micro- and nano-plastics can still be a large burden discharged to the marine and freshwater environment. This is without any consideration of how stormwater discharges with macro- and micro-plastic content are also contributing to this contamination of our environment.

This session, of significance to wastewater systems and stormwater operators and managers, will bring together a range of national and international research to discuss the current state of knowledge on microplastics, and where our research and management efforts on microplastics should be focused in the near and long-term, as well as implications for treatment process selection and standardisation of analytical techniques. In New Zealand, we can expect in the near future higher standards for our WWTP discharges – and probably our stormwater discharges - and we must be in a position to determine the optimum liquid stream and solid stream processes to ensure that microplastics do not become any more of a problem in our aquatic and land environments than they are now.

KEYWORDS

Microplastics, nanoplastics, biosolids, treatment process, WWTP, discharge, water environment

PRESENTER PROFILE

Garry is widely recognised as an expert in wastewater engineering with over 43 years' experience in a wide variety of wastewater projects, both in New Zealand and abroad.

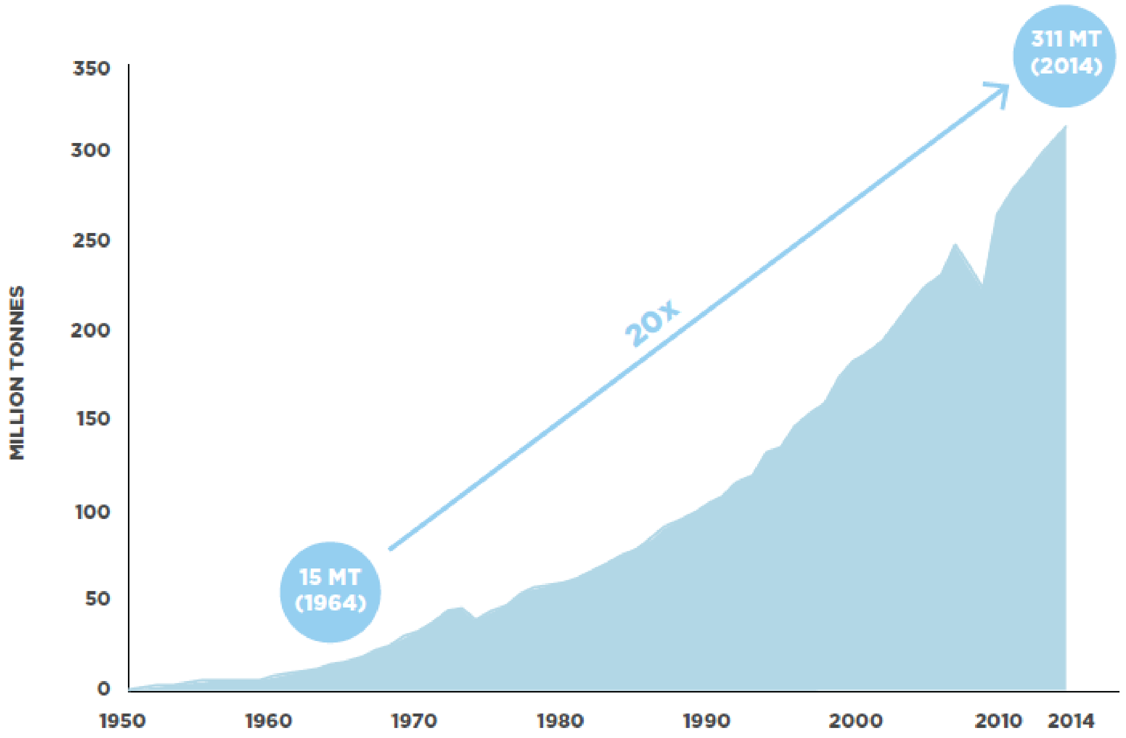
Bridget's background is in water and wastewater engineering, sustainability, and systems engineering. She specialises in applying design thinking to complex issues and is currently focussing on quantifying capital carbon and climate change impacts.

INTRODUCTION – WHAT IS THE PROBLEM?

Plastics in the 21st century are ubiquitous no matter where one is in the world. They have become a "necessary evil" in every facet of our everyday lives, from food and drink containers, to wrapping and packaging, to material use in all forms of construction. Unfortunately, plastics also comprise a large volume of "single-use" products and materials and therefore create a huge waste materials problem in every country. These features of, and issues with plastics, are compounded by the fact that many plastic products are extremely durable and chemically resistant, which has contributed to its mass accumulation in the biosphere, however, studies by Andrady et al. (2003) and Barnes (2004), have shown that plastic will fragment under photo-oxidative and mechanical stress. This fragmentation results in a subset of plastic pollution termed microplastics (MPs), characterized by plastic particles smaller than 5mm.

The volume of microplastics is accelerating at much the same rate in which the production of plastics has exploded in the last 60 years, growing at some 4% per annum (refer to Figure 1). Microplastics represent a unique environmental threat due to the potential transport of toxic chemical and additives through their bioavailability and complex interaction within the food-web. This paper will illustrate how microplastics enter wastewater systems and where they are removed or accumulate in treatment plants.

Figure 1: Growth in global plastics production from 1950 to 2014. Production from virgin petroleum-based feedstock only (does not include bio-based, greenhouse gas-based or recycled feedstock). Source: thecivilengineer.org

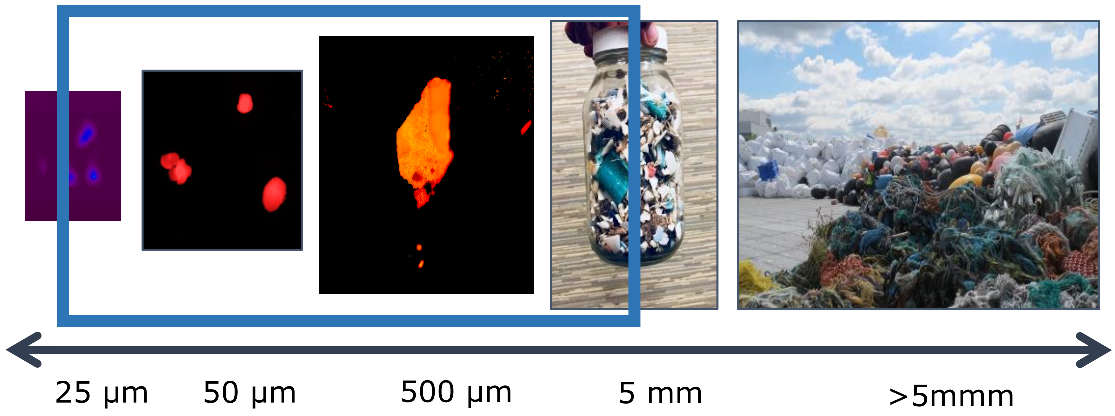


MICROPLASTICS – WHERE DO THEY COME FROM?

Microplastics are very small pieces of plastic that enter the environment in a number of ways which are addressed in more detail below. Microplastics are categorized as either primary or secondary microplastics. Primary microplastics are any plastic fragments or particles that are already 5.0 mm in size or less before entering the environment. These include microfibers from clothing, microbeads, and plastic pellets (also known as nurdles). Secondary microplastics results from the fragmentation mentioned before and have less uniform shape.

A useful size comparator of macro and microplastics is shown in Figure 2, with the conventionally accepted range of microplastics highlighted.

Figure 2: Size range of plastic particles.



MICROPLASTICS – WHERE DO THEY END UP?

The majority of macro- and microplastics end up in the water environment, being transported through sewerage and stormwater systems to lakes, streams and rivers, and thence into the oceans. Macroplastics are, by their nature, more visible, as well as highly buoyant, and much has been written and featured about the vast "plastic rafts" in the world's oceans (University of Florida, 2016) caused by the accumulation of plastic wastes flushed from the land or disposed directly to sea.

In comparison, microplastics are less visible but more highly mobile, and therefore of more concern to global ecosystems as they are ingested by animals, fish and other marine organisms such as zooplankton, thereby finding their way into the human food chain. There are predictions (thecivilengineer.org, 2016) that this situation will worsen in the next decades (refer Figure 3 and Figure 4) unless the use of plastics decreases and the amount of microplastics discharged to the environment is controlled and curtailed.

Figure 3: Predicted growth in plastic production 2014 to 2050. Source: thecivilengineer.org

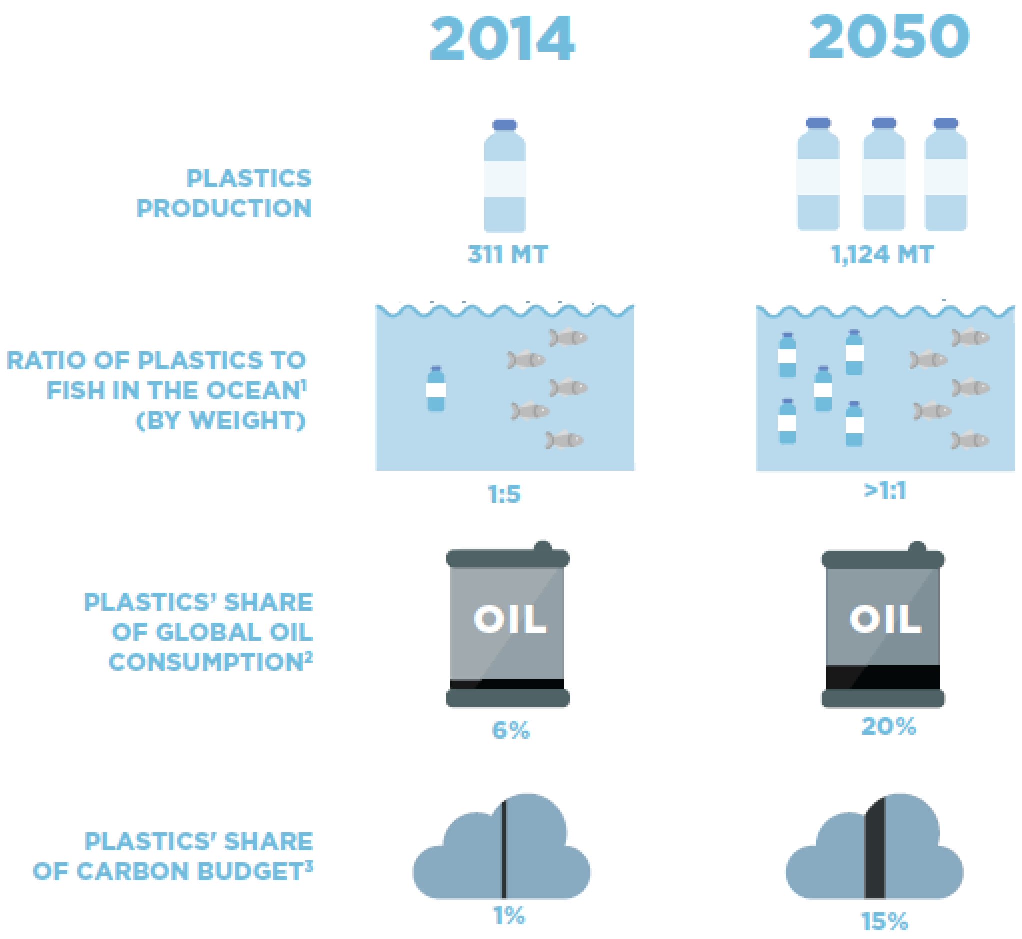
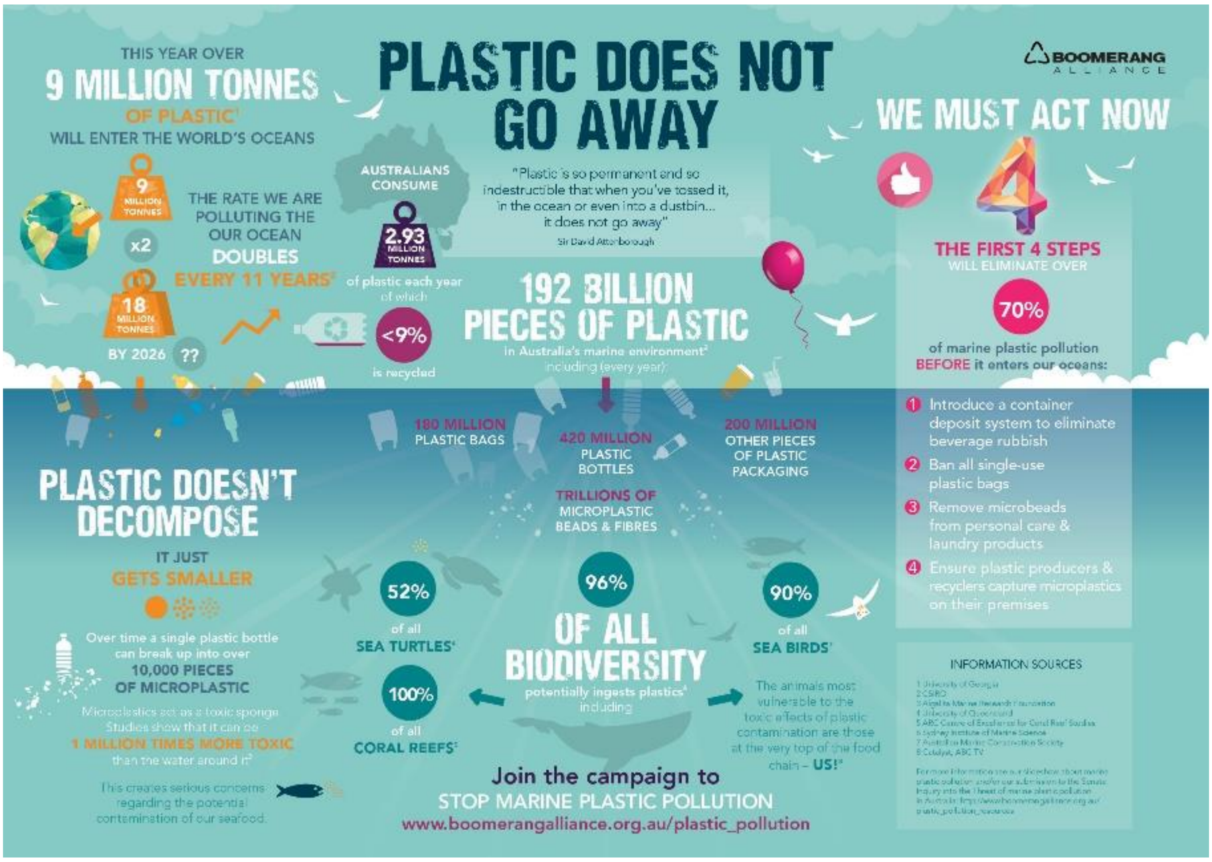


Figure 4: Infographic showing scale of plastic pollution in the marine environment. Source: Boomerang Alliance



HOW DO WE ANALYSE FOR THEM?

The analyses for microplastics starts with their separation from the environment from which they are being sampled. Clearly, separation from aqueous samples is easier than from soil or sludge substrates, and is most frequently undertaken through a series of washed sieves in the 25 μm to 5mm range (Enders et al. 2020).

Microplastic quantification can be undertaken in a number of ways, with the main distinction being between "visual/physical" and "analytical chemical" methods. Each has their own features, benefits and issues and these are summarized in Table 1. For more information on these methods, the reader is referred to Hidalgo-Ruz et al. (2012).

Table 1: Comparison in visual / physical versus analytical chemical methods for microplastic identification and quantification. Source: Hidalgo-Ruz et al. (2012)

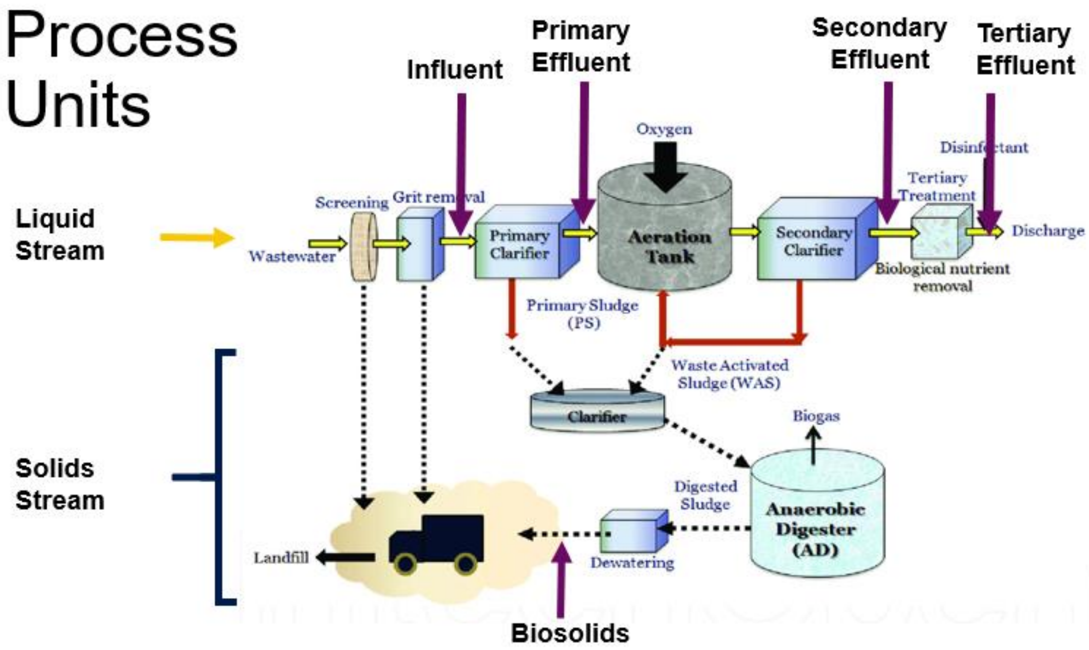
Visual/Physical Methods for MPs - **Quick processing times** - **High-throughput** - **User Bias**	**Analytical Chemical Methods for MPs** - **Direct identification** - **Identification confidence** - **Time consuming**
Visual Identification of Plastics - User bias - Human error	Fourier Transform Infrared Spectroscopy - Widely used in microplastics research - FPA: plane imaging removes bias - Time consuming - Largely unavailable to our lab
Small Anthropogenic Litter (SAL) - Removes user bias - True MP loading/impact unknown	X-Ray Photoelectron Spectroscopy - Inexpensive to run & available - Point imaging introduces user bias - Uncommon for microplastics
Nile Red Staining - Removes user bias - More accurate MP loading rates - Some contaminants are also stained - Validation by chemical analysis needed for future use	Energy-Dispersive X-Ray Spectroscopy - Less expensive to run - Moderate Availability - Claims of high-throughput capacity - Uncommon for microplastics

WHAT HAPPENS IN WWTPS?

TYPICAL MICROPLASTICS REDUCTIONS IN WWTPS

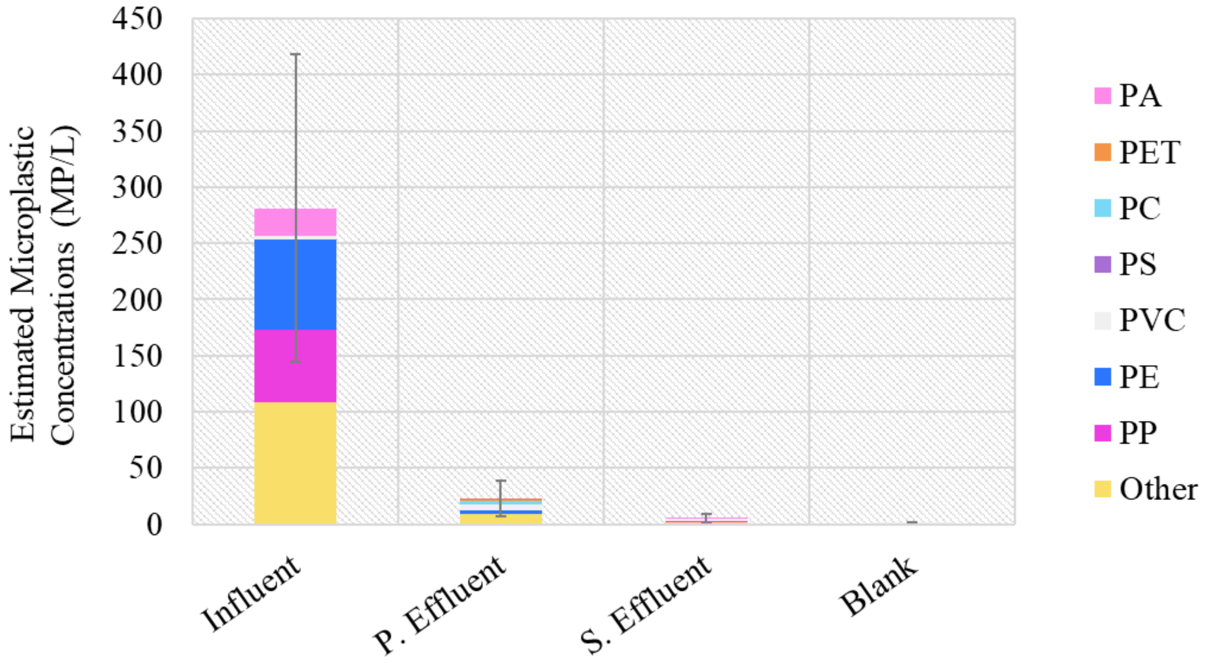
Figure 5 shows a typical schematic of a wastewater treatment facility. Fahrenfeld et al. (2019) selected sampling locations to examine the inflow and outflow of microplastics from unit processes. In addition to sampling the wastewater, a 20 L blank sample of DI water was run at the end of the sample event to quantify contamination. Preliminary results from FTIR analysis showed 91% removal of total microplastic concentrations in the primary clarifier and an additional 7% removal in secondary treatment.

Figure 5: Typical schematic of a wastewater treatment facility showing sampling locations to examine the inflow and outflow of microplastics from unit processes. Adapted from Koye, Elbeshbishy, and Elsayed (2019)



As shown in Figure 6, WWTPs have been shown to be reasonably effective at removing microplastics, with removal efficiencies in the 75%–100% range for conventional WWTPs utilizing activated sludge processes and secondary clarification.

Figure 6: Concentrations of microplastics in a WWTP – influent vs primary and secondary effluent. Source: Macdonald, G. J., Sturm, B., and Fahrenfeld, N. (2020)



High polyethylene and “other” microplastics fractions were observed throughout the plant. The other microplastics category represented biodegradable, copolymer, additive microplastics. Future clustering analysis aims to further delineate this group based on more advanced speciation.

Primary and secondary effluent sampling relied on grab samples for simplicity, the large variance in microplastics composition in the samples support switching to 24-hr composite samples for more representative triplicate samples.

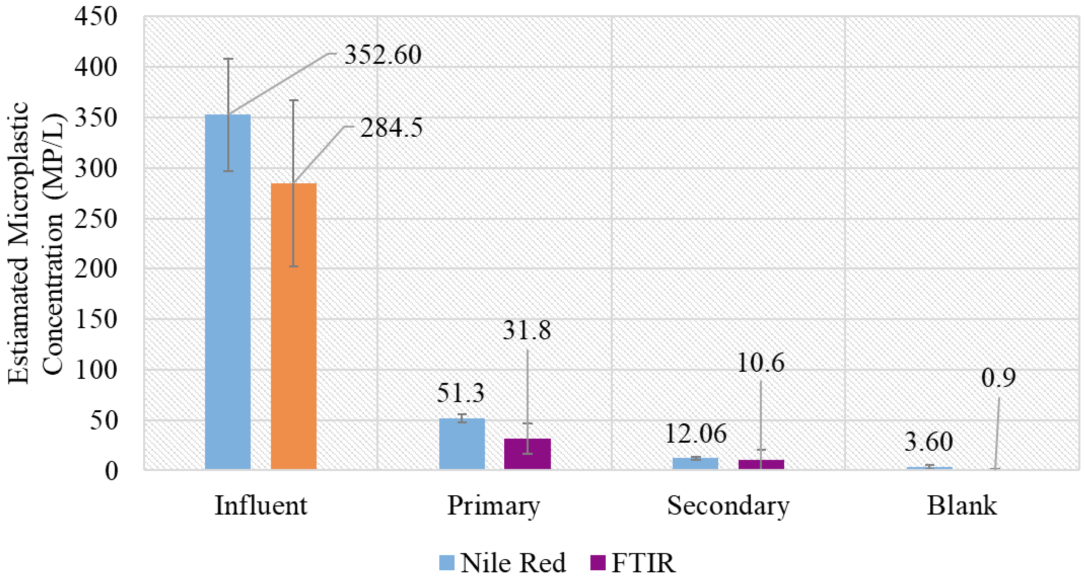
Very little evidence of cross-contamination existed in the blank samples and it is worth pointing out that these microplastics concentrations were estimated using a conservative identification criterion so we would expect these numbers to be underestimates.

Not all WWTPs perform this way – in New Zealand, some advanced plants (such as in Christchurch) are able to achieve 97% removal (Ruffell et al., 2021). On the other hand, some plants do not result in a change to the level of microplastics (measured by number), as big pieces tend to get broken up as they pass through the system, especially if they go through the recycled activated sludge (RAS) stream and come back through the process.

DIFFERENCES IN ESTIMATED MICROPLASTICS REDUCTIONS BY MEASUREMENT METHOD

Using the Nile Red method, microplastic concentrations in a WWTP were observed to decrease an average of 85.4% due to primary clarification and 11.1% due to secondary treatment for a total removal of 96.6% (Macdonald, G. J., Sturm, B., and Fahrenfeld, N., 2020), as shown in Figure 7. However, the results using FTIR measurements were consistently lower than those found by the Nile Red method.

Figure 7: Nile Red vs FTIR microplastic concentrations. Source: Macdonald, G. J., Sturm, B., and Fahrenfeld, N. (2020)



In this case, cross-contamination observed in the Nile Red method was significant and would be factored in before comparing results from other facilities. Results from the same triplicate samples showed statistical similarity to the FTIR results (0.86 correlation and P=0.1566). There still is a need to research the impacts of common contaminants on Nile Red Staining methods, however these results suggest Nile Red concentrations could still serve as a conservative parameter for microplastic concentrations.

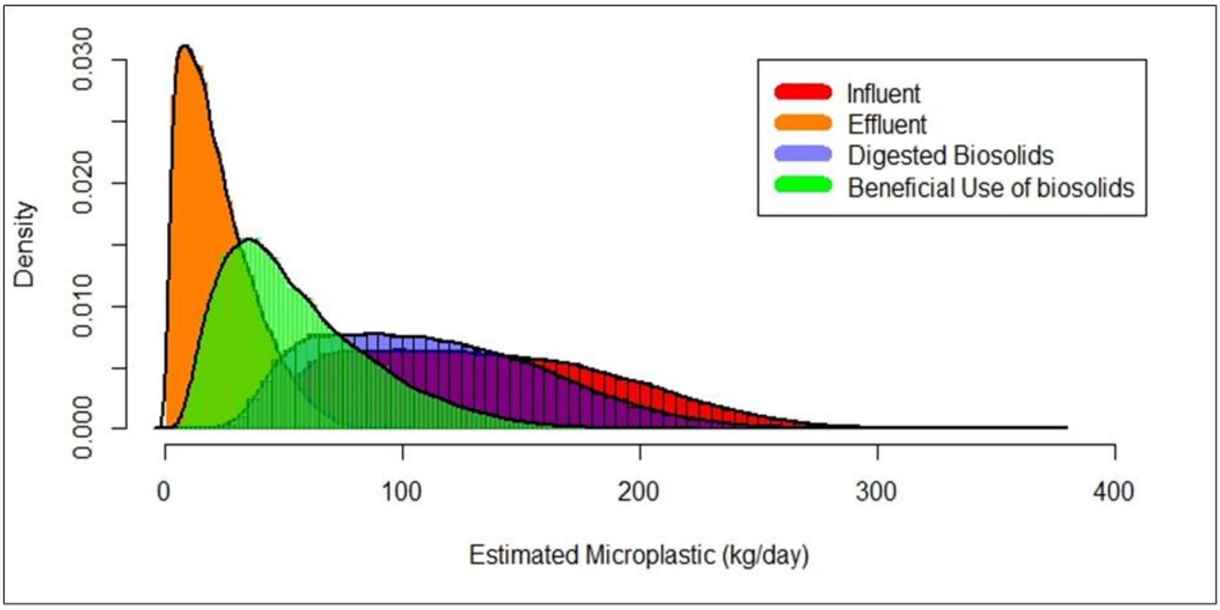
SO WHERE DO THE MICROPLASTICS GO?

While microplastics concentrations in effluent are lower than in influent indicating that they are effectively being removed from the liquid stream in WWTPs, they are not being digested – and so they need to go somewhere. Where they are removed from the liquid stream, they end up in the biosolids, and become a pollution problem associated with the solids' disposal method.

Influent concentrations of microplastics have been found at 16 plastic particles / L effluent. In the US, where 160 trillion liters of wastewater are generated per day, an estimated 256 trillion plastic particles / day are passing through wastewater treatment plants (WWTPs) (Macdonald, G. J., Sturm, B., and Fahrenfeld, N., 2020). WWTPs on the East and West Coasts of the USA will receive the majority of plastic contaminants due to the greater population density in these areas.

Fahrenfeld (2019) found that 99.7% of microplastics settled in digested sludge, which could be disposed for beneficial use. Figure 8 shows the typical concentrations found in biosolids, in comparison to WWTP influent and effluent. Only 0.02-0.3% of the microplastic remains in the effluent.

Figure 8: Typical load and density of microplastics found in WWTP liquid and solid streams. Source: Macdonald, G. J., Sturm, B., and Fahrenfeld, N. (2020)



In New Zealand, nearly one quarter (23.4%) of biosolids produced are disposed of by open land application (refer Figure 9 and Table 2, excluding quarry rehabilitation which is presumably covered by clean fill), from where it can be returned to the water environment either via stormwater runoff, being made air-borne by wind, or entering the food chain through agricultural processes and ending up back at the WWTP.

Given the high concentrations of microplastics found in biosolids, disposing of biosolids through types of land application where they are then exposed to the elements and able to re-enter the water cycle, implies that a significant part of the microplastics problem is simply being temporarily diverted from the water environment rather than permanently removed.

Figure 9: Sankey Diagram showing wet tonnes of end product of surveyed NZ utilities by treatment processes and end-fates. Source: Tinholt (2019)

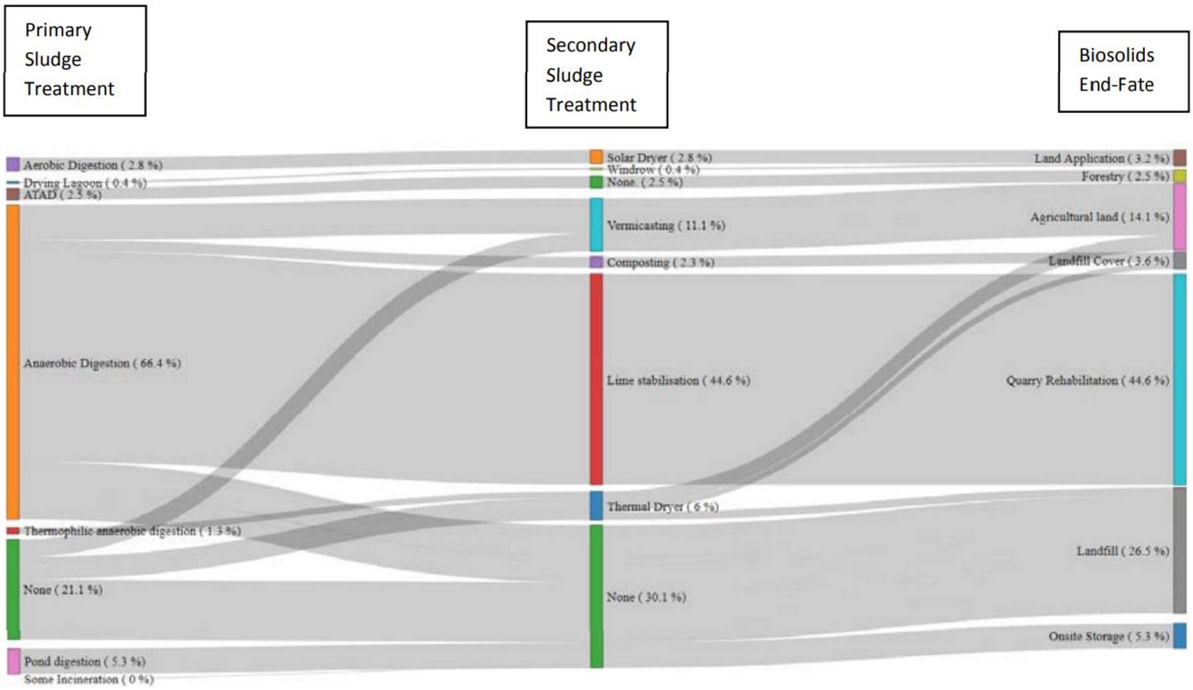


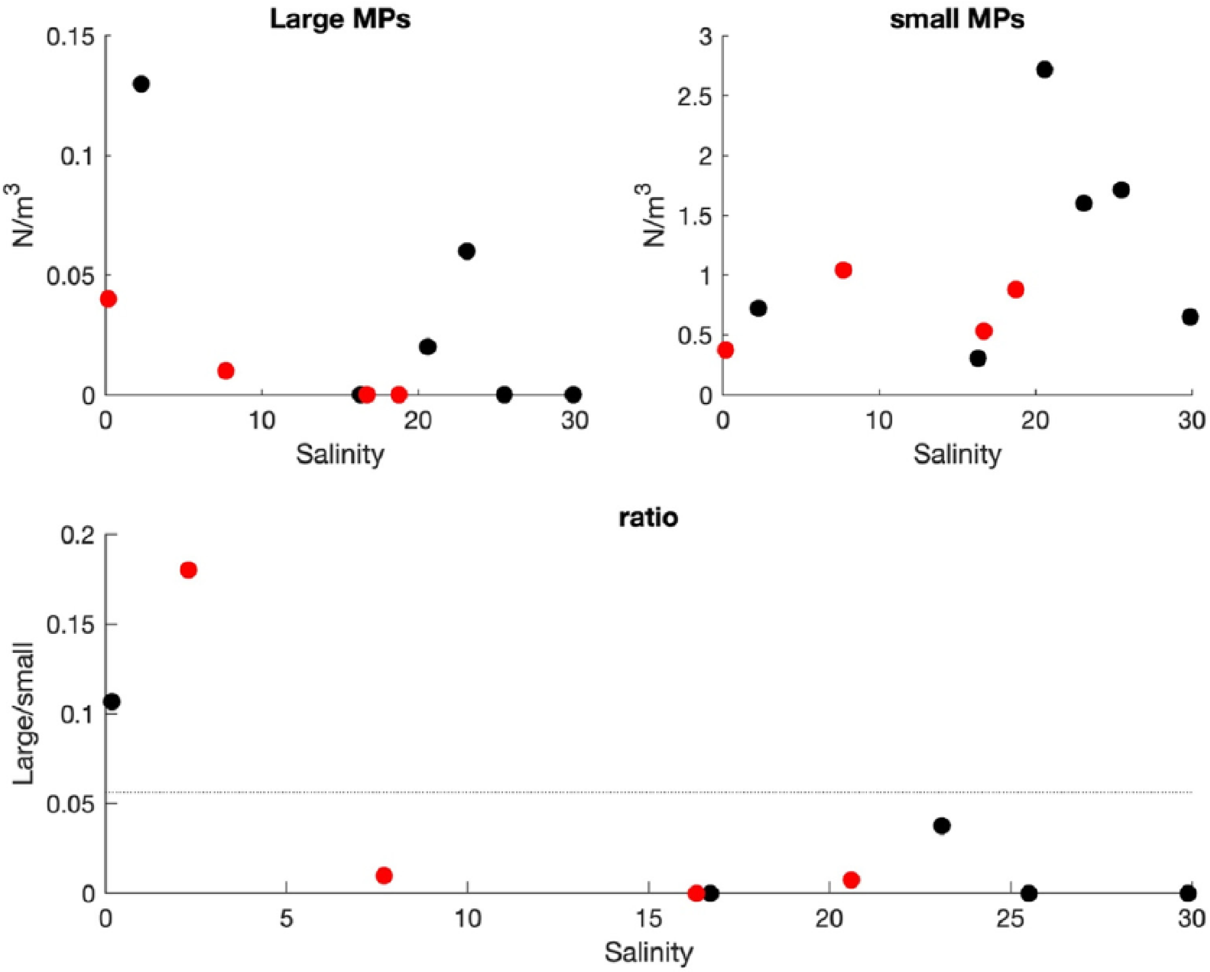
Table 2: End product percentage (dry solids) of surveyed NZ utilities by end-fate. Source: Tinholt (2019)

End Fate	**End product percentage by wet tonnes for surveyed WWTPs**	**Classification**
Land Application	3%	68% Resource Recovery
Forestry	2%	
Agricultural Land	14%	
Landfill Cover	4%	
Quarry Rehabilitation	45%	
Landfill	27%	32% Waste
Onsite Storage	5%	

FRESHWATER AS A NEW SOURCE OF MICROPLASTICS

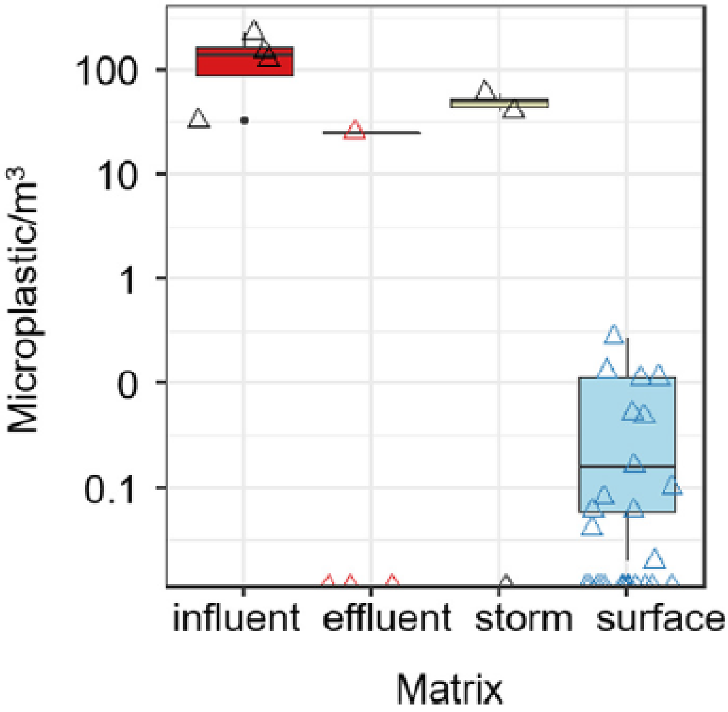
A recent study by Bailey et al. (2021), which looked at the size and concentration of microplastics along a river environment (at locations of increasing salinity) and in the plume of riverine sediment encroaching into a bay in the coastal marine environment of the East Coast of the USA, found that microplastics size decreased between the freshwater and saltwater samples, with concentration of large particles higher in less saline water and concentration of smaller particles higher in higher salinity (refer Figure 10) – indicating that the river is a source of microplastic pollution in that area.

Figure 10: Large microplastics (MPs) (upper left) and small microplastics (upper right) as a function of salinity on April 2019 surveys. Black dots are for 4/11 and red for 4/16. Lower panel shows results of Fragmentation model. Dashed horizontal line shows the ratio of large microplastics to small microplastics based on fragmentation. Source: Bailey et al. (2021)



Microplastics were observed in every sample type (surface water, storm water, wastewater), as shown in **Error! Not a valid bookmark self-reference.**. In surface water samples, microplastic concentrations for the 500-2000 µm particles were the highest in the river and lowest in the samples collected in the highest salinity water where Raritan Bay meets the coastal ocean – suggesting the river is a source that is diluted as it enters the estuary. In contrast, the highest estimated MP concentrations for the 250-5000 µm samples were located in the mid-Raritan Bay in the vicinity of the Hudson River.

Figure 11: Boxplot with jitter (open triangles) of 500-2000 µm microplastic concentration on log scale of wastewater influent ("influent", N = 4), wastewater effluent ("effluent", N = 4), stormwater (N = 3), and surface water (N = 26). Data points intersecting the x-axis had <1 microplastic per cubic metre. Source: Bailey et al. (2021)



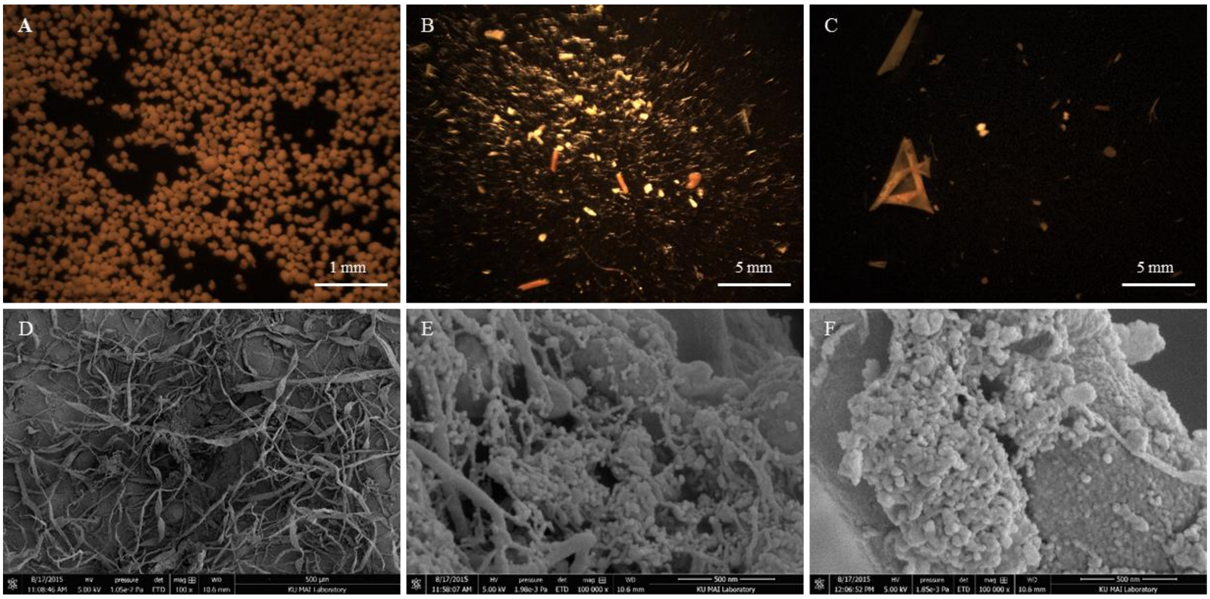
One striking result is the tendency for large microplastics to be present in the freshwater end of the Raritan River while the smaller size class of microplastics was most prevalent in the mid-Raritan Bay River plume. The ratio of large microplastics to small microplastics was significantly lower than predicted by a fragmentation model. Thus, this suggests that the source of the smaller microplastics is the Hudson River.

FTIR and/or Raman analyses demonstrated that polyethylene, polypropylene, and rubber were predominant polymer classes observed in the bay.

MICROPLASTICS AS A VECTOR FOR PATHOGENIC BACTERIA?

The presence of microplastics in WWTPs means they are in contact with pathogenic organisms and could thus act as a vector for pathogens to enter the environment by attaching to the microplastics' surfaces. Figure 12 shows biofilm on microplastics sampled from a WWTP's influent and effluent.

Figure 12: Light microscopic images (top) and scanning electron microscopic images (bottom) of microplastic (A) PVC control pellets (B) Influent (C) Activated sludge (D) microplastic thread like structure and bacterial biofilm on the plastic surface in (E) influent and (F) activated sludge samples taken from Lawrence WWTP. Source: Macdonald, G. J., Sturm, B., and Fahrenfeld, N. (2020)

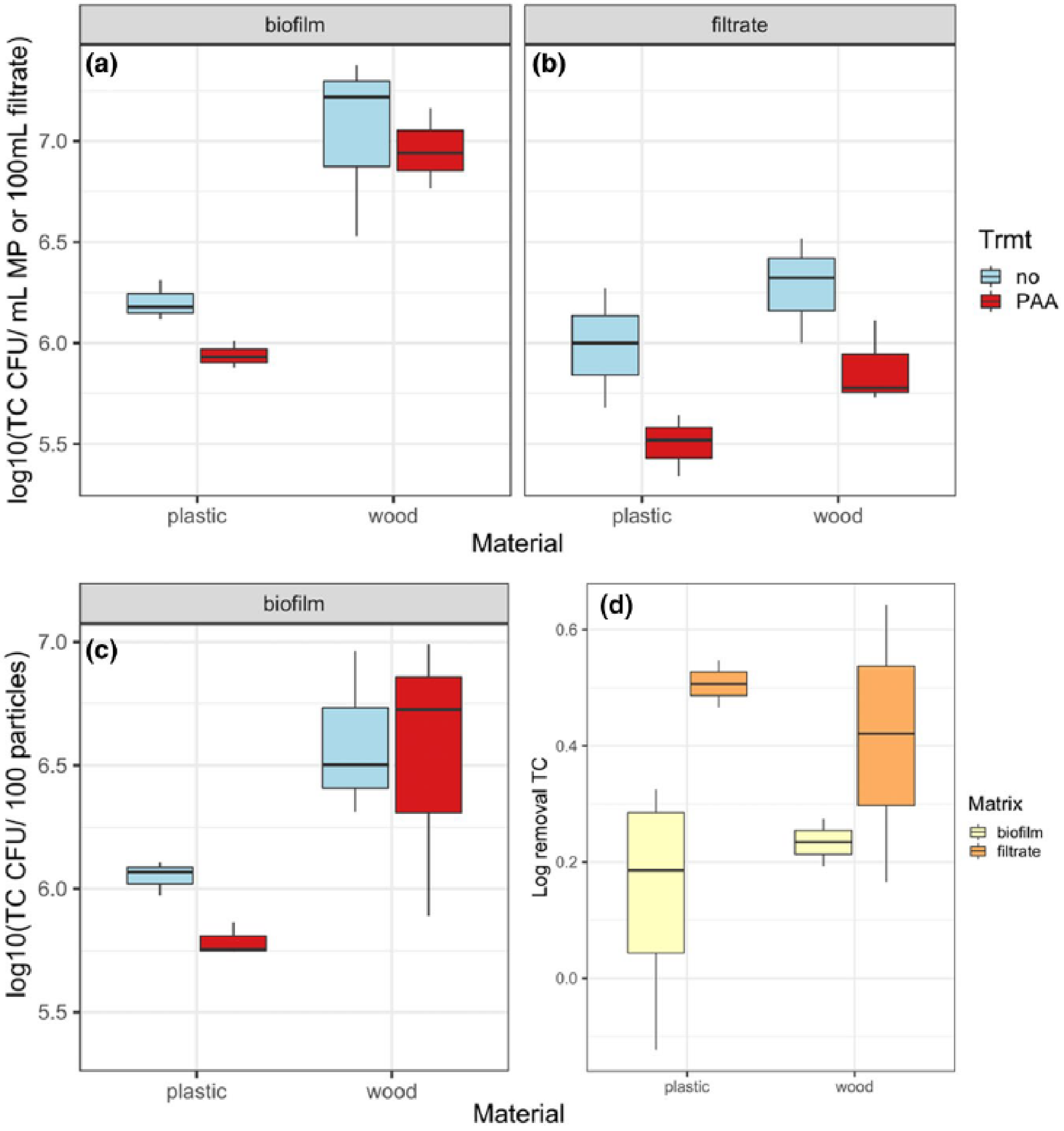


Boni et al. (2020) incubated microplastic particles and wood chips in either municipal wastewater influent or pre-disinfection secondary wastewater effluent and reactors were either disinfected with PAA or not treated. PAA is considered a green disinfectant because it has not been reported to form regulated disinfectant by-products and was chosen due to its status as a disinfectant that will likely see increased use in the coming years.

Results of the disinfection study (Figure 13) demonstrate that biofilm microbes were more resistant to disinfection than planktonic microbes, but that faecal indicators in MPs biofilm did not have different log-inactivation compared to wood microparticle biofilms. The first observation was expected: biofilms are generally considered to be more difficult to disinfect than planktonic organisms.

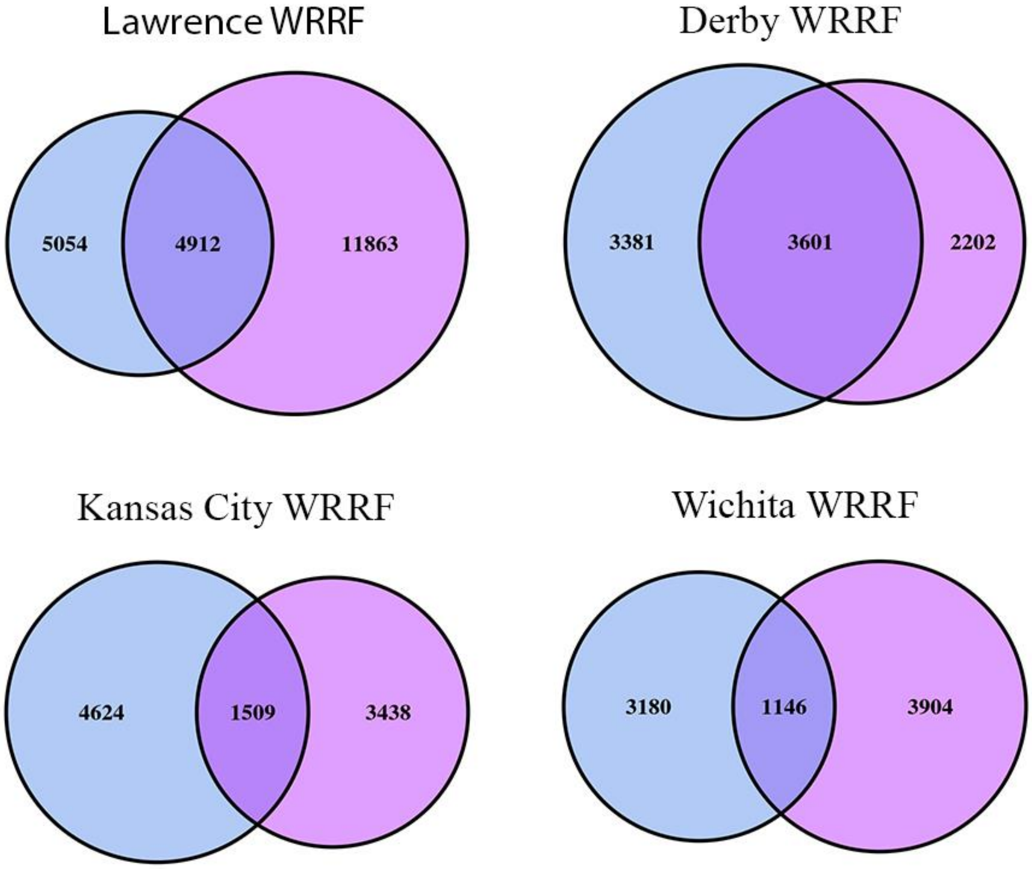
However, biofilms dislodged from wood microparticles grew the most total coliform and *E. coli* of the substrates studied, likely due to surface texture and availability of nutrients. Given that the MP biofilms behaved similarly to other microparticles with regard to disinfection, one may be able to rely on the literature for disinfection of biofilm faecal indicators on other particles when predicting MPs behavior.

Figure 13: (a) Log total coliform (TC) CFU for the dislodged biofilm (TC/mL of particles) and (b) filtrate (TC/100mL filtrate) grown in wastewater influent. (c) Log total coliform (TC) CFU for the dislodged biofilm on a per-particle basis (TC/100 particles). Results are shown for reactors with microplastic (LDPE) or control microplastics (wood chips) with peracetic acid disinfection ("PAA") and without ("no"). (d) Log removal of TC for both matrices (biofilm and filtrate) and particle types (LDPE or wood). N = 3. Source: Boni et al. (2020)



The fact that biofilms on MPs and wood were equally resistant to disinfection (i.e., had similar log inactivations) indicates that understanding the relative concentration of MPs compared to other buoyant microparticles in WW effluent would help indicate which particle type is contributing most to the bypassing of disinfection by biofilm faecal indicator organisms. This is significant because it highlights the importance of optimizing wastewater treatment processes for the removal of neutrally buoyant particles such as MPs and/or removing biofilms during disinfection. While WWTPs are not thought to be the only source of MP in the freshwater environment (Fahrenfeld et al., 2019), they are not 100 percent effective at removing MPs, allowing a path for pathogenic organisms from wastewater to bypass disinfection processes (refer Figure 14 showing overlap in bacteria species between activated sludge and microplastics in activated sludge in four US WWTPs). Nonetheless, wastewater treatment processes that in general remove particulates that carry harder to disinfect biofilms will reduce the loading of faecal microbes to effluent receiving water bodies (Boni et al., 2020).

Figure 14: Venn diagram showing bacterial OTU overlap between activated sludge (blue) and activated sludge microplastic (purple) in (A) Lawrence WRRF, (B) Derby WRRF, (C) Kansas City WRRF and (D) Wichita WRRF. Source: Macdonald, G. M., Sturm, B., and Fahrenfeld, N. (2020)



Overall, >22 % of the OTUs (~species) observed were shared between activated sludge and microplastic biofilm.

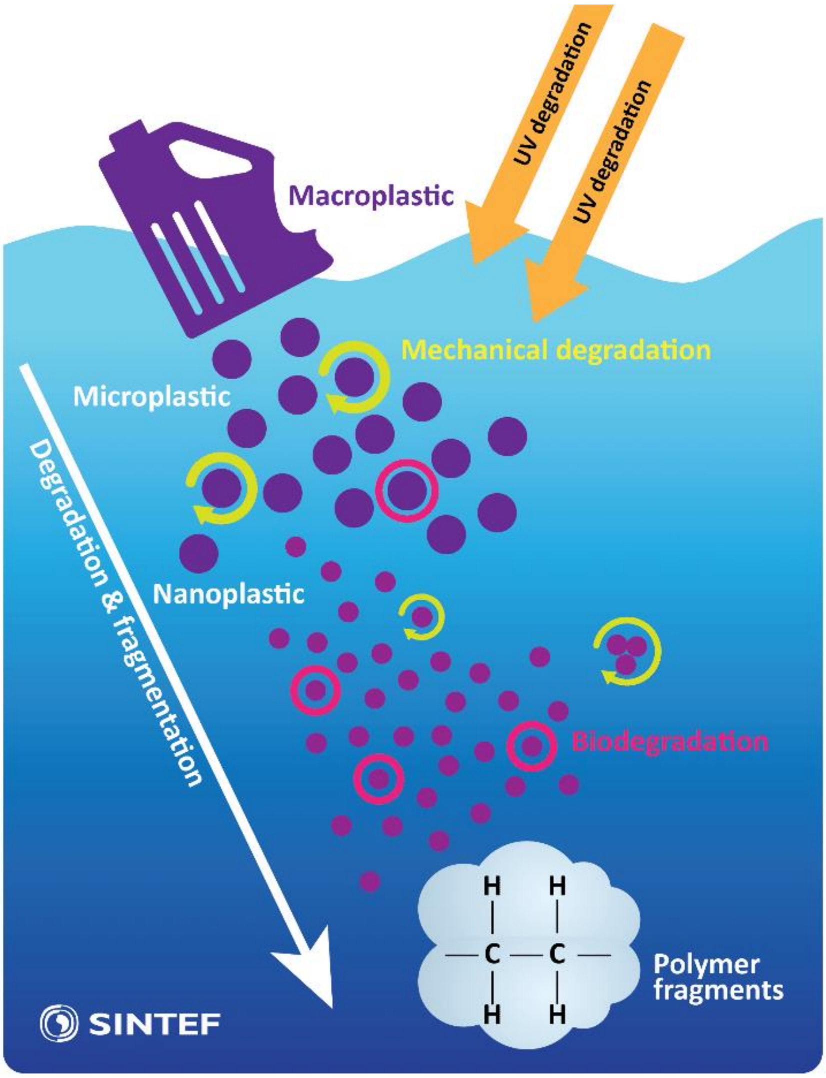
WHAT NEXT?

Further work is needed on improving analytical methods to measure for microplastics which would enable better comparison between studies, and on other pathways for microplastics to enter the water environment via WWTPs and stormwater.

Given the prevalence of plastics in certain WWTP processes and their tendency to slowly break down over their lifespan simply into smaller pieces of themselves (i.e. as per Figure 15), more research is needed to understand how WWTPs themselves may be inadvertently contributing to microplastic pollution, for example via biological trickling filter media, especially those containing UV stabilisers and black colour. Another possible but un-investigated source could be antioxidants etc. from moulding.

Furthermore, considering that a primary vector for plastics in soils is through biosolids (Ruffell at al., 2021), more work is urgently needed on the extent of potential microplastic pollution entering topsoil via land application and contaminating the food chain.

Figure 15: Degradation pathways for macroplastics into microplastics. Source: SINTEF



Research into microplastics in the NZ water environment is ongoing through ESR and others, particularly the impacts on and from stormwater and WWTPs. And there is evidence that the freshwater environment may have a few tricks up its sleeve when it comes to dealing with microplastics – the 2020 winner of the

Bjorn von Euler Award for Innovation in Water, Junzhi Xie, developed a phytoremediation method for removing microplastic particles from wastewater via duckweed, wherein most recovered particles were absorbed onto the surface roots and fronds, and showed minimal evidence of accumulation inside the plant.

CONCLUSIONS

Conventional WWTPs utilizing activated sludge processes and secondary clarification have removal efficiencies in the 75% to nearly 100% range. Less than 1% of the microplastics entering wastewater treatment plants are discharged to waterbodies... but this small percentage is a significant number of particles. Activated sludge accumulates much of the plastic load. >22 % of the OTUs (~species) observed were shared between activated sludge and microplastic biofilm. Mass majority of environmental release of microplastics from WWTPs will occur through land application of stabilized biosolids.

The bacterial community on the microplastic surface can be transported into waterbodies. Biofilm microbes were more resistant to disinfection than planktonic microbes, but faecal indicators in MP biofilm did not have different log-inactivation compared to wood microparticle biofilms. Wastewater treatment processes that in general remove particulates that carry harder-to-disinfect biofilms will reduce the loading of faecal microbes to effluent receiving water bodies. WWTPs are hotspots for antimicrobial resistance, organic contaminants, and horizontal gene transfer – and now concentrated microplastics.

Nonetheless, wastewater treatment processes that in general remove particulates carrying harder-to-disinfect biofilms will reduce the loading of faecal microbes to effluent receiving water bodies.

We urgently need standard methods for microplastic identification that are relatively quick, and accurate. This underpins the larger research goals of better understanding how sludge structure and property effects microplastics fate within treatment plants, the role of microplastics as a microbial carrier in environment, the fate of microplastics in biosolids, and how microplastics show up in stormwater.

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