

Microplastics in wastewater

Quantities and hazards associated with their release into the marine environment

Mike Williams, Khoa Pham, Roger Mulder, Nikita Pring, Mark Hickey, James Mardel

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Introduction

Issue

Plastics are widely used by societies throughout the world. The global production of synthetic polymers, or plastics, is around 300 million tonnes per year. Unfortunately, many discarded or end-of-life plastic products flow into the marine environment. These products include plastic from fragmented plastic waste, microbeads used in personal care products and medicines, road tyre wear, manufacturing wastes, or microfibres from clothing manufactured from synthetic polymer fibres. When these plastic materials are below a certain size (5 mm) they are known as microplastics. A significant pathway of microplastics to the marine environment is through wastewater treatment plants (WWTPs).

WWTPs reduce the environmental impacts of wastewater contaminants, such as nutrients and organic/inorganic contaminants. They can also effectively remove a large proportion of microplastics present in wastewater prior to discharge from the WWTP into the environment. Despite this, WWTPs are a significant pathway of microplastics entering the marine environment. There is increasing evidence that the ingestion of microplastics can cause physical damage to aquatic organisms (e.g. damage to gill surfaces, blocking normal food digestion). In addition, microplastics can transfer other contaminants (such as industrial chemicals) present in wastewater to marine organisms (Figure 1).

Plastic pollution is considered to be a 'wicked problem' because:

- The sources and breadth of contamination are widespread and varied,
- Human attitudes and behaviour drive contamination, and
- The science contributing to solutions, including sampling and analysis of environmental microplastics, is still maturing.

All these factors make source control and mitigation of microplastic pollution extremely challenging. Extensive resources and strong, unified leadership are needed to manage the problem.

Existing regulation

The NSW and Commonwealth Governments led the development of a voluntary industry agreement (VIA) to phase out microplastics in personal care products. This will lead to regulatory measures if there is evidence that the VIA has not been effective in reducing the number of microbeads in products (NSW EPA, 2016).

Assessment of compliance

There are two ways to assist compliance with the VIA, namely:

- By analysing the products available on the market in Australia; for example, 94% of products do not contain microplastics as of February 2018 (O'Farrell, K, 2018).
- By analysing wastewater flows through reliable, rapid and accurate methodology to measure the loads of microbeads, and microplastics more broadly, in wastewater.

Contaminant assessment

Microplastics can also bind chemical contaminants from industrial waste in wastewater prior to marine life exposure and ingestion. This can potentially increase the hazard of contaminants for marine organisms (Figure 1).

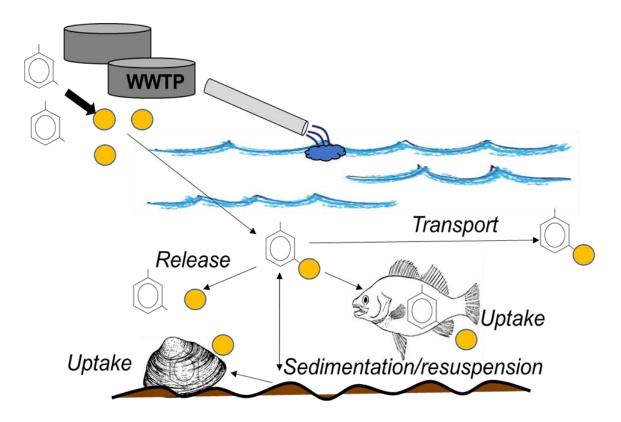


Figure 1. Conceptual overview of microplastics and associated wastewater contaminants moving throughout marine systems and their exposure to marine organisms following discharge from wastewater treatment plants (WWTPs). Chemical contaminants are represented by hexagons and microplastics are represented by yellow spheres.

The Project

CSIRO made two separate assessments of microplastics in wastewater for their release into marine environments.

1. Measurement of the number and types of microplastics (including microbeads) in wastewater inflows and outflows. This was measured by collecting samples over a 24-hour period for 10 months from two WWTPs servicing Sydney, namely Malabar and Cronulla. Additionally, biosolids produced in seven Sydney WWTPs (Cronulla, Malabar, Quakers Hill, Rouse Hill, St Marys, Winmalee and West Camden) were analysed in a once-off assessment.

The results were used to:

- a) Assess the effectiveness of the VIA
- b) Compare the effect of wastewater treatment methods for the removal of microplastics between the two WWTPs over a 10 month period in 2018 and 2019

c) Assess the removal of microplastics from wastewater into biosolids during wastewater treatment and their transfer to terrestrial environments.

The methodology and detailed results are in **Attachment 1**.

2. Assessment of wastewater contaminants binding to microplastics in wastewater. This was done by mixing common industrial chemicals found in wastewater with microplastics and simulating the effect in seawater and in a marine organism's gut.

The results were used to:

a) Estimate the extent of contaminant release from marine-released microplastics and how this impacts marine organisms.

The methodology and detailed results are in **Attachment 2**.

Summary of Results

Assessment 1 – Microplastics removal from wastewater and discharge into the Marine **Environment**

- It was estimated that Cronulla WWTP is discharging between 86 million to 350 million microplastic particles each day and that Malabar WWTP is discharging between 5.4 billion to 120 billion microplastic particles each day into the marine environment.
- Spherical microbeads from personal care products were infrequently detected in wastewater. When detected, between 1-3 microbeads per litre were measured in influent from Cronulla and Malabar WWTPs.
- At Malabar WWTP removal of microplastics during wastewater treatment ranged from very little to a large fraction (0-79%) through primary (screening and settling) treatment, while the Cronulla WWTP removed more than 98% of microplastics through tertiary (primary treatment plus biological treatment and disinfection) treatment.
- Most microplastic particles in all biosolid samples were fragments and the loads ranged from 45,000 to 323,000 microplastics/kg, consistent with international studies (typically 3,373 to 187,000 microplastics/kg).

<u>Assessment 2 – Contaminants in wastewater binding to Microplastics</u>

- An assessment of three common wastewater contaminants (benzalkonium chloride, bisphenol A and triclosan) indicated they were strongly bound to polyethylene (PE) and polyethylene terephthalate (PET) microplastics.
- After binding to PE and PET, the release of the contaminants was minimal in seawater but release of bisphenol A and triclosan under conditions simulating gut conditions of marine organisms was greatly enhanced; this suggests microplastics can transport contaminants and can release contaminants once ingested.
- Despite this, there are low concentrations of contaminants (ng/L or parts per trillion) in wastewater and low concentrations of microplastics in wastewater (ng/L to µg/L or parts

- per trillion to parts per billion), relative to naturally occurring organic matter (mg/L or parts per million).
- The hazard for marine organism from contaminants associated with microplastics discharged from WWTPs is therefore likely to be very low, since only a small fraction of contaminants in wastewater will associate with microplastics and marine organisms are likely to be exposed to a small fraction of these contaminated microplastics.

Conclusions

- 1. Tens of millions to hundreds of billions of microplastics are released to the marine environment each day from two Sydney WWTPs. These levels are comparable with other published studies in Australia, Asia, North America and Europe.
- 2. There is a low level of use of personal care products containing microbeads in the Cronulla and Malabar catchment areas, indicating that the VIA is effective in these areas. Future monitoring could be used to confirm this trend in other areas and to support the results of industry surveys.
- 3. The Cronulla WWTP was more effective at removing microplastics from wastewater than the Malabar WWTP. Malabar WWTP uses primary wastewater treatment, which includes removal of particulates through screening, surface skimming and sedimentation. The Cronulla WWTP uses tertiary wastewater treatment, which includes primary treatment plus biological digestion of nutrients and particulates and UV disinfection prior to release of effluent.
- 4. The concentrations of microplastics in biosolids from WWTPs, although very high, are unlikely to adversely impact terrestrial organisms based on existing evidence related to terrestrial effects assessments. The amount of microplastics, however, may increase over time in soils through ongoing biosolid application and microplastic accumulation. The potential impact needs to be balanced against the many benefits of biosolids reuse for soil improvement (e.g. carbon emission reductions, reduced use of synthetic fertilisers).
- 5. Contaminants within wastewater can bind to microplastics, which can then be ingested by marine organisms. The hazard associated with microplastic-bound contaminants, however, is likely to be relatively low. This is due to the extensive dilution of microplastics in the marine environment (reducing the potential for exposure) and the very low proportion of microplastics in wastewater relative to other particulate matter present that can also bind contaminants.

Microplastic analysis method development

Quantification and characterisation of microplastics in wastewater and biosolids was undertaken using a Fourier transform infrared (FTIR) spectroscopy technique combined with a microscope (Figure 2). The development of additional software by CSIRO allowed the semi-automated characterisation of polymer type, particle size and total microplastic numbers. User input was required for cross-checking data generated from software analysis. Reference spectra of 14 common polymer types were used for the automated analysis of microplastics, with additional reference spectra also included for further user analysis where required.

For FTIR analysis, it was essential to remove the high loads of organic (through oxidation reactions) and inorganic (through density separation) matter in the wastewater and biosolid samples. Overall, this methodology represents a relatively rapid (<2 days for preparation and analysis), data-rich analysis of each sample.

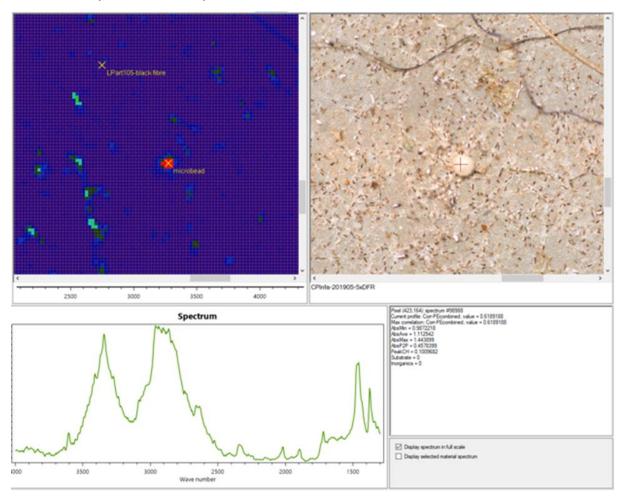


Figure 2. An example of a polyethylene (PE) microbead detected at Cronulla WWTP using FTIR microscopy. A correlation heat map for the PE polymer (top left) and the visual image of the PE microbead (top right) is generated from library matching of PE polymers (bottom left). Each square in the correlation heat map is 25 μm², which also corresponds to the scale of the visual image.

Microplastics quantification and characterisation in wastewater

Wastewater samples were collected over a 24-hour period at licenced EPA sampling points at Malabar and Cronulla WWTPs. Malabar WWTP has two influent streams from its South Western suburbs ocean outfall sewer (SWSOOS) catchment:

- 1. SWSOOS 1 (S1); which comprises 30% of total inflow
- 2. SWSOOS 2 (S2); which comprises 70% of total inflow

and one effluent stream, treating approximately 450 ML (or million litres) per day. Cronulla WWTP receives and discharges approximately 50 ML/day. Samples of 1 L (or 10 L for Cronulla effluent) were processed for microplastics analysis.

Over the 10-month period (from November 2018 to September 2019), 19–236 microplastics/L were quantified in the influent (Cronulla and Malabar WWTPs), 11-597 microplastics/L in the Malabar WWTP primary-treated effluent and 1.9–6.6 microplastics/L in the Cronulla tertiarytreated effluent (Figure 3 and 4). This is equivalent to 8.7×10⁸–1.4×10¹⁰ (870 million to 14,000 million) microplastic particles entering Cronulla WWTP and 2.4×10¹⁰–6.1×10¹⁰ (24,000 million to 61,000 million) microplastic particles entering Malabar WWTP each day. It was estimated that between 8.6×10⁷–3.5×10⁸ (or 86 million to 350 million) and 5.4×10⁹ and 12×10¹⁰ (5,400 million to 120,000 million) microplastic particles were being discharged from the Cronulla and Malabar WWTPs per day, respectively (

Figure 5). This is comparable with other published studies in Australia, Asia, North America and Europe (1).

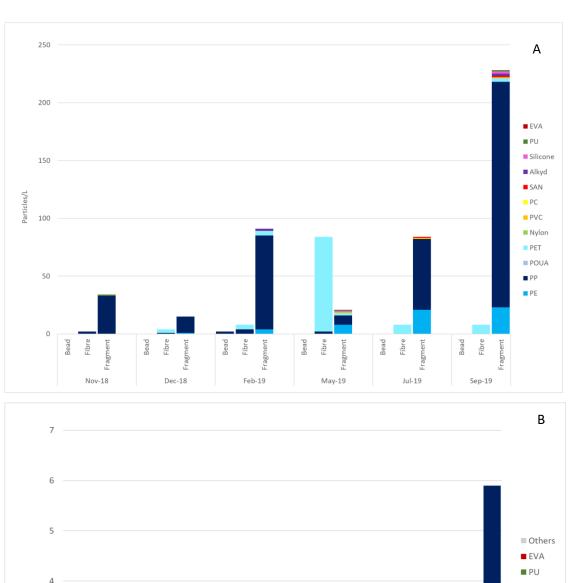
Polypropylene (PP) was the predominant polymer type and microplastics typically had a fragment morphology. Polyethylene (PE) and polyethylene terephathalate (PET) were other common polymer types detected in wastewater, with silicone, PC, PU, alkyd, SAN and EVA infrequently detected at very low numbers (Figure 3 and 4).

Microbeads

Compared with microplastics with a fragment morphology, spherical microbeads were detected infrequently in Cronulla and Malabar WWTP wastewater. When detected, between 1-3 microbeads/L were measured in influent from Cronulla (1 sample) and Malabar (8 samples) WWTPs. These microbeads were identified as PE (55%), PP (33%) and PET (12%) polymers.

This would indicate that microbead containing products are likely to still be in use, although the relatively low concentrations and infrequency of detection suggests this is not common. To monitor whether the expectations of the VIA are being met, additional sampling of wastewater in the future would be able to confirm an ongoing decline in community use to support market surveys.

(1)Blair et al., 2019; Carr et al., 2016; Conley et al., 2019; Dyachenko et al., 2017; Gies et al., 2018; Lares et al., 2018; Leslie et al., 2017; Long et al., 2019; Magni et al., 2019; Magnusson et al., 2014; Mason et al., 2016; Michielssen et al., 2016; Mintenig et al., 2017; Murphy et al., 2016; Simon et al., 2018; Talvitie et al., 2017, 2015; Ziajahromi et al., 2017



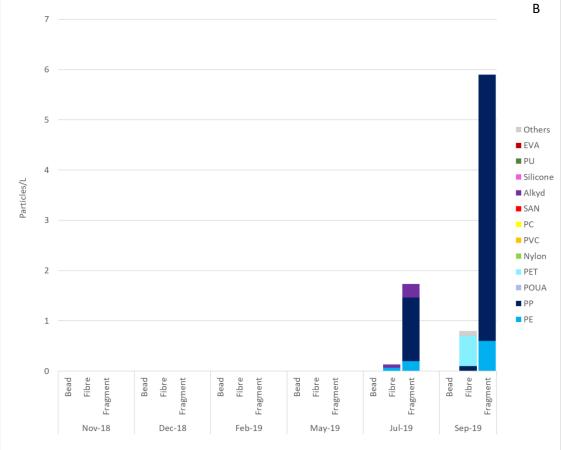
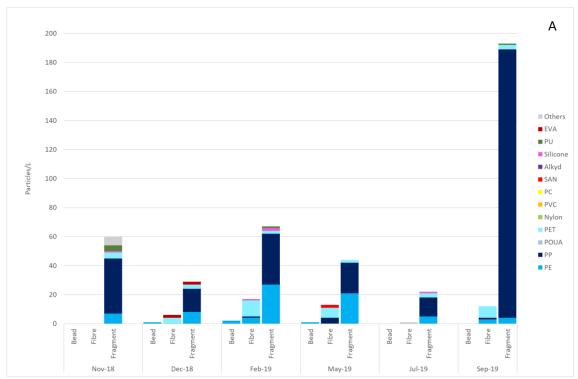
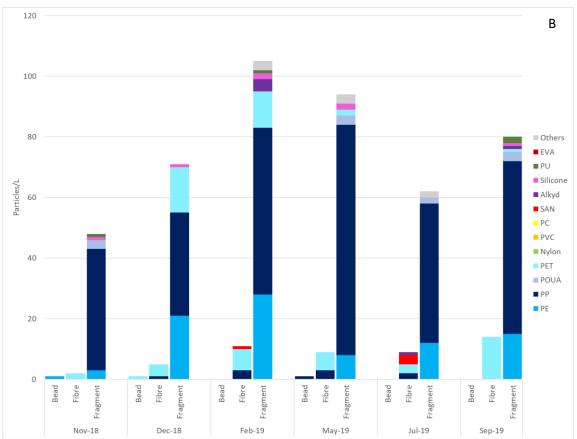


Figure 3. Summary of microplastic quantities (particles/L), polymer type and morphology (fragment, fibre or microbead) measured in Cronulla WWTP (a) influent and (b) effluent over a 10 month period. Note contamination of effluent samples from Nov18 to May19 led to their exclusion.





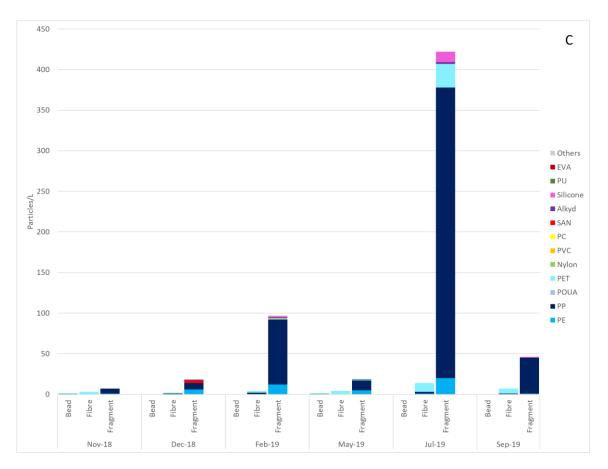


Figure 4. Summary of microplastic quantities (particles/L), polymer type and morphology (fragment, fibre or microbead) measured in Malabar WWTP (a) S1 influent, (b) S2 influent and (c) effluent over a 10 month period.

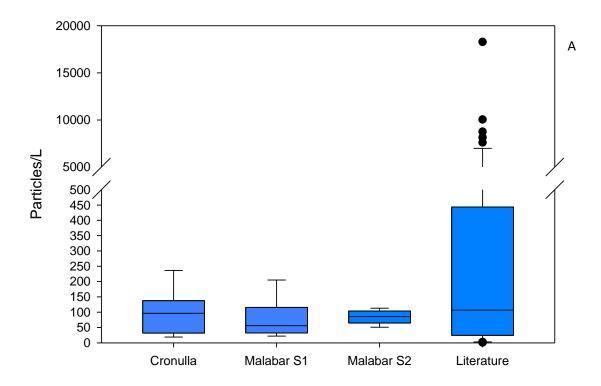
Removal of microplastics from the wastewater

Based on the relative concentrations of microplastics in the influent and effluent of the two WWTPs, Malabar WWTP had between 0 and 79% removal, while Cronulla WWTP had >98% removal. It should be noted that physical fragmentation during wastewater treatment can potentially make concentrations in effluent higher relative to the influent, which would reduce apparent removal rates. This may account for some of the high variability seen in the removal of microplastics at Malabar WWTP. Malabar WWTP uses primary wastewater treatment, which includes removal of particulates through screening, surface skimming and sedimentation. Cronulla WWTP uses tertiary wastewater treatment, which includes primary treatment plus biological digestion of nutrients and particulates and UV disinfection prior to release of effluent.

Biosolids

Biosolids are stabilised sewage sludge, a by-product of wastewater treatment, and are mainly composed of organic matter derived from microorganisms that are essential to the treatment process. Due to the chemical stability of microplastic polymers detected here, the most likely removal process from wastewater is through their attachment to organic matter in wastewater, which is collected as sludge and further treated to produce biosolids. A once-off collection of biosolids from Cronulla, Malabar and five other WWTPs (Quakers Hill, Rouse Hill, St Marys,

Winmalee and West Camden) in the Sydney area was used to measure microplastic loads in these biosolids using the same processing and analytical technique applied to wastewater.



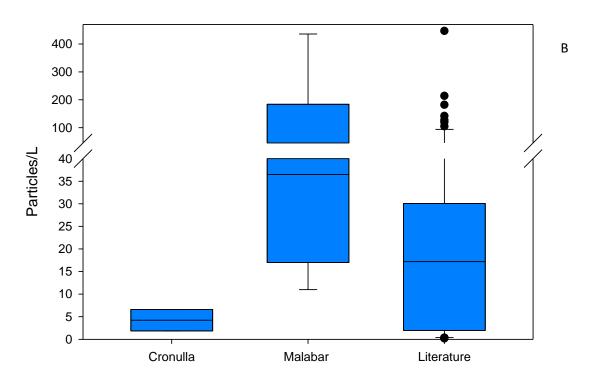


Figure 5. Comparison of the range of microplastic concentrations (particles/L) measured in Cronulla and Malabar WWTP (a) influent and (b) effluent with other published studies (n=13) measuring wastewater concentrations of microplastics. Bottom and top line of box indicate 25% and 75% of values, respectively, middle line indicates median of values, bottom and top of whisker lines represent 10% and 90% of values and dots represent outlying values.

The majority of microplastic particles in all biosolid samples were PP fragments (<50 µm in size), which was consistent with microplastics quantified and characterised in wastewater collected from Malabar and Cronulla WWTPs. Based on the number of microplastics measured in one gram of biosolid samples, loads of microplastics in biosolids ranged from 45,000 to 323,000 microplastics/kg. This is broadly consistent with concentrations of microplastics in biosolids published in international studies that range from 3,373 to 187,000 microplastics/kg (Figure 6) (2). The majority of biosolids produced in Australia are reused as a soil amendment for land application due to their high nutrient loading. The presence of microplastics in biosolids should be further assessed to ensure that adverse levels do not occur in terrestrial systems, especially over the longer term where microplastics may accumulate from ongoing applications of biosolids. This needs to be carefully balanced, however, against the current practice of beneficial reuse of biosolids. Current reuse practices are considered the most sustainable option for biosolids, in terms of economics, reduction in carbon emissions, reduced reliance on non-renewable soil amendments (e.g. synthetic fertilisers) and the associated environmental benefits.

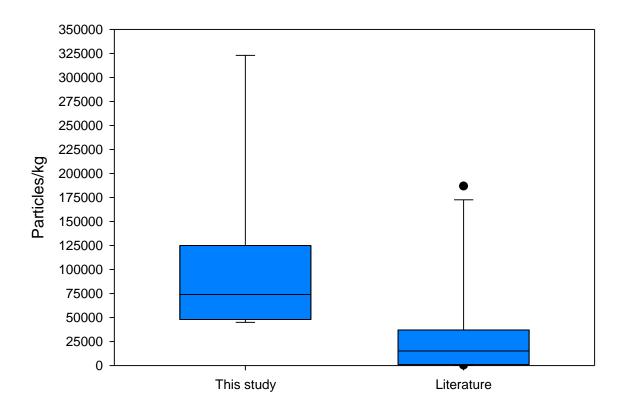


Figure 6. Comparison of the range of microplastic concentrations (particles/kg) measured in this study for biosolids collected from seven WWTPs within the Sydney catchment with other published studies (n=11) measuring biosolid concentrations of microplastics. Bottom and top line of box indicate 25% and 75% of values, respectively, middle line indicates median of values, bottom and top of whisker lines represent 10% and 90% of values and dots represent outlying values.

(2) (Carr et al., 2016; Gies et al., 2018; Lares et al., 2018; Leslie et al., 2017; Li et al., 2018; Magni et al., 2019; Magnusson et al., 2014; Mahon et al., 2017; Mintenig et al., 2017; Murphy et al., 2016; Talvitie et al., 2017)

Hazard characterisation of microplastics in wastewater

Sorption and desorption of contaminants from microplastics

The wastewater contaminants selected for the assessment of extent of their association (sorption) with and release (desorption) from microplastics, along with how this related to their potential hazard to marine organisms, were:

- Benzalkonium chloride (BAC), a widely used disinfectant
- Triclosan (TCS), an antimicrobial found in personal care products; and
- Bisphenol A (BPA), a plasticiser used in the manufacture of certain plastics

These contaminants are widely used industrial chemicals and commonly measured at ng/L (parts per trillion) concentrations in wastewater. The microplastics used in this assessment were:

- Polyethylene (PE), in the form of microbeads, fibres and fragments; and
- Polyethylene terephthalate (PET), in the form of fibres.

Although PP was most commonly detected in the wastewater samples from the survey of microplastics in Cronulla and Malabar WWTPs, it has a very similar chemical composition with PE and would therefore interact with contaminants in a similar manner.

Contaminants were mixed with microplastics at considerably higher contaminant (mg/L or parts per million) and microplastic (g/L or parts per thousand) concentrations than would be expected in the environment to ensure sorption and desorption were quantifiable. Contaminants were loaded onto microplastics in wastewater solution before being transferred to seawater solutions to assess the extent of contaminant release from the microplastics. Three seawater solutions were selected:

- Natural seawater
- Natural seawater containing 15 mM sodium taurocholate
- Natural seawater containing 15 mM sodium taurocholate, adjusted to pH 4 and 38°C

The first scenario was used to simulate the discharge of microplastics containing contaminants into seawater. Sodium taurocholate is a naturally occurring surfactant used to mimic the gut conditions within a marine organism and the second and third scenarios were used to simulate digestion of microplastics containing contaminants. Higher temperatures and acidic conditions at pH 4 can be more favourable for the release of contaminants from microplastics.

The extent of sorption of contaminants was in the order TCS >BAC >BPA, with the contaminants generally showing the greatest affinity for PE. The contaminants had a moderate to high degree of affinity with the microplastics, with association (measured by sorption coefficient, K_d) ranging from 160-8,800 L/kg. Release (or desorption) from the microplastics into seawater was minimal over a 24 h period, suggesting microplastics have the potential to transport these contaminants

throughout marine environments. The release of BAC and TCS in particular (up to 100% of the sorbed amount) was greatly enhanced under the simulated biological conditions, suggesting the majority of these microplastic-associated contaminants can be released once ingested. Release of BPA from the microplastics was typically low (<7% of sorbed amount) in all solutions.

Following the discharge of microplastics into the marine environment, the extent of transport throughout marine environments will be dependent on a number of factors related to the microplastic and receiving environment that can influence whether a microplastic stays on the surface, settles into sediments or is cycled between the two. For example, environmental factors can include wave action, tides, nearshore currents, water temperatures, nutrient interactions, particulate matter concentrations and salinity. The polymer type can influence whether a microplastic sinks or binds to particulate matter that can then drop out into sediments, known as sedimentation. Particulate matter can therefore have a strong influence on the amount of microplastic remaining in the water column or sediments. Although large numbers of microplastics are released in WWTP discharges, there is likely to be a considerable degree of dilution once they enter the marine environment. Previous work has indicated that nearshore concentrations of microplastics are typically <1 particle/L, although concentrations in sediments can be much greater (Burns and Boxall, 2018).

Surveys of a range of marine organisms show corresponding particle loads in organisms to be reasonably low (typically <5 particles/organism), with the majority of surveyed organisms (i.e. >50%) typically not containing microplastics (Burns and Boxall, 2018; Jamieson A. J. et al., 2019; Lusher, 2015).

Hazard assessment

The hazard associated with these contaminants is related to concentrations of contaminants that an organism will be exposed to and the concentration that would be expected to cause an effect in an organism.

In Figure 5, the highest microplastic counts in scientific literature for WWTP effluent were found in a study where 447 microplastics/L were estimated, equivalent to 12 μg microplastics/L (Simon et al., 2018). This very high measurement of microplastics is still more than a thousand times less than the ~60 mg/L of organic carbon measured in wastewater used for this study. Only a very small fraction (< 0.01%) of the total amount of contaminant being released in WWTP effluents are likely to become associated with the microplastics (Table 1).

The 447 microplastics/L found by Simon et al. (2018) were greater than the amounts found in the Cronulla and Malabar WWTP wastewater (Figure 5) but the equivalent mass of 12 µg/L could be applied to the sorption data (Table 1) as a worst-case scenario. Up to \sim 1 μ g of the contaminants was measured in each litre of wastewater, of which <0.01% would be expected to associate with the microplastics. Overall, the proportion contaminants associated with microplastics represents a minor fraction of the contaminant that is available to an organism, relative to the contaminant present in solution, and is consistent with a previous analysis of the relative importance in the uptake of contaminants from ingested microplastics (Koelmans et al., 2014). This would be even further reduced in the case of BPA, where the majority of the amount bound to the microplastics is not released back into solution.

Table 1. Estimate of percentage of BAC, BPA and TCS measured in wastewater effluent that could become associated with microplastics present in wastewater at a concentration of 12 $\mu\text{g}/\text{L}$

CONTAMINANT	SORPTION CAPACITY RANGE	WASTEWATER CONCENTRATION	AMOUNT SORBED TO PLASTIC	% TOTAL SORBED TO PLASTIC
	K_d (L/kg) ^a	(ng/L)	(ng) ^b	
ВАС	316±63-2048±666 ^c	388±38-998±228 ^c	0.001-0.025	0.0004-0.0025
ВРА	134±84-3150±3104	196±34-1111±201	0.0003-0.04	0.0002-0.004
TCS	1429±451-8793±1548	164±18-221±19	0.003-0.023	0.002-0.01

^abased on K_d values for PE particles; ^bbased on 12 μg/L microplastic in effluent (Simon et al. 2018), mean effluent contaminant concentration range and mean K_d value range for PE particles; value represents mean value \pm standard deviation of 3 replicates

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For further information

Land and Water
Dr Mike Williams
+61 8 8303 8515
mike.williams@csiro.au
csiro.au/landandwater

Manufacturing Dr Roger Mulder +61 3 9545 2550 roger.mulder@csiro.au csiro.au/manufacturing