Microplastics in a UK Landfill: Developing Methods and Assessing Concentrations in Leachate, Hydrogeology, and Release to the Environment

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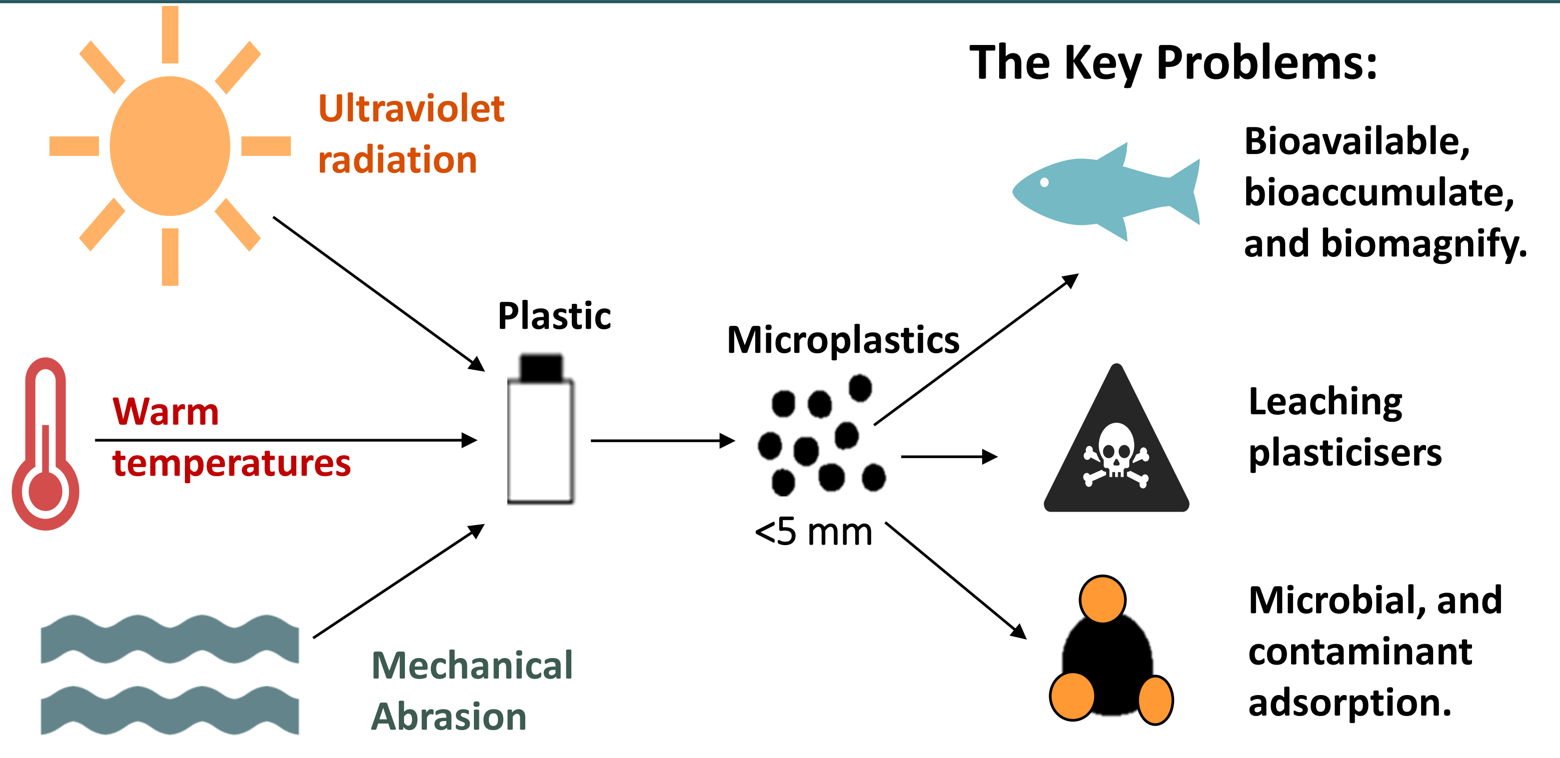
Plastic in freshwater environments

ROYAL HOLLOWAY UNIVERSITY OF LONDON

Small Plastics, Big Problem



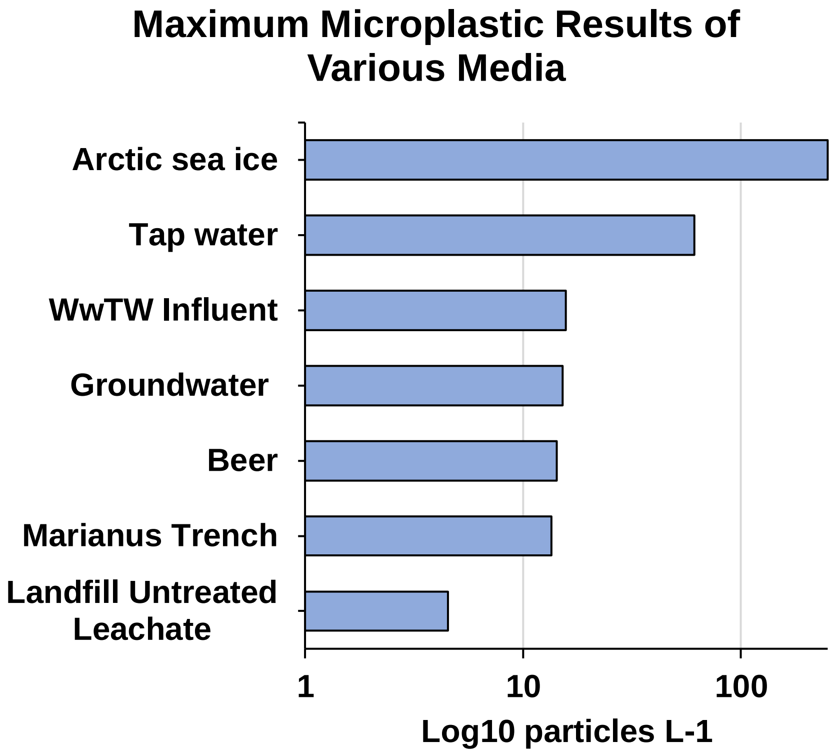


Figure created by Waddell (2019), using information by Cole et al. (2011). Microplastics as contaminants in the marine environment: A review. *Marine Pollution Bulletin*, **62**(12), pp.2588-2597. Graph created using data from various sources.

Microplastics (< 5 mm particles) are ubiquitous in the environment, and it is unusual for a study not to find microplastics. Warm temperatures and ultraviolet radiation can oxidise plastic material (E.g., adrift in the ocean) making it brittle. Coupled with mechanical abrasion the plastic fragments into smaller and smaller particles. Once a particle's size is <5 mm it may be considered a 'microplastic', of which causes concern over its bioavailability to organisms, leaching of chemical constituents, or adsorption of contaminants. The present study aimed to address a knowledge gap of microplastic's occurrence in the environment both within and around an active landfill site, with a focus on water samples. The study took place throughout Summer 2019.

A Need to Characterise Sources and Loads

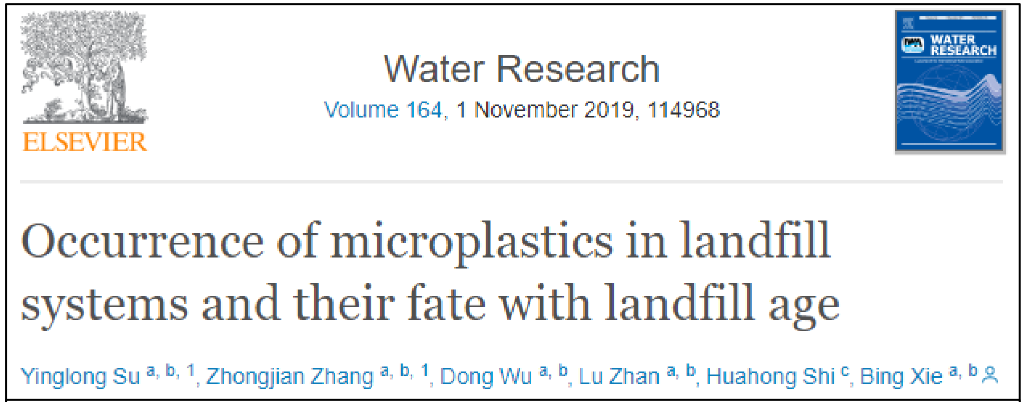
The question arises as to what the key sources of microplastics are and the loads released to the environment. One possibility highlighted are landfill sites.

There is very little research into microplastics at landfill sites, but since they are still a fundamental disposal option for plastic waste, they remain a possible reservoir of microplastics from the breakdown of said plastic. By mass, plastic can make up 7% of landfill waste composition in the UK (Letcher and Vallero, 2019).

Waddell (2019)

Microplastics could be generated by one or a combination of factors:

- Landfill fluctuating temperatures, reaching as high as 60 to 90 °C.
- Ranging pH values (4.5 to 9).
- Deep-seated fires.
- Physical stress and compaction.
- Microbial degradation.



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Water Research
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Occurrence of microplastics in landfill systems and their fate with landfill age

Yinglong Su [a, b, 1], Zhongjian Zhang [a, b, 1], Dong Wu [a, b], Lu Zhan [a, b], Huahong Shi [c], Bing Xie [a, b]

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Water Research
Volume 159, 1 August 2019, Pages 38-45

Municipal solid waste (MSW) landfill: A source of microplastics? -Evidence of microplastics in landfill leachate

Pinjing He [a, c, d], Liyao Chen [a, b], Liming Shao [c, d], Hua Zhang [b], Fan Lü [a, b]

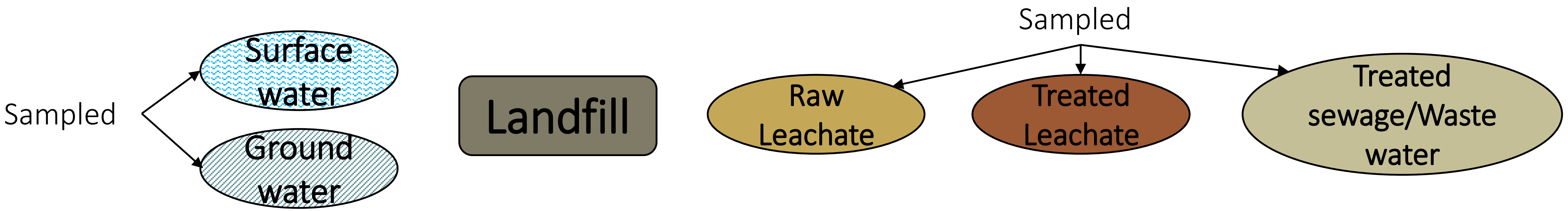
The Project – What Was Done

NOTE: The landfill site in question is required to be anonymous at the request of the waste operator.

The project investigated five key facets of landfill; **raw leachate, treated leachate, treated sewage, groundwater and surface water**. Due to the lack of previous work in some of these areas, and standardisation of sampling and analysis methods, the aims were:

1) To develop an effective method for sample collection and analysis of landfill mediums, with reliable recovery and identification of microplastic particles.

2) Use the developed method to undertake a preliminary investigation of MPs in leachate and surrounding hydrogeology of a UK landfill site, characterising their occurrence, concentrations, and potential implications for the wider environment.



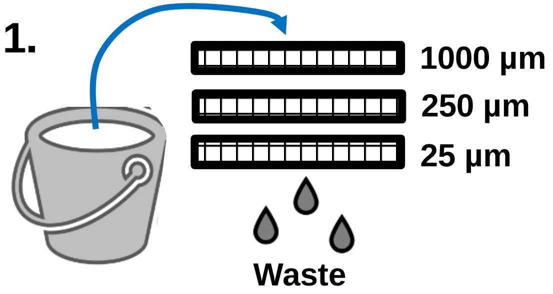
Leachate Well 4


Leachate Treatment Plant


Surface Water Outflow

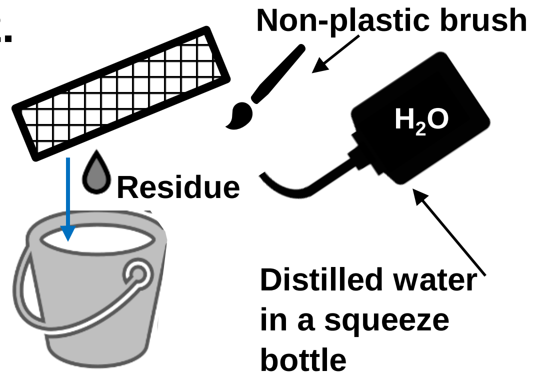
Method Development and Sampling

1.



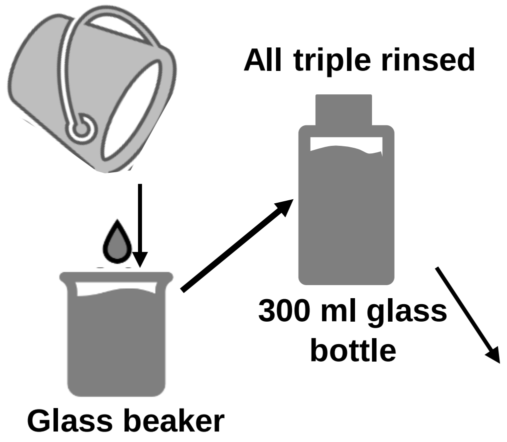
1. 10 L of sample was collected in a pre rinsed bucket and filtered in the field through three stainless steel mesh sizes. The smallest mesh had an aperture size of 25 µm. This step was taken to reduce the volume of sample taken to the laboratory.

2.

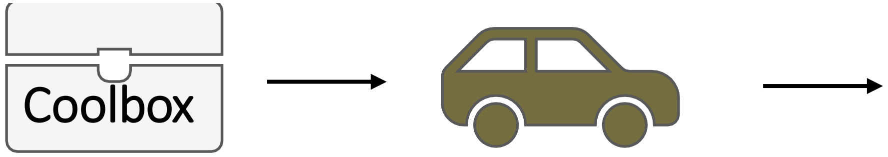


2. The meshes were rinsed with distilled water back into the bucket and each mesh brushed while using the water to dislodge any particles caught in the mesh pores. The brush was also rinsed.

3.



3. The volume reduced contents now in the bucket, which included all residue collected on the meshes, was transferred into a glass beaker to facilitate careful pouring into triple rinsed 300 ml glass sample bottles. The bottles were labelled, with a date and time, stored in a coolbox, and transported to the RHUL laboratory, and then stored in a refrigerator until sample preparation began.

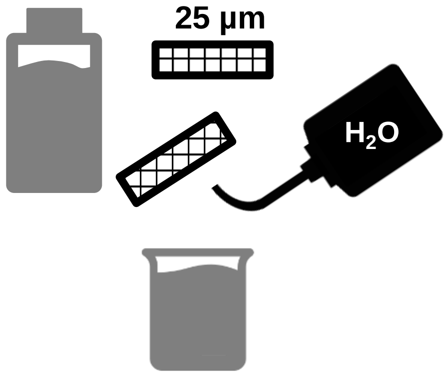


RHUL Geochemistry Laboratory

QA/QC:
- Repeat sampling over different days (at least three samples per site).
- Equipment blank samples.
- All equipment pre washed and rinsed with distilled water.
- Bucket calibrated to 10 L.
- Cotton clothing worn, aside from mandatory PPE.

Method Development and Laboratory Work

1.



2.

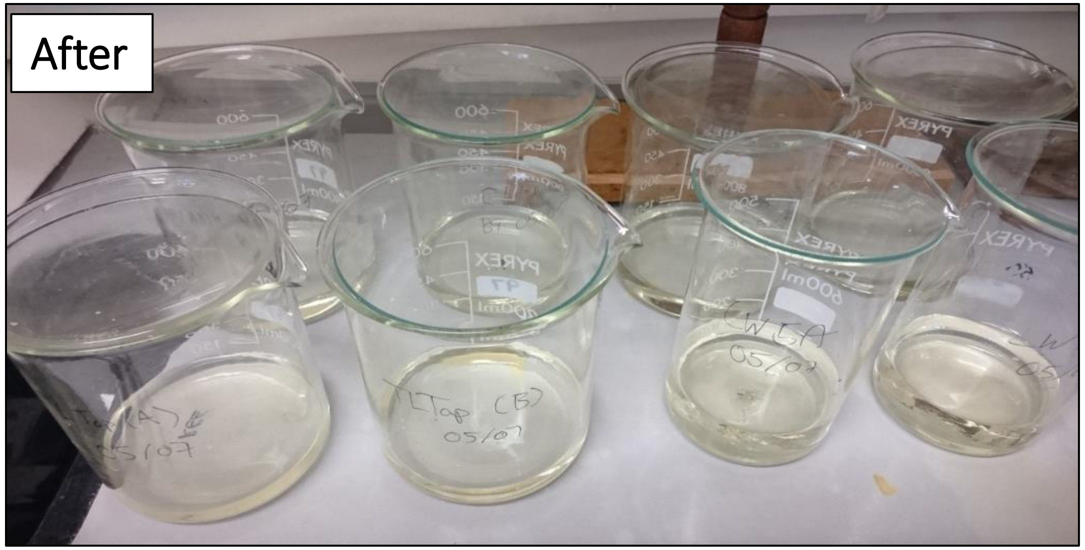
40-80 ml H_2O_2

40-80 ml $(NH_4)_2Fe(SO_4)_2$

1. In the laboratory, samples were filtered once more with a smaller 25 µm sieve, and residue rinsed into a glass beaker. By this method, most of the liquid fraction was removed and thus reduced the volume of Fenton's reagent required in the next step.

2. Fenton's reagent was added to the sample residue in the beaker. 40 ml of 30% H2O2 and 40 ml of ammonium iron(II) sulphate solution was added. The beakers were covered with a watch glass, placed in the fume cupboard, and reactions left for one week to react.

Before



After

QA/QC:
- All glassware and utensils cleaned with distilled water before use.
- Laboratory operatives to only wear clothing of natural fibres.
- Laboratory blank samples made during each batch of samples.
- Samples placed in the fume cupboard to prevent secondary contamination.

Hurley et al. (2018). Validation of a Method for Extracting Microplastics from Complex, Organic-Rich, Environmental Matrices. *Environmental Science & Technology*, **52**(13), pp.7409-7417.

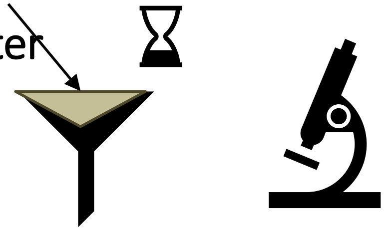
Method Development and Laboratory Work

3. Density Separation by NaCl

3. For some samples, density separation was used by NaCl but only when absolutely necessary since this additional step may introduce further error. This was mostly used for groundwater samples due to sediment.

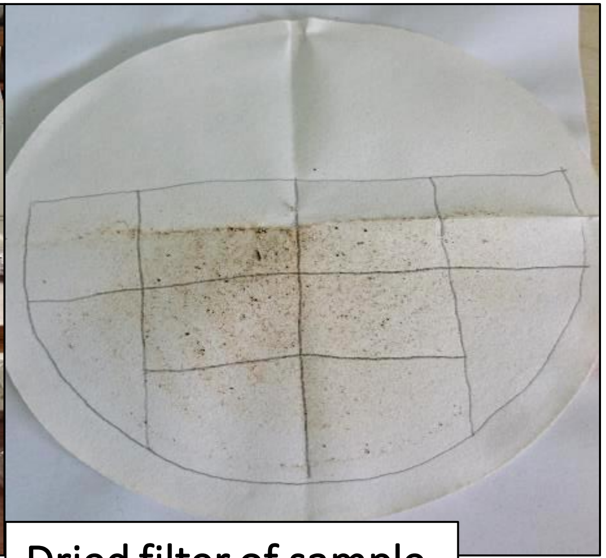
4.

Filter



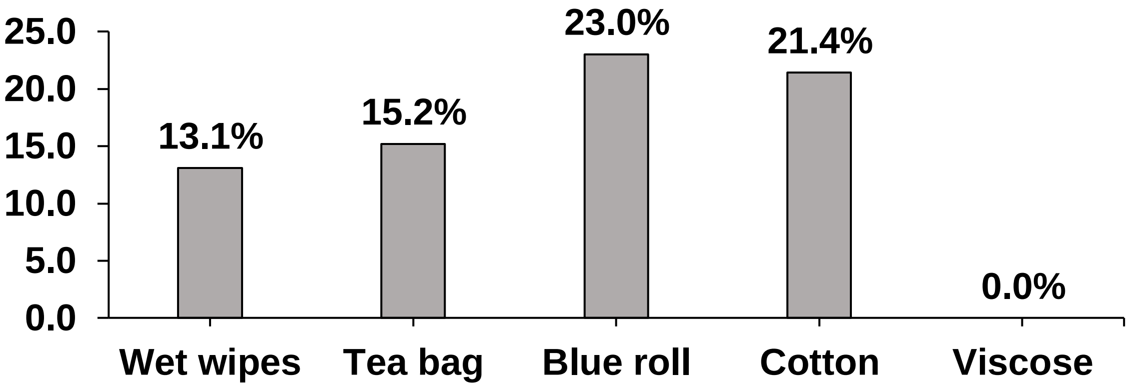
4. The final preparation step was to filter the Fenton's reagent reacted samples, and density separated samples through Whatman 20 µm paper filters. The filters were allowed to dry flat, covered by watch glasses. Microplastics were then counted by aid of a microscope, by shape and colour.

Density Separation



Dried filter of sample

Material % Reduction by Fenton's Reagent



Method development tests:
Additional tests were taken to test some of the steps of the method. Fenton's reagent was unable to fully digest some materials such as cotton and blue roll. An open filter was left in the fume cupboard and in the laboratory to check for aerial deposition. Both tested positive for fibres, suggesting fume cupboards do not prevent contamination.

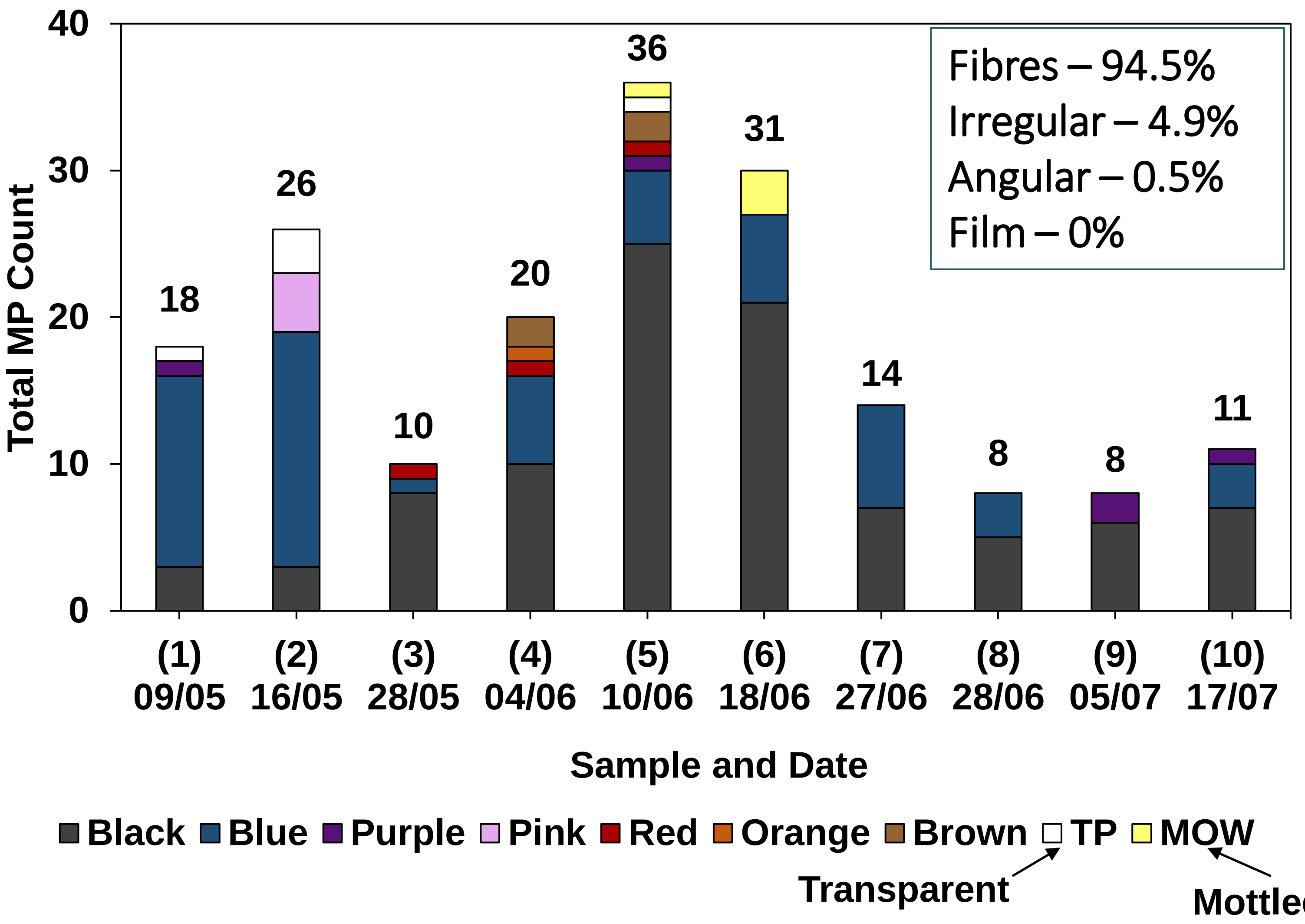
Dyachenko et al. (2017). Extraction and identification of microplastic particles from secondary wastewater treatment plant (WWTP) effluent. Analytical Methods, 9(9), pp.1412-1418.

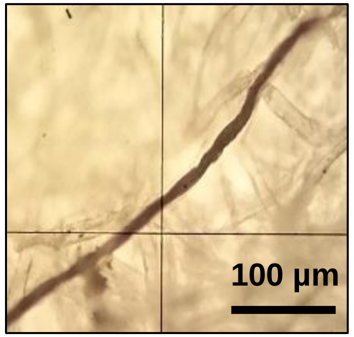
Microplastics were indeed recovered from landfill samples…

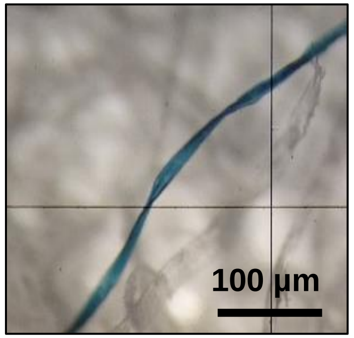
Raw Leachate   Treated Leachate   Waste Water   Groundwater   Surface Water

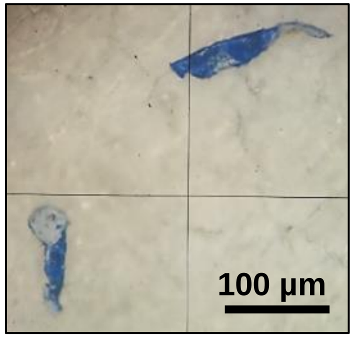
Laboratory Blanks

Microplastics Total Count and Particle Colour for Laboratory Blank Samples





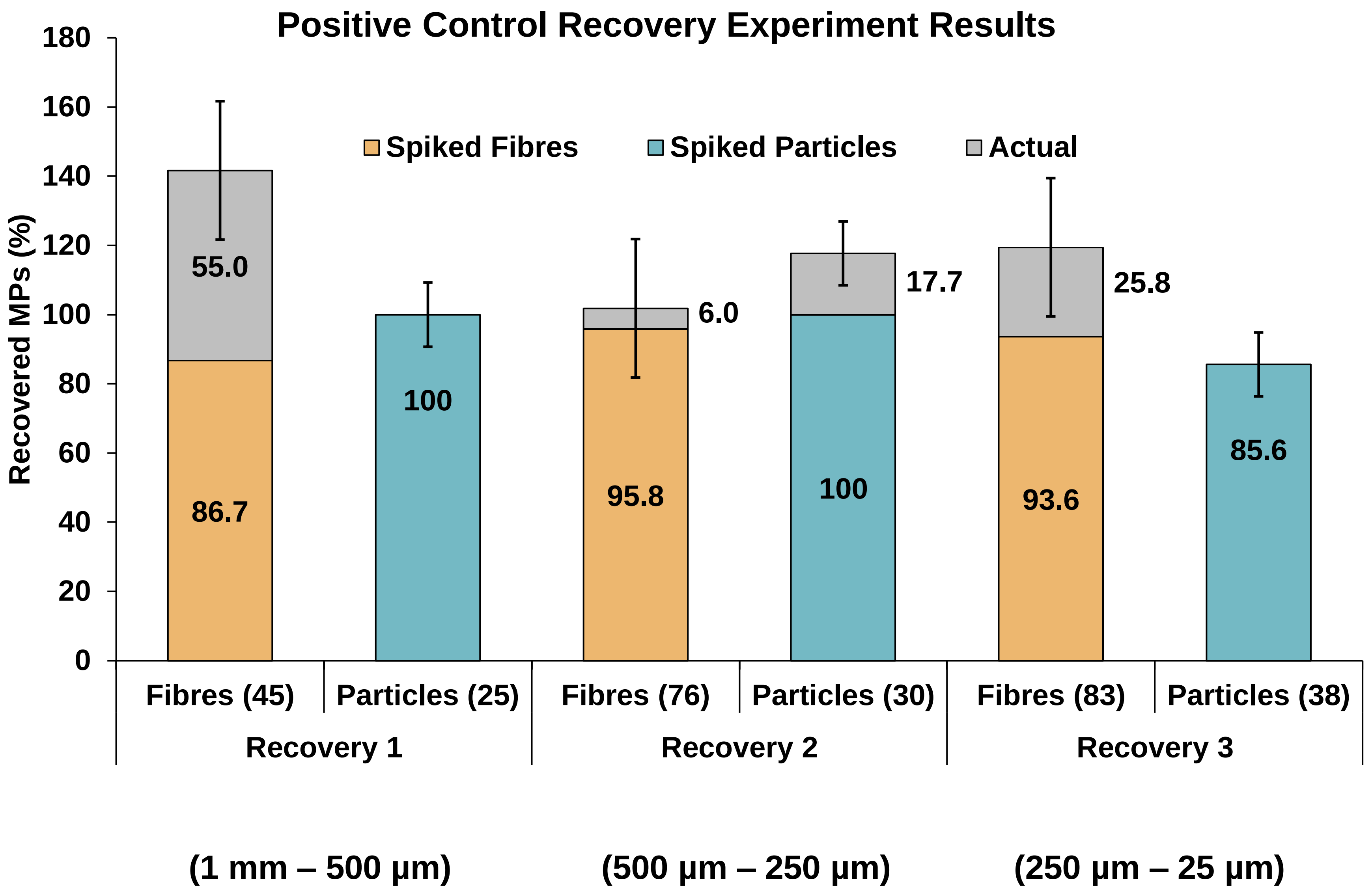




Laboratory blank samples were taken for each batch of samples and used to correct the data if the same colour and shape particle was found in both the blank and the sample.

It is possible that more counts were observed within the middle of July 2019 when there was a greater number of students also using the laboratory, and so it is possible this greater traffic of users may have attributed to some partial secondary contamination.

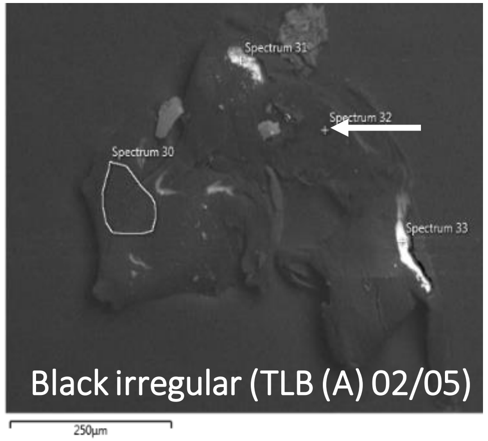
Microplastic Recovery Experiments



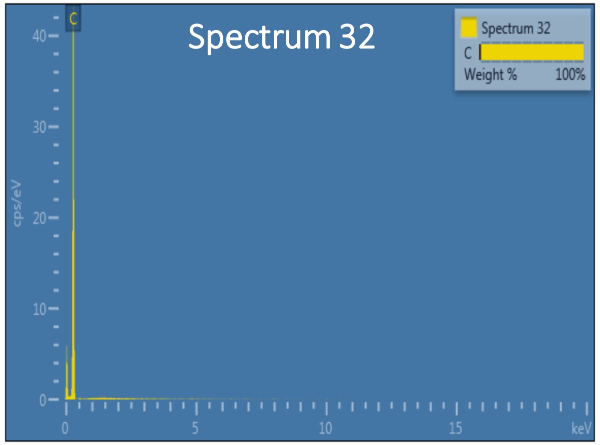
Recoveries were set up by spiking a beaker of distilled water with PP/PE fragments, and fibres (mostly polyester fibres). Particles were sieved using the stacked sieves to select size fractions that reflect those found in landfill samples. Three recoveries were performed. The procedure involved spiking the stainless-steel bucket, filled with distilled water, and proceeding exactly as with all other samples through the methodology.

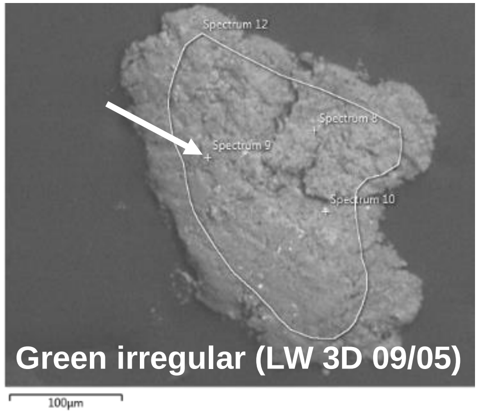
The quantity recovered is compared to the quantity spiked, to produce a recovery quotient. The orange and blue bars represent the % recovery of fibres and particles respectively for the sample. The grey bars represent the % of particles that were not of the spiked material and so must stem from secondary contamination.

SEM-EDS Results and ‘Determinacy’

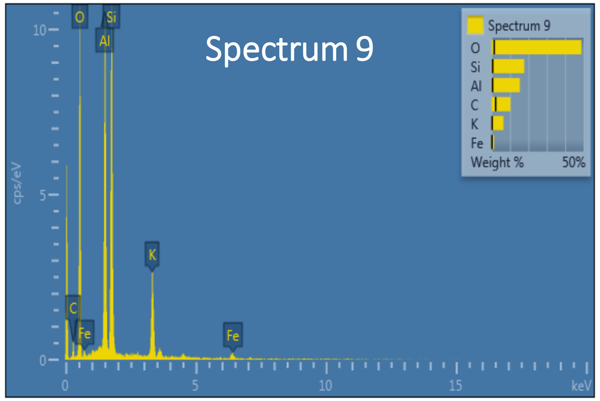


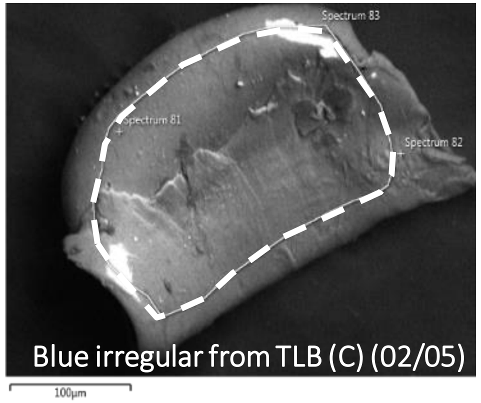
Black irregular (TLB (A) 02/05)



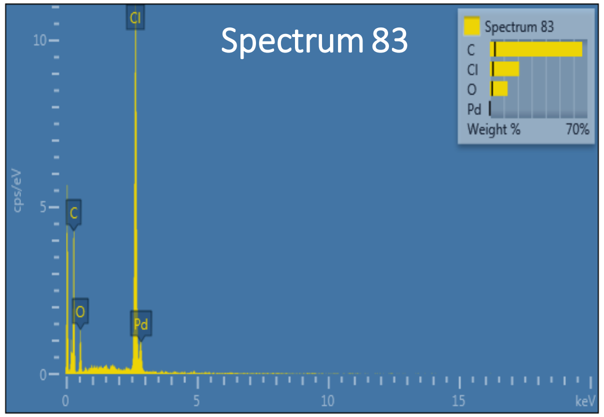


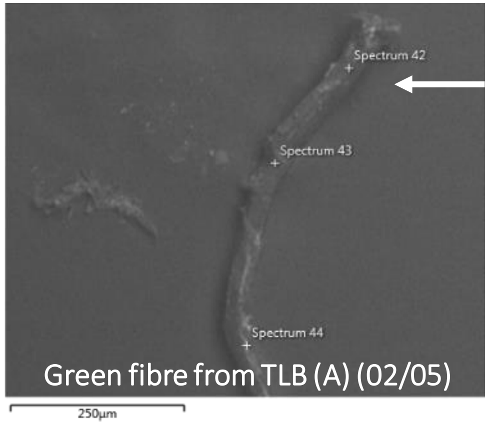
Green irregular (LW 3D 09/05)



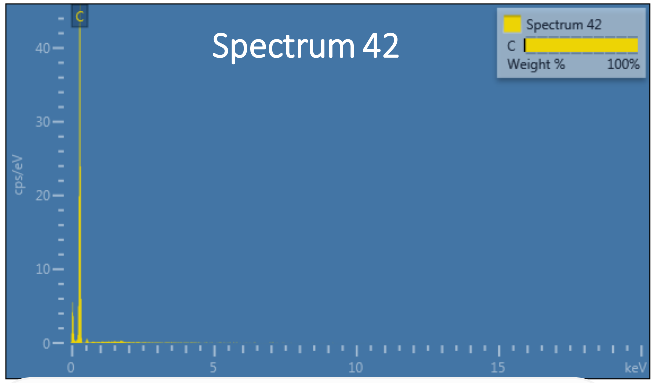


Blue irregular from TLB (C) (02/05)





Green fibre from TLB (A) (02/05)



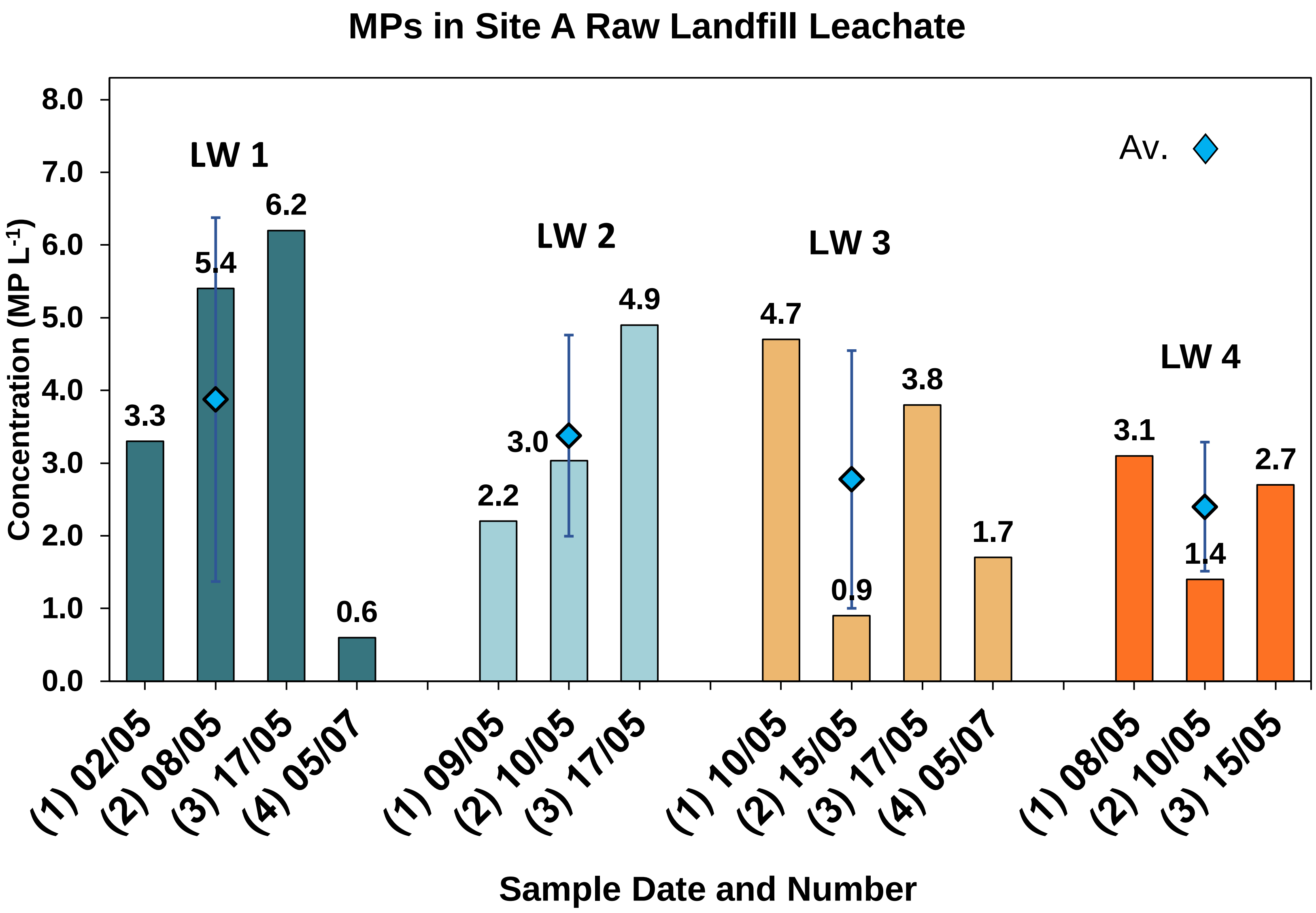
Considered MP - 28
EDS Intense C/Cl - 22
Determinacy - ~79%

Wagner, J., Wang, Z., Ghosal, S., Rochman, C., Gassel, M. and Wall, S. (2017). Novel method for the extraction and identification of microplastics in ocean trawl and fish gut matrices. Analytical Methods, 9(9), pp.1479-1490.

28 particles examined by SEM (Hitachi S3000) with an associated Link Isis EDS system. Their spectra were interpreted to assess degradation/fragmentation features of particles, and whether they were likely of plastic material, to indicate how determinate the analysts’ MP selection was.

Particles that were characterised by a strong carbon peak, without a large diversity of other elements in their spectra, were considered as likely plastic material. Note that to confirm whether the particles are plastic, and the polymer type, FTIR or Raman would be required.

Results - Raw Leachate



Microplastics (MPs) were indeed recovered from all landfill samples. The lowest average concentration of MPs was found within LW 4 (~1-2 years old), and the highest found in LW 1 (~20 years old).

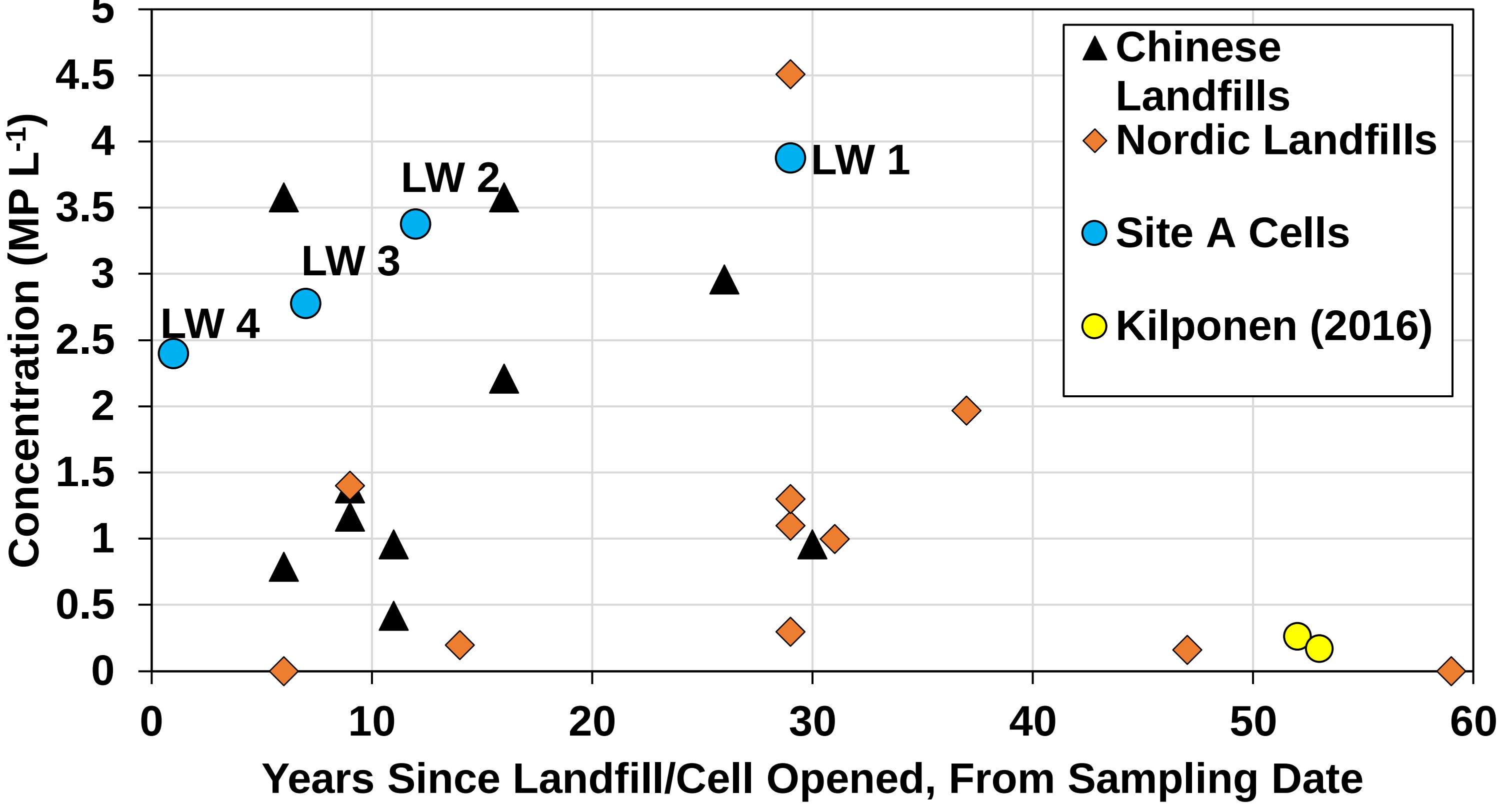
The concentration of MPs is going to depend primarily on two factors: (1) where MPs are distributed within the leachate of the well, and (2) the height of the pump and the aperture of its filter.

Mostly fibres were discovered, but greater variation of particles were observed in leachate from recent landfill cells than older cells.

As most MPs were less than 500 µm, the fragmentation process within landfills is likely severe.

Raw Leachate

MP Concentration vs Years Since Landfill Operation Commenced



Results from He et al. (2019), van Praagh et al. (2018), and Kilponen (2016) are graphed alongside the present study's average raw leachate results. A similar conclusion can be drawn to Su et al. (2019), that landfill age has little effect on the concentration of MPs. However they did find that landfill age influenced MPs found in the solid waste refuse samples.

Related literature:

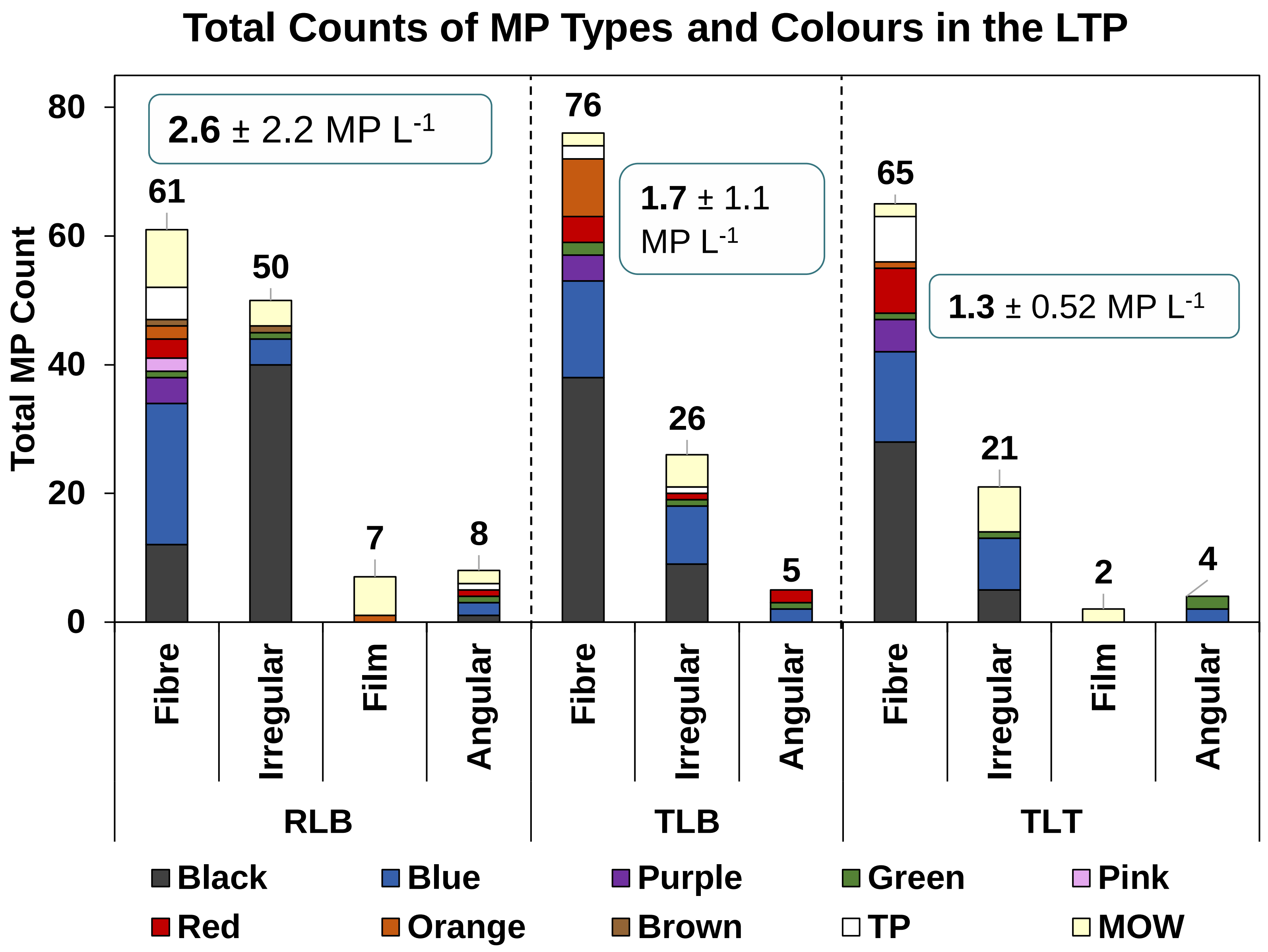
He et al. (2019). Municipal solid waste (MSW) landfill: A source of microplastics? - Evidence of microplastics in landfill leachate. Water Research, 159, pp.38-45.

Kilponen, J. (2016). Microplastics and Harmful Substances in Urban Runoffs and Landfill Leachates: Possible Emission Sources to Marine Environment. Lahti University of Applied Sciences, pp.1-84

Su, Y., Zhang, Z., Wu, D., Zhan, L., Shi, H. and Xie, B., 2019. Occurrence of microplastics in landfill systems and their fate with landfill age. Water Research, 164.

van Praagh et al. (2018). Microplastics in Landfill Leachates in the Nordic Countries. Denmark: Nordic Council of Ministers, pp.1-53.

Treated Leachate

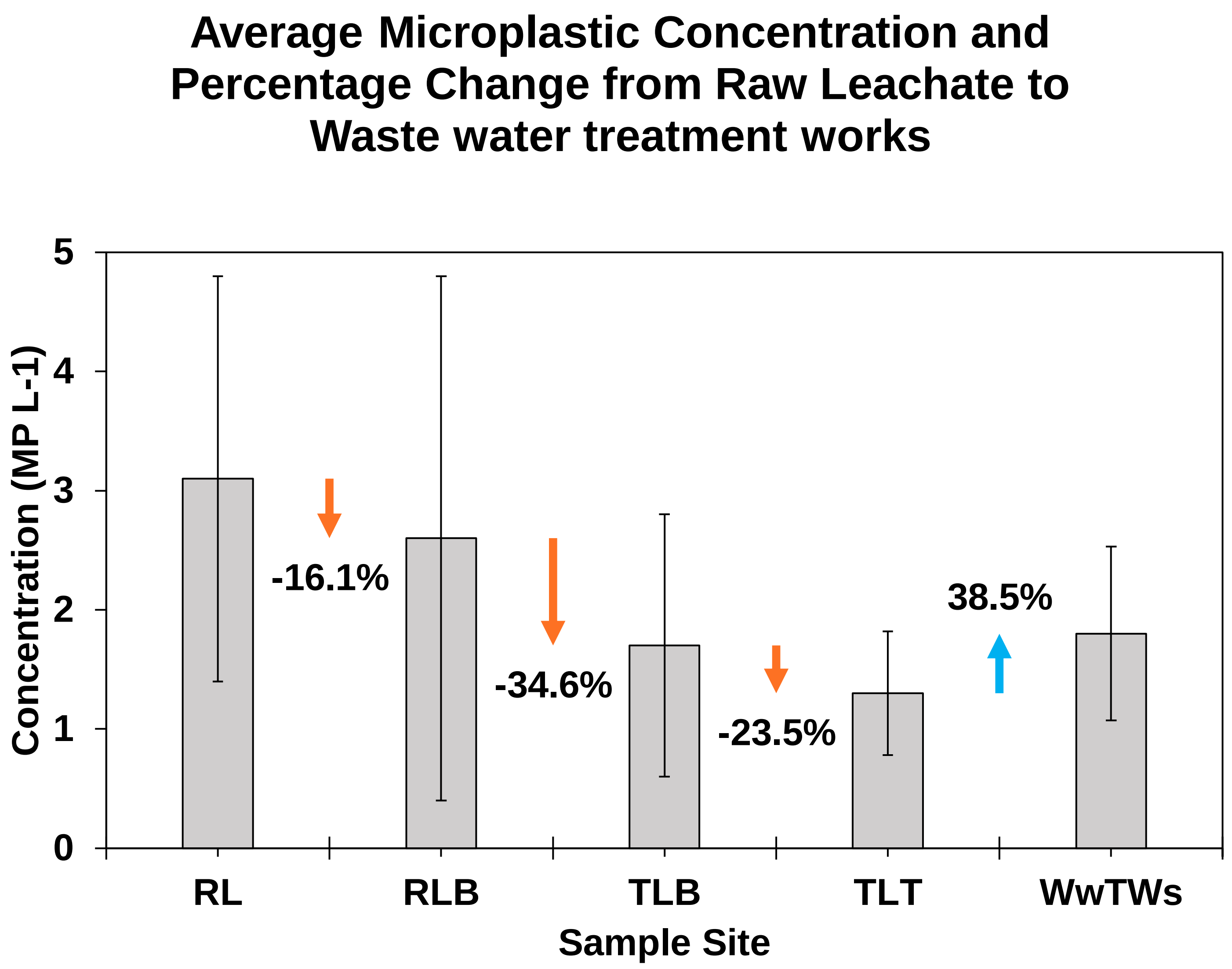


MPs were investigated across a sequence batch reactor (SBR) leachate treatment plant (LTP) from raw leachate (RLB) to treated leachate (TLB) to that discharged from the LTP to sewer (TLT). A decrease in MP concentration across the process suggests that MPs are retained within the LTP process, most likely the sludge.

Leachate fed to the LTP was predominantly from more recent landfill cells. The LTP is open to air and so is exposed to potential aerial deposition of MPs. Bright fluorescent fibres are likely from PPE worn by site staff.

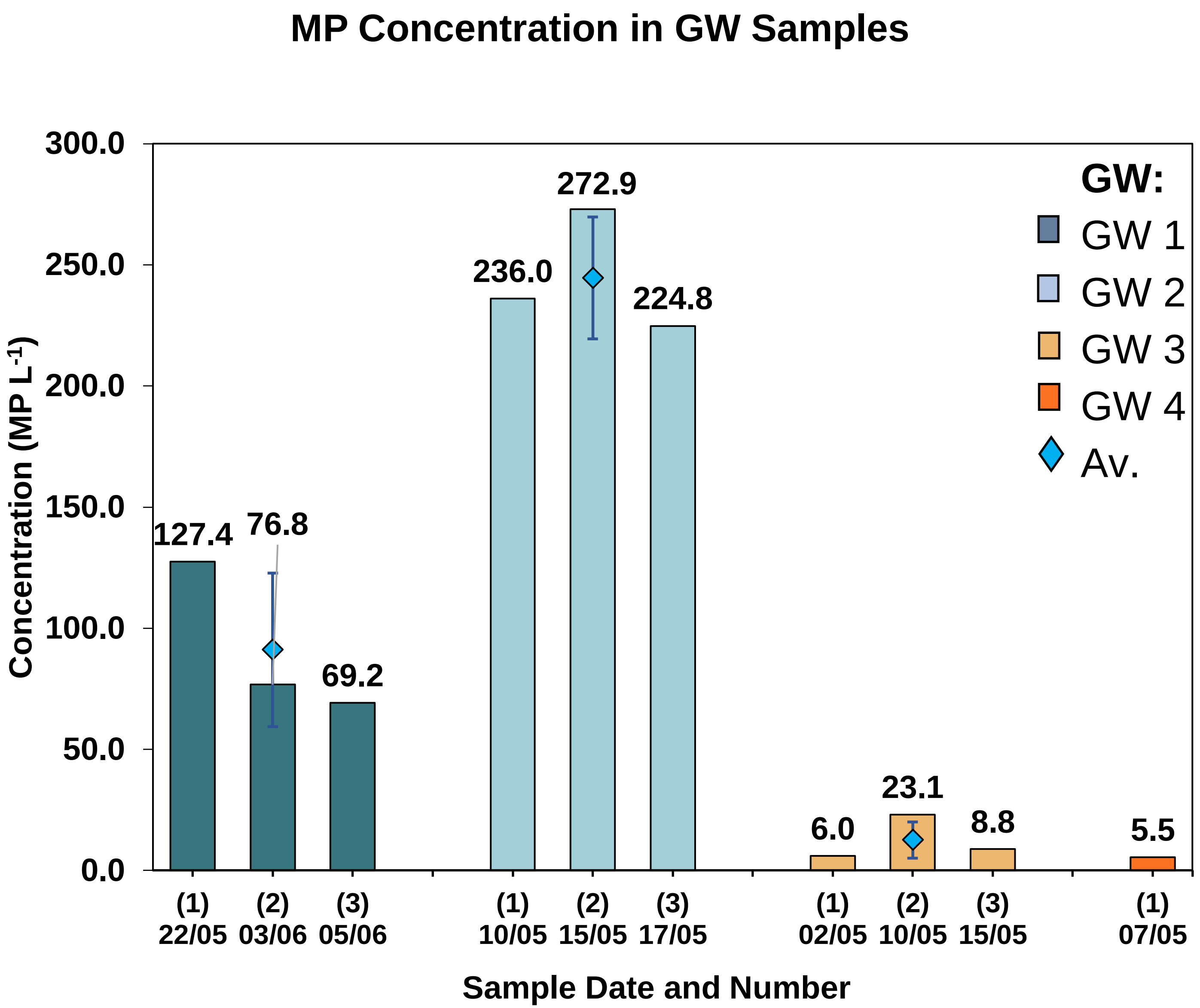
Comparing the average concentration of MPs in raw leachate from the leachate wells, there is a 58.1% decrease in MP concentration by the point leachate is discharged from site to the sewer.

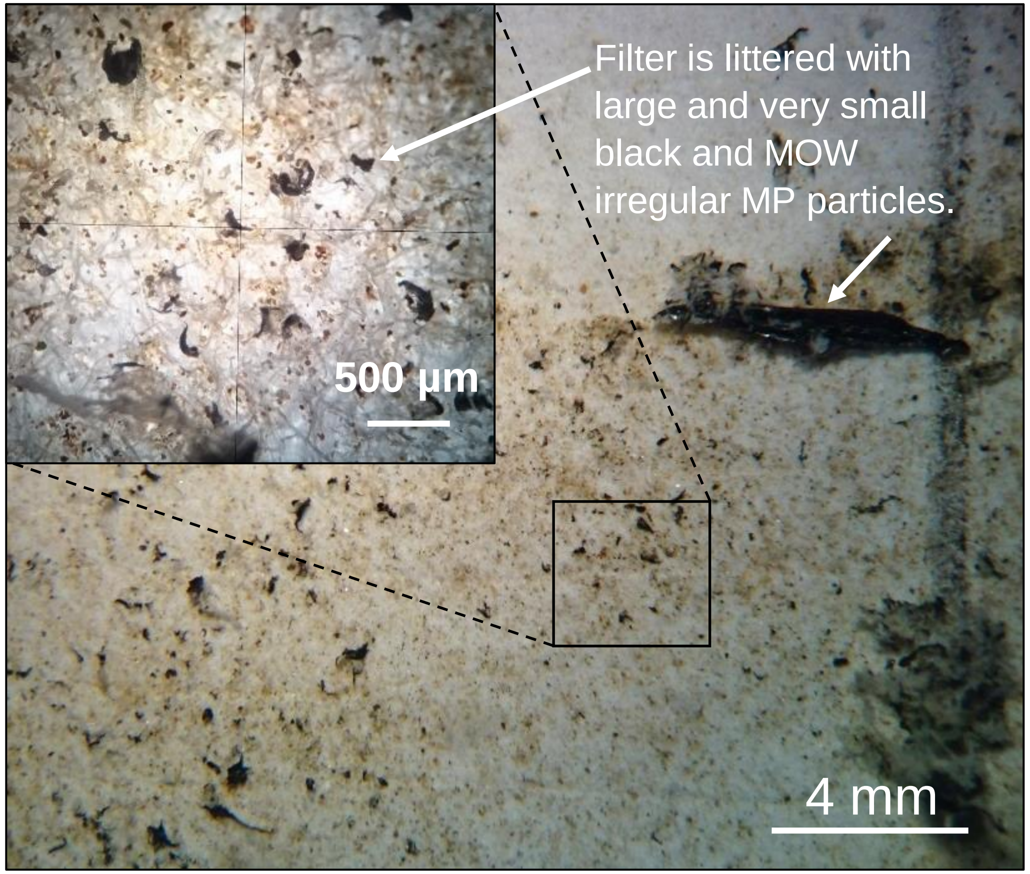
Reductions Across LTP Process



MP concentration decreases from raw leachate to the discharged treated leachate. Treated leachate is then sent to a sewage treatment works for further treatment before discharged to a surface water stream. At this surface water outlet, the concentration of MPs increased suggesting MPs were also being released from the waste water treatment works (WwTWs).

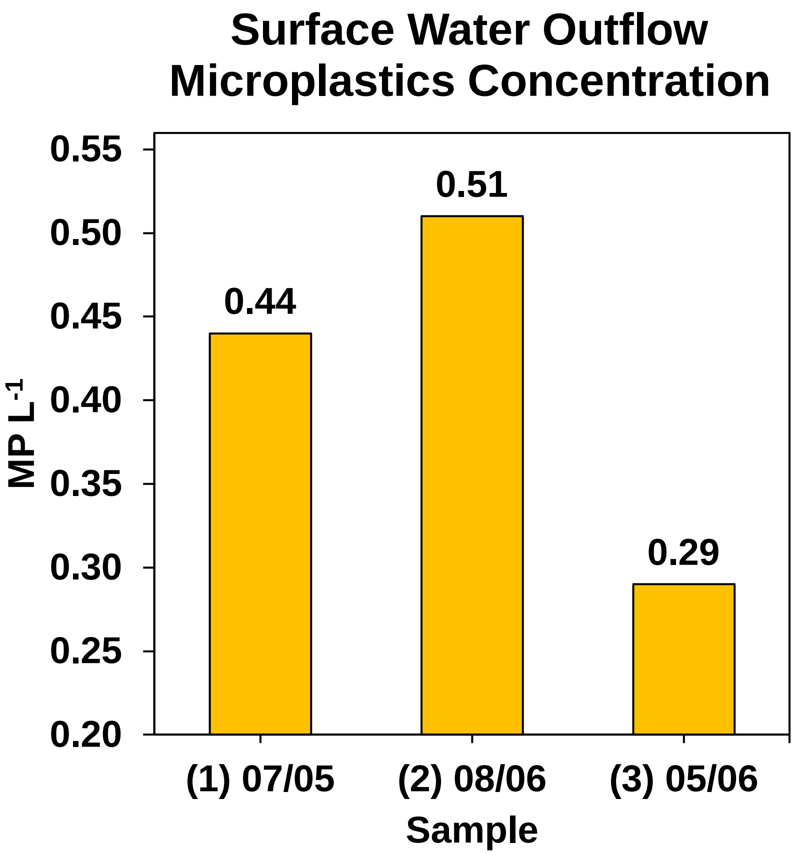
Groundwater





Considerable number of irregular shaped black and mottled opaque white microplastics in groundwater, very likely originating from the sampling tubing and borehole themselves due to the abrasive nature of purging groundwater. Microplastics are unlikely to stem from the landfill itself.

Surface Water



Surface Water Outflow (Waddell, 2019)

100 L of surface water was filtered per sample at the landfill surface water outflow (SWO). Concentrations were the lowest experienced per litre of sample.

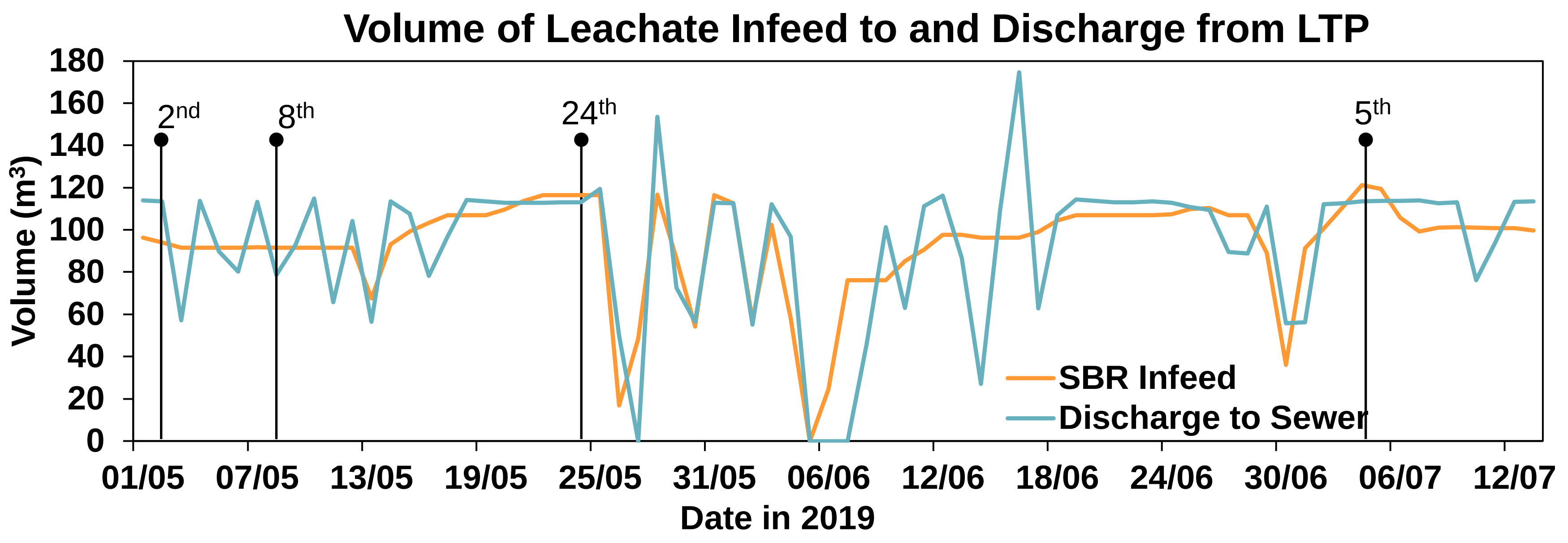
However, with flow rate and, subsequently, discharge calculated, the potential 'load' of MPs could be estimated.

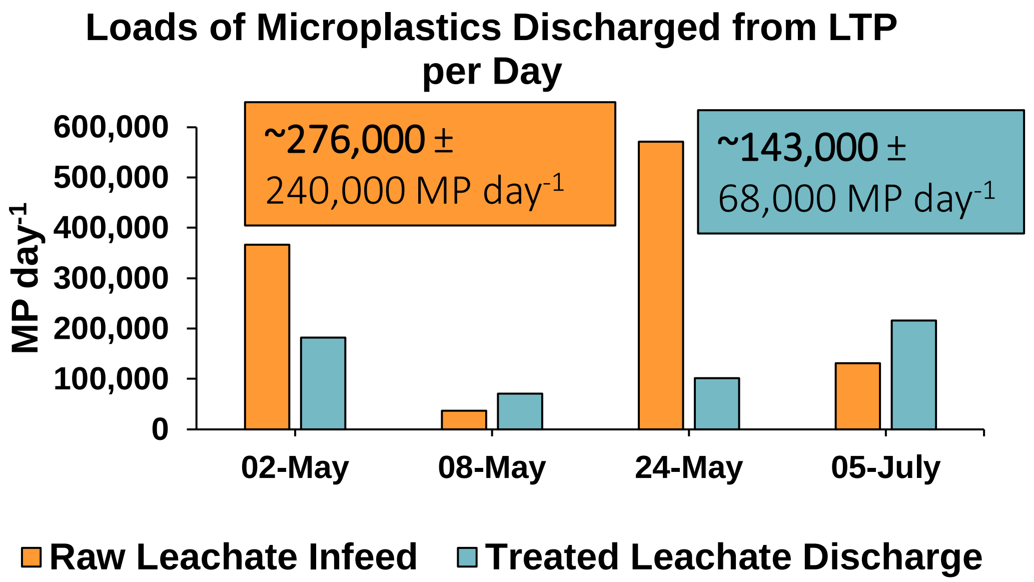
SWO: 5858 m^3 day^{-1} multiplied by average of 413 MP m^{-3} =

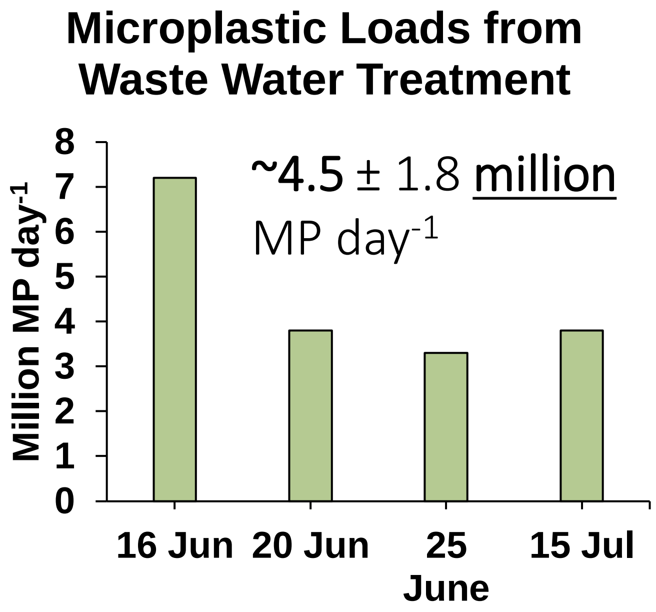
Approximately 2.4 million MP day^{-1}

Discharge was calculated using area of water released from the outflow pipe, multiplied by velocity. The same calculation was applied for the WwTWs outflow.

Microplastic Loads Released to the Environment







This 'load' of MPs discharged was also calculated for the leachate treatment plant effluent. Using discharge values taken from the LTP computer system, approximately 143,000 ± 68,000 MP day^{-1} discharged to the WwTWs per day.

This value is dwarfed by comparison to the calculated discharge from the WwTWs outflow: Approximately 4.5 ± 1.8 million MP day^{-1}

Limitations, Conclusions and Recommendations

Limitations:

Landfill sites are complex and dynamic. Leachate is at times recirculated or transferred between cells which casts doubt over the accuracy of data for particular landfill cells/age. Almost all landfill infrastructure (liners, piping) is made from HDPE, so some MPs may have originated from this. Secondary contamination in the laboratory was highlighted as an issue. A greater sample volume (E.g., 100L) for leachate samples could have produced more precise results. Due to the mesh size, particles < 25 µm were omitted. There was potential misidentification of 21% by the analyst as suggested by SEM-EDS results.

Conclusions and Recommendations:

- A cost effective, relatively rapid method of good recovery and determinacy has been established for landfill samples, but could be more robust with the use of FTIR analysis for confirmation of plastic material. **(Aim 1)**
- Many new features of landfill investigated for microplastics previously unassessed in the literature. **(Aim 2)**
- The landfill is a source of microplastics (>25 µm), but is unlikely a significant one. **(Aim 2)**
- Leachate treatment appears to reduce concentrations by the point it is discharged, likely retaining particles in the sludge.
- Waste water results are comparable with other studies, and may be highlighted as key discharge points of microplastics.
- Ground/surface water results are concerning, but require further study to see if their microplastics originate from the landfill.
- Further studies should make use of a clean room and positive pressure fume cupboard to avoid secondary contamination.