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Microplastics in groundwater: a literature review

Environmental Change, Adaptation & Resilience Programme

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BRITISH GEOLOGICAL SURVEY

ENVIRONMENTAL CHANGE, ADAPTATION & RESILIENCE
PROGRAMME

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1 Introduction

Attention on microplastic (MP) pollution has increased greatly over the past decade, particularly in aquatic systems such as the marine and freshwater environments, where publications focus on both sources and fates of MPs, as well as the ecological/toxicological impacts of their presence on organisms within those environments. Literature exploring the extent to which MPs occur and interact with groundwater systems, however, remains limited. Within this limited body of research an even smaller subset of publications focussing on specific studies with experimental designs exist. The potential significance of MP occurrence in groundwater from a perspective of risks to drinking water supply, groundwater dependant ecosystems and their toxicological potential to be enhanced by their ability to vector both organic and heavy metal pollutants, is slowly becoming clearer as new research emerges, but more research is needed on this topic.

Using the following keyword search terms (groundwater + microplastics + FTIR) and search engines such as google scholar and Web of Science, this short literature review identified groundwater studies alongside wider literature on MPs. It explores the methodological designs, limitations of sampling, isolation and analytical techniques employed and identifies aspects for consideration in future groundwater research on this topic.

2 Groundwater focussed studies

2.1 KARST AQUIFER SYSTEMS

There are only a limited number of field-study based publications identified which look at MPs in groundwater, one of which is Panno, et al. (2019) which investigated MP contamination in Karst groundwater systems. Karst systems are identified as being of significant interest due to their geographical extent globally and because they constitute approximately 25% of the world's drinking water sources (Ford and Williams, 2007). Karst systems are also documented to be of particular interest due to their vulnerability to surface ingress of dissolved and particulate pollutants. The study focussed on determining the presence of MPs in this environment by sampling groundwater wells and springs in a Karst region in Illinois, USA and on whether MPs were detected alongside anthropogenic contaminants including those associated with pharmaceutical and personal care products (PPCPs). The rationale for the study was that since the presence of enteric bacteria and PPCPs had already been found in this environment and was linked to sources such as septic discharge, road runoff and agricultural activities, the presence of MPs could also be expected (particularly originating from septic discharge) The findings of this study confirmed the presence of MPs, particularly MP fibres, with no microbead or fragments, indicating the prevalence of smaller MPs in groundwater systems. Additionally, MP particle concentrations measured in samples were noted to be much lower than those found in surface waters (with maximum concentrations of 15.2/L compared to maximum surface water samples of ~90/L). Pyrolysis (py)-Gas Chromatography Mass Spec. (GCMS) chemical analysis of some of the samples identified MPs as being polyethylene. No other plastic types were identified, but the study documents that other samples were destroyed during the process of py-GCMS analysis, indicating that further sample collection may have been beneficial as it would have provided more opportunities to identify other source materials of plastics. The paper concludes that whilst septic discharge appears to be a main contributor to the presence of MPs in groundwater samples and surface ingress is the primary source, there is insufficient evidence to solidly link their presence. It highlights the need for further investigation into MPs in groundwater to better understand their dynamics within the system and potential impacts on its ecology.

2.2 OTHER AQUIFER/GROUNDWATER SYSTEMS

Along with Karst systems, behaviour of MPs in sandy and alluvial sedimentary aquifers have also been documented (Samandra et al., 2022; Song et. al. 2019). The Song et al. (2019) study, like the Panno et al. (2019) from the same year, builds upon previous research by the research group looking at organic pollutant and enteric microbe contamination of groundwater to incorporate an

assessment of MP contamination. However, in this study, MPs in the form of polystyrene bead nanoparticles (50-200nm) with variable surface functional groups are measured as they move through the sand/aquifer column. Unlike Panno et al. (2019), the introduction of MPs to the aquifer media is carried out in a laboratory environment. The study documents that organic matter influences the stability and increased mobility of MPs in aquifers, both with particle size and electrical charge associated with surface functional groups, and in turn the potential behaviour and fate of weathered micro/nano-plastics in aquifer environments. The findings of this study are replicated elsewhere, for example in Zhang et al. (2020), where biofilm formation on micro-nanoplastics (180 nm–125 µm) greatly reduced the ability of flocculation in drinking water treatment processing to remove MPs. The Samandra, et al. (2022) study found on average ~38 MP particles/L persample of PET, PP, PE and polystyrene composition. Samples have however been collected from an alluvial sedimentary aquifer, and it unclear as to what extent agricultural practices have influenced MP contamination in this system. The average size of MP particles found (89µm) is however concurrent with sizes found in other studies such as Panno et al. (2019).

A further study carried out in 2019 and published in 2021 (Selvam et al.2021), also documented the potential for MPs, both particulate and fibrous in nature, to act as vectors for heavy metals. Unlike the Song et al. (2019), Selvam et al. (2021) found MPs in both surface and groundwater samples taken from the same hydrological system, rather than a specific MP being introduced. Analysis of these MPs showed a differential in size profile of MPs between surface and groundwater samples (smaller MPs being present in groundwater, with some with diameters down to between 6-30µm), and polyethylene (>50% of MPs sampled), polyamide (nylon) and polyester fibres dominating the plastic composition of MPs in the groundwater samples (compared to surface water, where a wider range of MP plastic types including PVC and polypropylene were also found). Whilst not exploring the issue as a subject in the paper, Selvam et al. (2021), cites biofilm formation on MPs as a potential influencing factor in an MP's ability to vector heavy metals (and organic contaminants) and suggests further research is needed on this topic.

2.3 DRINKING WATER

A limited number of groundwater-specific studies exist for drinking water were identified (e.g. Mintenig et al. 2019; Johnson et al., 2020). Most notable of these is Mintenig et al. (2019). This study focuses on determining the presence of MPs in ground water and drinking water (>20 µm) using Fourier transform infrared spectroscopy (FTIR) imaging. The study focussed on five sample locations in North West Germany, taking water from pre-treatment abstracted ground water and post-treatment drinking water sampling points at water treatment work sites as well as one in-network sample point. The rationale for this study was to identify MPs in drinking water sourced from groundwater and to assess if and where contamination with MPs might occur within the treatment process. Whilst the study acknowledges the potential for MP contamination in raw groundwater, it is not the primary focus of the study. MPs were identified as 50-150 µm in size, and polyethylene, polyamide, polyester, and PVC in composition. Whilst the study did identify MP particles in pre-treatment samples, the results were not completely conclusive in demonstrating the presence of MPs in groundwater due to the low levels of MPs found (MP particles ranged from 0-7 particles/m³ with an overall sample mean of 0.7 particles/m³), and compared to the near identical mean concentrations found in blank samples, indicating that MPs found may have also been as a result of sample contamination. The study, being the first of its kind, has been cited in several wider review publications such as Koelmans et al. (2019) and Erkes-Medrano et al. (2019) as evidence of MPs in groundwater sourced drinking water. It is also worth noting that this was the first MP study to sample large volumes of water (300–1000L of raw water and 1200–1500L of drinking water), taking water from both the intake and outflow of a water treatment works (Koelmans et al.2019), giving an indication of the potential volumes of water required for sampling when attempting to detect low levels of MPs.

3 Wider reviews and studies

Alongside the specific primary studies mentioned above, there are several recent review and focus publications regarding MPs in groundwater. These include Re (2019) which highlights the growing concern over MP fibres as potential contaminants on groundwater. The paper cites Panno et al. 2019 study as one of its sources alongside multiple primary studies assessing MPs in both soils and surface freshwater environments. Re (2019) highlights the overall lack of body of knowledge on MP fibres and how they may pose a potential threat to human health via groundwater in the same way that asbestos has been documented to (Wallis, et. al, 2020; Willenbring, 2016), given potential for their mobility. As mentioned above, reviews into the presence of MPs in drinking water include Koelmans et al. (2019) and Erkes-Medrano et al. (2019), which highlight the need for further research into the potential for MPs to be introduced into drinking water from groundwater sources.

Further publications by Kim and Lee (2020) set out more geographically specific concerns for potential MP contamination in South Korean groundwater. The paper highlights the difficulties in assessing and quantifying MPs in groundwater (an issues discussed later in this paper) and the importance of the hyporheic zone in freshwater systems as a potential transmission point for MPs into groundwater. This is also covered by Drummond, et al. (2020). Whilst technically a field based/primary paper, little is explained of the methodology utilised for physical assessment of MPs, instead focussing more on the hyporheic exchange rate of MPs into groundwater from river sediment environments. The paper highlights that potential transmission of low-density polymers such as polyethylene are much higher than other plastic types; a finding concurrent with some of the other studies explored in this paper that also identify the dominance of this plastic type in MP sample composition.

Finally, studies such as Wanner (2021), Zhang et al. (2022) and Huang et al. (2022) highlight the role of soils and the terrestrial environment as a source of MP contaminants to groundwater. These can act as a significant potential source of MPs to groundwater, both through the vertical migration of MPs through soil horizons as a pathway, but also via surface runoff into waterways, and then via hyporheic zones into groundwater (Schell, et. al, 2022). Soil type, along with polymer type and polymer size have the potential to significantly govern how and when MPs might migrate to groundwater (Dong et al. 2022; O'Connor et al. 2019; Wu et al. 2020; Zhang et al. 2022;). Wanner (2021) also highlights the role of MPs entering groundwater from this source to vector organic pollutants of concern such as pesticides into groundwater and suggests that for a key area of further research, more effort needs to be put into understanding the influence of particle size on the transport of organic pollutants, the influence of eco-corona (covering of MPs by organic material such as humic acids, polysaccharides and proteins) on sorption of organic pollutants and better modelling of MPs through agricultural soils into groundwater. Significant sources of MPs into soils include biosolid application, addition of composts/digestates and mulching (Bandopadhyay et al. 2018; Crossman, et. al, 2020; Hurley and Nizzetto, 2018; Vithanage et al. 2021). Unlined historical landfills from gravel extraction could also be a potentially important source of MP and could circumvent natural attenuation in the soil and subsurface and lead to direct groundwater contamination (Wan et al. 2022).

3.1 PLASTICISERS

While investigation into the presence and behaviours of MPs in groundwater has only taken place recently, a slightly larger and more well-established body of literature exists on the presence of plasticisers in water. Studies such as Lapworth et al. (2015), Stuart et al. (2014), Manamsa et al. (2016a), Manamsa et al. (2016b) and Yang et al. (2017) highlight the presence of plasticisers in groundwater as part of wider studies into the presence of micro-organic contaminants in groundwater. A wider body of literature also exists looking at the presence of plasticisers, particularly phthalates, in drinking water and bottled drinking water, where concern over human health has been more prominent (for example, Kim and Kannan, 2018).

Groundwater studies such as those mentioned above are potentially useful in providing a framework/approach for addressing experimental requirements to sample for MPs in the same environment. However, in many cases, plasticisers themselves are not the main focus of the

studies conducted, with a wider focus instead being placed on broader suites of microcontaminants. Plasticiser risk to both human (both in terms of endocrine disruption and carcinogenicity) and ecosystem health (e.g. disruption of soil C/N nutrient cycling, with the potential impact this could have on groundwater quality in addition to more direct disruption on groundwater ecology) are identified by studies such as Yang et al. (2017), Lu et al. (2020) and Qi et al. (2020), with the latter considering plasticisers as potentially being the 'primary pollutant load' of MPs themselves. Specific plasticisers such as Bisphenol A (BPA), PAE (phthalic acid ester-based plasticisers) and P based plasticisers such as tributyl-phosphate are identified in some of the primary papers reviewed.

Aspects which are not explored in these papers include the potential interactions between plasticisers and MPs in the groundwater environment. For example, the presence of microplastics in groundwater environments where plasticisers have been identified and sampled, may influence concentrations detected and influence estimations of concentrations anticipated to be present in those systems. Additionally, the potential for plasticisers to be used to detect or predict the presence of MPs in groundwater samples has not been identified, either as an indication of a previously unidentified pool of MPs in the subsurface or for predicting the movement of MPs through less well-connected aquifers where the greater solubility of plasticisers may allow for their faster movement. A limited body of research has however identified the potential use of plasticisers as tracers for MPs in marine trophic systems, suggesting that this approach may be transferable to groundwater studies (Gugliandolo et al. 2020). Plasticiser detection may prove to be a useful tool in attempting to assess the presence of smaller MPs closer to the limit of reliable detection where more conventional spectroscopic techniques such as those discussed later in this document, may not be as effective.

4 Quality control of MP studies

Alongside reviewing publications looking at groundwater MP studies and wider literature, another objective of this initial review has been to look at methodological design and quality of MP studies. Notable papers that stood out as being important to mention include Provencher et al. (2020) and Viaroli et al. (2022).

Whilst there have been many studies and publications exploring the various aspects of MPs in different environments ranging from polar oceanic systems to terrestrial agricultural systems, a growing concern has emerged over quality control and standardisation of MP studies. Aspects of this include the selection of standardised analytical assessment techniques, the use of a standardised approach to address contamination of samples (something that will be of particular importance when looking at environments with potentially low concentrations of MPs such as groundwater) and consistent use of language/nomenclature when describing results of MP research in publications. Whilst a large body of knowledge and publications already exists for MPs in other environments, the relatively recent advent of research into MPs in groundwater gives an opportunity to implement some of these recommendations into research in this subfield going forward. Given that no single approach may be applicable to assessing MPs, particularly in groundwater (Provencher et al. 2020; Re, 2019), and in different hydrogeological settings (Viaroli et al. 2022), a documented standardised sample preparation and analysis approach early in the development of this research area could prove highly beneficial to facilitate greater assurance of findings and also enable better comparability of results between studies. It is also noted that papers relating to other environments such as Qi et al. (2020) do also call for a standardisation of MP detection and analysis; in this case for analysis of MPs in soils where similar challenges have also been identified.

5 Key considerations for groundwater sampling, isolation and analytical techniques

In reviewing literature aspects of various stages of the process of assessing MPs (i.e. sampling, isolation and analytical approaches) have been identified. These are documented here to be captured and considered in any methodological design to be used for future groundwater assessments.

5.1 SAMPLING

In the field studies reviewed, whilst varying volumes of water have been extracted for capture of MPs, the overall impression has been that large volumes of water have been required for extraction. Thought will need to be given to the selection of suitable equipment to achieve this, particularly if groundwaters are turbid and may rapidly block filters with small cut-off sizes. Additionally, where groundwater is sampled at differing depths, thought will need to be given to borehole construction and the materials/equipment used in obtaining water samples, for example stainless steel rather than plastic tubing (such as that used in the Johnson et al. (2020) study assessing surface water for drinking water supply rather than groundwater), to ensure minimisation of contamination of samples collected. An example of where this has been an issue which has thrown into doubt findings, is the detection of plasticisers in Manamsa et al. (2016b), where the authors cannot be sure that the detection of plasticisers in some samples collected is not because of leaching from the equipment used.

5.2 SAMPLE STORAGE AND CONTAMINATION

In some of the studies reviewed, details on sample storage and transport of samples have been outlined giving consideration of possible risks of MP contamination. These include considerations over the materials used to hold samples as well as temperature-controlled storage to minimise biological degradation of MP samples. It is also worth noting in this section that some studies also used blank samples to address the issue of contamination, both for handling and storage of samples in materials that may introduce an element of contamination (for example Manamsa et al. (2016b) and Mintenig et al. (2019)).

5.3 FILTRATION OF GROUNDWATER SAMPLES

Several of the primary studies discuss filtration of groundwater samples collected. In each, descriptions are given of the materials of filters used. Avoidance of plastic based filters (and plastic filter casings) will be particularly important with groundwater sampling, given the anticipated low concentrations of MPs. Additionally, the use of full method blanks will again be crucial.

5.4 PREPARATION OF SAMPLES TO REMOVE BIOFILMS

This is particularly important where samples are going to be analysed by visual microscopy or FTIR/Raman spectroscopy, as rough surfaces and biofilms can provide false readings. In carbonate and other sedimentary groundwater systems the removal of calcite by treatment with acidic solutions is essential due to the interference of calcite MPs with analytical methods. Methods for removing biofilms include the use of KOH, peroxide and Fenton's reactions. Concern has been raised however in some literature (e.g., Kühn et al. 2017; Thiele et al. 2019) that the use of some oxidising agents to remove biomatter can also dissolve some plastic types, or distort their surface chemistry sufficiently (e.g. Polyamide) to make them undetectable via spectroscopy when compared to known databases. Although this is not thought to be a significant issue with larger particles, (Tagg et al. 2017), this may be more of an issue for MPs within the size range relevant to groundwater samples. One potential solution for this may be to collect sufficient sample material to analyse both with py-GCMS and spectroscopy approaches, and to undertake a cross-comparison afterwards.

5.5 SUITABILITY OF CURRENT ANALYSIS TECHNIQUES FOR GROUNDWATER STUDIES

The following analytical techniques have been identified in literature, a brief overview/critique is provided for each.

5.5.1 Visual microscopy

This technique has generally been used to assess larger sized MPs in marine/aquatic samples. With the anticipated (small) size of MPs likely to be encountered in groundwater samples, it seems unlikely that this approach will be appropriate for sample analysis.

5.5.2 FTIR & Raman Spectroscopy

FTIR and Raman spectroscopy techniques have been used in several of the papers reviewed to assess both loading and composition of MPs in samples. Both methods work by measuring the intensity of light reflected from different plastic samples across a range of spectra. In general FTIR is better with larger MPs and a homogenous distribution of plastic types (due to assessing a wider spectra) whereas Raman spectroscopy is better for smaller MP sizes (Cabernard et al. 2018; Thermofisher 2018;), however the latter is limited in terms of higher concentrations of MPs (which may not be as problematic when assessing anticipated MP concentrations in groundwater samples). Thought needs to be given to which technique or combination of techniques to use when sampling different environments with suspected variations in MP pollutant concentrations, although initial assessment appears to favour Raman spectroscopy. Both FTIR and Raman can be limited by MP rough surfaces and biofilms which may prove problematic when assessing groundwater MP samples. On a more pragmatic level, spectroscopic analysis of MP samples can be very time consuming, with analysis potentially taking in excess of 40 hours per sample without automation (De Frond et al. 2022). Raman spectroscopy has an advantage over FTIR in terms of having a lower size limit of detection (Käppler et al. 2016; Xu et al. 2019), 10 μm versus 25 μm (under practical settings for the FTIR with a 1.5-hour scan time). This however may be offset by FTIR spectroscopy being automated, meaning potentially requiring far fewer man hours per analysis of individual samples – something which could potentially be very significant when financially planning any investigatory or regulatory sampling regime.

5.5.3 GCMS /Pyrolysis GCMS (py-GCMS)

py-GCMS, sometimes abbreviated to 'pyrolysis' has been utilised in a couple of the studies reviewed. The technique involves the thermal destruction of MP samples and analysis of the by-products evolved by GCMS. Py-GCMS is capable of analysing both MP load as demonstrated in studies such as Funck et al. (2020) and Fischer and Scholz-Böttcher (2019). Plasticisers have also been detected using GCMS without the use of prior pyrolysis.

py-GCMS has the advantage of bypassing issues around biofilms and weathered surfaces which plague surface spectrometry. It also has the advantage of being a less time-consuming process than spectroscopy as samples potentially need less preparation. For the technique to be effective however, reference samples or a library of compounds are required to compare readings against. For some plastics, these already exist, potentially negating the need to produce replicate results, however further investigation is required to assess how feasible it may be to obtain these. The process is also potentially limited by MP size, which may prove problematic in physical preparation of groundwater samples to be analysed (Strungaru et al. 2019), given the potentially very low numbers and small size of nano-MPs which may be sampled. There is also the issue of detection limits for py-GCMS, it is as yet uncertain how much MP needs to be loaded on to a sample to generate a positive detection above background levels and this may limit the use of py-GCMS to more contaminated samples. In this sense it could potentially be useful as a screening technique to prioritise further analysis by FTIR.

A multi-analytical approach is likely required to fully assess samples taken (Re 2019), most likely FTIR and/or Raman spectroscopy coupled with py-GCMS, however this may be dependent on the nature of the investigation/experiment and if quantitative or qualitative information is being sought. Further harmonisation between analytical techniques is still required (Primke et al. 2020).

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