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Tracing sources and pathways of micro-organic contaminants in a complex peri-urban floodplain aquifer

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Peri-urban floodplains include a diverse range of potential contaminant sources and complex surface water-groundwater interactions which control the transport of emerging contaminants in underlying groundwater systems. The impact of an unlined legacy landfill located within the floodplain of the River Thames in southern United Kingdom is assessed with two downgradient multi-depth piezometer transects, and results compared with a control piezometer site previously shown to be unaffected by the landfill leachate plume. Groundwater samples collected from two monitoring rounds in 2017 and 2018 from a network of sites showed the landfill to be a heterogeneous source of contaminants. Over 580 micro-organics (MO) including pharmaceuticals, pesticides, lifestyle products, surfactants and per- and polyfluoroalkyl substances (PFAS) were screened in groundwater and surface water using an semi-quantitative LCMS screen. Use of sucralose (an artificial sweetener) post-dates the closure of the landfill. Sucralose found within the landfill plume, together with contaminants found within the control site, suggests a more recent source of contamination also contributes to groundwater contamination within the floodplain. As these samples do not match the fingerprint of the River Thames, it is proposed that the additional contaminant source could be infiltration from either a canal or train wastewater discharge upgradient of the landfill. Spatial variation shows contaminant attenuation down gradient of the landfill with higher concentrations found in the deeper part of the alluvial gravel aquifer system possibly due to local recharge diluting shallow groundwater. Although closed 40 years prior to this study, groundwater sampling has demonstrated that the landfill continues to be a source of persistent, mobile contaminants.

KEYWORDS
artificial sweeteners, groundwater, PFAS, pharmaceuticals, tracers

1 Introduction

A wide range of anthropogenic organic compounds are now being detected in the environment. These can be used as tracers of different sources of pollution from a range of human activities including waste disposal in landfills (Busch et al., 2010; Masoner et al., 2020; Masoner et al., 2016; Propp et al., 2021), wastewater discharge (White et al., 2019), storm water discharge (Masoner et al., 2019), agriculture (Lapworth et al., 2015; Moreau et al., 2019; Yadav et al., 2015) and industry (Earnshaw et al., 2014; Lapworth et al., 2015; Loos

et al., 2010). Landfills have historically contained waste from a broad range of human activities (Busch et al., 2010) and contaminants within landfills can persist for decades (Barnes et al., 2004; Gooddy et al., 2014; Propp et al., 2021) with a direct pathway for groundwater contamination below unlined legacy landfills. There are an estimated 4,500 legacy landfill in southeast England alone (Environment Agency, 2024) and these do not conform to the environmental standards required for modern landfill sites, but rather use a dilute and disperse approach and are a potential risk to downstream water bodies. This was a widespread practice in England, and other regions of Europe, continuing until the early 1980s.

Rivers and groundwater can act as both sources and receptors of contamination within floodplain settings (Lapworth et al., 2018; White et al., 2019). River water is contaminated by run-off from a range of sources including agricultural and industrial land, wastewater treatment effluent or untreated wastewater (Bradley et al., 2017; Earnshaw et al., 2014; Loos et al., 2013; White et al., 2019). River contaminants can enter groundwater where the rivers lose water or where there is groundwater pumping in adjacent alluvial gravels (Ascott et al., 2016) and floodplain settings can have multiple sources of contamination (Stuart et al., 2014).

Concentrations of different micro-organic (MO) contaminants in groundwater are controlled by a range of factors; different sources, residence time in the soil/aquifer, sorption onto organic matter/clays, chemical and microbial breakdown as well as other more specific contaminant/usage properties (Belgiorno and Rizzo, 2012; Lapworth et al., 2012; Li et al., 2019).

Many pesticides have a long history of use and subsequent occurrence in natural waters due to a wide variety of uses. Pesticides are soluble and many studies have looked at their mobility (Boivin et al., 2004; Romero et al., 1996) and degradation in soil and groundwaters (Bradley et al., 2023; Gooddy et al., 2002; Johnson et al., 2000; Kolpin et al., 2000; Lapworth and Gooddy, 2006).

Pharmaceuticals are commonly detected in groundwater (Fram and Belitz, 2011; Lapworth et al., 2012; Lapworth et al., 2019) and incorporate compounds that are either prescribed or purchased over the counter. Lifestyle compounds include artificial sweeteners, stimulants and their breakdown products (Bunting et al., 2021) and are primarily used in food and drink and medications. Pharmaceuticals and artificial sweeteners can occur in historic landfills due to disposal of unused products (Masoner et al., 2016; Musson and Townsend, 2009), pharmaceutical industry waste (Holm et al., 1995) or from sewage sludge disposal.

Per- and polyfluoroalkyl substances (PFAS), surfactants and flame retardants are a large group of compounds incorporated in many products disposed of in landfill and have been detected downgradient of landfills (Busch et al., 2010) in groundwater (Lapworth et al., 2018; Manamsa et al., 2016; Moreau et al., 2019) and surface waters (Loos et al., 2017; Loos et al., 2007).

MOs have been detected in many different aquifers globally and their concentrations quantified to assess drinking water quality and used to assess connectivity to surface ingress (Sorensen et al., 2015), assess impacts of intensive pumping (Lapworth et al., 2018), urban sewerage leakage (Wolf et al., 2012; Vystavna et al., 2019) to fingerprint contamination in contrasting landuse types (Stuart et al., 2014) and to understand urban vs. rural sources of nitrate (B). However, there are by comparison few studies which have sought to apply them as tracers within floodplain settings to assess connectivity and sources of contamination (Wilske et al., 2020). This paper specifically builds on initial research undertaken by Stuart et al. (2014), who used non-polar GCMS screening methods and found more limited evidence for landfill impacts in this setting, through the application of polar MOs as tracers to understand sources of contamination within a peri urban floodplain setting. Polar MOs are more mobile within groundwater compartments compared to non-polar MOs, which tend to be attenuated due to attachment and sorption on to the surface of soil and aquifer matrices (Sanchez-Vila et al., 2015).

The aim of this study is to use a source-pathway-receptor framework to assess the sources and fate of polar MOs in a peri-urban floodplain setting, typical of lowland Europe. Specific objectives are to assess the occurrence of MO in the floodplain aquifer system and use polar MOs to trace the sources and fate of MOs within a complex alluvial aquifer system. Critically, these objectives are underpinned by a detailed understanding of the complex hydrogeology of the site through previous work (Sturat et al., 2014; Gooddy et al., 2014; Macdonald et al., 2018).

2 Methods

2.1 Study site

Port Meadow is a peri-urban floodplain to the north-west of Oxford comprising common land with grazing cattle and horses, and allotments (Figure 1). The Oxford canal, to the east of Port Meadow, is fed by streams to the north of Figure 1. Studies show that canal water recharges the floodplain aquifer (Stuart and Lapworth, 2011). A train track forms the eastern border of the study site. Approximately 25% of trains still discharged toilet waste directly onto railway tracks in 2017 (Butcher, 2017) and historically pesticides have been used to keep tracks clear (Lapworth and Gooddy, 2006).

Burgess Field, the largest of the landfill sites (Figure 1; Supplementary Figure S1), was in use between 1937 and 1980 and is now a nature reserve. The Burgess Field landfill is unlined, and the broad categories of waste disposed are heterogeneous (see Supplementary Figure S1). It is common for groundwater levels to be within the base of the landfill material (White and Macdonald, 2015).

The groundwater flow in the floodplain aquifer is, for the majority of the time, in a north-west to south-east direction towards the River Thames channel (Figure 1). The gradient may reverse during periods of high river stage, although it is unlikely that river water penetrates any more than a few metres into the floodplain sediments (Macdonald et al., 2018). The river commonly overbanks several times a year and this flood water recharges the floodplain aquifer in localised low-lying areas (Macdonald et al., 2018). Groundwater levels are generally within a metre of the ground surface beneath the floodplain. Groundwater levels were higher in 2017 than 2018 by approximately 17 cm. Groundwater levels within GBH3 and GBH9 were within the landfill material during both sampling rounds (Supplementary Table S4).

The floodplain sediments are composed of fine-grained silty clay alluvium, underlain by highly permeable sands and gravels which in

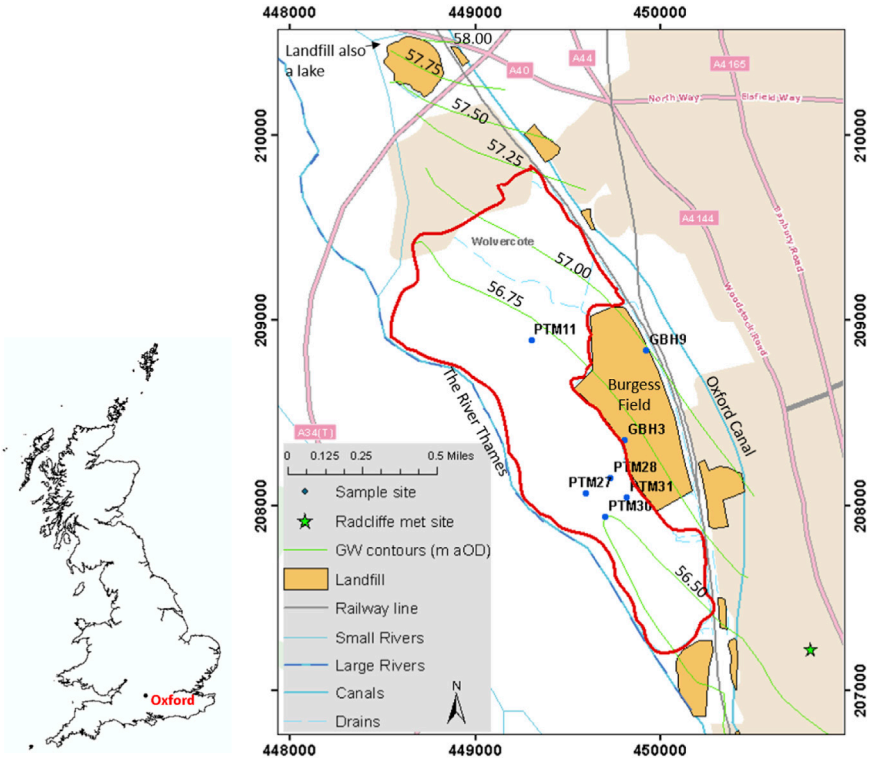


FIGURE 1
Study area including Port Meadow, Burgess Field and adjacent locations and sources of contamination. The area surrounded in red is Port Meadow. Groundwater level contours are shown for the area to the east of the River Thames. Contains Ordnance Survey data © Crown copyright and database rights 2021.

turn are underlain by a mudstone bedrock (Gooddy et al., 2014; Macdonald et al., 2012a). The alluvium is permeable enough to allow groundwater recharge to occur to the floodplain (Gooddy et al., 2014).

An established conceptual model developed through earlier research at this site (Stuart et al., 2014; Gooddy et al., 2014; Lapworth et al., 2013; Macdonald et al., 2012b) is used as the basis to test the utilty of a range of polar MOs to evaluate and descriminate between a range of different sources of contamination within this setting; the Burgess Field landfill, upgradient sources from Oxford such as the canal or tain track (using evidence from Stuart et al., 2014 and earlier work), and the River Thames.

2.2 Antecedent hydrological conditions

Daily precipitation and temperature data from the Radcliffe Meteorological Station (Burt and Burt, 2019) for the year preceding both sampling rounds were analysed. There were comparable temperatures and precipitation for the year preceding each sampling round, with average daily temperatures of 11.5 °C and 11.6 °C and total rainfall of 560.3 mm and 552.2 mm for the period 14/09/2016 to 13/09/2017 and 12/09/2017 to 11/09/2018, respectively. However, the month and week prior to the commencement of sampling were wetter in 2017 (56.5 mm and 23.1 mm, respectively) than in 2018 (43.3 mm and 0.1 mm, respectively).

The groundwater levels were slightly higher in 2017 than 2018. Groundwater levels within GBH3 and GBH9 in September 2017 and 2018 were both within the landfill material at Burgess Field at the time of sampling.

2.3 Sampling

Twelve sampling sites were selected from the groundwater monitoring network to include: a site up-gradient of the landfill (PMT11), landfill piezometers (GBH3 and GBH9), nested piezometers distributed along the flow-path between Burgess Field and the river (North transect PMT27C and D, PMT28C and D; South transect PMT30C and D, PMT31C and D) (Figure 1). The bottom 0.5 m of the floodplain piezometers are screened in the floodplain gravels. The piezometers at Burgess Field are screened in both the gravel and the overlying landfill layer.

Sites were chosen to capture any potential downgradient plume emanating from the landfill, these comprised two multi-level transects completed in the alluvial deposits as well as a control site up-gradient to the landfill. All accessible landfill monitoring wells were also sampled to characterise the landfill itself. The surface water sample from the River Thames was collected downgradient of the landfill site.

Groundwater samples were pumped using a peristaltic pump after purging piezometers to withdraw the equivalent of >3 standing volumes of borehole water. Prior to sampling, stable field

measurements (pH, dissolved oxygen (DO), conductivity, temperature) were obtained using a flow-through cell.

A surface water sample was taken from the River Thames which regularly floods Port Meadow (Figure 1). River Thames samples were collected by holding the sample bottle under the surface until the bottle was full. Samples were collected during two field campaigns (11th – 14th September 2017 and 10th – 14th September 2018) to assess variability. Flow conditions were similar during both sampling events.

2.3.1 Quality assurance

To check the repeatability of the method, two samples from the same sample bottle for PTM30D (both years) were processed through separate solid phase extraction (SPE) cartridges. The results are reported separately. Earlier evaluation of this method is reported by White et al. (2017), which showed that this method was suitable for use for both groundwaters and surface waters with comparable matrices (i.e. DOC and other dissolved constituents). The landfill site waters are very dilute and have comparable chemical compositions with those within the downgradient wells and other monitoring wells so large matrix effects are not likely. Samples are also spiked with internal reference standards and AQC standards are used to quantify recovery and assess matrix effects and these are evaluated as part of the QA and reporting procedure, see White et al. (2019), Supplementary Material for further details.

Blanks were collected to test different stages of the method; these included field blanks, field sample carry over blanks and laboratory blanks. Further information and results for the duplicates and blanks are in the Supplementary Material.

2.3.2 Preparation and analysis

Pre-conditioned resin cartridges in sealed centrifuge tubes were provided by National Laboratory Services (NLS) and returned to NLS for analysis after pre-concentration. The cartridges were kept refrigerated before and after use and were sent back to the laboratory packaged with ice blocks. Pre-concentration of the samples onto the resin cartridges took place within 5 days of sampling at the BGS laboratories in Wallingford. Samples were refrigerated prior to SPE, apart from this there was no sample pre-treatment prior to SPE The method of pre-concentration onto pre-conditioned SPE cartridges is detailed in previous studies (Richards et al., 2021; White et al., 2019; White et al., 2017), please consult these for details on the SPE method used.

2.3.3 LCMS-MS Q-TOF screen

Broad screening for MOs was carried out using pre-concentration by SPE followed by target based Ultra- High-Definition (UHD) Accurate-Mass Quadrupole Time-of-Flight (Q-TOF) liquid chromatography/mass spectrometry (LC/MS) using a semi-quantitative method (White et al., 2019; White et al., 2017). A blank and a single point calibration standard (typically 1 µg/L) is used to establish a response factor and quantify target substances. The calibration is known to be linear from zero to several orders of magnitude higher than the concentration of the samples. A limit of detection (LoD) was defined for over 580 compounds including a large number of pharmaceuticals (n = 193) lifestyle products (5), pesticides (366), surfactants and flame retardants (21). The LoD of compounds ranged from 0.001 µg/L to 0.1 µg/L.

Results were blank corrected by removing compounds detected in blanks. In 2017, cycluron, dichlorvos, saccharin and triclosan were detected above the LoD in multiple samples as well as the blanks; carbaryl was detected in the control site (PTM11 Figure 1) and the blanks; and Trinexapac was only detected in the lab blank (Supplementary Table S1). No compounds were detected in the blanks above the LoD in 2018. Compounds with no LoD were removed. Statistical analysis was carried out on duplicate data to look at repeatability of results (see Supplementary Material 'Method validity checks'). For further details on the analytical method and QA/QC used by the NLS see Richards et al. (2021), White et al (2017) and White et al. (2019).

3 Results

3.1 MO detections in floodplain settings

The full results are tabulated in Supplementary Table S5–S8 in Supplementary Material. The compounds detected by the LCMS have been split into four groups (pesticides, lifestyle, pharmaceuticals, surfactants and flame retardants). Figure 2 is a graphical summary of total concentrations (Figures 2a,b) and total number of compounds within the compound groups during both sampling events (Figures 2c,d).

3.1.1 Pesticides

A total of 19 pesticides and 2 degradation products were detected above LoD, the number of detections varies between the different years (Figure 2; Supplementary Table S5). The maximum number of pesticides detected was in the River Thames, (n = 14 in 2017 and n = 10 in 2018), although their total concentrations were low (0.097 µg/L in 2017 and 0.094 µg/L in 2018). Eleven pesticides were unique to the River Thames. By comparison, 1–5 compounds were detected in the groundwater samples. Bentazone was the only pesticide detected at the control site (PTM11), the only other detection was in the River Thames. Five pesticides were unique to Burgess Field and the transects. Mecoprop was common to all sites except PTM11 and had the highest pesticide concentration (13 µg/L at PTM28D in 2018). A wider range of compounds were detected in 2017 (n = 18) than in 2018 (n = 10) across all sites except PTM11.

3.1.2 Lifestyle compounds

The 3 lifestyle compounds make up a relatively high proportion of the total concentration of contaminants in every sample (Figure 2). Cotinine, an alkaloid found in tobacco and the predominant metabolite of nicotine, was detected above the LoD in the River Thames during both sampling rounds but in no other sample (Supplementary Table S7). The artificial sweeteners sucralose and acesulfame-K were both detected above the LoD; during both sampling rounds sucralose was only detected in GBH9,

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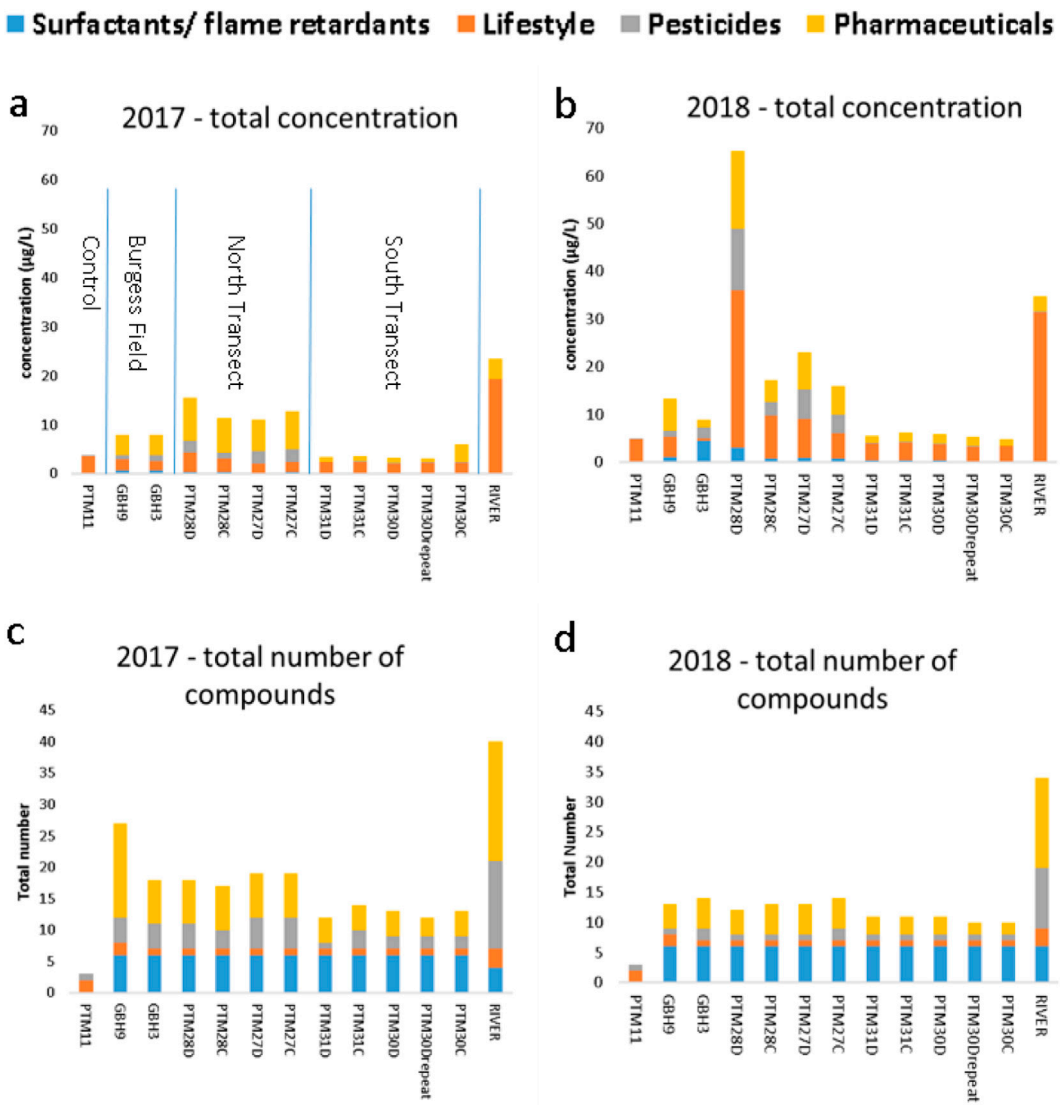


FIGURE 2
Total concentration (µg/L) of compounds within each group found during the **(a)** 2017 and **(b)** 2018 sampling rounds. Total number of compounds within each group found during the **(c)** 2017 and **(d)** 2018 sampling rounds. Note: all data below LoD removed and all compounds found in the blanks were removed during data QA.

PTM11 and the river and not the transects (Supplementary Table S7). Acesulfame-K was above the LoD in every sample (Supplementary Table S7).

3.1.3 Pharmaceuticals

During the study 37 pharmaceuticals were detected above the LoD (Figure 2; Supplementary Table S6). In total, 25 pharmaceuticals were detected in the River Thames during both sampling rounds (n = 20 in 2017; n = 15 in 2018 Supplementary Table S6); 20 (n = 16 in 2017 n = 13 in 2018) were exclusive to the river. A total of 17 pharmaceuticals were detected in Burgess Field (n = 17 in 2017 and n = 5 in 2018); 6 pharmaceuticals were solely detected in Burgess Field (Figure 2; Supplementary Table S6). No pharmaceuticals were unique to the transect (Supplementary Table S6). No pharmaceuticals were detected above the LoD at the control site. Results for pentobarbital (Figure 3) had the highest concentration (up to 15 µg/L) of all pharmaceuticals.

3.1.4 Surfactants, PFAS and bisphenols

Seven compounds (mostly PFAS compounds along with 1,4,5,6,7,7-hexachloro-5-norbornene-2,3-dicarboxylic acid and bisophenol S) were detected in every sample except the control site in both the 2017 and 2018 sampling event (Supplementary Table S8). No compounds in this group were solely detected in the river or the transect, however bisophenol S was only detected in GBH9 in 2018. The compound with the highest concentration in this group is 1,4,5,6,7,7-hexachloro-5-norbornene-2,3-dicarboxylic acid (Supplementary Table S8). The sum of PFAS concentrations by site and year is shown in Figure 4. PFOS and PFOA accounted for

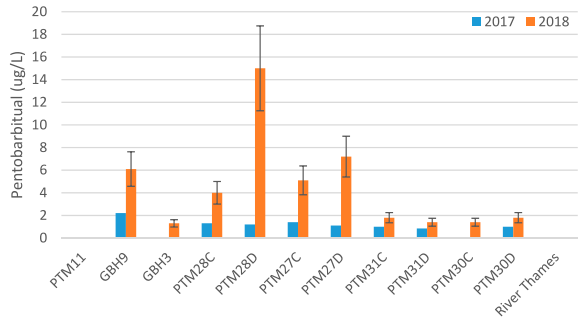


FIGURE 3
Pentobarbital detections (µg/L) for both sampling rounds. Error bars reflect average % error based on repeat sampling at PTM30. Control: PTM11, Burgess Field Landfill: GBH9, GBH3, North transect: PTM27 and PTM28, South transect: PTM30 and PTM31.

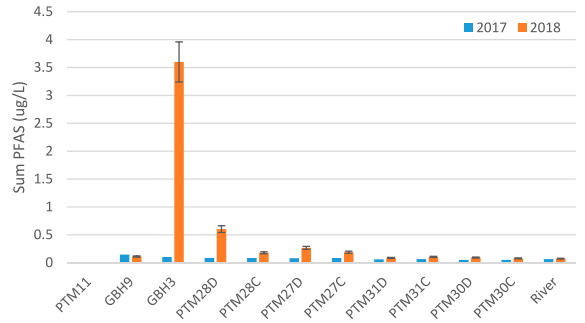


FIGURE 4
Sum PFAS (µg/L) for both sampling rounds. Sum PFAS is sum of Perfluoro Heptanoic acid (PFHpA), Perfluoro Hexanoic acid (PFHxA), Perfluoro Octanoic acid (PFOA), Perfluoro Pentanoic acid (PFPeA) and Perfluorooctane sulfonate (PFOS). Error bars reflect average % error based on repeat sampling at PTM30. Control: PTM11, Burgess Field Landfill: GBH9, GBH3, North transect: PTM27 and PTM28, South transect: PTM30 and PTM31.

the highest proportion of PFAS detections in landfill and groundwater samples (See Supplementary Material).

4 Discussion

The concentrations of contaminants in samples across the floodplain in 2017 are lower compared to 2018, this is likely due to dilution as a result of increased recent recharge and higher groundwater levels in 2017. Figure 5 shows the proportional (in terms of total concentrations) for each contaminant group.

4.1 Occurrence of MOs within the floodplain system

4.1.1 Pesticides

Out of the 14 pesticide compounds detected above their LoD within the River Thames, 6 are still approved for use, 2 are pesticide degradation products and the rest are legacy compounds (Figure 2; Supplementary Table S5), the sources are likely to include agricultural runoff, WWTW effluent and groundwater baseflow inputs to the Thames (White et al., 2017). The following pesticides; mecoprop (0.1–13 µg/L), dichlorprop (0.008–0.2 µg/L), Schradan (0.03–0.5 µg/L) and monuron (0.003–0.01 µg/L) were detected within the landfill and down gradient wells but were not detected in the upgradient well. Mecoprop was the most frequently detected and had the highest concentrations and was found in both transects but with much higher concentrations in the north transect, detections in the River Thames (0.005–0.007 µg/L) were 2–3 orders of magnitude lower than in the downgradient wells.

In both sampling rounds, bentazone (LoD 0.001 µg/L) was the only pesticide detected above the LoD at the control site (PTM11, Figure 3), the only other site in which it was detected was the river. Bentazone, an agricultural pesticide, is unlikely to have been used on the meadow, the railway tracks or in the allotments. Bentazone is a highly soluble and leachable compound (Romero et al., 1996) minimally sorbed by soil particles (Carrizosa et al., 2000). Over time sorption increases in clay soils but desorption is still possible (Boivin et al., 2004). The possible original sources for bentazone at PTM11 are overbank flooding from the River Thames, or from up gradient infiltrating canal water (Figure 5).

Mecoprop was detected above the LoD in every sample except the control site and was the highest concentration pesticide (or metabolite) in the study with overall concentration higher in 2018 than 2017 (Supplementary Table S5). Mecoprop is resistant to hydrolysis, reduction, oxidation, and chemical reactions with organic or inorganic compounds are very slow (Zipper et al., 1998). Biodegradation in anaerobic conditions, like those found in a landfill and contaminant plume, is negligible (Lyngkilde and Christensen, 1992) but aerobic biodegradation has been shown (Heron and Christensen, 1992; Müller and Buser, 1997; Zipper et al., 1996). Zipper et al. (1998) assessed concentrations of mecoprop in landfill leachate and within a downgradient plume in groundwater and found high and variable concentrations within downgradient monitoring sites (0.001–975 µg/L). It was also found that sorption of mecoprop to aquifer material was minimal with the highest proportion found in the dissolved phase (Zipper et al., 1998) and as such sorption does not retard the movement of mecoprop. The groundwater concentrations for mecoprop found in this study were significantly higher than Loos et al. (2010) a regional European groundwater study, while Buss et al. (2006) report that mecoprop was the most frequently detected pesticide above 0.1 µg/L in England between 1996–2003.

The low concentration of mecoprop in the river, the lack of detection at PTM11 and the higher concentrations at Burgess Field and north transect (PTM28 and 27 D Figure 2) suggest Burgess Field as the source. The significantly lower concentrations in 2017 may be a dilution effect due to higher groundwater levels. Previous studies have also found that mecoprop concentrations are greater in groundwater down-gradient than within the landfill due to periodic release of mecoprop from leaking containers in the landfill (Zipper et al., 1998).

Diuron, a currently used pesticide, was detected above the LoD (0.001 µg/L) in the River Thames in both rounds and in 2017 at GBH9 (Figure 5). The other pesticides found predominantly at Burgess Field and the transects are legacy compounds; the likely

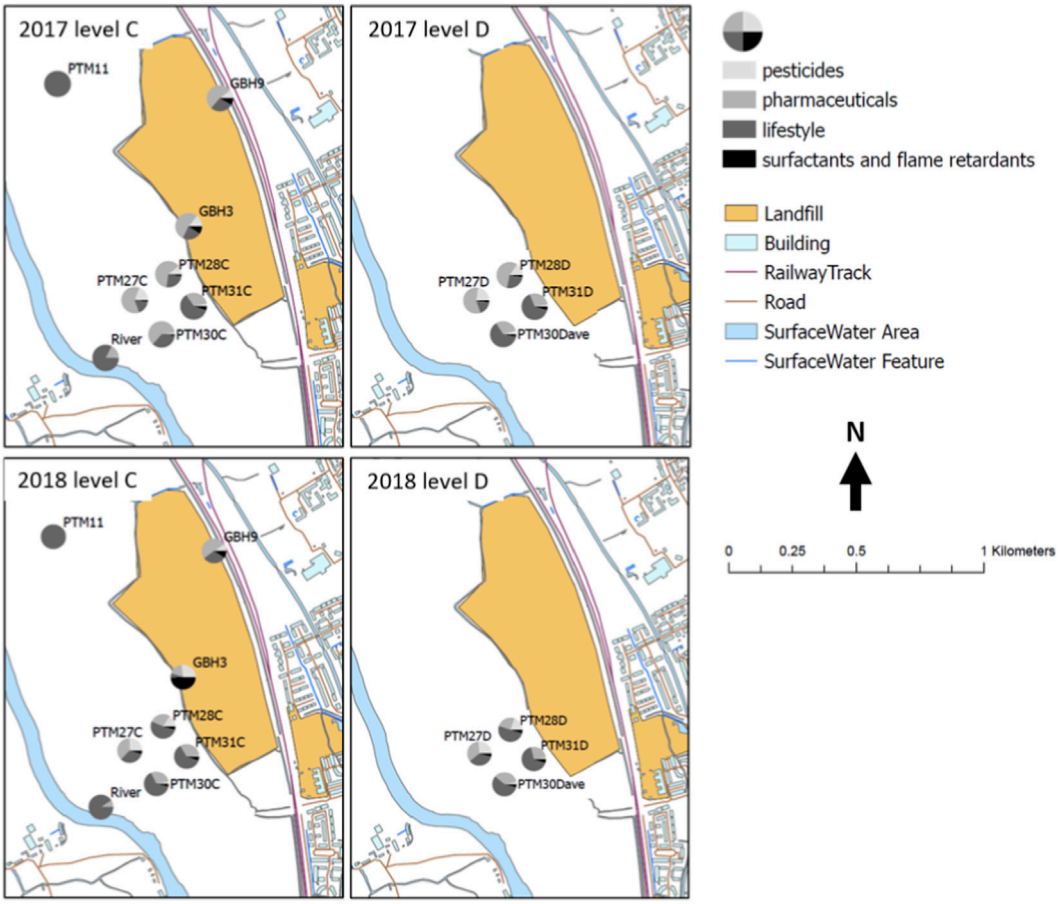


FIGURE 5
Proportion of the total concentration of each contaminant group for 2017 and 2018 for both piezometer depths. Piezometer completion depths: GBH3 5.1 m bgl; GBH9 5.2 m bgl; PTM 11 3.4 m bgl; all C piezometers 1.8 m bgl; PTM27D and PTM30D 2.9 m bgl; PTM28D 3.6 m bgl; PTM31D 3.9 m bgl. Contains Ordnance Survey data © Crown copyright and database rights 2023.

source being Burgess Field and not overbank flooding as they were not detected above the LoD in the River Thames.

4.1.2 Artificial sweeteners

Sucralose concentrations detected within the River Thames during both sampling rounds were an order of magnitude above those detected in GBH9 and two orders of magnitude above those detected in the control site (PTM11); it was not detected in the transect (Supplementary Table S7). The source of sucralose within the River Thames is likely to be WWTWs (Loos et al., 2009; Oppenheimer et al., 2011; Scheurer et al., 2009; Yang et al., 2017; White et al., 2019). Sucralose has been on the European market since 2004 (Loos et al., 2009) and has been in industrial use since the 1990s (Neset et al., 2010); this post-dates the closure of Burgess Field. Possible sources of sucralose at GBH9 and PTM11 are the Oxford canal or the rail track that runs behind Burgess Field. The River Cherwell receives WWTW effluent up stream of its confluence with the canal. Stuart and Lapworth (2011) showed water loss from the canal into the adjacent sand and gravel aquifer. Another possible source is human waste discharged onto the train tracks behind Burgess Field infiltrating into the aquifer.

Concentrations of acesulfame-K are higher within the control site (3.3–4.3 μg/L) than they are in the River Thames (1.2-.13 μg/L) or Burgess Field (0.4–2.3 μg/L). The highest concentration in both years in the tansect is at PTM28D (33 μg/L). It does not biologically degrade, is highly soluble and has been used as a landfill leachate tracer (Roy et al., 2014). It is likely that an acesulfame-K plume has been migrating down-gradient from Burgess Field and is now within the floodplain aquifer and that the source at PTM11 is the Oxford Canal or the train tracks (Figure 5). Roy et al. (2014) reported a range of artificial sweeteners in comparable concentrations to this study detected in landfills and landfill impacted groundwaters across Canada. Belton et al. (2020) undertook a global review of acesulfame-K in waters and found that groundwater detections ranged greatly (<LOD-9.7 μg/L), with overall higher concentrations found in this study likely due to the presence of sweeteners in the landfill plume at this site.

4.1.3 Pharmaceuticals

4.1.3.1 Antimicrobials

Six antimicrobials were detected above the LoD in the study. Sulfamethoxazole only occurred in the River (Supplementary Table

S6) and the remaining five antimicrobials were found in groundwater from Burgess Field and the transects; sulfadiazine and sulfamerazine were only detected in Burgess Field in 2017.

Sulfathiazole was detected above the LoD in GBH9, GBH3 and the north transect in both years, and at PTM28D in 2017 with the highest concentrations at GBH 9 (both years). This suggests a plume of sulfathiazole originating at Burgess Field that only intersects the north transect. Overall, the highest concentrations of sulfanilamide and sulfamethazine occur in 2018 (Supplementary Table S6) with the peak of the concentrations in the north transect in that year; both were detected in GBH3 and 9. Burgess Field is an obvious source of these 5 antimicrobials as it received medical waste. However, in terms of ecological risks from sulfamerazine, which was detected at significantly higher concentrations compared to sulfamethoxazole and sulfadiazine, the concentrations detected in this study in groundwater and surface waters (0.02–1 μg/L) were several orders of magnitude below the lowest experimental freshwater PNEC concentration of 30 μg/L (NORMAN, 2026).

4.1.3.2 Non-steroidal anti-inflammatories

Diclofenac and codeine were both detected above the LoD in the river but not the groundwater and ibuprofen was only detected in GBH9 in 2017. The highest concentrations of lidocaine occurred at GBH 9 with the second highest in the River Thames in 2017; it was below detection in the River Thames in 2018. Lidocaine was detected in the north transect in 2017 and 2018 and the shallower piezometers of the south transect in 2017. There appears to be a source of lidocaine at Burgess Field and the plume travels through the flood plain (Supplementary Table S6). Lidocaine has been frequently detected in other landfill leachates (Masoner et al., 2014) and the River Thames (White et al., 2019).

4.1.3.3 Anti-convulsants

Carbamazepine was detected above the LoD at GBH9 in 2017 and at concentrations orders of magnitude higher in the River Thames during both years; it is absent from all other samples. Carbamazepine has been found in a high proportion of landfill leachate in modern (Masoner et al., 2014) and historic landfills (Propp et al., 2021), but WWTW effluent is another source. Gabapentin was only detected in the River Thames in 2017, and a degradation product of carbamazepine and oxcarbazepine (10,11-dihydroxycarbamazepine) was detected in the River Thames in 2018.

The corticosteroid methylprednisolone was detected in 2017 at GBH9, GBH3, the northern transect and PTM30C but is not detected at all in 2018. Similarly, phenobarbital is detected within GBH9, GBH3, the transects and the river in 2017 but not at all in 2018. Concentrations are higher within the north transect. This shows the rapid change in concentrations of certain MOs within the downgradient plume throughout the transects. This indicates the high spatial variability in sources within the landfill, narrow plume dimensions and the complex interplay between this and the groundwater levels which may dilute sources as they rise within the base of the landfill. However, this should not be over-interpreted as this study only shows results from two rounds of sampling and more detailed seasonal sampling is required to understand temporal variations in more detail.

4.1.3.4 Barbituates

The highest concentration pharmaceutical quantified during the study was the barbiturate pentobarbital which was detected above the LoD within Burgess Field and the transects (Supplementary Table S6; Figure 3). In contrast to other contaminants in this study concentrations were overall higher in 2018, notably so in the north transect (4 μg/L to 15 μg/L in 2018 compared to 1.1 μg/L to 1.5 μg/L in 2017) (Figure 3). In both years the concentrations within the transects were higher than those at GBH3. In 2017 the concentration at GBH9 was above that in any of the transects, however in 2018 concentrations at the deeper piezometers of the north transect are higher than GBH9. The source(s) of pentobarbital are within Burgess Field and concentrations are still high even approximately 40 years post closure. Pentobarbital was also detected at high concentrations (maximum 39.8 μg/L, median 5.9 μg/L) in leachates from modern landfills in the United States (Masoner et al., 2014) highlighting the high concentrations in this studies groundwater compared to modern landfill leachate. The concentrations of Phenobarbital detected in groundwaters within the downgradient landfill plume in this study were comparable or exceeded the lowest experimental and modelled freshwater PNEC of 4.6 μg/L (NORMAN, 2026), while there was no detection of Phenobarbital in the River Thames itself.4.1.4 Surfactants and flame retardants (PFAS).

During 2017 and 2018 the highest total concentration of surfactants and flame retardants compounds was detected in GBH 3; the concentration within the River Thames was an order of magnitude lower (Supplementary Table S8). The highest total concentrations within the transects in both years are seen in the northern transect, with PTM28D always being higher; none were detected at PTM11 (Supplementary Table S8). The high concentration at PTM28D and the large variation of concentrations within Burgess Field and the transect can be accounted for by multiple sources within the landfill as well as dilution and dispersal within the aquifer system.

Perfluorooctane sulfonate (PFOS), perfluoro heptanoic acid (PFHpA), perfluoro hexanoic acid (PFHxA), perfluoro octanoic acid (PFOA) and perfluoro pentanoic acid (PFPeA) were detected in all surface and groundwater samples except the control in both years. The sum of PFAS (Figure 4) shows an order of magnitude higher concentrations at GB3 in 2018 compared to 2017 and compared to GBH9 and the other groundwater sites (3.5 μg/L compared to 0.1–0.5 μg/L). PFOS and PFOA accounted for the highest proportion of PFAS in the majority of monitoring sites. It was noticeable that the ratio of PFOS/PFOA was more consistent (1.2–2.4 in GBH3 and the transect boreholes while for GBH9 and the river the ratio was much more variable (4.7 in 2027 and 0.4 in 2018 and tracked with the river samples (3.8 and 1.2 respectively). Together these results suggests that while the landfill is the source of PFAS in downgradient boreholes, there is significant variability temporally within the landfill and the contamination plume is narrow and limited to the north transect, PTM28 and PTM27, with highest concentrations in the deeper piezometers, consistent with other contaminants emanating from the landfill.

The lower groundwater levels in 2018 compared to 2017 and higher concentrations could indicate that rising groundwater levels leads to dilution of the landfill source emanating from the base of the

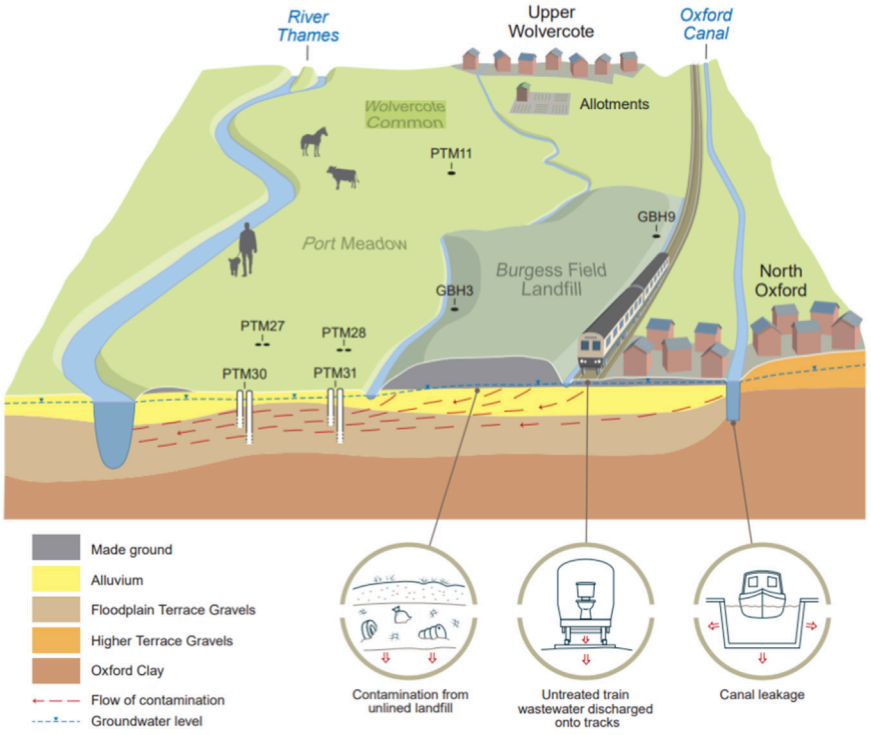


FIGURE 6
Conceptual model of the migration of different sources of organic contaminants within the alluvial groundwater system at Port Meadow, Oxford.

landfill. No PFAS were detected in PTM11, the control site, this supports the inference that the landfill is the major source of PFAS in this study area. PFAS are frequently detected in landfills (Hepburn et al., 2019; Masoner et al., 2020; Propp et al., 2021). However, there has been limited monitoring in legacy 'dilute and disperse' landfills such as Burgess field which have been shown to be an important source of PFAS in this study.

Groundwater and surface water concentrations of PFOS, PFOA and PFHpA in this study exceeded the lowest experimental freshwater PNEC values (NORMAN, 2026) by an order of magnitude on at least one occasion. This suggests that landfills and adjacent groundwaters may be a significant source of selected PFAS and may contribute to ecological impacts of dependant freshwater bodies such as rivers and wetlands.

4.2 Tracing sources and pathways within the complex alluvial aquifer system

Using a range of MO contaminants as tracers has allowed us to better characterize the different sources and pathways within this complex floodplain setting where multiple sources and pathways for contaminants exist. Figure 6 presents a general conceptual model of the different sources of organic contaminants and their movement within the alluvial aquifer system beneath Port Meadow. The insights gained from this tracer approach for understanding sources and pathways for each domain sampled within the floodplain are discussed in detail below.

4.2.1 The River Thames

In this context, the River Thames is a potential source of groundwater contaminants due to infiltration of overbank flooding due to its known significant contaminant load (White et al., 2019). A total of 60 compounds were detected above the LoD in the whole study, and of these 32 were unique to the River Thames. Pharmaceuticals made up the highest proportion of detects in the River Thames (Figure 2). However, artificial sweeteners (lifestyle compounds) contributed the highest concentration (Figures 2, 5). Artificial sweeteners would have been a good tracer for the influence of the River Thames due to overbank flooding had they not also been found in every other sample; some at higher concentrations. No one compound was found to be a unique tracer for the River Thames.

The River Thames contains a multi-compound contaminant load (Lu et al., 2017; Neal et al., 2000; White et al., 2019; Yamaguchi et al., 2003); it is impossible to attribute contamination of the river from Burgess Field especially as many compounds are already found within the River Thames. However, there is likely to be limited hydraulic connectivity between the gravels and the Thames at this location (Gooddy et al., 2014) (Figure 6). The MOs found at the control site (PTM11) are unlikely to come from flood water originating from the River Thames due to the lack of pharmaceuticals.

4.2.2 Burgess Field landfill site

This study confirms that Burgess Field is a key source of contamination for the alluvial gravel aquifer and has a significant influence on down-gradient groundwater chemistry with attenuation of leachate-derived elements along the groundwater flow path (Figure 3; Gooddy et al., 2014; Macdonald et al., 2012b; Stuart et al., 2014). However, this study also shows that groundwater at Burgess Field is a receptor for contaminants from upgradient sources, which has not been shown by previous contaminant studies at the site (e.g. Stuart et al., 2014).

The landfill material is heterogeneous and the spatial spread of sampling points will never give full confidence that all the contaminants present in the landfill are detected. However, in the case of Burgess Field, sucralose was detected in the landfill samples and the manufacture of sucralose post-dates the closure of the landfill; clear evidence that groundwater flowing through/beneath Burgess Field from the east is adding to the contaminant load. This underlines the complexity of different sources and pathways within this, and other similar floodplain settings where unlined landfills are located, and the need for a clear hydrogeological conceptual model of groundwater flow and groundwater-surface water connectivity (Gooddy et al., 2014) (Figure 5).

Due to the hetergeneous nature of the Burgess Field landfill,the two piezometers sampled have been summarized separately. At GBH3, a total of 19 compounds were detected above the LoD, and of these, sulfadiazine was only found in GBH3 in 2017 (Supplementary Table S5–S8). Similarly, at GBH9 a total of 28 compounds were detected above the LoD, and of these 4, compounds were unique to GBH9 (Supplementary Table S5–S8). The higher total number of contaminants and higher concentration of lidocaine and sulfathiazole at GBH9 suggests an up-gradient source, either within the landfill or upgradient of the landfill, e.g. the Oxford canal or the rail tracks (Figure 5). This is supported by Stuart et al. (2014) which reported results from a site close to the Oxford canal (OX14) which contained a much wider range of compounds (e.g. gabapentin, carbamazepine, t-octyl phenol, DEET) similar to that of the Thames River composition which is known to receive wastewater inputs, and this was considered to imply that the canal was likely leaking and contributing to groundwater contamination in this region upgradient of the landfill.

It appears that the landfill was the original source for pentobarbital, sulfamethazine, mecoprop, and PFAS but the plume has since migrated into the floodplain aquifer, with highest concentrations found at PTM28D, the lower floodplain gravels (Figure 2). While mecoprop is preferentially degraded under aerobic conditions (Heron and Christensen, 1992), PFAS, pentobarbital and sulfamethazine are more recalcitrant even to aerobic biodegradation (Peschka et al., 2006; Pan and Chu 2016), which explains why these and other compounds persist in the sub-oxic conditions of the landfill and landfill plume (Gooddy et al., 2014; White and Macdonald, 2015). Burgess Field, specifically the area close to GBH 3, is the obvious source of the five perfluorinated compounds in 2018 but not in 2017. However, there may well be other PFAS compounds that occur within the landfill plume as the list of PFAS compounds in this screen were by no means extensive. The variation between the 2 years may be due to a number of factors, the over low concentrations, dilution when groundwater levels rise seasonally as well variable inputs from different parts of the landfill site. However more temporal sampling is required to explore this further and confirm initial findings.

4.2.3 Control site up-gradient of the landfill

In both years, only bentazone, sucralose and acesulfame K were detected at PTM11. Previous studies have shown that PTM11 is unaffected by the leachate plume from Burgess Field (Gooddy et al., 2014; Stuart et al., 2014). The source for these contaminants could be: river water, either directly through overbank flooding or from alluvial sediments left on the flood meadow; infiltration of canal water; or human waste discharged to railways tracks up gradient of the site. The River Thames is discounted as pharmaceutical contamination would be expected when comparing sucralose concentration at PTM11 to the concentrations in the river (Figure 3).

4.2.4 Floodplain aquifers impacted by the landfill

The north transect has a longer groundwater flow line through Burgess Field than the south transect. The difference between them in terms of MO contaminants is noticeable (Figures 2, 3). PTM28D is the first deep site down-gradient of Burgess Field and is the site with the greatest overall concentrations in both years (Figure 3) (Stuart et al., 2014). Previous work on the north transects has shown that the deeper piezometers are affected to a greater extent by the landfill leachate plume (Lapworth et al., 2013). A study of NH_4 concentrations concluded that those in the shallow gravels (depth C) were diluted relative to the deeper gravels (depth D), increasingly so down flow-gradient towards the River Thames (Gooddy et al., 2014). This was explained as a combination of mixing and dilution between groundwater leachate from Burgess Field (high NH_4) and local shallow recharge to the floodplain gravels. Concentrations of mecoprop, pentobarbital and acesulfame-K are elevated within the deeper gravels (depth D) and corroborates the findings of Gooddy et al. (2014). Using the residence time indicator SF_6 (Macdonald et al., 2012b), showed that the groundwater within the gravels is older at depth than in the shallower part of the aquifer. Consistent with previous observations (Gooddy et al., 2014; Macdonald, 2019), the number and concentration of contaminants is lower in the south compared to the north transect (Figure 2) and is likely due to shorter groundwater flowlines and residence time within Burgess field (Figures 3, 5).

5 Conclusions and future research perspectives

A wide range of polar organic compounds were used as tracers within a peri-urban floodplain setting to elucidate different sources of anthropogenic contamination found within the alluvial groundwater system.

The major source of contamination to the floodplain groundwater was confirmed as the unlined Burgess Field landfill, closed 40 years prior to the study. Groundwater in the north transect had a higher concentration and number of contaminants due to a longer pathway through Burgess Field. Attenuation was seen down gradient of the landfill, except at PTM28D where high concentrations of contaminants are attributed to possibly greater

permeability and hence enhanced solute migration in the deeper gravels.

There was spatial variability of contaminants within Burgess Field and at least one contaminant, sucralose, was shown to originate from an up-gradient source due to manufacture/use postdating landfill closure. Burgess Field is still an active source of contamination to the adjacent aquifer system 40 years after its closure and will continue to be for a considerable time based on our observations of polar MOs within the transect boreholes in this study, and earlier studies which employed other tracers (e.g. Gooddy et al., 2014; Stuart et al., 2014; Lapworth et al., 2013).

Contaminants found up-gradient of the landfill did not have the MO signature of either the River Thames or Burgess Field. It is therefore likely that these are sourced from either infiltration to groundwater from the adjacent canal or from wastewater discharged to the railway tracks upgradient of the floodplain. This highlights how useful a multi-tracer technique can be in settings with multiple overlapping contaminant sources.

The River Thames contained the highest number of contaminants in the study and is a potential source of groundwater contamination via infiltration of floodwater. However, it is likely to be a minor source of contamination within the floodplain aquifer compared to the landfill. Given the limited exchange between the groundwater and the River Thames at this location, the contaminant plumes within the alluvial aquifer are also not likely to be a significant source of contamination in the River Thames.

This study shows the value of using MOs as a means to better understand complex source-pathway-receptor systems when combined with a conceptual model of groundwater-surface water flow. It also highlights the risks to groundwater from the disposing of solid waste on floodplains. There has been limited research to date on the use of MOs as tracers within peri-urban floodplain settings. However, these tracers are highly effective in discriminating different sources of contamination and as hydrological tracers in their own right, particularly when used in combination with other tracers such as isotopes of water or groundwater age dating tracers. The screening approaches used in this study demonstrates the utility of these methods for characterising and tracking pollution within complex settings with multiple sources of contamination. The high concentrations detected within the landfill and downgradient monitoring wells within the alluvial aquifer demonstrate the role of legacy landfills as sources of a range of regulated and emerging contaminants such as PFAS and potential risks for a range of downstream receptors (e.g. aquifers, rivers, wetlands, springs and water abstraction users).

Data availability statement

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author.

Author contributions

DW: Formal Analysis, Investigation, Visualization, Writing – original draft, Writing – review and editing. DM: Conceptualization, Project administration, Visualization, Writing – review and editing. WC: Data curation, Formal Analysis, Methodology, Writing – review and editing. MM: Investigation, Resources, Writing – review and editing. BT: Investigation, Writing – review and editing. DL: Conceptualization, Funding acquisition, Investigation, Project administration, Resources, Supervision, Visualization, Writing – original draft, Writing – review and editing.

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Conflict of interest

The author(s) declared that this work was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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Supplementary material

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fbuil.2026.1985554/full#supplementary-material

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