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Potential contamination of the coastal zone by eroding historic landfills

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ABSTRACT

Historically solid waste was commonly landfilled in the coastal zone in sites with limited engineering to isolate waste from adjacent coastal environments. Climate change is increasing the likelihood that these historic coastal landfills will erode releasing solid waste to the coastal zone. Historic coastal landfills are frequently located near designated ecological sites; yet, there is little understanding of the environmental risk posed by released waste. This research investigated inorganic and organic contaminant concentrations in a range of solid waste materials excavated from two historic coastal landfills, and the potential ecological impact should eroded waste be released to the coastal environment. Contaminant concentrations in the analysed waste materials exceeded sediment quality guidelines, indicating erosion of historic coastal landfills may pose a significant environmental threat. Paper and textile wastes were found to make a significant contribution to the total contaminant load, suggesting risk assessments should consider a wide range of solid waste materials.

1. Introduction

Historically estuaries and the coastal zone have frequently been used for solid waste disposal in landfills due to low economic land values and proximity to industrial and population centres. Waste has also been used for coastal land reclamation and/or to prevent flooding, e.g. land raise and waste filled coastal defences (Cooper et al., 2013). For example, in England there are > 1200 historic coastal landfills, i.e. historic landfills that have a 0.5% annual probability of coastal flooding if not adequately defended (Brand et al., 2017). Many of these historic coastal landfills are currently protected by coastal defences to ensure that the solid waste is confined and isolated from the coastal and marine environment. However, climate change and coastal management strategies now present a number of scenarios by which solid waste buried for decades could be exposed.

Climate change predictions for the coastal zone include increases in sea level and in the magnitude and frequency of coastal storm surge events; it is generally believed the risk of coastal flooding could increase by a factor of ten by the 2080s and with it coastal erosion (IPCC, 2013; Thorne, 2014). Therefore, it is increasingly likely that coastal landfills will be inundated by saline waters, or that erosion or catastrophic failure of the landfill sites and/or their coastal defences will occur resulting in the release of solid waste to the coastal zone. Indeed, erosion and release of solid waste materials from historic landfills to the coastal zone has already been documented around the world (Pope et al., 2011; Alaska Department of Environmental Conservation, 2015; Brand et al., 2017) and 1 in 10 of England's historic coastal landfills are expected to start eroding by 2055 if no interventions are made (Brand et al., 2017). In addition, there is increasing economic and legislative pressure on those managing the coastal zone to seek alternatives to 'hold the line' strategies and remove hard engineering, such as de-embankment (managed realignment). This could have significant consequences when the defence structures are providing a physical barrier between waste and coastal environments.

The majority of England's historic coastal landfills pre-date modern environmental regulations and are no longer managed by their original operator, as a result there are limited or non-existent records regarding the volume or nature of the waste and contaminants they contain (Pope et al., 2011; Brand et al., 2017). Therefore, to inform landfill management decisions, and to help understand the impacts of climate change on the coastal environment, there is a need to investigate the chemical characteristics of degraded waste and the potential consequences should such waste be released to the coastal environment. This is a significant challenge given the large numbers of historic landfills that may be at risk of erosion.

There has been very limited examination of the physical or chemical characteristics of degraded waste previously as the challenges associated with excavating a closed landfill are considerable. For example, financial costs and potential health and safety impacts to workers are high. Most research has focussed on leachate (e.g. Robinson and Maris, 1979; Robinson et al., 1982; LaGrega et al., 1994; Robinson, 1995, 2007; Ziyang et al., 2009) as this is the most likely pathway for

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contaminants to enter the wider environment if the landfill is structurally intact, and leachate sampling is relatively easy via boreholes. A few studies have examined degraded solid waste for the purpose of resource recovery, but did not consider environmental impact of waste release (e.g. Hull et al., 2005; Zhao et al., 2007; Prechthai et al., 2008; Quaghebeur et al., 2013). These typically only analysed waste matrix material (fine and medium grained soil-like particulate materials) and did not consider other waste materials, e.g. paper, textiles, plastics and wood, which can form over 60% of the waste (Parfitt, 2009) and if released may also pose an environmental risk.

The aim of this study was to assess inorganic and organic contaminant concentrations in excavated degraded solid waste (including matrix material, wood, paper and textiles) and to determine the potential impact in the coastal zone if solid waste materials were to erode (either accidently through the effects of flooding and coastal erosion or deliberately through managed realignment). The study focusses on two defended historic coastal landfills that are currently at risk of flooding. One is also at risk of erosion and was considered as a potential candidate for managed realignment in coastal management plans (Environment Agency, 2009).

2. Methodology

2.1. Sample collection

Two landfill sites on the north bank of the Thames Estuary, Essex, UK (Fig. 1) were identified for excavation and waste sampling. These sites were selected because they represent two common scenarios, a landfill forming a flood defence and a landfill protected by a flood defence, and gaining access and permissions/consents for sampling them was relatively straightforward. Hadleigh Marsh is a coastal flood embankment constructed between 1980 and 1987 filled with household and commercial waste capped with puddled clay. It is approximately 4 km long by 65 m wide and is estimated to contain 500,000 m^3 of solid waste (Essex County Council, n.d.). The site was previously identified as a candidate for managed realignment (de-embankment) (Environment Agency, 2009). Leigh Marshes landfill is approximately 0.25 km^2 containing an estimated 800,000 m^3 of waste (Halcrow Group Ltd, 2012) and was constructed between 1955 and 1967. It is currently protected by flood embankments and contains industrial, commercial and household waste (Environment Agency, 2015). Both sites fall within the Environment Agency's Flood Map for Planning flood zone 3 (0.5% annual probability of tidal flooding) and Hadleigh Marsh is on an eroding coastline. No detailed information exists for the waste deposited at either site as they pre-date legislation requiring records to be kept (Secretary of State, 2002). Both Hadleigh Marsh and Leigh Marshes landfills fall within a Ramsar, Special Protection Area, Site of Special Scientific Interest, and a Marine Protected Area, and Leigh Marshes is also within a National Nature Reserve. Both sites are within the Bathing Water Zone of Influence Catchments of eight public beaches on the Thames Estuary (Environment Agency, 2017), and there are bivalve

Fig. 2. Waste extracted from Leigh Marshes trial pit 6 included a sealed poison bottle containing an unidentified liquid (insert).

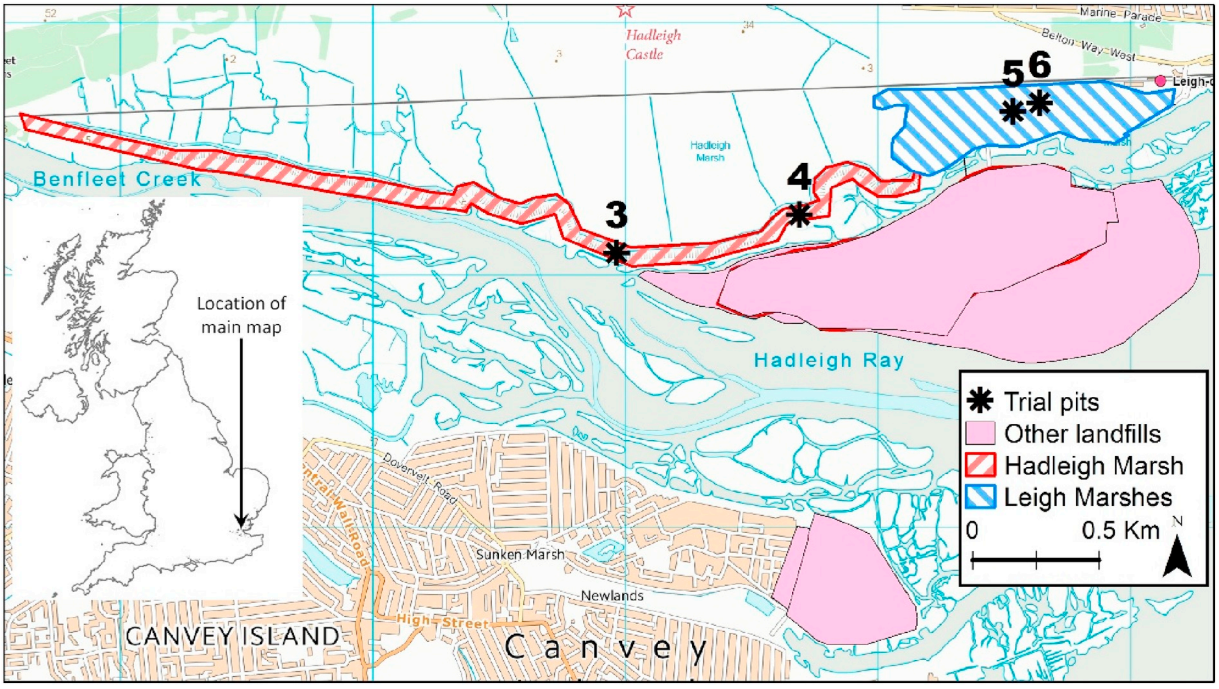

Fig. 1. Hadleigh Marsh flood embankment, Leigh Marshes recreational area, and the trial pit locations. No sampling was undertaking at locations 1 or 2, not shown; they were in the area with the highest ecological designations and were contingencies in case the supervising ecologist vetoed the preferred locations.
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Fig. 3. Waste in Hadleigh Marsh flood embankment, trial pit 3.

mollusc production areas located immediately downstream of the landfills (Cefas maps of bivalve mollusc production areas, O. Morgan, personal communication, email, November 2, 2015).

Four excavator-dug trial pits, two at each study site (Fig. 1), were excavated to a maximum depth of 2.1 m in Leigh Marshes and 1.5 m in Hadleigh Marsh. The trial pit locations were selected to avoid historic ground investigation works, areas protected by the highest ecological designations, areas with vegetation that may provide habitat for protected species, private property, underground services and areas covered with concrete or tarmac. The number of sampling locations was limited by trial pit excavation costs. At each trial pit approximately 360 l of waste was collected in 8 × 45 l plastic containers. The excavator bucket was jet washed between each sampling location to minimise cross-contamination. Four containers for each trial pit were foil lined to preserve samples for organic contaminant analysis. The containers were sealed to maintain field moisture levels, returned to the laboratory, and refrigerated (inorganic contaminant analysis) or frozen (organic contaminant analysis) until required. This sampling method oxygenated the waste during its collection and is representative of waste materials being eroded onto the foreshore.

2.2. Sample preparation and analyses

Due to the hazardous nature of the material, all sample handling took place in a fume cupboard. All plasticware, ceramic ware, and glassware were acid washed, rinsed three times in de-ionised water and dried prior to use. In addition to the matrix material sampled from both sites, paper, textiles and wood were sampled from Hadleigh Marsh as they were present in large quantities in the decomposed waste. For each sample container, a minimum of three subsamples of approximately 10 g of matrix material, sieved to < 2 mm, were air dried overnight and then on a hotplate for 6 h at 105 °C in a fume cupboard until constant mass was achieved. Three subsamples of textiles, paper and wood sieved to < 10 mm (Riber et al., 2007) were dried from each container that contained sufficient material of that type (totalling 15 wood, 18 paper and 9 textile samples). These materials were cut from larger samples where necessary and care was taken to select samples with minimal quantities of attached matrix material.

2.2.1. Inorganic analyses

0.5 (± 0.025) g subsamples of dried waste were heated in 12 ml Aqua Regia (3:1 HCl:HNO_3) for 5 h. The resulting solutions were then filtered and made up to 50 ml volume with deionised water. Each dried subsample was analysed in triplicate.

ICP-OES was used to determine concentrations of Cr, Cu, Pb and Zn in the matrix material, wood and textile extracts and ICP-MS was used to determine concentrations of Cd in the matrix material extracts and Cd, Cr, Cu, Pb and Zn in the paper extracts.

Certified reference materials (BCR143R, LGC6137 and LGC6187) were used to assess data quality; mean recoveries ranged from 85 to 122% with an average relative standard deviation (%RSD) of 11%. Method precision was tested using replicate extractions of the certified reference materials (CRMs) and achieved an average %RSD of 11.8%. 10% blanks were used to check for background contamination, and concentrations of metals reported for the waste samples were adjusted accordingly.

2.2.2. Organic analyses

PAHs were identified for analysis as they are persistent organic pollutants which have previously been identified at the site (Halcrow Group Ltd, 2012). Three matrix material subsamples of approximately 250 ml from each of the organic (i.e. foil lined) sample containers that contained sufficient matrix material were analysed by GC–MS, totalling 21 samples for Hadleigh Marsh and 24 for Leigh Marshes. Wood, paper and textiles are not suitable for commercially available PAH analysis methods and, therefore, were not analysed.

2.3. Calculation of the total metal loads in the landfills

To assess the contribution of metals associated with non-matrix materials to the total contaminant load, total dry masses of the matrix material, paper and textiles in Hadleigh Marsh were estimated (insufficient data were available for wood) by substituting typical values

Table 1
Inorganic contaminant concentrations in matrix material samples (mg/kg) excavated from two historic coastal landfill sites in Essex.

	Leigh Marshes landfill							Hadleigh Marsh landfill						
	Trial pit 5 (n = 36)			Trial pit 6 (n = 35)			Trial pit 5 + 6	Trial pit 3 (n = 35)			Trial pit 4 (n = 36)			Trial pit 3 + 4
	Min	Max	Median	Min	Max	Median	Median	Min	Max	Median	Min	Max	Median	Median
Cd	2.1	4.5	3.0	1.6	11	6.8	4.1	0.3	21	1.0	0.4	28	1.8	1.2
Cr	39	106	62	13	159	84	64	26	81	63	39	371	52	59
Cu	245	1281	571	258	20,140	647	605	< LOD	103	25	24	322	39	33
Pb	625	2625	1083	886	14,936	2031	1332	27	508	100	22	199	56	63
Zn	667	3100	1232	936	5731	1866	1540	99	5128	314	35	2532	144	209

Table 2
Organic contaminant concentrations in matrix material samples (μg/kg) excavated from two historic coastal landfill sites in Essex.

	Leigh Marshes landfill							Hadleigh Marsh landfill						
	Trial pit 5 (n = 12)			Trial pit 6 (n = 12)			Trial pit 5 + 6	Trial pit 3 (n = 12)			Trial pit 4 (n = 8)			Trial pit 3 + 4
	Min	Max	Median	Min	Max	Median	Median	Min	Max	Median	Min	Max	Median	Median
Acenaphthene	26	237	73	112	1020	354	142	8.9	581	113	2.0	31	23	30
Acenaphthylene	44	1100	188	45	208	82	95	16	273	92	2.5	52	15	44
Anthracene	162	1450	580	339	3780	614	606	23	1020	362	10	189	76	186
Benzo(a)anthracene	887	6850	2180	1070	15,700	1740	2010	139	3970	1480	41	675	318	743
Benzo(a)pyrene	696	8530	2190	1280	15,700	1935	2080	200	3930	1640	51	719	303	761
Chrysene	353	6410	2040	1350	24,400	2245	2040	166	4290	1685	42	699	342	771
Dibenzo(ah)anthracene	248	1480	468	367	5560	441	451	37	957	394	9.2	160	74	150
Fluoranthene	1840	8690	4650	2820	22,200	4210	4405	247	9420	2990	75	3550	806	2050
Fluorene	51	372	172	194	1160	484	291	5.0	545	154	5.0	99	31	54
Naphthalene	335	3370	812	861	8230	2665	1840	20	465	114	11	673	73	100
Phenanthrene	913	6960	2720	1290	10,300	2905	2770	84	4300	1100	23	1960	348	622
Pyrene	1660	12,100	3865	709	16,500	3275	3475	267	8200	2940	72	2430	823	1940

for household waste composition at the time of construction (Parfitt, 2009), landfill density (Leonard Sr et al., 2000) and waste moisture content (Tchobanoglous et al., 1993; Riber et al., 2009) into Eq. (1). No data were available relating to industrial waste composition at the time Hadleigh Marsh was constructed or the proportion of the site that contains industrial waste, it was therefore assumed that the industrial waste is similar to household waste (Defra, 2013). Total metal loads in each material were then calculated using the total dry mass and median metal concentration for each material.

Calculation of the dry mass of the material type of interest (after Brand, 2017)

$$m = V \times D \times \frac{P}{100} \times \left(1 - \frac{L_c}{100}\right) \quad (1)$$

where:

m = dry mass of the material type of interest (tonnes),

V = total volume of landfill waste at site of interest (m^3)

D = typical landfill density at construction, 0.54–0.72 $t\,m^{-3}$ (Leonard Sr et al., 2000)

P = percentage of typical household waste that was the material of interest at the time the landfill was constructed, estimated from (Parfitt, 2009). NB to obtain the dry mass of the matrix material the dry masses of fines, miscellaneous and putrescible materials must be calculated individually and summed.

L_c = moisture content (% by mass) in material at time of landfilling. Paper = 8%, textiles = 10%, fines and misc. = 8%, putrescible materials = 70% (Tchobanoglous et al., 1993; Riber et al., 2009)

3. Results and discussion

3.1. Composition of excavated samples

The waste extracted from Leigh Marshes landfill (constructed 1955–1967) was predominantly composed of a brown and black, fine-grained particulate matrix interspersed with broken bricks, glass, ceramics, and small quantities of paper, rubber, bones, plant materials and wood (Fig. 2). This composition of waste is consistent with household waste of the era (Parfitt, 2009) and is likely to be broadly representative of over 850 mixed waste industrial, commercial and household landfills operational in England at the time (Environment Agency, 2015).

The waste extracted from Hadleigh Marsh landfill (constructed 1980–1987) had the appearance of present-day waste (before separation for recycling), consisting of plastics, ceramics, textiles (shoes, carpets, clothes), paper, wood, batteries, soil (predominantly clay), and putrescible materials (Fig. 3). This composition of waste is consistent with household waste of the era (Parfitt, 2009) and is likely to be broadly representative of over 1400 other mixed waste commercial and household landfills operational in England at the time (Environment Agency, 2015).

3.2. Contaminant concentrations in matrix material

Inorganic and organic contaminant concentrations in the matrix materials were highly variable within each trial pit, within each site and between the two historic landfills from different eras.

At the local scale, i.e. within individual trial pits, there were differences of up to two orders of magnitude between the minimum and maximum metal and organic contaminant concentrations in Leigh Marshes, and in Hadleigh Marsh differences in contaminant concentrations of up to three orders of magnitude for metals and up to two orders of magnitude for organics (Table 1 and Table 2). Generally metal concentrations (Mann-Whitney U, $\alpha = 0.05$) were also significantly different between trial pits at both landfills, whereas PAH (Mann-Whitney U, $\alpha = 0.05$) concentrations were generally only significantly different in Hadleigh Marsh, although there were some exceptions.

There were also significant differences in contaminant concentrations between the two historic landfills. Concentrations of Cd, Cr, Cu, Pb, Zn and all of the PAHs were significantly higher in the older Leigh Marshes landfill compared to Hadleigh Marsh (Mann-Whitney U, $\alpha = 0.05$). This is likely to be predominantly due to changes in the types of materials being landfilled in the different eras. Wood and coal ash were more common waste constituents at the time Leigh Marshes was constructed (mid-20th century), and ash can contain high concentrations of Cd, Cr, Cu, Pb, Zn and PAHs (Smolka-Danielowska, 2006; Liu et al., 2008; Tsibart and Gennadiev, 2013). In addition to commercial and household wastes, Leigh Marshes also contains industrial wastes. In keeping with many historic landfills there is no information regarding the nature of this waste (Environment Agency, 2013) so further conclusions cannot be drawn as to the potential industrial sources of these metals. The metal concentrations are broadly comparable to those found in studies of fresh and excavated waste (e.g. Hull et al., 2005; Zhao et al., 2007; Prechthai et al., 2008; Quaghebeur et al., 2013) but no equivalent studies of PAH concentrations in waste have been identified.

No previous studies of contaminant variability in solid wastes in historic landfills could be found for comparison, but such variability is consistent with other studies of both landfill leachates and fresh solid wastes and is due to the high heterogeneity of the waste materials being disposed of and the lack of mixing as they are deposited (Allen, 2001; Blight and Fourie, 2005; Taylor and Ramsey, 2006; Sormunen et al., 2008). When assessing risk and environmental impact for contaminated land investigations, such heterogeneity is dealt with through intense sampling to ensure representative data collection, and a site the size of

Table 3
Inorganic contaminant concentrations in wood, paper and textile samples (mg/kg) excavated from Hadleigh Marsh historic coastal landfill site.

	Wood						Paper						Textiles					
	Trial pit 3 (n = 36)			Trial pit 4 (n = 9)		Trial pit 3 + 4	Trial pit 3 (n = 26)			Trial pit 4 (n = 27)		Trial pit 3 + 4	Trial pit 3 (n = 9)			Trial pit 4 (n = 18)		Trial pit 3 + 4
	Min	Max	Median	Min	Max	Median	Min	Max	Median	Min	Max	Median	Min	Max	Median	Min	Max	Median
Cd	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD	2.1	0.26	0.01	14	0.25	< LOD	< LOD	< LOD	< LOD	< LOD	< LOD
Cr	1.8	53	11	< LOD	24	2.3	1.0	28	4.9	1.4	90	13	8.6	39	19	4.8	875	35
Cu	8.2	126	41	5.9	141	24	1.7	107	13	3.9	497	13	20	96	37	83	1767	119
Pb	< LOD	15,271	164	11	308	39	4.0	133	22	4.4	734	16	174	459	275	34	1016	60
Zn	153	6300	1098	268	3218	510	35	4700	356	21	965	144	520	889	685	58	699	183

Hadleigh Marsh would require 750 sampling locations (Verstraete and Van Meirvenne, 2008; British Standards Institution (BSI), 2011). Such sampling density is not achievable for historic coastal landfills, first due to the large numbers of historic landfills and financial constraints, but more importantly, such invasive sampling could destabilise the landfills making them even more vulnerable to erosion. Some comparative risk assessment approaches have assumed that a single generic contaminant dataset is representative of all landfill sites (e.g. Laner et al., 2008, 2009; Neuhold and Nachtnebel, 2011; Neuhold, 2013). However, our data heterogeneity both within and between sites (even with similar waste streams and environmental conditions) clearly indicates that this approach has limited validity as contaminants in one area of a landfill can even be significantly different to those in other areas of the same landfill. A more practicable approach may be to undertake limited sampling and analysis to determine the types of materials present and to obtain indicative contaminant datasets which are likely to be within the same order of magnitude as those from more intensive site investigations and therefore provide a reasonable indication of the pollution potential of a site at a fraction of the cost. This approach is also likely to be resource intensive when dealing with large numbers of historic landfill sites and there clearly exists a need for a method to determine which sites should prioritised for invasive investigation (e.g. Brand and Spencer, 2018).

3.3. Contaminants in wood, paper and textile samples

Metal concentrations in the wood, paper and textile samples from Hadleigh Marsh were also found to be highly heterogeneous with differences of up to three orders of magnitude between the minimum and maximum metal concentrations in the wood and paper samples, and up to two orders of magnitude in the textile samples (Table 3). Metal concentrations in the materials were highly variable within individual trial pits and there were significant differences in Cr and Pb concentrations in wood, Cr and Zn concentrations in paper, and Cu, Pb and Zn concentrations in textiles between the trial pits (Mann-Whitney U, $\alpha = 0.05$).

Most assessments focus on the matrix material as it is assumed to contain the greatest contaminant concentrations due to its high specific surface area (Parizanganeh, 2007). However, in this study although metal concentrations were generally lower in wood, paper and textiles compared to the matrix materials, the highest solid waste contaminant concentrations were observed for Cu and Zn in textiles and wood respectively (Kruskal-Wallis, $\alpha = 0.05$, and Mann-Whitney U post-hoc analyses, $\alpha = 0.0083$ with Bonferroni correction, Table 4) suggesting it is not only matrix materials that may be a significant contaminant source in solid waste materials. These contaminants may have been present when the materials were landfilled, e.g. high concentrations of Zn in the wood may be attributed to a combination of natural occurrence in wood and its use in wood paints, preservatives and fire retardant coatings (Mahltig et al., 2008; Pereyra and Giudice, 2009; Sakthivel et al., 2012), or the result of sorption of metals from leachates moving through the waste.

To assess the contribution of metals associated with non-matrix materials to the total contaminant load, total metal loads in the matrix material, paper and textiles (insufficient data were available for wood) were then calculated using the total dry mass and median metal concentration (Table 5). Whilst metal concentrations in paper and textiles are generally lower than in the matrix material, the total load of metals (in kg) associated with these materials is considerable, particularly for Zn associated with paper. Paper is of particular concern as it has been a significant component of household waste since the 1960s (Parfitt, 2009) and, therefore, is likely to be present in significant quantities in circa 20% of historic coastal landfills (Environment Agency, 2015). This suggests that in addition to the matrix material other waste materials may also present a chemical hazard if waste is released and should be considered within risk assessments.

Table 4
Ranking inorganic contaminant concentrations in wood, paper and textile samples using Kruskal-Wallis tests and Mann-Whitney U post-hoc analyses.

	Matrix vs wood	Matrix vs paper	Matrix vs textiles	Wood vs paper	Wood vs textiles	Paper vs textiles	Summary
Cd	Medians < LOD	M > P	Medians < LOD	Medians < LOD	Medians < LOD	Medians < LOD	Insufficient data to determine ranking
Cr	M > W	M > P	M > T	W = P	T > W	T > P	M > T > W = P
Cu	M = W	M > P	T > M	W > P	T > W	T > P	T > W = M > P
Pb	M = W	M > P	M = T	W > P	T = W	T > P	M = W = T > P
Zn	W > M	M = P	M = T	W > P	W > T	P = T	W > T = M = P

Table 5
Total metal loads in Hadleigh Marsh landfill by material type (range of values determined using lowest and highest typical landfill densities in the calculations).

	Matrix material	Paper	Textiles	Total
Proportion of the site by volume	~38%	~35%	~4%	~77%
Dry mass of material (tonnes)	57,480–76,650	86,940–115,920	9720–12,960	154,140–205,530
Cd (kg)	68–91	22–30	4–5	94–125
Cr (kg)	3398–4531	810–1080	184–246	4393–5857
Cu (kg)	1914–2552	1121–1494	991–1321	4026–5368
Pb (kg)	3622–4830	1620–2159	1691–2254	6933–9244
Zn (kg)	12,037–16,049	14,636–19,515	5075–6767	31,748–42,331

3.4. The potential ecological impact of solid waste release to the coastal zone

To assess fully the potential impact of released waste, e.g. through erosion, and to determine whether any physical or chemical deterioration to the environment would occur, it would be necessary to predict the rate and the total mass of waste released. Whilst it is understood that landfill instability and the slope failure are often caused by excessive water infiltration there are currently no suitable modelling tools for assessing the rate of waste erosion (Blight and Fourie, 2005; Dixon and Jones, 2005; Peng et al., 2016). However, the worst case scenario, where an entire site catastrophically fails rapidly releasing all of its waste, can be assessed (Neuhold, 2013). If the entirety of Leigh Marshes or Hadleigh Marsh landfills were to erode, the Cu and Zn inputs to the Thames Estuary would respectively be equivalent to 6.4 times and 3.3 times (Leigh Marshes), and 0.03 times and 0.05 times (Hadleigh Marsh) the total annual input of those metals to the estuary from all known sources (based on data for metal inputs in Stevenson and Ng, 1999).

The solid waste material has the potential to physically and chemically alter the estuarine environment if released. The potential ecological impacts of releasing waste materials from Leigh Marshes and Hadleigh Marsh landfill sites to the foreshore has been assessed using sediment quality guidelines in the absence of specific waste related standards (Figs. 4 and 5). Where concentrations exceed Interim Sediment Quality Guidelines (ISQGs) this indicates that there will be occasional adverse biological effects and exceedance of Probable Effects Levels (PELs) indicates that there will be frequent adverse biological effects (Canadian Council of Ministers of the Environment, 2001). The majority of Hadleigh Marsh matrix material samples exceed the ISQGs for Cd, Cr, Cu, Pb, Zn and PAHs and the PELs for PAHs, but are below the PELs for metals. In Leigh Marshes all matrix material samples exceed the ISQGs for Cd, Cu, Pb and Zn and the majority also fail for Cr. All Cu, Pb and Zn, and approximately half of the Cd concentrations also exceeded the PELs. In all of the Leigh Marshes matrix material samples all of the PAH concentrations exceeded the ISQGs, and the majority exceeded the PELs. Sediments in the Thames Estuary are already moderately contaminated with metals (Attrill and Thomes, 1995) and PAHs (Woodhead et al., 1999; Chesman et al., 2006); however, concentrations of most metals measured in the matrix material are significantly higher than those in adjacent estuarine surface sediments (Mann-Whitney U, $\alpha = 0.05$) (O'Shea, 2016; O'Shea et al., 2018). Therefore, release of matrix material to the adjacent saltmarsh would have a local and immediate deleterious impact on sediment quality and a long-term ecological impact on inter-tidal flora and fauna. The impacts of these effects are likely to be exacerbated by the presence of sensitive ecological sites, e.g. SSSIs, adjacent to the study sites, and similarly over one-third of England's historic coastal landfills are in or within close proximity of designated ecological sites (Brand et al., 2017).

Sediment quality guidelines may be used to assess the potential ecological impact of released matrix waste as this has similar physical and chemical characteristics (e.g. size, shape, behaviour) to coastal minerogenic sediments. Assessing the potential ecological impacts of contaminant concentrations in paper, textiles and wood is more problematic. The majority of the wood samples contain Cu concentrations that exceed the ISQGs, and Pb and Zn concentrations that exceed the PELs. The paper samples contain a substantial proportion of samples with Cu and Pb concentrations that exceed the ISQGs, and Zn concentrations that exceed the PELs. The textile samples contain a substantial proportion of samples with Cr concentrations that exceed the ISQGs, and the majority contain Pb and Zn concentrations that exceed the PELs. Pb and Zn in wood, and Cu, Pb and Zn in textiles are present in higher concentrations than in adjacent sediments (O'Shea, 2016; O'Shea et al., 2018). These materials could become incorporated into bed sediments or provide a food source. For example, wood-boring and ingesting crustaceans and molluscs can be adversely affected by the presence of contaminants (Sleeter and Coull, 1973; Cragg et al., 1999; Pati et al., 2012) and small paper/textile fibres have the potential to be consumed by filter feeders (Yusof et al., 2004; Cefas, 2008). These materials may take weeks (paper) to hundreds of years (man-made fibres) to decompose (DHEC, 2007). This suggests that although matrix material is usually the focus of investigations into the potential environmental impacts of landfills, eroded wood, paper and textiles also have the potential to cause adverse ecological effects.

These findings, based on Canadian sediment quality guidelines, highlight the need for the development of UK specific sediment quality guidelines and further investigations into the bioavailability of contaminants in historic landfill waste, e.g. sequential extractions, to fully understand the potential impact of eroding landfills on UK flora and fauna in the intertidal zone. This further work was beyond the scope of this study.

In addition to the potential for pollution from eroding waste materials, there is potential for the diffusional flow of contaminants from the waste to surrounding saltmarsh sediments due to the concentration gradient (Allen, 2001). It is likely that the clay linings of the landfills are attenuating this flow (United States Environmental Protection Agency, 2005); however, there is evidence of leachate plumes

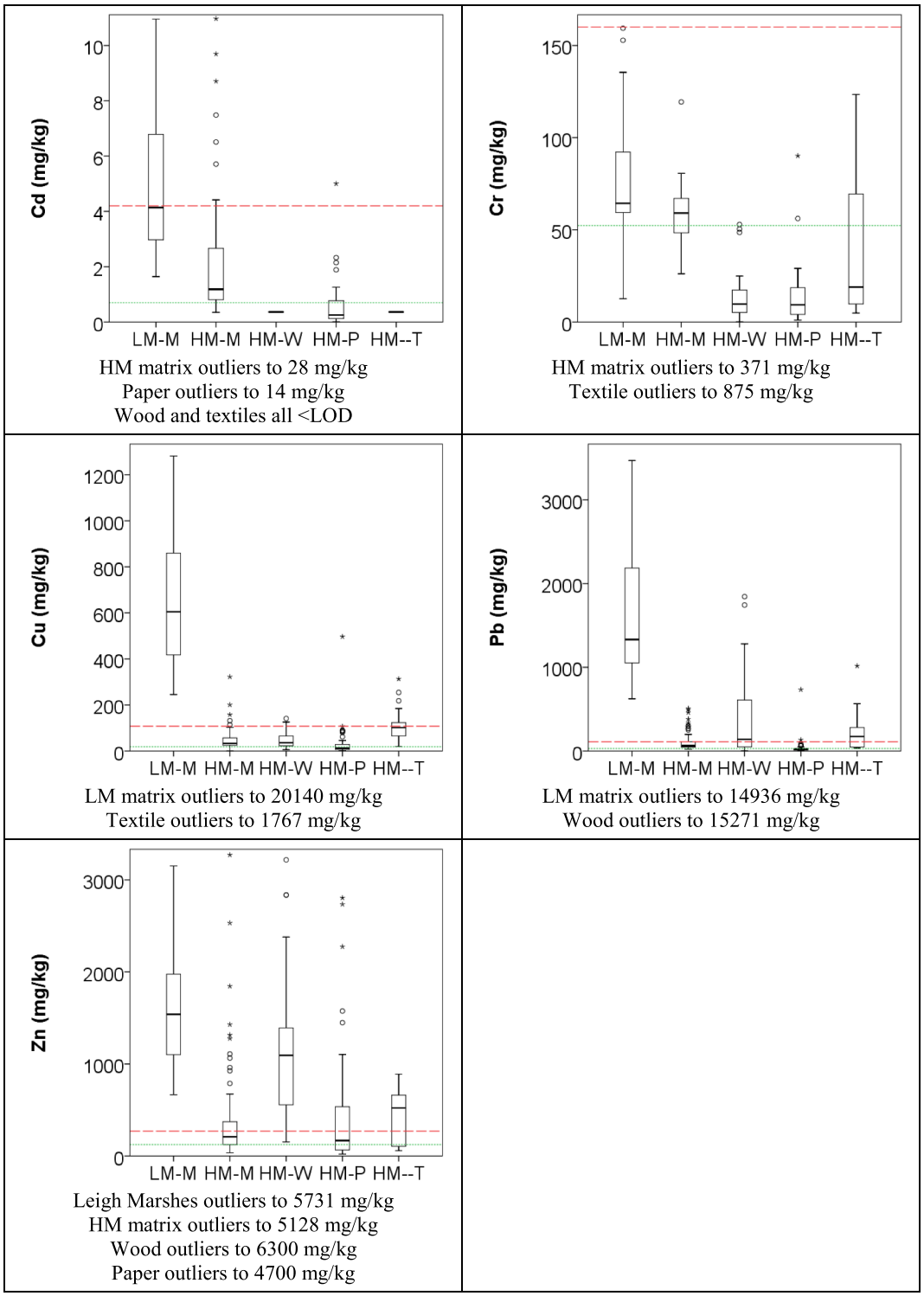

Fig. 4. Inorganic contaminant concentrations in Leigh Marshes matrix material (LM-M), and Hadleigh Marsh matrix material (HM-M) wood (HM-W), paper (HM-P) and textiles (HM-T) compared to Canadian sediment quality guidelines (··· ISQG Marine, ··· PEL Marine).

surrounding both sites at depths of circa 25 to 50 cm, and these contaminated sediments could become a secondary source of pollution if they were to erode (O'Shea, 2016; O'Shea et al., 2018).

4. Conclusion

Inorganic and organic contaminant concentrations in solid waste materials in two historic landfills in the Thames Estuary, Hadleigh Marsh and Leigh Marshes, are highly variable both within and between sites due to the heterogeneity of the landfilled waste and multiple contaminant sources. Generally, contaminant concentrations in the solid waste materials exceed sediment quality guidelines and may pose a significant threat to the environment if the site defences are not adequately maintained and waste materials are allowed to erode and are released to coastal environments. Soil and soil like materials (matrix) are the usual focus of contaminated land investigations, but here

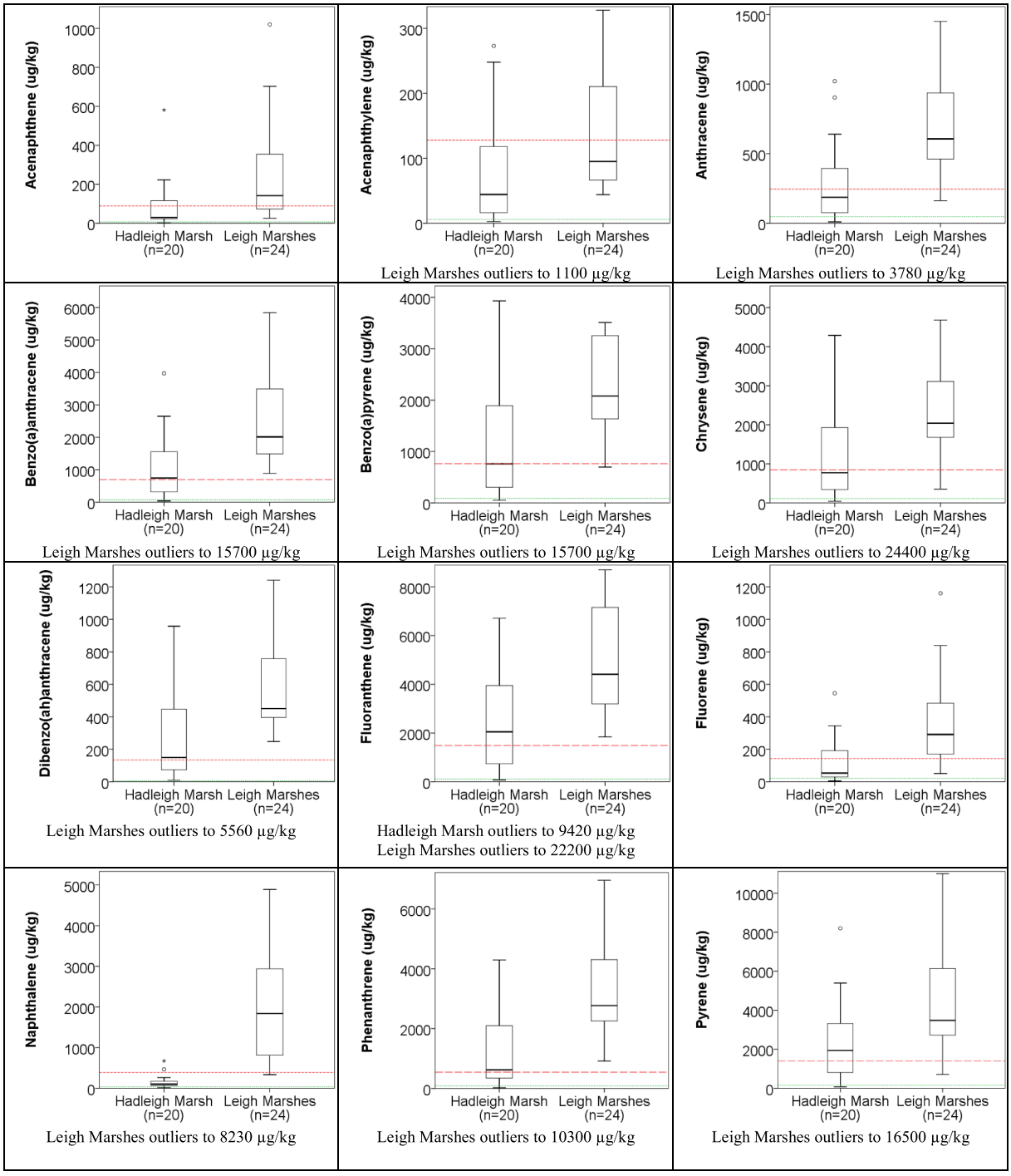

Fig. 5. Organic contaminant concentrations in the matrix material compared to Canadian sediment quality guidelines (··· ISQG Marine, ··· PEL Marine).

contaminant concentrations in wood, paper and textiles are also potentially hazardous and may be sources and sinks for contaminants in the waste stream. This highlights that historic landfill risk assessments should consider a range of waste materials.

Accurately determining the environmental impact resulting from eroded waste requires knowledge of the rate of waste erosion and dispersion, but currently there are no suitable tools for modelling landfill erosion and understanding of waste dispersion is limited (Browne et al., 2015). However, the landfills are within or adjacent to designated sites where it is an offence to allow any pollution to occur. The response strategy at landfill sites that have already released waste is to collect the largest waste materials as they erode. If this approach were used for Leigh Marshes or Hadleigh Marsh it would leave behind highly contaminated materials with implications for legislative compliance. This has implications for the other 443 historic coastal landfills in England that fall in or within 100 m of designated ecological sites (SAC, NNR, SSSI, SPA, Ramsar and MPA) (Brand et al., 2017).

Therefore, it is clear that in the short-term these landfill sites and their defences must be maintained and this places significant restrictions on the organisations responsible for the management of these sites – in this instance the local government authority and national Environment Agency. In the longer term, a more detailed understanding of the mechanical stability and life expectancy of historic coastal landfill sites, and the nature of waste, its impact and its eventual

fate in the coastal zone, are required in order to develop protocols for assessing risk. This will enable stakeholders to make evidence based, financially justified decisions on alternative sustainable coastal management options. Such options may include continued maintenance and protection of sites, relocation of waste, which is a strategy the US Government is already applying in Alaska (State of Alaska Department of Environmental Conservation, 2012), or managed release of waste materials to the coastal zone. It is also necessary to understand which landfill sites pose the greatest pollution risk in order that management resources can be prioritised and the authors recommend that a risk screening assessment method is developed for this purpose (e.g. Brand and Spencer, 2018).

Contributions

JHB carried out the research and performed all sample collection, preparation and analysis, except ICP-MS analysis was carried out by Plymouth University and GC–MS sample preparation and analysis was carried out by the Environment Agency's National Laboratory. JHB carried out all data collection and interpretation. KLS conceived the overall project and contributed to data interpretation. JHB and KLS wrote the manuscript. All authors have approved the final article.

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