Focus Article

Potential pollution risks of historic landfills on low-lying coasts and estuaries

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Historically, it was common practice to dispose of landfill waste in low-lying estuarine and coastal areas where land had limited value due to flood risk. Such 'historic landfills' are frequently unlined with no leachate management and inadequate records of the waste they contain. Globally, there are 100,000s such landfills, for example, in England there are >1200 historic landfills in low-lying coastal areas with many in close proximity to designated environmental sites or in/near areas influencing bathing water quality; yet, there is a very limited understanding of the environmental risk posed. Hence, coastal managers are more likely to select conservative management policies, for example, hold-the-line, when alternative more sustainable policies, for example, managed realignment, may be preferred. Some historic coastal landfills have already started to erode and release waste, and with the anticipated effects of climate change, erosion events are likely to become more frequent. Strategies to mitigate the risk of contaminant release from historic landfills such as excavation and relocation or incineration of waste would be prohibitively expensive for many countries. Therefore, it will be necessary to identify which sites pose the greatest pollution risk in order that resources can be prioritized, and to develop alternative management strategies based on site specific risk. Before such management strategies can be achieved there remain many unknowns to be addressed including the extent of legacy pollution in coastal sediments, impacts of saline flooding on contaminant release and the nature, behavior and environmental impact of solid waste release in the coastal zone.

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INTRODUCTION

Disposal of solid and hazardous waste through landfilling became common practice in Europe and the US toward the end of the 19th Century.[1] Initial regulatory guidance for waste disposal was highly variable with limited environmental considerations; however, as populations, waste production, and environmental awareness grew the guidelines for landfill operations became increasingly detailed and stringent.[1] Prior to modern waste disposal regulations in the latter half of the 20th Century (e.g., the Resources Conservation and Recovery Act in the US or the Waste Licensing Regulations in the UK), waste was frequently disposed of in sites with no impermeable lining, no leachate or gas collection/monitoring and limited or nonexistent reporting of waste materials, types (e.g., domestic, industrial, or hazardous), or volumes. Indeed, despite the more recent introduction of waste disposal regulations in the Global South, much waste is still disposed of in uncontrolled and poorly managed sites.[2–4] These landfills—as opposed to modern containment landfills—often rely

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on natural attenuation in surrounding soils and sediments to disperse and dilute the leachate contaminants to reduce the pollution impact on nearby surface and ground waters.[5] Most of these landfills are now closed, and are termed 'historic' or 'legacy' landfills, although legal definitions vary according to national regulatory authorities. This has left a widespread legacy of contaminated sites, for example, c. 100,000 historic landfills in the US,[6] c. 2000 in Flanders, Belgium,[7] c. 1000 in New Zealand,[8] c. 1000 in Austria[9] and c. 20,000 in England.[10]

Awareness of historic landfills and legacy waste is increasing and there is a growing body of evidence to demonstrate their potential to pollute the surrounding environment for decades following their closure. First, there is evidence that in the absence of linings or leachate management systems, sites may release dissolved nitrogen and metals to groundwater, floodplains, river water, and surrounding intertidal sediments.[11–13] Indeed, it is likely that some leachate will eventually escape all landfills, including those with impermeable and low permeability liners.[5,14] Second, some historic landfills are already eroding[15,16] and there may be release of solid waste to adjacent environments with potential implications for ecological health.[17] Finally, where historic sites are used for grazing, which is common practice for closed landfills, there is evidence that potentially toxic metals can translocate into above ground biomass.[18]

Historic landfills are frequently located on low-lying floodplains and coastal plains due to their proximity to population centers, easy access and the low value of the land. Hence, they are at risk of fluvial or coastal flooding[9,19,20] and these risks are likely to increase with climate change. Climate change is anticipated to cause higher sea levels, resulting in increased saline intrusion into estuaries, more frequent and intense storm events, higher storm surges and increased coastal flooding.[21–25] Where historic landfills are currently defended from the sea, coastal squeeze is likely to increase further the risk of flooding due to the loss of wave attenuating saltmarshes and may also increase the risk of erosion.[26,27] Flooding a landfill site will increase the volume of leachate generated by increasing percolation and the piezometric head of the leachate, which will cause the rate of leachate leakage to increase.[5] Neuhold and Nachtnebel[28] estimated that metal release during fluvial flooding of landfills may increase by up to four orders of magnitude through leaching or up to six orders of magnitude if matrix material is eroded. However, there is less understanding of how flooding with seawater and increased throughput of saline water will affect leachate contaminant loads. Over the next 100 years, many low-lying coastal locations are also expected to erode, and it is increasingly likely that more historic coastal landfills will begin to erode or catastrophically fail releasing solid waste and previously trapped leachates into the coastal zone. This is of particular concern due to the paucity of information regarding the waste materials present. Consequently, it is important to identify the potential receptors of any waste released and its contaminants to understand better the scale of the potential pollution risk and begin to identify the level of resources that would be required to address it.

This paper focuses on the potential pollution risks of historic coastal landfills sites in England. Historic landfills are defined by the Environment Agency as closed landfill sites that have '*no PPC [Pollution Prevention and Control] permit or waste management licence currently in force. This includes sites that existed before the waste licensing regime, if a site has been licensed in the past, and this licence has been revoked, ceased to exist or surrendered and a certificate of completion has been issued.*'[29] Historic coastal landfills are defined by this paper as historic landfills within areas with an annual risk of flooding by the sea of 0.5%, if not adequately defended, as shown in the Environment Agency's Flood Map for Planning (Rivers and Sea). Prior to the Control of Pollution Act 1974 (Secretary of State, 1974[30]) there were no requirements to keep records of waste disposed of in any landfill sites and it was not until the introduction of the Waste Management Licensing Regulations 1994 (Secretary of State, 1994[31]) that records were required for all landfill sites accepting controlled waste, i.e., household, industrial and commercial waste, or similar waste. The Waste Licensing Regulations 1994 only required records of estimates of the total quantities of biodegradable, nonbiodegradable, and special wastes within a site and the location of the special waste, i.e., controlled waste with special disposal requirements relating to its potential to pollute. The introduction of The Landfill (England and Wales) Regulations 2002 (Secretary of State, 2002[32]) made the keeping of detailed records of waste origin, type, volume, and disposal location mandatory, and introduced restrictions on disposing of different classifications of waste, i.e., hazardous, nonhazardous, or inert, within the same landfill. However, the majority of historic landfills predate the more stringent regulations and records for them are either incomplete or only specify whether there is evidence of the presence of inert, industrial, commercial, household, special, or liquids/sludge waste.[10] Records for individual landfills do not provide details of the materials deposited, and the composition of waste types changed greatly during the 20th century.[33,34] The Landfill (England and Wales) Regulations 2002 (Secretary of State, 2002[32]) also introduced the requirement for most modern-day landfills to have impermeable liners and leachate management systems to control leachate leakage.

Existing Environment Agency datasets for England are reviewed to determine the numbers of historic landfills that are potentially at risk of coastal flooding and/or erosion and the literature is reviewed to understand the potential pollution pathways and the environmental receptors that could be at risk. In addition, we propose a conceptual site model for contaminant transport pathways in the coastal zone that could inform the understanding of potential pollution pathways from historic landfills in a range of geographical contexts.

ASSESSING THE NUMBER OF HISTORIC LANDFILLS AT RISK OF FLOODING OR EROSION IN ENGLAND

The Environment Agency has recorded the locations of all 19,635 known historic landfills in England in the Historic Landfill Sites National Dataset, which consists of an ESRI shapefile containing the digitized boundaries of historic landfills and an attribute table which defines a unique Historic Landfill Database Reference Number for each site and contains (where known) data such as site addresses, site operator names, opening dates, closing dates, and waste types.[10] The datasets are updated frequently and were correct as of April 2017. To determine which historic landfills are at risk of flooding, ESRI ArcMAP was used to compare their locations to flood zone 3 as shown in the Environment Agency's Flood Map for Planning (Rivers and Sea) Dataset.[35] This showed 4759 historic landfills are located within flood zone 3, i.e., they have a 1% annual probability of fluvial flooding and/or 0.5% annual probability of coastal flooding if they are not adequately defended.

The focus of this paper are historic coastal landfills, of which, there are at least 1215 around the coast of England (Figure 1). Historic coastal landfills in England are predominantly clustered around estuaries with major cities, e.g., Liverpool, London, and Newcastle upon Tyne, but in southeast England there are also significant numbers in rural estuaries between Harwich and Ramsgate. As historic coastal landfills are often near major towns and cities, many are protected by flood defenses, and some form part of the flood defense network, e.g., Dengie, Hadleigh Marsh, and South Fambridge in Essex have flood embankments constructed from landfill waste capped with clay.[36] Climate change effects mean it is becoming increasingly likely that these sites will be inundated, although this increased risk differs around the UK due to variations in the effect of isostatic adjustment on relative sea level rise, e.g., between 1990 and 2095 increases in sea level of 21–68 cm are projected for London for medium emissions by UK Climate Projections compared to 7–54 cm for Edinburgh.[24]

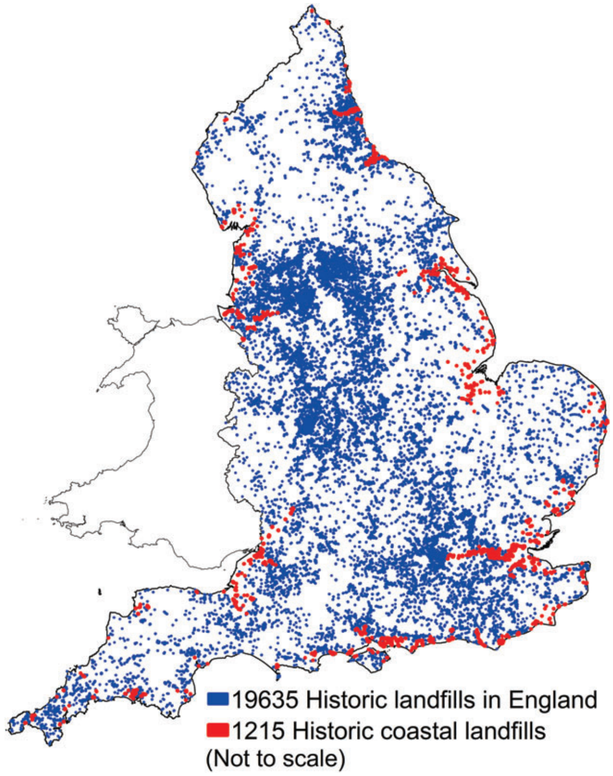

FIGURE 1 | Locations of historic landfill sites in England. (created using data © Environment Agency copyright and/or database right 2017. All rights reserved. Contains information © Local Authorities. © Crown copyright and database rights 2004 Ordnance Survey 100024198).

In addition, 28% of the coastline of England and Wales is eroding by at least 10 cm *per annum*.[37,38] To determine which historic coastal landfills are at risk of erosion, ESRI ArcMAP was used to compare their locations to the Environment Agency's National Coastal Erosion Risk Map's No Active Intervention 95% confidence scenario. Without intervention, 79, 122 and 144 historic coastal landfills are expected to start to erode into coastal waters in the short-term (by 2025), medium-term (by 2055) and long-term (by 2105) respectively.

MATERIALS AND CONTAMINANTS IN HISTORIC LANDFILLS

Municipal Solid Waste can contain hazardous substances including cleaning products (acids, alkalis, and solvents), batteries (heavy metals, e.g., lead, nickel, cadmium, and mercury), pharmaceuticals, pesticides and biocides, oils and fats, paints (solvents and fungicides), wood preservatives (e.g., creosote, tributyltin, and copper chrome arsenate), metal food containers (usually coated with Bisphenol A, an endocrine disruptor), and electrical and electronic equipment (e.g., mercury in fluorescent tubes, heavy metals, chlorofluorocarbons, and brominated flame retardants in plastics).[39,40] While there are numerous studies exploring leachate composition,[41–46] few studies have looked at contaminant concentrations in solid waste materials.

Typically there are no detailed records of the solid waste materials received in historic landfills.[1] In England, waste categories (e.g., household or industrial) for each site are recorded (where known) .[10] However, 42% of historic landfills contain waste from multiple categories with no data relating to the proportions of each category present, while there is no waste category information for 24% of the sites. General records of household waste types do exist, which show that the typical composition of waste includes glass, metals, paper, plastics, putrescibles, screenings (dust/ash), textiles, and other unclassified materials.[1,33] The proportions of these in landfills has changed significantly during the 20th century as legislation and availability of new materials has changed, e.g., there has been a reduction in screenings and increases in paper and plastics proportions since the mid-1960s,[33,34] however, the operational period of 50% of historic landfills is unknown.[10]

Studies of historic waste tend to have examined metals in matrix materials,[47,48] often focusing on the potential of the sites for ‘landfill mining’ and therefore overlooking metals contained in other waste materials and potential contaminants that have no recycling value such as organics or asbestos. Metal concentrations can vary up to two orders of magnitude between sites, with mean concentrations of up to 19 mg kg^{-1} of Cd, 5730 mg kg^{-1} of Cr, 5750 mg kg^{-1} of Cu, 2640 mg kg^{-1} of Pb, and 5600 mg kg^{-1} of Zn.[49–52] These concentrations are approximately two orders of magnitude higher than natural background concentrations in adjacent sediments.[53,54] The magnitude of contamination may pose significant environmental risks to surrounding coastal and estuarine environments; however, there have been few studies determining the potential impact upon sediment quality, and flora and fauna[51,52,55] should the waste be released to the coastal zone. The significant variability of contaminant concentrations, and the shortage of data relating to operational periods, waste categories, and material types present in historic landfills, makes the meaningful assessment of the environmental risks of historic landfills challenging.

POTENTIAL TRANSPORT AND FATE OF POLLUTANTS FROM HISTORIC COASTAL LANDFILLS

Qualitative conceptual models are presented (Figure 2) to show the possible present-day and future pollution linkages between solid waste stored within historic coastal landfills and sensitive coastal receptors. Such models can provide a framework to identify potential pollution risks and inform further investigations and remediation strategies. A number of sources of contamination exist within these environments including solid waste, contaminated leachate, and secondary sediment contamination in the natural attenuation zone.[13] Pollution pathways may include leachate migration, and erosion of solid waste and contaminated sediments and their release to the coastal zone.[20]

The composition of leachates, the water-based solution generated by infiltration of water and internal moisture in a landfill,[56] and, therefore, their contaminating capacity is strongly linked to the original waste material composition[57] and degradation state of the waste. When waste is first deposited in a landfill, conditions are oxic, resulting in a short period[58] of aerobic degradation classified by high temperatures and CO_2 production. Once oxygen has been consumed, anaerobic degradation becomes dominant,[59] where high concentrations of ammonia, CO_2, and carboxylic acid are produced and the waste temperature drops to 30°C.[60] Carboxylic acid is then turned into acetic acid, dropping the waste pH and significantly increasing the solubility of metals.[61] This is characterized by a discreet spike in heavy metal concentrations within the leachate.[56,62] Subsequent methane production restores the pH and represents methanogenesis, the longest degradation stage[63] with ammonia being produced throughout a landfill's lifetime.[43] The rate at which the landfill passes through each of these stages can be significantly affected by geological conditions at the site.[64,65]

Leachate composition and production from landfills has been thoroughly studied,[62,66] and in the absence of basal linings leachate will migrate through surrounding fine-grained sediments undergoing natural attenuation[67] and producing leachate plumes up to 1000 m in length.[66] Where historic landfills are situated

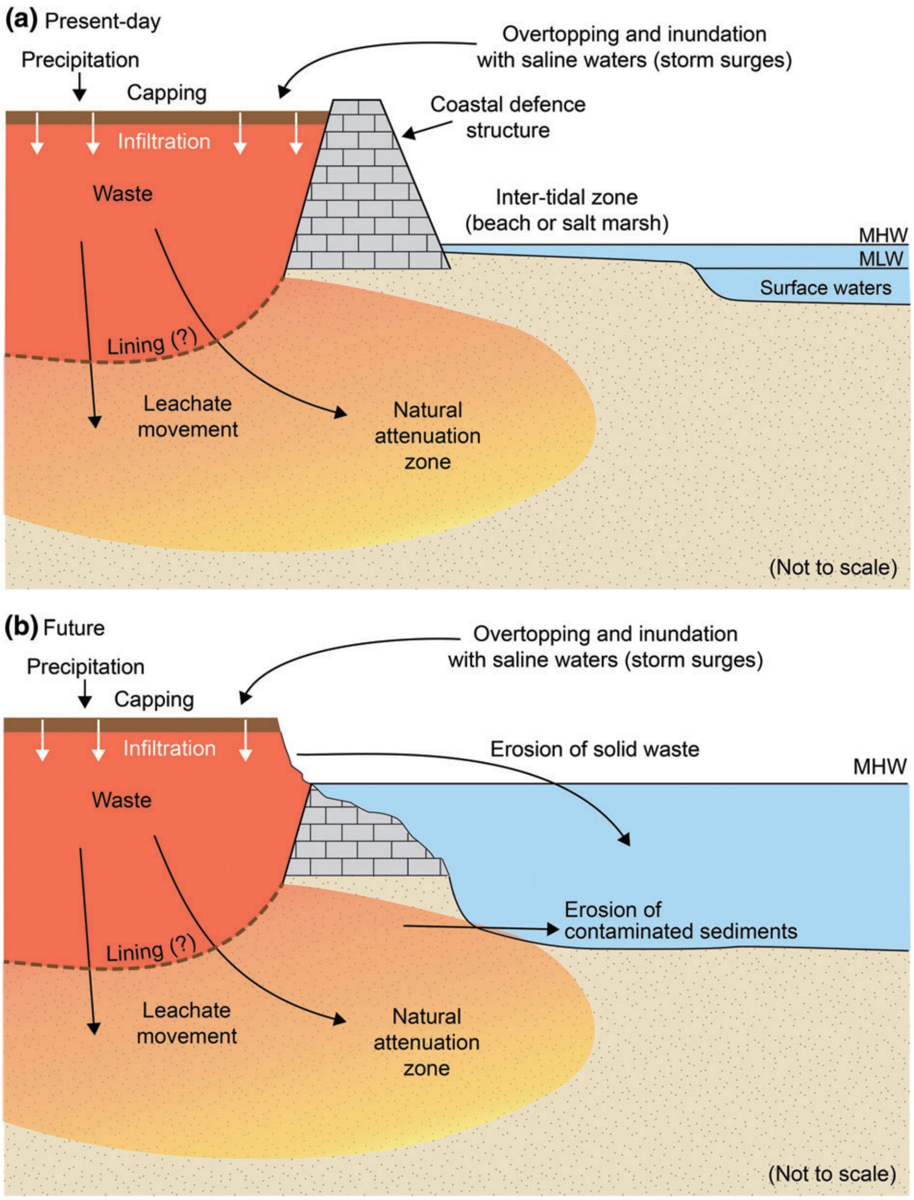

FIGURE 2 | Conceptual models showing leachate migration from a fully contained landfill under present-day conditions (a) and a potential future scenario (b) where sea level has risen, the landfill's defenses have been breached, and erosion of solid waste and contaminated sediments has occurred.

directly in the coastal zone, natural attenuation processes such as sorption and precipitation within coastal sediments will immobilize some contaminants, e.g., metals, resulting in elevated sediment metal concentrations surrounding historic landfills while reducing the impact of leachates on surface waters.[68] These areas of contaminated sediments extend beyond the landfill boundary and constitute a secondary contamination source, particularly where those sediments may be subject to erosion and remobilization.[12,13] Indeed, contaminated sediment surrounding historic landfills has been identified as a significant cause of *in situ* contamination in freshwater and coastal environments.[69] In addition, landfills still produce ammonia-rich leachates polluting rivers and groundwaters decades after closure[11] although this is unlikely to have a significant ecological impact in marine waters where ammonia levels are naturally higher. The three-dimensional extent of leachate plumes is dependent upon interaction with the water table, and leachate viscosity and density and their vertical movement can be unpredictable.[70] However, the extent of this natural attenuation zone in the coastal environment, which will be influenced by tidally fluctuating groundwater, has received little attention.

While the release of leachates as a potential pollutant pathway from landfills has been well studied, most of this research assumes normal operating conditions, i.e., sites are not inundated and waste is fully contained.[71] With sea level rise, and increased risks of storm surges, saline intrusion, and flooding events, there is an increasing risk that historic coastal landfills may be inundated. Waste decomposition rates are known to be largely controlled by moisture content and leachate cycling can be used to increase moisture content to speed up degradation[62]; however, the impacts of flooding on contaminant release have rarely been considered.[9] Inundation with seawater would increase the volume of leachate generated as more water would percolate through the waste[5] and would change the biogeochemical environment within the landfill, i.e., introducing oxygenated, fully saline waters, which may mobilize contaminants.[72]

The impact of increased salinity on waste is relatively well understood due to the recycling of leachate containing high total dissolved solids concentrations (between 2000 and 60,000 mg L^{-1}) in bioreactor landfills.[66] The goal of leachate recycling is to maintain the moisture balance and enhance biodegradation and methane production,[73] and in arid regions seawater or brackish waters are sometimes also used. However, at high salinity (> c. 3% dissolved solids) anaerobic bacteria are inhibited reducing biogas production increasing the time taken for waste to stabilize with potential impacts for the long-term management of historic landfills.[74,75] A few studies have examined biodegradation in landfill sites inundated with seawater but have not looked at contaminant mobility.[76,77] In the wider literature, it is commonly recognized that organic compounds are less soluble in seawater than freshwater due to the 'salting out' effect.[78,79] Therefore, it is likely that seawater intrusion into historic landfills will not increase the release of soluble organic contaminants. However, there is no such consensus for inorganic contaminant solubility. During waste degradation metals are likely to have been immobilized by a number of processes including sorption to soil particles and organic matter in the waste.[62] Studies of contaminant release from sediments and soils following increases in salinity generally report an increased metal solubility resulting from cation exchange processes and the availability of complexing ions such as chloride and sulfate.[80–82] The effects of salinity changes on the release of metals from other materials are rarely studied, but Schäfer et al.[83] found the solubility of metals in urban particles decreased as salinity increased. However, ultimately the behavior of metals with increasing salinity is complex,[83,84] due to differences in metal speciation and organic content of the materials.[85,86] Therefore, inundation of waste with seawater may result in an increased flux of dissolved metals. The effects of saltwater intrusion into waste could be modeled, e.g., using PHREEQC[87]; however, this requires data characterizing solid waste including information on metal speciation, contaminant sorption characteristics, and organic matter content which may not always be available for historic sites.

Under both current and future scenarios (Figure 2), there is potential for solid waste to erode, and erosion of landfills has been observed at a number of sites,[17,88,89] e.g., at East Tilbury in the Thames Estuary (Figure 3). In addition to the mobilization of contaminants, flooding increases the probability of erosion due to the movement of water over the site[90] and because infiltration of high volumes of water can adversely affect the structural integrity of the waste increasing the likelihood of mechanical failure of the landfill.[91] In addition, rising sea levels and increased storminess may increase the likelihood that defenses currently containing the waste fail. This could lead to the physical mobilization of pollutants and solid waste including glass, metal, plastics, and asbestos. In addition, the waste will be released to oxidizing environments and as precipitation with sulfides is a key mechanism for immobilizing metals[62] there is again potential for contaminant release.

ASSESSING THE RISK TO RECEPTORS

Although there have been few studies of the effects of landfill waste on estuarine and marine environments,[12,17,92] it is known that leachates can contain contaminants that may adversely affect flora

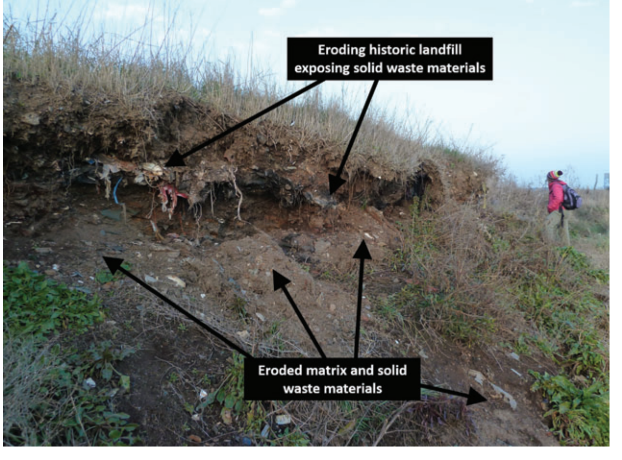

FIGURE 3 | Erosion of solid waste materials from East Tilbury landfill in the Thames Estuary (Source: Dr. J. H. Brand, January 23, 2017).

and fauna in coastal environments, e.g., through deoxygenation, eutrophication, direct toxicity, or toxicity as a result of biomagnification/bioaccumulation.[12,93] However, until data are obtained relating to historic coastal landfill leachate volumes and contaminant concentrations in saline environments, it is not possible to determine the specific effects there may be on receptors. In addition, it is not possible to assess the effects of eroded solid waste until data are obtained relating to the material types, contaminant concentrations, and rates of erosion, but it is known that there is the potential for mechanical impacts on ecology in addition to chemical ones if waste erodes, e.g., through ingestion of plastics.[94,95]

While there is still a need for further data to quantify the potential release of contaminants from historic coastal landfills following inundation and/or erosion, it is clear that there may be significant impacts in the coastal zone to humans, ecology, and surface waters. The Historic Landfill Sites National Dataset,[10] Flood Map for Planning (Rivers and Sea) Dataset,[35] and ESRI ArcMAP were used to assess the proximity of environmentally sensitive areas to historic coastal landfills, and where the receptors were within 100 m they were considered 'at risk.' It should be noted that this assessment does not consider the potential for buoyant eroded waste materials, such as wood and plastics, to carry contaminants to vulnerable receptors in remote locations.[96] Table 1 shows the number of landfill sites that pose a risk to each receptor type assessed, and the types and numbers of potentially vulnerable receptors are shown in Table 2.

TABLE 1 | Number of Historic Coastal Landfills in or within 100 m of Sensitive Environmental Areas in England

Site Type	Number of Landfills in or within 100 m of Sensitive Sites	Proportion of the 1215 Historic Coastal Landfills (%)
SSSI	411	34
National Nature Reserve	33	3
SAC	169	14
SPA	302	25
Ramsar	305	25
OSPAR Marine Protected Areas	246	20
Bathing Water Catchments	579	48
Bivalve mollusc production areas[1]	47	4

[1] Some locations have multiple bivalve production areas designated for different species.

TABLE 2 | Number of Sensitive Environmental Areas on or within 100 m of Historic Coastal Landfills in England

Site Type	Number on or within 100 m of Coastal Landfills	Proportion of the Total Number of Sites in England (%)
SSSI	120	3
National Nature Reserve	21	9
SAC	28	11
SPA	39	46
Ramsar	37	51
OSPAR Marine Protected Areas	47	39
Bathing Water ZOI Catchments	128	32
Bivalve mollusc production areas[1]	137	31

[1] Some locations have multiple bivalve production areas designated for different species.

Humans may be exposed to contaminants in eroded landfill waste through direct contact with debris on the foreshore, e.g., through handling, accidental cuts and inhalation (e.g., asbestos), or through bathing in water that may be contaminated by either leachate or eroded waste. Approximately one-third of England's bathing water ZOI (zones of influence) catchments[97] are within 100 m of historic coastal landfills (Table 2).

Eroded waste material has the potential to harm flora and fauna by physically and chemically altering the estuarine environment, e.g., by increasing localized suspended particulate matter concentrations and nutrient loads, reducing dissolved oxygen concentrations, and physically damaging benthos in the estuary by crushing or smothering them, but there have been no studies of the impact of landfill debris on the marine environment.[17,98] Physical harm to fauna in the estuary could also result from the release of plastics from landfill sites. The mechanical impacts of plastics on marine organisms include starvation or suffocation due to entanglement, and injury due to ingestion, which can result in reduced feeding, internal injuries, gastrointestinal blockages, and death.[94,95] Contaminants in the solid waste and leachates may be taken up through the roots of flora, or ingested by filter feeding fauna, which can result in either direct toxicity or biomagnification/

bioaccumulation leading to toxicity[12,93,99] and can result in trophic transfer of contaminants through the food web. Approximately one-third of bivalve mollusc production areas are within 100 m of historic coastal landfills (Table 2) (Cefas maps of bivalve mollusc production areas, O. Morgan, personal communication, email, November 2, 2015).

The potential exposure of a range of other environmentally designated sites to landfill contaminants was assessed using JNCC[100] and Natural England[101] datasets and it was found large numbers of designated sites are potentially at risk of contamination if historic coastal landfills are not adequately maintained, including over 50% of England's Ramsar sites (Table 2).

CONCLUSION

In England alone there are at least 1215 historic landfills in low-lying coastal areas that are at risk of flooding, and 79 sites that are at risk of erosion (by 2025) if they are not adequately defended, but there is a very limited understanding of the environmental risk posed by these historic coastal landfills. There are limited data available for the assessment of the risk of pollution from eroded solid waste, and while seawater intrusion into historic coastal landfills is likely to mobilize inorganic contaminants, this is supported by few detailed studies. Yet, over one-third of historic coastal landfills in England lie in close proximity to designated environmental sites and half are in/near areas influencing bathing water quality. This knowledge gap means coastal managers are more likely to select conservative management policies and continue defending the sites, e.g., hold-the-line, in order to ensure compliance with legislation that prohibits pollution of surface waters, when alternative more sustainable and cost-effective policies, e.g., managed realignment, may be preferred.

Although this paper has focused on the status of historic coastal landfills in England, the issues raised are equally applicable to vulnerable landfill sites elsewhere in the world, some of which have already started to erode and release waste.[102,103] With the anticipated effects of climate change, erosion events are likely to become more frequent. Strategies to mitigate the risk of contaminant release from historic landfills such as excavation and relocation or incineration of waste are being used in some locations, e.g., Alaska[104] and Switzerland.[105] However, these strategies would be prohibitively expensive for countries which have high numbers of large capacity historic landfill sites in vulnerable coastal locations. Therefore, it will be necessary both to identify which sites pose the greatest pollution risk in order that resources can be prioritized accordingly, and to develop alternative management strategies based on site specific pollution risk. Before such management strategies can be achieved, there remain many unknowns to be addressed including the extent of legacy pollution in coastal sediments, the impacts of saline flooding on contaminant release, and the nature, behavior, and environmental impact of solid waste release in the coastal zone.

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FURTHER READING

Further information and an interactive map can be found here: http://www.geog.qmul.ac.uk/research/historiclandfill

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