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Regadío, M. [orcid.org/0000-0002-0406-6416](https://orcid.org/0000-0002-0406-6416), Cargill, A., Black, J.A. [orcid.org/0000-0002-3529-6708](https://orcid.org/0000-0002-3529-6708) et al. (1 more author) (Submitted: 2020) High Attenuation Recycled Materials as landfill liners (the HARM project) – a new concept for improved landfill liner design. EarthArXiv. (Submitted)

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# High Attenuation Recycled Materials as landfill liners (the HARM project) – A new concept for improved landfill liner design

Mercedes Regadío <sup>\*1</sup>, Alex Cargill <sup>2</sup>, Jonathan A. Black <sup>2</sup> and Steven F. Thornton <sup>1</sup>

<sup>1</sup> Groundwater Protection and Restoration Group, Department of Civil and Structural Engineering, University of Sheffield, Broad Lane, Sheffield, S3 7HQ, United Kingdom

<sup>2</sup> Geotechnical Engineering Group, Department of Civil and Structural Engineering, University of Sheffield, Mappin Street, Sheffield, S1 3JD, United Kingdom

\* Corresponding author: Mercedes Regadío, [mregadio@mregadio.com](mailto:mregadio@mregadio.com)

**ABSTRACT:** A new approach in landfill liner design which combines hydraulic containment of leachate with contaminant attenuation to improve the performance of these environmental control systems at landfills is described. The idea is to re-use readily available industrial waste residues (construction and biomass waste) as additives for natural clay liners, wherein the additives have specific properties which enhance the attenuation of contaminants by the mixture. The aim is to (1) evaluate the contaminant attenuation capacity of these mixtures, (2) develop design guidelines to construct liners for waste containment systems and similar applications, and (3) interpret their performance using numerical modelling. This is evaluated in permeation studies using a geotechnical centrifuge, which enables the performance of liner compositions to be tested for representative time-scales (100 years), pressures and temperatures at realistic experimental time-scales of days-weeks in the laboratory. The permeation experiments include liner compositions flushed with leachate to deduce contaminant transport and attenuation mechanisms, followed by rainwater to assess the potential for release of attenuated contaminants. This experimental methodology is illustrated with depth profiles from permeation studies conducted on different clay-additive compositions. The concept will be applicable for liner design at other waste disposal facilities and is a timely improvement which addresses the problem of managing large quantities of industrial residues. Instead of disposal these can be recycled as an additive in host clay to construct these liners, thus conserving natural resources (clay) and reducing construction costs. It also provides an effective and more environmentally sustainable basis to reduce risks from leachate leakage.

*Keywords: diffusion, HARM project, landfill, leachate attenuation, recycled liners.*

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## 1. INTRODUCTION

Waste production continues to increase in response to a growing global population and modern lifestyles (EEA 2007; Eurostat Data Centre on Waste 2012). The 2 billion tonnes of waste produced across the world in 2016 are expected to increase by 70% in 2050 (Kaza et al., 2018). Globally, over two-thirds of waste is disposed to landfill, of which between 23% (high-income countries) and 63% (low-income countries) is unlicensed dumping (Kaza et al., 2018). As a result, the large volume of toxic and polluting leachate produced poses a high risk to human health and the environment. Environmental control measures such as liners to minimise leachate migration (containment function) and reduce pollutant loads (attenuating function) are required at landfill sites. The containment function relies on impermeable liners to confine polluting emissions from waste and engineered systems to collect these emissions for treatment. The attenuation is based on the selection of suitable geological strata, which function as reactive materials able to reduce the flux of contaminants in leachate by natural attenuation processes (Allen, 2001; Allen and Taylor, 2006).

Modern landfills use a combination of both *engineered (geo)synthetic materials* as containment liners and *compacted clay* as attenuating liners. Their primary aim is to minimise leachate production and escape from a site. Regulations establish a maximum hydraulic conductivity (i.e permeability to fluids) and minimum thicknesses for the clay liners to achieve containment, but no guidelines or criteria on their attenuating capacity (Council Directive 1999 Annex I). A major limitation of the containment strategy is that final stabilisation of the waste is prolonged and the contaminating potential of leachate is maintained, potentially beyond the design life of the control systems, to create a long-term environmental risk. Failure of the liner components in <10 yrs by different mechanico-chemical mechanisms is common (Louati et al., 2018; Rowe and Sangam, 2002; Yesiller et al., 2000). Diffusion of contaminants through clay liners due to the high concentration gradient and persistent source in the leachate remains a key problem causing liner failure in the long term (Chen et al., 2005).

A novel way to improve the environmental safety at landfills is to integrate the attenuation and containment function in liner design. In this way, the risk of environmental pollution and related cost and duration of aftercare monitoring could be reduced. Natural attenuation is already used in the management of contaminated groundwater but not in landfill liner design. The concept uses naturally-occurring processes (dilution, dispersion, biodegradation, redox processes, precipitation, sorption, ion exchange reactions) to mitigate environmental risk (National Research Council, 2000). Previous studies have shown the suitability of natural clays as natural attenuation barriers to contaminants in landfill liners (Regadío et al. 2012; Thornton et al., 2001). This is largely due to biogeochemical interactions which occur between clay minerals and contaminants in leachate (Regadío et al., 2015). While this concept offers a promising way to increase the environmental security of landfill sites, it has not been widely implemented in current liner design, and there is also a need to ensure it is environmentally sustainable.

The HARM project (*High Attenuation Recycled Materials as sustainable barriers for waste disposal sites*), described in this paper, attempts to address this issue. The research evaluates the performance of compacted clays mixed with non-hazardous industrial residues as new low-cost liner materials, for enhanced attenuation of contaminants in landfill leachate. The use of industrial wastes in this context is novel. Often the plasticity and shrink/swell potential of clays may cause significant and irreversible volume changes, differential settlement, cracking and enhanced pollutant fluxes through barriers. Cohesionless granular soils such as sand can be added to clays to solve this. Instead of sandy soils, the HARM project proposes to recycle non-hazardous residual materials from industrial processes as additives in compacted clay liners, as these wastes are freely available in large quantities and their reuse decreases the quantities requiring disposal. In addition, the potential saving of natural resources (clay, sand) will counteract the increasing demand for, and dependency on, raw materials in the EU-28 (European Commission, 2011; 2013).

The approach followed in the HARM project involves the evaluation of contaminant attenuation by clay-waste mixtures in leachate permeation experiments using a geotechnical centrifuge to simulate relevant operational timescales for landfill liner systems. The most appropriate composition(s) of the

liner(s) will be selected for studying their performance under field conditions, over and beyond the design lives (>100 yrs) of landfills. The permeation experiments include a leachate flushing phase followed by a rainwater flushing phase to assess the reversibility of contaminant attenuation and risk of contaminant release under different environmental conditions that may affect liner performance.

The potential of centrifuge modelling has rarely been explored for the purpose described in this study (Kumar, 2006, 2007; McKinley et al., 1998; Timms and Hendry, 2008; Shu et al., 2018), being widely used to study failure mechanism of slopes, earth and barriers retaining structures, and for building or bridge foundation (Edelmann et al., 1999; Indrasenan Thusyanthanet al., 2006a, 2006b; Jessberger and Stone, 1991; Taylor 2003). This research will produce design guidelines enabling the selection and engineering of liner compositions from these materials which maximise contaminant attenuation for appropriate operational timescales (i.e. decades) of landfill sites. The aim of this paper is to illustrate the technical and experimental approach underpinning this new concept in landfill liner design.

## **2. OBJECTIVES**

The overarching research objectives of the HARM project are to:

- Experimentally determine the attenuation of inorganic and organic chemicals in landfill leachate for recycled additive/clay mixtures and derive quantitative descriptions for these processes;
- Quantify reductions in contaminant diffusion through liners using these mixtures;
- Determine the changes in mechanical properties (especially strength and deformation) and hydraulic conductivity of the most effective material mixtures after use as a liner
- Evaluate the potential for release of attenuated contaminants in material mixtures after use as liner
- Develop a numerical model for quantitative prediction of contaminant transport, attenuation and engineering design of dual containment-reactive liners using these mixtures

The important stages which enable the achievement of these objectives include:

1. Characterisation, selection and testing of clays and non-hazardous industrial waste materials for use in mixtures as low-permeability liners;
2. Performance evaluation of clay-waste mixtures as candidate liners in laboratory permeation studies over representative design timescales and operational conditions;
3. Translation of the experimental results into practitioner-focused technical guidance for the engineering design of containment-attenuation liners based on the HARM concept for implementation at landfill sites

This paper focuses on describing the experimental strategy adopted for (1) and (2). As an output from these two stages, the guidance (3) will be reported in a separate paper.

## **3. SELECTION AND CHARACTERISATION OF EXPERIMENTAL MATERIALS**

An initial sample size of at least 100 kg each material and 100 L of leachate is recommended for all the characterization and experimental tests.

### **3.1 Natural clay substrata**

Clays have important properties (e.g. high specific surface area (SSA), cation exchange capacity (CEC), and absorption) which enhance the attenuation of inorganic contaminants in landfill leachate by filtration, sorption, biodegradation and precipitation reactions (Allen, 2001; Allen and Taylor, 2006). However, clays can have high plasticity, which may result in large volume changes due to swelling and

shrinkage upon uptake and removal of water or contact with high ionic strength liquids, such as leachate (Louati et al., 2018; Yesiller et al., 2000). This can lead to increased leakage through compacted clay liners. To assess the effect of these properties and develop selection criteria for the use of clays in liner mixtures, several candidate clays were sampled from the following sources in the UK, based on a survey of the literature (Fannin 2006; Freeman, 1964, Hudson and Martill, 1994; Kemp and Wagner, 2006; McEvoy et al., 2016; Scotney et al., 2012):

- London Clay (North Essex) is a marine, smectite-rich clay of Eocene age, with the highest porosity and plasticity due to its content of expandable clay minerals (smectite). This provides the highest CEC and total SSA. Porosities were compared and determined in each clay at the maximum peak point of the compaction curve of British Proctor BS 1377:4:3.3 (1990).
- Oxford Clay of the Peterborough Member (Northwest Buckinghamshire) is a marine, illite-rich clay of Middle Jurassic age, with the highest particulate organic matter content. It also has a high content of iron sulphide minerals and calcium carbonate (from fossils), providing a high pH buffering capacity.
- Coal Measures Clay (West Yorkshire) is a fluvatile kaolin-rich clay of Upper Carboniferous age with an acidic pH due to its relatively high content of iron sulphides and lack of fossiliferous material providing alkaline pH buffering capacity. It has the highest content of clay minerals, giving the highest external SSA and lowest CEC due to the prevalence of kaolinite. It has the lowest porosity and plasticity and highest water void saturation after optimum compaction.

This analysis shows that the three clays differ significantly in their shrinkage potential, sorption capacity and permeability to fluids. The pore water of the clays is dominated by dissolved sulphate, calcium and sodium, while potassium and ammonium are negligible. The electrical conductivity measured after pore water extraction with a 2:1 liquid to solid ratio is <math>1-3\text{ mS/cm}</math>, except for Oxford clay which is ca.  $5\text{ mS/cm}</math>. After optimum compaction to achieve the maximum (bulk) dry density and minimize any settlement risk, both the London and Oxford clays retain more water than the Coal Measures Clay. However, the void saturation achieved with water is lower for the London and Oxford clays compared with the Coal Measures Clay, decreasing the overall air content from 8 and 7 to 2 vol% respectively, and thus the permeability to fluids. The physico-chemical, mineralogical and geotechnical properties of the clays related to their feasibility as landfill liners are described in more detail in Regadío et al. (2020).$

### 3.2 Additives recycled from industrial process residues

The high plasticity (CH in the Unified Soil Classification System, USCS) when applying the BS 1377:2 (1990) of the Oxford and London clays implies a susceptibility to cracking and slope and landsides failure; thus they are unsuitable as a bottom liner *per se* (Louati et al 2018, Wagner 2013; Yalcin 2007). However, London Clay is the best material regarding the sorption capacity and low erodibility, and Oxford Clay is the best regarding the acid buffering and organic degradation properties. Coal Measures Clay is the best material regarding the plasticity (intermediate) and the impermeability (due to its lowest air void saturation), but it has poorest sorption capacity. No single clay has an optimum range of properties supporting its use in isolation. This can be addressed by including a granular, non cohesive, industrial process residue in a mixture with the clays to improve the overall characteristics of the liner.

Two industrial waste residues with no plasticity were selected as additives in the liner mixtures: fine mixed aggregates (FMA) and biomass bottom ashes (BBA). After applying standard water-leaching methods (section 5.1), these recycled additives are classified as inert waste (Council Decision 2003). Thus, they do not release elements at concentrations above hazardous levels after leaching with water, with no risk in their intended use. Both additives are reported to improve the bearing capacity of clays for road infrastructures (Agrela et al., 2012; Cabrera et al., 2014). Furthermore, FMA and BBA may also enhance the attenuation of contaminants in landfill leachate due to their high SSA, absorption capacity and chemical composition (Cabrera et al., 2016):



- FMA (from [GECORSA](#), the waste and recycled aggregates management company in Andalusia, Spain) are masonry residues of various origin from building, reparation and demolition works. They mostly contain calcite/magnesium carbonates, hydrated calcium sulphate and calcium/iron/silica oxides.
- BBA (from [Sacyr](#) Industrial, the biomass power plant of Puente Genil in Andalusia, Spain) is unburnt olive agricultural waste produced in the renewable power plant. They mainly contain amorphous phases, calcium hydroxide and carbonate, potassium nitrate, magnesium/silica oxides and calcium phosphate hydroxide.

Overall the FMA and BBA have a much higher organic matter and carbonate content than the clays. The additives have a higher content of water soluble ions (based on a water extract with a 2:1 liquid to solid ratio) than the clays. Dissolved sulphate and calcium dominate the FMA extracts, while BBA releases relatively high concentrations of water soluble potassium and hydroxides, followed by chloride, sodium and sulphate. The electrical conductivity slightly increases from clays (0.16 mS/cm) and FMA (0.14 mS/cm) to BBA (0.92 mS/cm), while the pH increased significantly from 7.0 to 9.0 to 12.5, respectively (solid:water 1:10). However, when in contact with the landfill leachate, the pH decreased (7.7, 8.0 and 10.2, respectively) due to the leachate buffering capacity. At optimum compaction, when the dry density is a maximum and thus the risk of settlement is minimal, the BBA holds more water than the clays or FMA, but the water void saturation is the lowest. This results in the highest air content (11 vol% of total) and lowest bulk density ( $< 2 \text{ g/cm}^3$  20°C) of all materials. This reflects the more friable, highly porous nature of the BBA, with a high water absorption potential and external SSA. Overall, the additives consist of larger particles than the clays: 33 and 43 wt% of FMA and BBA particles are  $< 500 \mu\text{m}$  in diameter, respectively, whereas 90 vol% of the clays are particles of  $< 60\text{-}760 \mu\text{m}$  diameter.

### **3.3 Municipal solid waste landfill leachate**

A methanogenic leachate and 50% CO<sub>2</sub> and 50% N<sub>2</sub> compressed gas mixture (BOC Ltd in Guildford, UK) are used for the permeation experiments. This gas mimics the CO<sub>2</sub> content of methanogenic landfill gas. It is necessary to both preserve the anaerobic status of the leachate and minimise changes in the speciation of bicarbonates/carbonates in the leachate from degassing of CO<sub>2</sub> that may affect the inorganic alkalinity and solubility of other species (e.g. Ca, Mg, metals). The landfill leachate was sampled from two cells at a domestic waste site in Manchester (UK). Both samples are methanogenic leachate and thus represent the phase of waste stabilisation that dominates for most of a landfill site operation. The leachate "South" was selected for its higher ionic strength (Table 1).

The leachate is an alkaline potassium-sodium-chloride-bicarbonate liquid with relatively high ammonium content (ca. 1 g/l) and recalcitrant organic matter and low metal content, as is typical of mature landfills of municipal solid waste (Kjeldsen et al., 2002; Regadio et al. 2012, Table 6). Therefore, the leachate was spiked with three heavy metals with different sources in domestic waste and geochemical properties to study their attenuation by the liner mixtures: zinc (Zn) with batteries, fluorescent lamps, food waste and burnt tyres as a typical source, chromium (Cr), typically originating from steel waste deposits and nickle (Ni) typically found in disposed batteries. A multi-metal solution was made from chloride salts of these metals (2 g/L Zn and Cr, 1 g/L Ni, and pH 2.5) and added to the leachate, resulting in equilibrium concentrations of  $\approx 1$ , 7 and 11 mg/L of Zn, Cr and Ni, respectively. These are higher than the initial concentrations in the leachate (Table 1), but lower than the theoretical stoichiometric calculation (30, 30 and 15 mg/L), due to metal precipitation in the alkaline leachate. Rainwater collected in Sheffield (pH 7.6) was used to permeate liner mixtures previously permeated with landfill leachate. This was done to study the potential for contaminant emissions in the event of water reaching the liner.

Table 1. Composition of selected parameters in landfill leachate for the permeation experiments (mg/L except pH and EC).

	South site	North site
pH	7.56	7.66
EC, mS/cm	20.95	16.57
TOC	871	n.m.
TIC	112	n.m.
K <sup>+</sup>	3323 <sup>a</sup>	n.m.
Na <sup>+</sup>	3111 <sup>a</sup>	n.m.
Cl <sup>-</sup>	3262 <sup>a</sup>	n.m.
NH <sub>4</sub> <sup>+</sup>	1054 <sup>a</sup>	n.m.
SO <sub>4</sub> <sup>2-</sup>	449 <sup>a</sup>	n.m.
Ca	47.24 <sup>a</sup> , 98.86	68.51
Mg	44.46 <sup>a</sup> , 70.16	55.35
B	10.595	7.025
Fe	5.910	5.117
Ba	1.246	0.882
Sr	0.972	0.671
Rb	0.614	0.402
Mn	0.582	0.406
Cr	0.480	0.284
Al	0.320	0.196
Li	0.312	0.188
Ni	0.189	0.130
Se	0.170	0.116
As	0.175	0.106
V	0.098	0.064
Zn	0.053	0.030
Co	0.042	0.028

Elements measured by inductively coupled plasma-mass-spectrometry after acidification 1/100 HNO<sub>3</sub>/sample, except ionic species<sup>a</sup> which were measured by ion chromatography. TOC: total organic carbon, TIC: total inorganic carbon, EC: electrical conductivity (neat), n.m: not measured.

## 4. EXPERIMENTAL APPARATUS

### 4.1 Design and operation of centrifuge facility

The permeation tests are performed in a geotechnical centrifuge, with a radius of 2 m and a maximum payload capacity of 0.5 ton accelerated up to 100 *g* (50 *g*-ton) (Black et al., 2014). This is a single basket beam with an 0.8 m wide by 0.8 m long platform. A sealed stainless steel fluid reservoir (for leachate or rainwater) and 12 specimen test cells with all cabling are clamped to a test platform attached to the swing basket (0.8 x 0.8 m) of the centrifuge (Figure 1). The test fluids are distributed to specimen cells using a manifold and a pressurised gas system (50%CO<sub>2</sub>/50% N<sub>2</sub> for leachate or compressed air for rainwater). The headspace overlying the liner mixture in each cell was filled with these fluids under a pressure of 1.1 bar to ensure continuous permeation of the specimens during a centrifuge run (Figure 1). Every 1 to 5 days, the centrifuge is slowed to vertically introduce a paper-wrapped metal rod through the air outlet hole of the reservoir. The wet mark in the paper serves to determine the fluid head, to estimate the hydraulic conductivity averaged over the 12 cells during the effective spinning time of the centrifuge run.

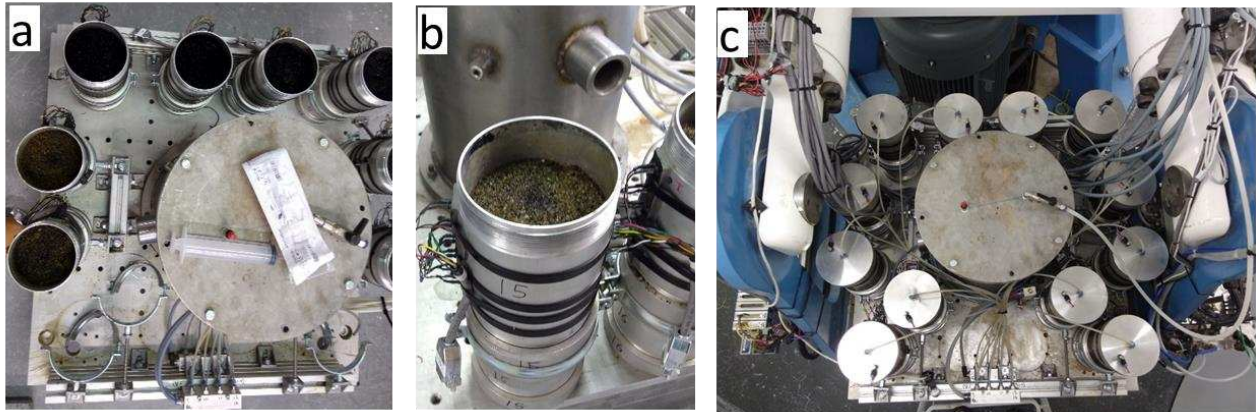


Figure 1. a) Reservoir and permeation cells clipped to the test platform to be attached to the swing basket, b) Headspace overlying the liner mixture inside the cell, that will be filled with the fluid in the reservoir, c) Assembled permeation cells (12) with fluid and gas cabling and reservoir in the centre.

#### 4.2 Design and construction of centrifuge test cells

Each centrifuge test cell comprised a 26 cm long cylinder, able to accommodate a 5 cm radius by 10 cm height compacted test specimen (Figure 2). The cells (2.2 kg) have five threaded parts made from standard U100 sample tube aluminium T6-6082 grade: 1) an upper cap with connections to a fluid (leachate or rainwater) inlet and gas valve, 2) a main central tube with 6 voltage sensors spaced at intervals of 2 cm along its length except for the top two which are 3 cm apart, 3) a base cap with 5 drainage holes, 4) a catchment tube with an air outlet hole at 6.8 cm height, and 5) a lower cap. Pipe clips M8-M10 x 108-116 mm are used to attach the cells to the test platform.

The voltage sensors installed along the aluminium tube are used to record the variation in electrical conductivity (EC) of the porewater from the output readings in voltage simultaneously at all depths, at minute intervals during a permeation experiment. This enables real time profiling of leachate migration in the specimen, based on the contrast in EC between the leachate (high) and ambient pore fluid in the specimen (lower) before the start of the leachate permeation.

Layers of coarse sand (1.18 – 2.36 mm particle diameter), 1 cm and 2.5 cm in thickness are placed on the top and bottom of the specimen, respectively, to homogenise fluid flow across the specimen and as a support (Figure 2). A 4.5 cm radius disc and two 5.25 cm radius discs made from 316 stainless steel mesh (100  $\mu\text{m}$  size) are used in each cell to cover the perforated base cap, and for framing the specimen (Figure 2). These ensure an even distribution of the sand and fluid permeation across the the top of the specimen and prevent transfer of particulate material into the fluid in the base cap.

A rubber gasket is used to prevent fluid leakage from the five threaded parts of the cell (Figure 2) and small amounts of silicone grease are used in the threads and on the base of gaskets to ensure a seal. Silicone sealant is applied to the outside of the junction of each component for final closure. The cells are deconstructed for sampling the specimen after a centrifuge run using a hinged pipe vice clamp and an adjustable rubber strap wrench to loosen the threads of each component.



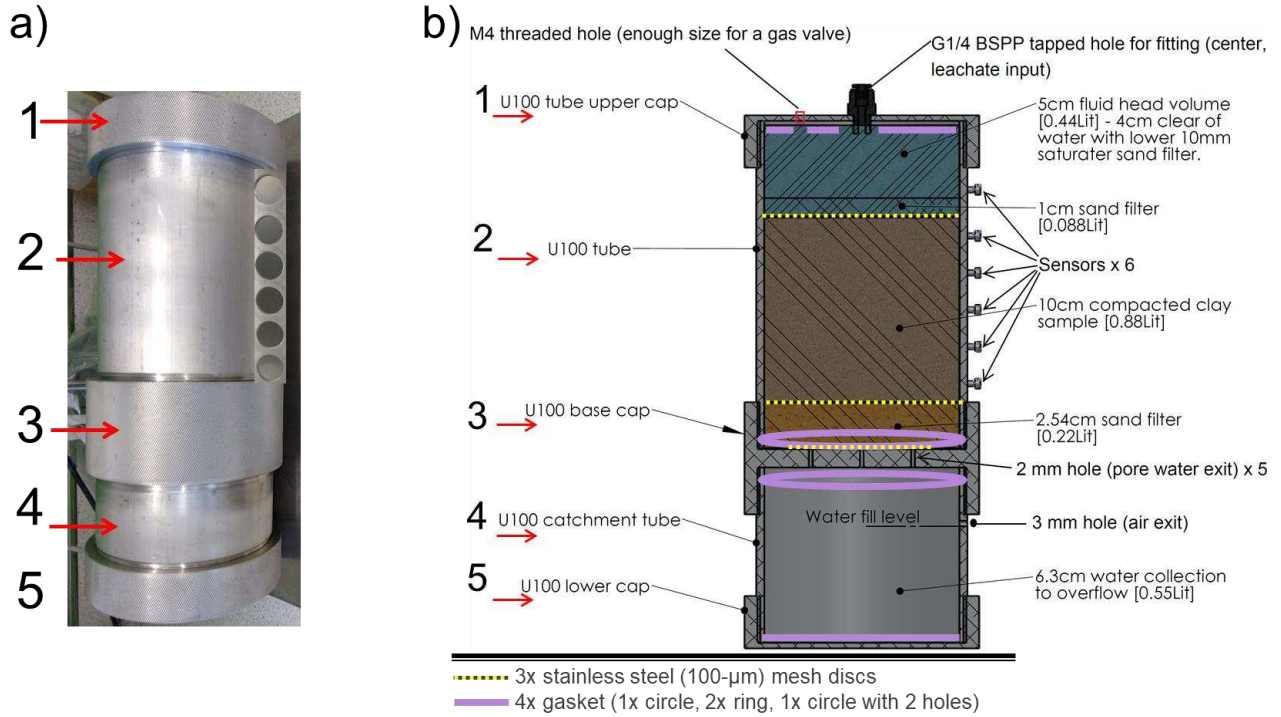


Figure 2. a) Permeation cell sections 1 – 5 (cross section). b) Manufacturing design of a permeation cell (cross section, SolidWorks software).

#### 4.3 Preparation of liner mixtures for centrifuge experiments

Different blends of FMA/clay and BBA/clay with optimized content of tap water are compacted in the specimen test cells according to the British Proctor BS 1377:4:3.3 (1990) to achieve maximum (bulk) dry density corresponding to that of field-scale clay liners, with minimum susceptibility to settle. To apply the same compaction pressure to the new final volume of the test specimens, the corresponding number of blows ( $B$ ) in each of three compaction layers was adjusted according to Eq. 1:

$$B = \frac{P V}{m_p d_p L} \quad (1)$$

where  $P$  is the pressure applied during the Proctor compaction laboratory test (6.075, Kg/cm<sup>2</sup>, Eq. 2),  $V$  is the desired volume of the compacted specimen (785 cm<sup>3</sup>, Eq. 3),  $m_p$  is the mass of the 25-mm radius rammer (2.5 kg),  $d_p$  is the drop height of the rammer (30 cm) and  $L$  is the number of layers to compact (three).

$$P = \frac{m_p d_p B_p L}{V_p} \quad (2)$$

where  $B_p$  is the number of blows per layer during the Proctor compaction laboratory test (27) and  $V_p$  is the volume of the Proctor mould (1000 cm<sup>3</sup>, Eq. 3).

$$V = \pi r^2 h \quad \text{where } r \text{ is the radius (cm) and } h \text{ the height (cm)} \quad (3)$$

All specimen liner mixtures prepared in this way were temporarily stored at 4 °C in sealed polythene bags prior to a centrifuge run, to prevent changes in moisture content and dessication.

## 5 EXPERIMENTAL DESIGN

### 5.1 Preparation of solid materials

The clay samples consisted of large irregular blocks while the recycled additives consisted of loose granular materials (Figure 3). Several small subsamples of the clays were used for the analysis of pore water soluble ions (undried material) and CEC (air-dried, powdered and 3 x 10 sec ultrasonificated material). For the remaining tests the clays were reduced to <20 mm aggregate lumps after oven drying, using a rammer (Wickes) and several perforated screen trays, and then to <2 mm using a bench soil grinder (Humboldt Co attached to a larger hooper with a 20-mm mesh), in which lumps between 2 and 20 mm were broken until passing a 2-mm mesh plate. To minimize dust inhalation, a headtop connected to a powered air turbo unit; was worn and a fume extraction unit with a hood at the end of adjustable arms (Nederman Ltd) was used. The recycled additives were sieved to <4 mm and the larger fraction discarded. Samples of clays and additives were crushed to a powder for elemental chemical analyses, while samples for BET surface analysis were gently ground (at least 10 cycles) to <0.4 mm and the 0.064-mm fraction discarded. The material passing the 0.425-mm sieve was used for consistency tests.

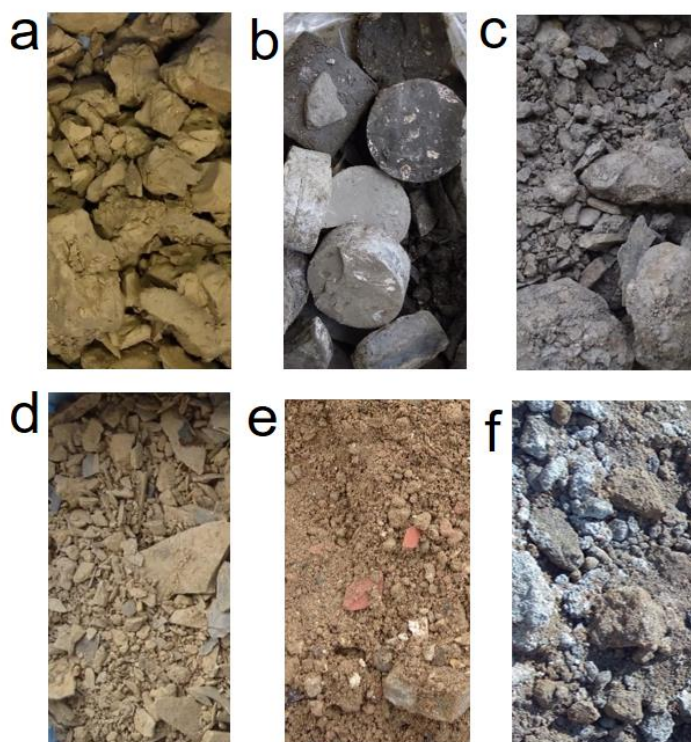


Figure 3. Appearance of a) London clay, b) Oxford clay, c) Coal Measures Clay (shallow) d) Coal Measures Clay (deep), e) Fine mixed aggregates, f) Biomass bottom ash.

The recycled additives were tested for compliance with relevant standards to identify if they present a risk to the environment or human health. Leaching tests with water percolating through the materials when *packed* (but not compacted) in columns (NBN EN CEN/TS 14405:2004) and water mixing with the *loose* materials in containers on a rotating agitator (UNE–EN 12457:2003) were completed for this. The analysis of filtered water after contacting the materials during these tests compared the release of 12 metals (chromium, nickel, copper, zinc, arsenic, selenium, molybdenum, cadmium, antimony, barium, mercury and lead) with regulation threshold values obliged by the Council Decision (2003). This showed that the FMA and BBA residues were classified as inert and non-hazardous.

## 5.2 Centrifuge permeation experiments

The time and speed of each centrifuge test is set to model a given landfill scenario (e.g. 30 years of operation and a leachate head of upto 2 m over a 1 m thick clay compacted to a hydraulic conductivity of  $10^{-9}$ cm). In practice this can be achieved with a centrifuge (model) run time of days and test specimen of a few cm in size (up to a maximum of 10 cm). Leachate is first permeated through the specimens, followed by rainwater (if appropriate) after changeover of the fluid and pressurised gas composition in the fluid reservoir. Using the centrifuge the gravity force ( $n$ ) on the model is increased, to recreate (through self-weight) stresses and strains in the laboratory which would exist at the field-scale (prototype) (Figure 4). The relationship between the gravity force and speed (revolutions per minute) of the centrifuge is given by Eq. 4. By knowing  $n$ , the results from the centrifuge (model) can be linked to results from a full-scale real case (prototype) using the scaling laws (Ng, 2014). The solved and unsolved scaling-related questions are kept up to date in the catalogue of Garnier et al. (2007).

$$n = 1.12 r \left( \frac{rpm}{1000} \right)^2 \quad (4)$$

where  $n$  is the gravity force (also termed relative centrifugal force, RCF),  $r$  is the radius of the centrifuge beam arm (mm) and rpm is the revolutions per minute (rpm) of the centrifuge rotor.

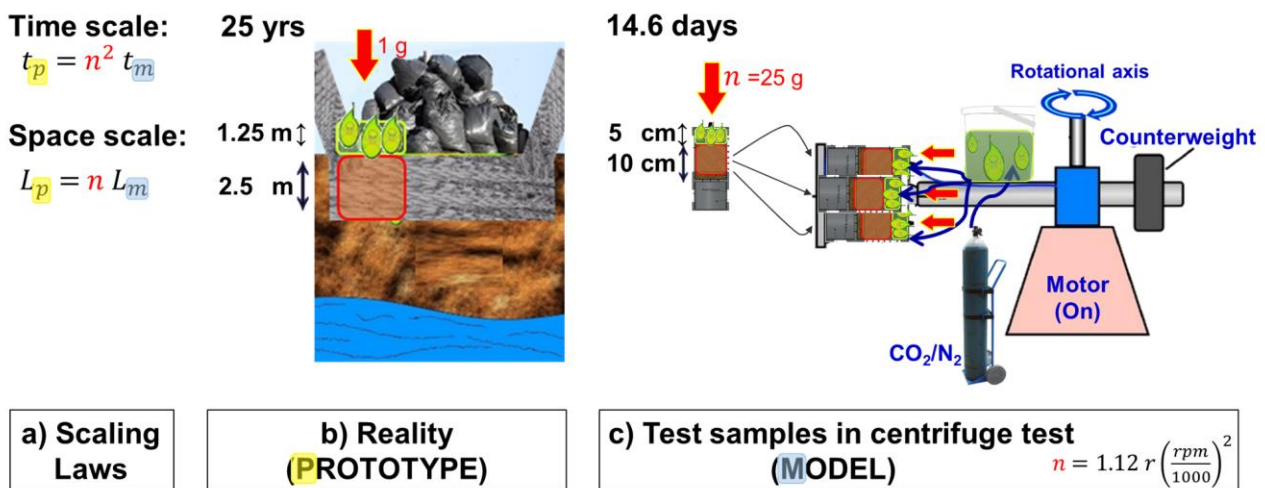


Figure 4. Centrifuge model for simulating the transport of leachate (green shading) through tested barriers (red shading). a) Space and time scales to calculate dimensions, periods of time, hydraulic conductivity, advective, diffusion and dispersion in the prototype from the model data. Cross sections of: b) a landfill and c) the experimental devices. Order of magnitudes of meters and years in the prototype ( $L_p$  and  $T_p$ ) correspond to days and centimetres in the model ( $L_m$  and  $T_m$ ) at a higher gravity force ( $n$ ).  $r$  is the radius of the beam arm (mm) and rpm the revolutions per minute of rotor.

The gravity selected should be chosen as a compromise between the experiment duration and sample size. Increasing the gravity conveniently decreases the experimental time but also the physical size of the specimen. For example, modelling a 100 years of transport of leachate pollution through a compacted clay liner would require a run time of only 7 days at 75  $g$ . After 100 years contaminants would move between 0.1-3 m in the fieldscale prototype, which corresponds to a very restricted model distance between 0.1-4.0 cm. However, at 25  $g$ , 0.1-3 m in the prototype would be equivalent to 0.4-12 cm in the model, but with a much longer run time of 58 days to achieve an equivalent period of 100 years. Lower gravities increase the waiting time to obtain samples, but allow increased sampling resolution (more

samples per specimen). In addition, there is a risk of introducing an artefact when working at extremely high gravities of artificially increasing the compaction of the specimens. Duplicate experiments are run in each centrifuge test to assess method precision and provide spare specimens for comparison of leachate and rainwater permeation phases.

This method is ideal for scoping studies over large areas and long periods of time. In our case, working at 25 g for a prototype of 33 years enables reliable results to be obtained in acceptable time frames (19 days centrifuge modelling). The centrifuge modelling provides results that are reproducible and comparable (Figure 5), and consistent with theoretical predictions (section 5.3) and field measurements (Regadío 2012). Nevertheless, care should be taken as some processes (e.g. biological and geochemical), that also affect the contaminant transport, are difficult to model.

At the end of the centrifugation, test samples from landfill leachate (obtained after loosening section 1 of the cell), from effluent leaving the cell (collected in section 4 of the cell) and from specimens (extruded from the double-open-ended section 2 of the cell) are recovered (Figure 2.a). The specimens are extruded from the test cell with a hydraulic pump and sliced to provide 10 samples of  $\pm 1$  cm thickness from which moisture contents are determined and the pore water is obtained. Pore water from each specimen slice is sampled by equilibration with deionized water at a liquid:solid ratio of 2.5 using the modified method described in BS 1377:3 (1990) (1 min vortex agitation, 3 cycles of 10 sec of ultrasonification, at least 8 h settling and reagitation for pH analysis). This is followed by analyses of electrical conductivity ( $EC_{2.5}$ ), major ions and metals using standard methods, after (bench) centrifugation and 0.45- $\mu\text{m}$  filtration. The detailed solute distribution obtained from the pore water chemical analyses enables leachate transport and contaminant attenuation process to be interpreted over extended operational time-scales.

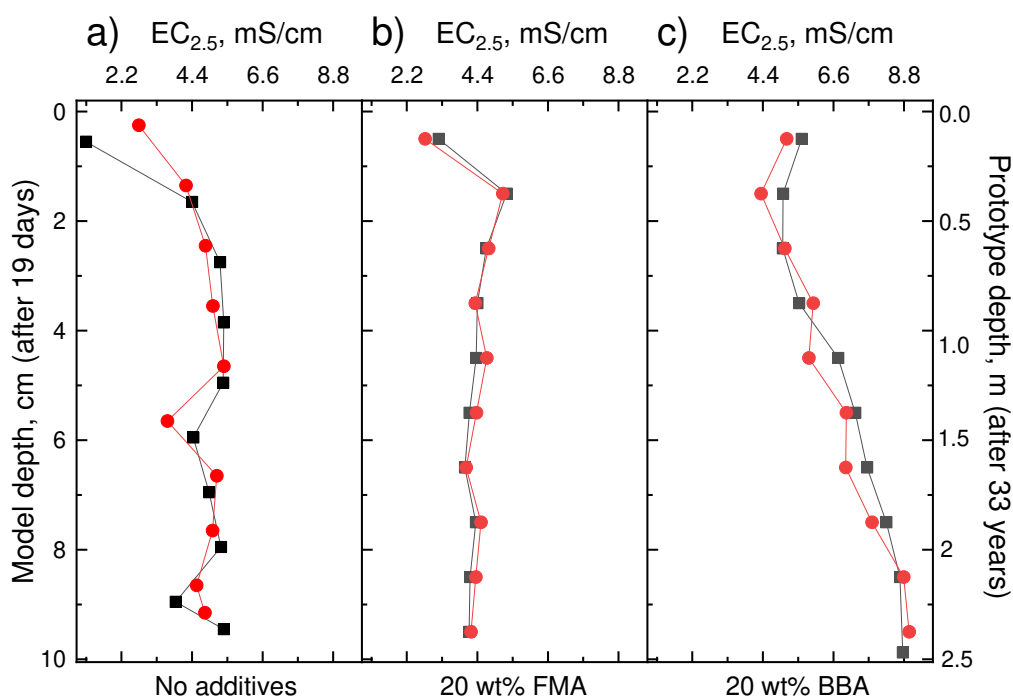


Figure 5. Depth profiles of electrical conductivities ( $EC_{2.5}$ ) through duplicate samples of Oxford clay without additives (a) and with 20 wt% fine mixed aggregates, FMA (b) or biomass bottom ash, BBA (c). Samples were centrifuged at 25 g with the landfill leachate and 50% of  $\text{CO}_2$ . Left Y-axis represents sample depth and right Y-axis its corresponding real depth.



### 5.3 Numerical modelling

The experimental approach using the permeation tests and centrifuge procedure yields solute-depth profiles in the liner mixtures for equivalent timescales of decades of operation at a landfill site. These results will be analysed using the geochemical model PHREEQCi (Parkhurst and Appelo, 2013), to (1) interpret the geochemical processes which control contaminant transport and attenuation in the liners during leachate and rainwater flushing phases, (2) derive parameter values and quantitative descriptions between leachate chemistry and liner material properties to predict contaminant attenuation, and (3) understand how the outputs can be scaled to develop full-scale liner designs. The numerical model will include diffusion and kinetic expressions that describe non-linear processes. This activity will also explore liner failure scenarios, to predict potential impacts on groundwater quality and environmental risks from contaminants released from the liner. The results will be reported in a separate paper.

## 6. CONCLUSION AND FORWARD LOOK

The HARM project will enable improved, sustainable landfill liner designs to be developed that reduce costs, mitigate impacts and ensure long-term environmental protection. This is critical given that, firstly, landfill restoration and aftercare typically costs  $\approx 838\,000$  €/yr (Hogg, 2002), and, secondly, restoring potable water polluted by leachate is almost impossible. It often means water supply wells and aquifers must be abandoned as lost assets (US EPA, 1991). This creates acute socio-economic and environmental problems where groundwater resources are heavily used and under threat, as in the Mediterranean countries. To the best of our knowledge, no adequate measures exist today to address such impacts and so the project would provide valuable technical innovation and guidance in this regard.

The performance of FMA and BBA in clayey liners for *in situ* treatment of leachate pollutants is unknown, with no quantitative basis for use in landfills. Mixing clays with these recycled additives may stabilize the consistency of London and Oxford Clay-based landfill liners and improve the attenuation of Coal Measures Clay liners. In addition, it would result in the recycling of industrial residues, that would otherwise end up in large stockpiles landfilled. This unique alternative re-use of waste products for environmental security, decreases: (1) waste disposal, (2) construction costs of landfill liners, and (3) consumption of natural resources; it improves: (1) the long-term performance of reactive natural liners, and (2) provides equivalent (i.e. time-scaled) long-term data for the engineering design of such liners. The findings will be informative to policy makers and the society in integrating natural attenuating within current landfill liner design to mitigate pollutant impacts from leachate. The attenuating strategy reduces the risk of leachate pollution and provides cheaper protective measures, an important consideration for low-income countries where  $>90\%$  of waste is openly dumped (Kaza et al., 2018).

The combined experimental approach (permeation by centrifugation of compacted columns at representative time and space scales) and numerical modelling (performance interpretation-design evaluation) provides new and valuable insight in related fields, such as geotechnical barrier design or reactive transport code development, which would otherwise not be possible. This is because contaminant migration in low permeability porous media can be studied with accelerated time-scaling for fluid flow over extended spatial and time scales. This is a unique advantage over conventional permeameter tests, because the test time and physical size of the specimen is significantly reduced for the corresponding conditions represented.



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