Altered chemical evolution in landfill leachate post implementation of biodegradable waste diversion

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Abstract:

Within the UK implementation of the EU Landfill Directive (1999) has led to the diversion of Biodegradable Waste (BW) from municipal solid wastes away from landfills. It has been widely anticipated, but thus far not verified, that the diversion of BW and consequent reduction in BW reaching landfill would lead to a change in the degradation processes occurring within landfills and that this would be reflected in an altered evolution in leachate chemistry compared to pre-Directive landfills. This paper provides evidence based on leachate chemistry from two operational landfills together with calculations of the reduced BW content, that demonstrate the acetogenic phase that characterised pre-directive landfill leachates is missing and is now more typical of methanogenic phase leachate. The paper demonstrates how data from national datasets and detailed landfill records can be used to constrain likely, and upper estimates of the amount of BW going into post-directive landfills and the observed change in the evolution of leachate chemistry which has resulted from a decrease in BW content from typical values of BW (pre-landfill directive) of 22% to an inferred 12% in the case-study landfills. Data provided here adds to the growing literature that estimates the amount of BW in recent post-directive landfills which importantly allow the quantitative linkage between a decrease in landfilled BW and observed changes in leachate chemistry to be established such that future landfill operators can increase confidence in the effect of Directive implementation on landfill operational parameters.

Keywords

Municipal solid waste, leachate, acetogenesis, methanogenesis, recycling, biodegradable waste.
1. Introduction

The European Union (EU) has legislated to reduce and control the volume of waste disposed into landfill. Thus, both landfilling practices and the materials disposed into landfills have been regulated and controlled to accommodate developing objectives in the reduction of polluting landfill leachates and greenhouse gas emissions (Brennan et al, 2016). The Landfill Directive required member states to separate biodegradable wastes (BW) and develop recycling (European Commission (EC), 1999). Initially BW included any waste capable of degradation either aerobically or anaerobically. Latterly, BW is defined by the EU as garden and park waste, food and kitchen waste from households, restaurants and caterers, retail premises and food processing plants (EC, 2008) and forms a rapidly degrading component of municipal solid waste (MSW). In addition to BW, MSW contains fractions where the projected biodegradation time, when occurring within landfills, extends to many centuries until complete stabilisation is achieved (Belevi & Baccini, 1989). These wastes are hereafter described as biodegradable municipal wastes (BMW). The Landfill Directive obliged member states to progressively reduce BW from 75% of their respective 1995 levels to 35% by 2016 (European Commission (EC), 1999). The current EU target for all recycling is 50% of MSW by 2020 (DEFRA, 2016). MSW generation in the United Kingdom has increased over the period 1995 – 2015 from 28,900,000 to 31,567,000 tonnes per annum (EUROSTAT, 2017). However, the annual quantity landfilled has reduced from 23,990,000 to 7,124,000 tonnes (EUROSTAT, 2017).

As a result of UK waste policy drivers which include landfill taxation, waste recycling and recovery, significant compositional changes to those wastes entering landfills
has ensued (Frank et al, 2016; Brennan et al, 2016; Fischer et al, 2012).

Furthermore, the UK Environment Agency (EA) (P1-494/SR1 2004) proposes that, with the implementation of the Directive’s standards and particularly the removal of the BW fraction from the landfill mass, then established degradation processes occurring in landfills would be different. To this point, the authors believe no data have been published that confirm or refute this, particularly where well constrained studies integrate reasonable estimates of post-directive BW that are compared to developing leachate chemistry. Recent leachate composition research continues to identify the complex but nonetheless established evolution arising during waste degradation and, for the most part, proposes innovations in leachate treatment (Adhikari et al, 2014; Hubert et al, 2016; Moody & Townsend, 2017; Naveen et al, 2017).

In this paper evidence is presented where leachate samples, taken over the first five years of landfill operation, confirm this evolution in leachate chemistry and attribute it to the diversion of BW away from post-directive landfills.

In order to address this question specific objectives of this paper are as follows: (i) to present data on the evolution of landfill leachate chemistry (composite leachate and well point) in two well characterised case-study landfill sites and compare these to characteristics of “conventional” landfills using two further pre-Directive landfills and data ranges from Ehrig (1983). (ii) To use national datasets and waste import data from specific case-study landfills to constrain an upper and best-estimate of the reduction in BW.

To establish the context for the study, a brief review of the evolution of chemistry within pre-Directive landfills and the UK perspective on diversion of wastes from landfill is outlined below.
2. Background

2.1 Critical landfill processes

Materials consigned to landfills undergo a series of physical, chemical and biological processes (Christensen & Kjeldsen, 1989). Landfill leachate occurs when the field capacity of the waste mass is exceeded due to continued water infiltration. Landfilling methods generate a brief aerobic period where oxygen becomes trapped within the waste matrix. With the onset of the biodegradation of the putrescible content, the rapid consumption of oxygen by bacteria leads to anaerobic conditions which develop into a sequence of consecutive phases: (1) the acetogenic phase, (2) an initial methanogenic phase, (3) a stable methanogenic phase (Christensen & Kjeldsen, 1995). Christensen and Kjeldsen (1995) and Bozkurt et al. (2000) have also reported an additional final aerobic phase. Each phase is identifiable by a distinct set of chemical and biological processes which influence the chemistry of the leachate. During the acetogenic phase, fermentative and acetogenic bacteria generate carboxylic acids, carbon dioxide, hydrogen and alcohols from proteins, carbohydrates and lipids occurring mainly in putrescible wastes (Rees, 1980, Christensen & Kjeldsen, 1989). For the acetogenic phase and following initial disposal into landfill, BOD and COD levels are greatest (Doedens & Cord-Landwehr, 1989) and the acidity of the leachate can reduce pH to 4.5 (Ehrig, 1983). In UK climatic conditions, transition from the acetogenesis phase to a stable methanogenic phase may not have occurred until after three years (Robinson, 2005). This phase is followed by the methanogenic phase where solid phase organic materials are microbially degraded producing methane as a significant product. Leachate pH typically increases above neutral to an average value of 8 (Ehrig, 1983).
2.2 Biochemical indicators for acetogenesis and methanogenesis

Where landfills receive domestic and commercial MSW, the landfilling practices employed and the chemical, physical and biological processes that result have led to leachates comprising four groups of components: heavy metals, inorganic macrocomponents, dissolved organic matter and xenobiotic organic compounds (Christensen et al., 1994). Established key process indicators in landfill leachate are: pH, $\text{BOD}_5$/COD (ratio) and sampled concentrations in mg/litre for sulphates, calcium, magnesium, manganese, iron and zinc (Ehrig, 1983). Identification of a particular phase of landfill degradation relies primarily upon comparisons made between leachate samples with concentrations lying within specific ranges which are then determined as being indicative of either the acetogenic or methanogenic phases (Ehrig, 1988; Kjeldsen et al., 2002). Their usefulness as indicators relies upon the fact that respective concentrations change significantly during the landfill decomposition process resulting from the two contrasting chemical environments occurring first during acetogenesis and second, during methanogenesis. Those components that do not change, which include the inorganic fractions of ammonium and the chloride ion content, have been excluded by Ehrig. Ehrig’s published concentrations are contained in Table 1. The sample average and sample range are provided.

<table>
<thead>
<tr>
<th>Key indicator</th>
<th>Acetogenic phase</th>
<th>Methanogenic phase</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>6.1</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>4.5 – 7.5</td>
<td>7.5 - 9</td>
</tr>
<tr>
<td>$\text{BOD}_5$</td>
<td>13000</td>
<td>180</td>
</tr>
<tr>
<td></td>
<td>4000 – 40000</td>
<td>20 - 550</td>
</tr>
</tbody>
</table>

Table 1
Sample concentrations for ten key indicators in the acetogenic and methanogenic phases after Ehrig (1988). All data values in mg/l except for pH and unitless ratio $\text{BOD}_5$/COD.
<p>| | | | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>COD</td>
<td>22000</td>
<td>6000 – 60000</td>
<td>3000</td>
<td>500 - 4500</td>
</tr>
<tr>
<td>BOD₅/COD</td>
<td>0.58</td>
<td>-</td>
<td>0.06</td>
<td>-</td>
</tr>
<tr>
<td>Sulphates</td>
<td>500</td>
<td>70 – 1750</td>
<td>80</td>
<td>10 - 420</td>
</tr>
<tr>
<td>Calcium</td>
<td>1200</td>
<td>10 – 2500</td>
<td>60</td>
<td>20 – 600</td>
</tr>
<tr>
<td>Magnesium</td>
<td>470</td>
<td>50 – 1150</td>
<td>180</td>
<td>40 – 350</td>
</tr>
<tr>
<td>Iron</td>
<td>780</td>
<td>20 – 2100</td>
<td>15</td>
<td>3 – 280</td>
</tr>
<tr>
<td>Manganese</td>
<td>25</td>
<td>0.3 – 65</td>
<td>0.7</td>
<td>0.03 – 45</td>
</tr>
<tr>
<td>Zinc</td>
<td>5</td>
<td>0.1 – 120</td>
<td>0.6</td>
<td>0.03 - 4</td>
</tr>
</tbody>
</table>

2.3 Disposal dynamics since the Landfill Directive – an overview

For the UK, the Landfill Directive 31/1999 represented a major shift in waste disposal strategy where landfilling now resides at the base (least favoured) of the waste management hierarchy. Implementation at the local level required that the many fractions from municipal solid waste (MSW) streams should now be regarded as recyclable and required diversion away from landfills. MSW can be categorized as waste collected at the kerbside and includes household and similar wastes generated by commercial, educational and governmental organisations (Burnley 2006). Reported statistics issued by the Department for Environment, Food and Rural Affairs (DEFRA) identify MSW as comprising food and garden waste, paper and packaging, wood, metal, WEEE, furniture and sanitary (solid) waste (DEFRA, 2009). Many of these wastes are classified as biodegradable however, respective degradation periods are substantially different with some having significant durations when compared to EU defined BW which occurs within a one to three-year period.

Data published by EUROSTAT reflects the United Kingdom’s (UK) obligation under the Landfill Directive to redirect wastes away from landfill noticeably the diversion of BW containing wastes that have led to food and garden wastes becoming a
feedstock for energy production via digestion and composting to the point where weights of materials recycled now exceed those landfilled. Figure 1 compares MSW generation and evolving disposal strategies over the period 1995 - 2015.

![Figure 1. Comparison of changing UK MSW disposal methods 1995 – 2015. Data from Eurostat.](image)

3. Methodology and data sources

3.1 Data sources and site description

Leachate data has been provided by the UK waste management company, Viridor Waste Management Ltd [Viridor]. It should be noted that Viridor provided significant data which is currently being used for a wider study and the site and data labelling reflects that used in the wider study. All data has been taken from engineered landfill sites where waste disposal operations commenced in 2005 (identified as Site C) and 2007 (identified as Site B). Viridor provided a range of data which includes individual sampling point or well-point data (B11, B12, C2 and C3), composite leachate data together with deposited waste records. These data are compared to Ehrig’s key indicators together with data from two homologous and established engineered
landfill sites, Sites E and Site F. Samples from these sites are composite leachates which firmly reflect the methanogenic phase of degradation when compared to Ehrig’s values.

Table 2 summarises the operational commencement dates, primary waste source, the sample range and current site status for the case-study landfills. The four sites are located in the South West Region of England. Herein the South West region comprises the UK counties of Cornwall, Devon, Dorset, Gloucestershire, Somerset and Wiltshire.

Table 2
Operational commencement, waste source and date range for the four Viridor sites.

<table>
<thead>
<tr>
<th>Site</th>
<th>Operation commenced</th>
<th>Waste source</th>
<th>Sample range</th>
<th>Status</th>
</tr>
</thead>
<tbody>
<tr>
<td>Site B</td>
<td>2007</td>
<td>urban</td>
<td>2007–2015</td>
<td>open</td>
</tr>
<tr>
<td>Site C</td>
<td>2005</td>
<td>rural</td>
<td>2006–2015</td>
<td>open</td>
</tr>
<tr>
<td>Site E</td>
<td>1988</td>
<td>urban</td>
<td>1995–2015</td>
<td>closed</td>
</tr>
<tr>
<td>Site F</td>
<td>1994</td>
<td>rural</td>
<td>2004–2015</td>
<td>closed</td>
</tr>
</tbody>
</table>

1. Waste deposition commenced 2006 and was only 10,157 tonnes

Sites B and C have been chosen specifically because old waste was not present at either site. This removes the possibility of new leachates passing through old waste layers. Where underlying waste layers are in the methanogenic phase, leachates generated from overlying new wastes, that pass through these methanogenic wastes, will reflect the characteristics of the older (methanogenic) leachates (Kjeldsen et al., 1998; Assmuth, 1992).

Wastes imported into Sites B, C, E and F are summarized in Table 3. Imported waste data for all sites commences from 2005. Viridor categorise imported wastes
using the *List of Waste* codes identified in *Technical Guidance WM3 (EA 2015)* having (first) separated waste imports into those high-level categories identified within the table. Waste imports are recorded in tonnes.

Table 3
1. Summary of waste sources for the four Viridor sites.

<table>
<thead>
<tr>
<th></th>
<th>Site B</th>
<th>Site C</th>
<th>Site E</th>
<th>Site F</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>tonnes ’000</td>
<td>tonnes ’000</td>
<td>tonnes ’000</td>
<td>tonnes ’000</td>
</tr>
<tr>
<td>Total waste imported</td>
<td>2,077.10</td>
<td>1,264.00</td>
<td>1,136.20</td>
<td>58.2</td>
</tr>
<tr>
<td>Cover materials</td>
<td>682.9</td>
<td>142.7</td>
<td>317.2</td>
<td>11.9</td>
</tr>
<tr>
<td></td>
<td>(32.9%)</td>
<td>(11.3%)</td>
<td>(27.9%)</td>
<td>(20.4%)</td>
</tr>
<tr>
<td>MSW - domestic &amp; non-domestic</td>
<td>1012.0</td>
<td>983.1</td>
<td>450.8</td>
<td>35.8</td>
</tr>
<tr>
<td></td>
<td>(48.7%)</td>
<td>(77.8%)</td>
<td>(39.7%)</td>
<td>(61.6%)</td>
</tr>
<tr>
<td>Difficult wastes</td>
<td>1.4</td>
<td>6.2</td>
<td>1.6</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>(0.1%)</td>
<td>(0.5%)</td>
<td>(0.1%)</td>
<td>-</td>
</tr>
<tr>
<td>Sludges</td>
<td>46</td>
<td>0.97</td>
<td>1.011</td>
<td>9.6</td>
</tr>
<tr>
<td></td>
<td>(2.2%)</td>
<td>(-)</td>
<td>(0.1%)</td>
<td>(16.4%)</td>
</tr>
<tr>
<td>Non-hazardous waste</td>
<td>16.2</td>
<td>5.8</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>(7.7%)</td>
<td>(0.5%)</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Other waste</td>
<td>174.6</td>
<td>126.1</td>
<td>365.6</td>
<td>0.9</td>
</tr>
<tr>
<td></td>
<td>(8.4%)</td>
<td>(10.0%)</td>
<td>(32.2%)</td>
<td>(1.5%)</td>
</tr>
</tbody>
</table>

Waste imported into each of the study sites can be classified as: *cover materials*, for the most part soils and transfer station fines; *MSW* comprising mixed domestic and non-domestic wastes, transfer station wastes and non-special clinical wastes; *difficult wastes*, which include sewage works screenings, printer toner waste and green wastes; *sludges* comprising mainly septic tank and sewage sludge residues; and *non-hazardous wastes* comprising contaminated (non-hazardous) soils. Site B
was also used to dispose approximately forty-six thousand tonnes of asbestos-containing materials which for the most part was contained within construction wastes.

3.2 Sampling and data

The leachate data provided forms part of the company’s leachate sampling and management systems as regulated by Schedule 2, Paragraph 2(1)(c) of the Landfill Regulations. Leachates were extracted from individual monitoring well-points. A list of the determinands used in this study and relevant analytical procedures is provided in Table 4. Specific site details are provided in Appendix 1.

Table 4
Analysis methods and reporting limits for leachate quality at the four Viridor sites.

<table>
<thead>
<tr>
<th>Determinant</th>
<th>Method Description</th>
<th>Reporting Limit</th>
<th>Reporting Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>WAS039 pH/ EC in Water by Electrode</td>
<td>1</td>
<td>pH units</td>
</tr>
<tr>
<td>BOD + ATU (5 day)</td>
<td>WAS001 BOD in Water</td>
<td>1</td>
<td>mg/l</td>
</tr>
<tr>
<td>COD (Total)</td>
<td>WAS040 COD in Water by Colorimetry</td>
<td>11</td>
<td>mg/l</td>
</tr>
<tr>
<td>Sulphate as SO₄</td>
<td>WAS036 Anions by Colorimetry</td>
<td>4.4</td>
<td>mg/l</td>
</tr>
<tr>
<td>Calcium, Total as Ca</td>
<td>WAS049 Metals in Water by ICP-OES</td>
<td>0.38</td>
<td>mg/l</td>
</tr>
<tr>
<td>Magnesium, Total as Mg</td>
<td>WAS049 Metals in Water by ICP-OES</td>
<td>0.6</td>
<td>mg/l</td>
</tr>
<tr>
<td>Manganese, Total as Mn</td>
<td>WAS049 Metals in Water by ICP-OES</td>
<td>0.007</td>
<td>mg/l</td>
</tr>
<tr>
<td>Iron, Total as Fe</td>
<td>WAS049 Metals in Water by ICP-OES</td>
<td>0.23</td>
<td>mg/l</td>
</tr>
<tr>
<td>Zinc, Total as Zn</td>
<td>WAS049 Metals in Water by ICP-OES</td>
<td>18</td>
<td>mg/l</td>
</tr>
</tbody>
</table>
4. Results and discussion

4.1 Analysis of leachate samples from Sites B and C

The leachate samples are presented as a series of boxplots (Figures 2 – 7). Specific chemical markers (the key indicators) pH, BOD, COD, BOD/COD together with the sulphate and calcium concentrations are compared to Ehrig’s ranges for both the acetogenic and methanogenic phases. Each boxplot represents the collated annual sampling data taken from individual well-points. MINITAB was used to prepare the boxplots and all data is presented as that supplied by Viridor with outliers determined by MINITAB. Outliers are defined as those observed values that exceed one and a half the interquartile range, with these being denoted with a star (*). For each variable, the median is indicated by the horizontal line within each interquartile range. The data range (excluding outliers) extends to the end of each whisker. the number of observations (n) is included as a footnote to each figure.

Metal concentrations for both sites were very low and are not included as boxplots. Mean values for Mg, Mn, Fe and Zn for Site B were 140 mg/l, 0.53 mg/l, 2.97 mg/l and 0.11 mg/l respectively. Mean values for Site C, in the same order, were 125 mg/l, 0.37 mg/l, 2.28 mg/l and 0.10 mg/l. Metal concentration in leachate increases with decreasing pH (Aucott M, 2006). Concentrations at these low levels seen in the data are typical of the methanogenic phase (Ehrig, 1988; Kjeldsen et al., 2002). Figure 2 compares combined pH data for both sites where the median pH exceeds 7 for all but one sample set.
Figure 2 (a & b). pH for Sites B & C from 2007 to 2011 compared to Ehrig’s data in the acetogenic and methanogenic phases. n for Site B 2007 – 2011: 13, 15, 28, 55, 53. n for Site C 2007 – 2011: 15, 18, 7, 9, 12

For these new waste deposits and the conditions prevailing in the UK, the pH of leachate samples should reflect weak acidic conditions throughout the transition of acetogenesis to the establishment of methanogenesis (Robinson, 2005). Individual
wellpoint (B11) pH data for Site B (Figure 3) where the sampled data highlights the initial period where pH values are less than neutral, 6.1, 6.4 and 6.6 for March, April and May 2007 respectively. By September 2007 and, following this initial reduced pH condition all values lie above 7 and generally reflect methanogenic values.

Figure 3 (a & b). pH for wellpoints 11 & 12, Site B from 2007 to 2011 compared to Ehrig’s data in the acetogenic and methanogenic phases. n for wellpoint B11 2007 – 2011: 3, 2, 10, 11, 10. n for wellpoint B12 2007 – 2011: 9, 9, 11, 9, 4
For the remaining key indicators, the leachate samples continue to reflect the methanogenic range identified by Ehrig (Figures 4 – 8).

**Figure 4 (a & b).** BOD₅ for Sites B & C from 2007 to 2011 compared to Ehrig’s data in the acetogenic and methanogenic phases. n for Site B 2008 – 2011: 2, 7, 18, 21. n for Site C 2007 – 2011: 5, 8, 7, 9, 12.
Figure 5 (a & b). COD for Sites B & C from 2007 to 2011 compared to Ehrig's data in the acetogenic and methanogenic phases. n for Site B 2007 – 2011: 13, 8, 7, 17, 21. n for Site C 2007 – 2011: 5, 8, 7, 9, 12.
Figure 6 (a & b). BOD/COD ratio for Sites B & C from 2007 to 2011 compared to Ehrig’s data in the acetogenic and methanogenic phases. n for Site B 2008 – 2011: 2, 7, 16, 22. n for Site C 2007 – 2011: 6, 8, 7, 9, 12.
Figure 7 (a & b). Calcium concentration for Sites B & C from 2007 to 2011 compared to Ehrig’s data in the acetogenic and methanogenic phases. n for Site B 2008 – 2011: 2, 7, 15, 17 (no data available for 2007). n for Site C 2007 – 2011: 5, 8, 7, 9, 12.
Figure 8 (a & b). Sulphates concentration for Sites B & C from 2007 to 2011 compared to Ehrig’s data in the acetogenic and methanogenic phases. n for Site B 2008 – 2011: 2, 7, 15, 17 (no data available for 2007). n for Site C 2007 – 2011: 5, 8, 7, 9, 12.
Whilst BOD$_5$ fully reflects the methanogenic range for Sites B and C some inconsistencies occur in the data sets for sulphates, COD values for Site C and BOD/COD for Site B. For the most part, COD sample values for Site C similarly reflect the methanogenic range with one exception. This exception applies to one data set only where COD is consistent for both sampled wellpoints (C2 and C3) at 1670 mg/litre, 1420 mg/litre, 1620 mg/litre and 1240 mg/litre however a single value of 11800 mg/litre for wellpoint C3 skews these lower values. BOD$_5$ for this sample is similarly high and equates to 6250 mg/litre, which has a similar effect. The BOD/COD ratios are conditional upon each parameter having been analysed from the same sample and this has reduced the number of observations. Furthermore, respective leachate samples have both low BOD$_5$ and COD concentrations and these impact on their ratio having a greater value.

Sites B and C are new operations and present a rare opportunity to examine two situations where newly generated leachates are uncontaminated through mixing with mature wastes. However, landfill leachate samples generally display the chemistry typical of mature wastes in the methanogenic phase. In addition, leachate recirculation does not occur at any of the sites examined in this study therefore any likely effect due to recirculation can be discounted.

4.2 Comparison with leachate samples from Sites E and F

Sites E and F provide direct comparison to Sites B and C in that the prevailing climatic conditions and waste types deposited share identical backgrounds. Their leachate pH values are similar (Figure 9) however, leachates generated at Sites B and C are from wastes not exceeding 4 years but those generated at Sites E and F
reflect leachates from mature wastes. BOD$_5$, and COD are similarly compared (Figure 10 and Figure 11) with the same result.


4.3 Waste diversion and its impact on MSW imported into sites B and C

The implementation of the Landfill Directive increased household recycling from 11.2% of total MSW disposal in 2000/01 to 32% in 2006/07 and then to 44% in 2014/15 (EA, 2016). Waste imports into Sites B and C were influenced by recycling strategies however recycling capture rates across England differ quite markedly. Records show South West recycling (the location of the case-study landfills) operations exceeded the national average by five percentage points during this period (EA 2016).

The UK trend in decreasing MSW to landfill (Figure 1) is replicated in sites B and C (Figure 12). MSW imports into Site B display a reducing trend after 2010. Domestic MSW deposition into Site C is reducing with non-domestic MSW decreasing and then remaining relatively constant. Whilst waste reduction is germane to the Landfill Directive, of critical importance is the redirection of BW into alternative waste
streams, away from landfill thus impacting on leachate chemistry by the removal of the acetogenic drivers found in BW wastes (Rees 1980, Christensen & Kjeldsen, 1989).

Figure 12 Imports of domestic and non-domestic MSW into sites B and C.

Wastes imported into Sites B and C (Table 3) identify MSW as forming a significant component of the imported waste. MSW deposits at each site include both rapidly degrading BW and slowly degrading organic wastes ranging from wood to packaging – plastic and otherwise through to mattresses and furniture which are shown to exist unchanged many years after deposition (Rathje & Murphy, 2001). However, the BW fraction is neither separately identified nor recorded.

Although the implementation of alternative waste strategies is having a significant impact on landfilling, it would be incorrect to suggest all rapidly degrading organic wastes have been diverted away from the study landfill sites. At the meso-scale of any landfill, pockets, possibly significant, of BW containing wastes will exist because of less well developed kerbside collections or inefficiencies in householders or
transfer station processing. Furthermore, the EU waste codes (EWC) used to record waste imports are generic descriptions and, as such, often encompass multiple components in respect of both rapidly and slowly degrading components. This is particularly so for MSW (EWC 200301) where it is necessary to identify a number of components which vary with changing preferences in consumer tastes.

4.4 BW reduction in the UK

Evidence that BW deposition into landfill has reduced substantially is provided by the Environment Agency’s UK Statistics on Waste (EA, 2016). The Landfill Directive obligated the UK to determine a 1995 baseline figure for the mass of biodegradable materials in MSW. The figure accepted by the EU in 2010 as the biodegradable municipal waste (BMW) content in MSW was 29,030,000 tonnes (EA, 2016) and is the figure subsequent landfilled BW is compared against. By 2010, recorded BW deposits into landfill had reduced significantly to 10,339,000 tonnes (35.6% of the 1995 figure) further reducing to 6,843,000 tonnes in 2014 (23.6% of the 1995 figure). Recorded MSW generated in 1995 was 28,900,000 tonnes and for 2010 was 31,955,000 tonnes (EUROSTAT, 2017).

UK specific MSW and BW data for the period before 2010 is unavailable and, where it exists, is unclear or contradictory. This point is clearly evidenced in the 1995 baseline figure for BMW content in MSW which exceeds the recorded quantity of MSW generated for that same year. Furthermore, waste composition analyses and respective BMW estimates contain variations that have resulted in understated organic content in domestic MSW. Parfitt (2002) concluded the percentage of biodegradable waste (BMW) in domestic MSW was as high as 59%. Adjusting for EU defined BW this equates to some 41% (Parfitt, 2002). The inference drawn from
these figures for Sites B and C would suggest for some 414,900 tonnes and 403,000 tonnes of BW or approximately 20 and 32 percent respectively of the entire landfill mass for each site for the MSW component alone.

It is important to establish quantitatively the decrease in BW going into landfills B and C so that this can be correlated with the changes in evolution of leachate chemistry, to answer the following question: what reduction in BW is required to impact the evolution of landfill chemistry? An analysis of waste imports into the case-study sites is required to estimate the likely BW content of the pre-directive as compared to post-directive landfills. This is achieved by reviewing national data and targets with recorded waste imports at the sites.

4.5 Waste imports and estimates of BW by fraction for Sites B and C

To estimate the BW content of wastes imported into the case-study landfills from the recorded tonnages the following methodology is adopted: (i) Identification/separation of the imported tonnages of wastes that contain biodegradable components from their EWC listings (summarised in Figure 13, a – d), (ii) Component analyses of these wastes to determine those with rapidly degrading fractions (Table 5) and (iii) An estimation of the proportion of BMW and BW in each fraction using best available data (Table 6).

For (i), recorded imports for respective EWCs was provided by Viridor for each site. Each EWC was then separated into either BMW, BW or non-degradable waste. BMW/BW containing wastes, where those components exceed 1000 tonnes annually, are summarised as Figure 13 a – d. MSW, domestic and non-domestic are, by far, the most significant waste imports.

Whilst the EWCs allow for the separation required for (i) above, there are no established practices for the final two steps and, as a result, different possible data sources are reviewed. For the component analysis required for step (ii), four compositional studies (Table 5) from the literature were reviewed as possible estimators from which to establish BW content for MSW imported into Sites B and C. The NHWAP study was undertaken before the introduction of the Landfill Directive and is considered as incomplete (Parfitt & Flowerdew, 1997) whilst the 2001/03 Burnley study is included for comparison purposes only. The later DEFRA study, undertaken in 2010/11 would not reflect the wastes disposed during the period 2005 – 2009. Therefore, for the purposes of this paper the DEFRA (2006/07) review is considered the most relevant for domestic and non-domestic MSW and includes both regional and BMW analyses (2008. Appendix 4).

Table 5
Compositional assessments for MSW in the UK 1991 – 2010/11
<table>
<thead>
<tr>
<th>MSW % composition</th>
<th>NHWAP(^1) (1991)</th>
<th>Burnley \textit{et al}(^2) (2001/03)</th>
<th>DEFRA(^3) (2006/07) Domestic</th>
<th>DEFRA(^3) (2006/07) Non-domestic</th>
<th>DEFRA(^4) (2010/11)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Paper and card</td>
<td>34.4</td>
<td>23.6</td>
<td>22.69</td>
<td>39.3</td>
<td>19.2</td>
</tr>
<tr>
<td>Kitchen and garden waste</td>
<td>20</td>
<td>35.1</td>
<td>33.65</td>
<td>18.3</td>
<td>33.3</td>
</tr>
<tr>
<td>Textiles</td>
<td>2.4</td>
<td>2.4</td>
<td>2.83</td>
<td>2.0</td>
<td>2.9</td>
</tr>
<tr>
<td>Plastics</td>
<td>10.9</td>
<td>10.2</td>
<td>9.99</td>
<td>14.4</td>
<td>3.8</td>
</tr>
<tr>
<td>Misc. combustible</td>
<td>3.7</td>
<td>4.6</td>
<td>2.37</td>
<td>2.6</td>
<td></td>
</tr>
<tr>
<td>Disposable nappies</td>
<td>4.2</td>
<td>3.6</td>
<td>2.51</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Fines</td>
<td>6.7</td>
<td>0.6</td>
<td>0.53</td>
<td>1.8</td>
<td></td>
</tr>
<tr>
<td>Wood</td>
<td></td>
<td>4.6</td>
<td>1.66</td>
<td>3.8</td>
<td></td>
</tr>
<tr>
<td>Furniture and mattresses</td>
<td></td>
<td>3.73</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Sanitary</td>
<td></td>
<td></td>
<td></td>
<td>1.6</td>
<td></td>
</tr>
</tbody>
</table>

1. The 1991 data was collected as part of the National Household Waste Analysis Programme (NHWAP). 2. The 2001-2003 data resulted from a study undertaken by Burnley \textit{et al}. (2006) on behalf of the Welsh Government. 3. The Defra study relied on a range of data most notably Resource Futures. 4. Defra (2015).

Step (iii) entailed the quantification of the BMW/BW content for respective EWCs. This was achieved by, first, determining the BMW content obtained from the DEFRA publication WR0119: A Review of Municipal Waste Component Analyses – APPENDIX 4 to each EWC and second, allocating the BW content as defined by the European Union in EC2008. BMW and BW content for each waste category is summarised in Table 6 which similarly includes the estimated percentages of BW contained within the wastes imported into Sites B and C. Confirmation of the BMW data is provided by Appendix 3 of the Resource Futures report WR1003 (DEFRA, 2012). Furthermore, the biodegradibility multipliers applied in Appendix 3 were used to complete gaps in the WR0119 data.
Table 6:

<table>
<thead>
<tr>
<th>Waste category</th>
<th>DEFRA WR0119 BMW Estimate (%)</th>
<th>EU BW Estimate (%)</th>
<th>Sites B &amp; C BW Estimate (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Food/kitchen waste</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>Garden waste</td>
<td>100</td>
<td>100</td>
<td>100</td>
</tr>
<tr>
<td>Other organic (pet bedding + excrement, unidentified putrescibles)</td>
<td>100</td>
<td>nil</td>
<td>50</td>
</tr>
<tr>
<td>Paper</td>
<td>100</td>
<td>nil</td>
<td>nil</td>
</tr>
<tr>
<td>Card</td>
<td>100</td>
<td>nil</td>
<td>nil</td>
</tr>
<tr>
<td>Glass, metals &amp; plastics</td>
<td>nil</td>
<td>nil</td>
<td>nil</td>
</tr>
<tr>
<td>Wood</td>
<td>100</td>
<td>nil</td>
<td>nil</td>
</tr>
<tr>
<td>Textiles</td>
<td>50</td>
<td>nil</td>
<td>nil</td>
</tr>
<tr>
<td>Sanitary (nappies &amp; clinical)</td>
<td>50</td>
<td>nil</td>
<td>50</td>
</tr>
<tr>
<td>Mattresses &amp; furniture</td>
<td>50</td>
<td>nil</td>
<td>nil</td>
</tr>
<tr>
<td>Miscellaneous combustibles</td>
<td>50</td>
<td>nil</td>
<td>nil</td>
</tr>
<tr>
<td>Miscellaneous non-combustibles</td>
<td>nil</td>
<td>nil</td>
<td>nil</td>
</tr>
<tr>
<td>Soils, builders waste &amp; asbestos</td>
<td>nil</td>
<td>nil</td>
<td>nil</td>
</tr>
<tr>
<td>Fines (typically &gt;20 mm)</td>
<td>50</td>
<td>nil</td>
<td>50</td>
</tr>
</tbody>
</table>

For a number of EWCs determining the BMW or BW content was straightforward. For example, where an EWC comprised a single waste with identifiable organic content and the imported quantity was low, or waste imports were naturally, rapidly biodegradable for example EWC 190503, off-specification compost, then these were treated as 100% BW. Furthermore, many waste imports at both sites are inert cover
materials or classed similarly and have no BMW or BW content. More challenging were those MSW components within Table 5 that comprise composite items and contain a mixture of both slowly and rapidly degrading wastes along with wastes that are excluded by EC2008 but nevertheless comprise rapidly degrading components. In defining BW, within EC2008, the European Commission removed a number of BMW components found in MSW, as such, paper and card, wood, textiles, furniture and mattresses have no BW content (EC, 2008 and reaffirmed in EC, 2016). Other excluded wastes, for example, disposable nappies and components of non-special clinical wastes which include hygiene waste and incontinence pads (EA, 2015), these, whilst composed of cellulose, some plastics and rubber, absorbent polymers and paper tissue (Rathje & Murphy, 2001, DEFRA, 2008) and considered to biodegrade extremely slowly, differ from their soiled content which comprise rapidly degrading organic constituents that need to be included in the BW estimate. Within the UK, the composition of disposable nappies is very similar (DEFRA, 2008) and it is assumed this similarity applies to some clinical wastes. The Waste and Resources Action Programme (WRAP) surveyed the excreta content in disposable nappies and estimated the weight of the soiled contents to be 727kg based upon a two-and-a-half-year use by a single infant (DEFRA, 2008). For this reason here some fifty percent of the mass of sanitary waste is included as BW as it is considered better to overestimate than underestimate. Miscellaneous combustibles contain carpets and underlay, rubber and unclassified combustibles. For these latter two items and due mainly to classification differences these remain unchanged that is 50% BMW but no BW content (2008, Appendix 4).

As a result of the diversion of kitchen and garden, paper and card waste and the wide-ranging definition of MSW, for both domestic and non-domestic MSW, the
component percentages are required to be renormalised to obtain the increased quantities of remaining MSW components still being landfilled. The given percentages in Table 5 are stated before any diversion occurs. As a result of diversion, other MSW components will increase as a result of the diversion strategy.

4.6 Estimated BW materials at Sites B and C

The total quantity of waste imported into Sites B and C together with respective MSW imports are presented in Table 7. MSW forms the major source of biodegradable materials imported into each site, for Site B a further 33% of imports comprise inert cover materials. In addition, Table 7 presents a series of estimated scenarios for the both the total BMW and likely BW content for different diversion capture rates for each site. The BMW estimate is included for comparison to Parfitt’s 2002 published estimate (section 4.4), although this is included as a percentage of total waste imported. For the three BW estimates, the first is a reconstruction of the no-recycle condition expected to exist either during the pre-Landfill Directive period or, had similar waste disposal practices continued, where the bulk of domestic and non-domestic MSW was discharged directly to landfill. The second estimates the situation occurring post 2005 where alternative waste strategies divert recyclable components in MSW into more favoured options – here the 50% estimate being representative of the minimum recycle rate for those components in the South West Region. The third is the estimated BW content where the maximum DEFRA diversion rate for kitchen and garden waste applies. The latter two scenarios provide a feasible range of diversion from which the reduction in BW can be estimated.

However, for scenarios two and three, it should be noted that the renormalisation of remaining wastes increases their mass when compared to those in the zero-
diversion scenario. This accounts for the fifty and seventy-five percent diversion rates not equating to that fraction of the BW content in the zero-diversion scenario.

Table 7:
Estimated BMW and BW content as weights and percentage of total waste deposits and MSW for landfill Sites B and C.

<table>
<thead>
<tr>
<th></th>
<th>Total imports 2006-9 in tonnes</th>
<th>Estimated total imported BMW &amp; BW as a proportion of total waste</th>
<th>Estimated MSW BMW &amp; BW as a proportion of total MSW</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Total imports 2006-9 in tonnes</td>
<td>BMW content in tonnes (%)</td>
<td>BW - zero diversion tonnes (%)</td>
</tr>
<tr>
<td>Site B</td>
<td>555,158</td>
<td>268,909 (48.4%)</td>
<td>115,809 (20.9%)</td>
</tr>
<tr>
<td>Site C</td>
<td>430,050</td>
<td>239,601 (55.7%)</td>
<td>103,298 (24%)</td>
</tr>
</tbody>
</table>

The waste imports used to estimate the BMW and BW content were restricted to those years encompassed by the 2006/7 DEFRA composition analysis. Furthermore, it is these wastes that will determine the resultant leachate chemistry. The literature identifies the completion of compositional analyses as providing a number of “challenges” (2008, Appendix 4). For this analysis a percentage of MSW remains
unclassified. From other MSW compositional studies these unclassified materials comprise glass, metals, WEEE, some hazardous wastes including batteries together with bricks, plaster soils and other building materials and are not specifically identified in the 2006/7 analysis. These wastes have no BMW/BW content (2008, Appendix 4, DEFRA, 2012). The fine fraction is accounted for. Despite these challenges, we feel the data provided is the best available for the period in question.

Given that MSW forms the major source of biodegradable waste at each site, the estimated range for landfilled BW, resulting from the 50% and 75% diversion scenarios, relates to a reduction when compared to the Parfitt’s (adjusted) estimate of 41% identified in section 4.4. Allowing for variations in methodology between Parfitt’s estimate and this approach this reduction is significant and is reflected in the chemistry of the leachates arising.

4.7 Proposing a contemporary waste phase in place of acetogenesis

The diversion of kitchen and garden wastes away from landfill has noticeably impacted on three of the key indicators; pH, BOD and COD such that each is a near representation of the methanogenic phase and not the expected acetogenic phase. The analysis suggests the absence of the classical “acetogenesis phase” where products of acetogenesis dominate leachate chemistry. However, as shown by the analysis above, a substantial amount of putrescible BW is still being deposited in the landfills so whilst acetogenic processes are still likely to be occurring within the waste mass it is at an insufficient scale to noticeably influence the landfill leachate chemistry. The chemical signature of landfill evolves directly to one resembling the established methanogenic phase and indeed each of the landfills is generating methane. Furthermore, the estimated BW reduction arising from diversion strategies
that generates this change is of the order of 40 – 60% of total BW deposited. It is acknowledged that further research is required to better understand and confirm these differences.

Ehrig further proposed that conditions arising during acetogenic phase degradation inhibit the development of methanogenic bacteria (Ehrig, 1983). Furthermore, he states that increasing pH is an indicator of methane production. The reduction in acetogenic conditions may lead to a more rapid pathway to methane production.

**Conclusions**

Two study sites have offered a rare opportunity to: (1) review and compare leachate samples from the post-Landfill Directive period to established research, undertaken before the Landfill Directives publication and (2) Estimate scenarios for the reduction in biodegradable materials deposited into landfills since the implementation of the Landfill Directive.

Results from leachate sampling corroborate the sites have circumvented the “classical” acetogenic phase. The leachate samples, for the most part, when compared to Ehrig’s data are representative of a site in the methanogenic phase and closely resemble Ehrig’s data values for this phase. The consistency or lack of variation in respective samples further reinforces this assessment.

The redirection of BW waste from landfills removes materials able to decompose rapidly. The lack of credible UK records for waste deposits, particularly in respect of the central biodegradable content within MSW waste streams, has led to the estimation of its likely content. Four scenarios are presented to explain the observed change in leachate chemistry. Whilst the estimate for total biodegradable materials
(BMW) represents some 49 – 56% of the materials deposited into the two study sites, the pre-Landfill Directive BW content represents 21 – 24% of deposits. Redirection to the end of 2009 has reduced this to approximately 10% with the concomitant change in leachates produced. It is noticeable that recycling strategies have continued to develop across the UK since this period. The authors consider the approach taken in this study lays the foundation for further research, particularly into the impact of changing MSW management strategies.

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References


November 1, 2017)
Environment Agency (2004) Improved definition of leachate source term from
landfills Phase 1: review of data from European landfills:
https://www.gov.uk/government/publications/improved-definition-of-leachate-
source-term-from-landfills-phase-1-review-of-data-from-european-landfills
(accessed May 1, 2016)
https://www.gov.uk/government/publications/waste-classification-technical-
guidance (accessed December 1, 2015)
Environment Agency (2016) UK Statistics on Waste:
management (accessed November 8, 2017)
Environment Agency (2016) Local Authority collected waste generation from April
2000 to March 2015 (England and regions) and local authority data April 2014 to
March 2015: https://www.gov.uk/government/statistical-data-sets/env18-local-
authority-collected-waste-annual-results-tables (accessed October 1, 2016)
Management and Research 1: 53-68.
Springer-Verlag
Eurostat (2017) Municipal wastes generated by households by year and waste
category: http://ec.europa.eu/eurostat/web/waste/municipal-waste-generation-
Eurostat (2017) Treatment by year and waste category.
Eurostat (2017) Municipal waste by waste operations:
http://appsso.eurostat.ec.europa.eu/nui/submitViewTableAction.do (accessed
October 21, 2017)
content/EN/TXT/?uri=CELEX:32008L0098 (accessed August 26, 2016)
European Commission (2011) Report from the Commission to the
European Parliament, the Council, the European Economic and Social
Committee and The Committee of the Regions on the Thematic Strategy on the
Prevention and Recycling of Waste SEC (2011) 70 final:
August 26, 2016)
http://eur-lex.europa.eu/legal-
content/EN/ALL/?uri=CELEX:31999L0031 (accessed on April 10, 2016)
Fischer C, Lehner M & Lindsay D (2012) Overview of the use of landfill taxes in
(accessed October 21, 2017)
Frank RR, Cipullo S, Garcia J, Davies S, Wagland ST, Villa R, Trois C & Coulon F
(2016) Compositional and physicochemical changes in waste materials and
biogas production across 7 landfill sites in UK. Waste Management. Ahead of
print October 2016,


Appendix 1 – Sites B and C background information

**SITE B**

**Site Design and Construction**

The Northern Extension has been operational since 2007 and is a fully engineered landfill site.

The 32-hectare site currently comprises a total of 10 sub-phases developed on the principal of engineered containment, with a basal lining system comprising an artificial liner comprising a geosynthetic clay liner (GCL) and a reworked and natural geological barrier of *in situ* Alluvium. The phases are subdivided into elongate sub-cells by permanent clay bunds of 2m height. Each sub cell is filled using a series of elongate ‘tipping areas’. It is proposed to cap the Northern Extension with a LLDPE geomembrane, overlain by 800mm alluvium and 200mm topsoil.

The design principles of the Northern Extension are based around both engineered and hydraulic containment of leachate within the landfill site during the operational and aftercare phases of the site life. During the site construction and development, groundwater underdrainage beneath the site will locally influence the naturally occurring groundwater regime that was present before site development, which will re-establish once dewatering operations have ceased.

The average waste depth is 13.0 metres with waste deposition undertaken in 2.5-metre layers.

**Leachate Management**

Leachate collection and control measures have been installed within each cell in the form of leachate monitoring and abstraction boreholes and aggregate drainage.
system. Leachate from both areas of the landfill is pumped to the raw leachate balance tank for on-site treatment. Each cell has a unique leachate sampling/extraction point identified on the site plan. Only leachate samples extracted from those well points indicated were used in the study. Furthermore, leachate recirculation was not installed at the time of sampling (2007 – 2011).

**Gas Collection System (GCS)**

The current GCS comprises vertical gas wells connected to gas mains and manifolds. Wells have typically been installed on twenty to forty metres spacing. Extraction is generally provided by a manifold system with wells connecting individually into inlets on the manifolds. Wells have generally been drilled to 375mm-450mm diameter and installed with a suitable standoff from the pit base.

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**Figure A1** Site B Northern Extension Plan. Individual landfill cells and site perimeter are indicated by the broken lines. Leachate sampling points/locations used in the study are included. The grid lines are at 100 metre spacings.
SITE C

Site Design and Construction

Site C has been operational since 2006 as a fully engineered landfill site.

The 26-hectare site currently comprises eleven engineered containment cells (Cells A to L). Cells A to E have been developed utilising 0.75m of low permeability clay plus a 0.25m protection layer, overlying in-situ Lias Clay. Cells E to J have been developed utilising 1m of low permeability clay with a maximum permeability of $2 \times 10^{-10}$ m/s. Where limestone bands have been encountered within a metre of the base of cells, then the upper 0.5m of Lias Clay has been replaced with clay that achieves a maximum permeability of $5 \times 10^{-10}$ m/s.

The average waste depth is 10.0 metres with waste deposition undertaken in 2.5-metre layers.

Leachate Management

Leachate collection and control measures have been installed within each cell in the form of leachate monitoring and abstraction. An installed drainage system delivers raw leachate to the remote sampling points before being pumped to the raw leachate balance tank for on-site treatment at the leachate treatment plant. Only leachate samples extracted from sampling points C1, C2 and C3 were used in the study. Composite leachate samples were not included. Furthermore, leachate recirculation was not installed at the time of sampling (2006 – 2011).

Gas Collection System (GCS)
The current GCS comprises vertical gas wells connected to gas mains and manifolds. Wells have typically been installed on twenty to forty metres spacing. Extraction is generally provided by a manifold system with wells connecting individually into inlets on the manifolds. Wells have generally been drilled to 375mm-450mm diameter and installed with a suitable standoff from the pit base.

Figure A2 Site C Plan. Individual landfill cells and site perimeter are indicated by the broken lines. The cell boundaries for cells D, E, F, G, H, I and L have been excluded to improve the plan’s reproducibility. Leachate sampling points/locations used in the study are included. For this study, composite leachate samples were not included. The grid lines are at 100 metre spacings.