From a Landfill Bioreactor to a Sustainable Storage.

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EXECUTIVE SUMMARY

For a landfill to be considered sustainable it should be in equilibrium with the environment within the time frame of one generation. In the framework of this project a sustainable landfill has been defined as a landfill that complies as soon as possible, but in any case within 30 years, with the leaching level of a landfill for inert solid wastes defined by EU-Landfill Directive regulations. The emission level from these landfills is considered that low that no upper/cover liner is mandatory and no aftercare is required. The project Sustainable Landfilling (NL) aims to assess the feasibility of three sustainable concepts, one of them is the bioreactor concept dominated by organic waste. The organic waste is a mix of municipal solid waste and comparable industrial waste. The 40% of waste, containing organic material in the mixture is not representative for the present Dutch situation, but will meet landfill mixtures in the southern and eastern EU countries and can be seen as a “worst case” situation in respect to leachate quality and landfill gas production en emissions.

The bioreactor has been constructed and filled with well-defined waste in 2001 and the operation started in spring 2002. The bioreactor pilot has a rectangular footprint of 55x80 meter, with an average depth of approximately 6 meters. In order to optimise the infiltration and recirculation of leachate, two separate horizontal systems for leachate infiltration were constructed, one horizontal system is situated more or less at a depth in the middle of the waste and a supplementary system is located below the top-cover. The landfill gas has been extracted. The top cover is permeable to permit rainfall entrance and is suitable for methane oxidation.

The (anaerobic and aerobic) flushing bioreactor pilot has been in operation five years and has additionally been simulated in a biological degradation and flushing model. The release of nitrogen seems to be delayed by the intermediate fixation in the form of biomass and stagnant zones. Because of the strict standards for the allowable nitrogen concentration from the viewpoint of emission control, the development of the nitrogen concentration is considered to be the main limiting factor to achieve final storage quality (FSQ) within the acceptable time-frame. Therefore the research has been extended to nitrification of the leachate recirculation flow and aeration of the bioreactor.

The pilot has been simulated with model calculations by assuming a triple porosity concept (preferential channels, slow mobile and stagnant zones in the landfill body). To verify and adjust the hydrological model additional (remote) geo-electrical research has been carried out.

After five years of operating the bioreactor landfill the organic waste have been stabilised for over 70% to approx. 90%.
INTRODUCTION

The Dutch sustainable landfill program

A sustainable landfill has been defined as a landfill that complies as soon as possible, but in any case within 30 years, with the leaching level of an inert landfill by EU-Landfill Directive regulations. The emission level from these landfills is considered too low that no upper/cover liner is mandatory and no aftercare is required. The Dutch project Sustainable Landfilling (NL) aims to assess the feasibility of three sustainable concepts, one of them is the bioreactor concept dominated by organic waste (see Figure 1.).

The project Sustainable Landfilling (NL) aims to assess the feasibility of three sustainable concepts: Bioreactor: organic dominated Final Storage Quality: a technology targeting stabilisation of organic matter/waste, applied at the landfills of Wijster and Landgraaf. Equifill: non-organic dominated Final Storage Quality: a technology targeting a landfill with stabilised organic matter, applied at Landfill Nauerna; Monolith: cold immobilisation: a technology aimed at the immobilisation of specific solid wastes applied at VBM-Maasvlakte landfill.

Final storage quality (FSQ) describes the desired stable end-picture of a sustainable landfill. The Equifill end picture is considered the final picture of all landfill processes. This means that the bioreactor will in the end also turn into Equifill characteristics. The intention is to reach this final state as soon as possible and in any case within 30 years (one generation cycle).

The landfill pilots Equifill and Bioreactor can be situated between the EU Landfill Directive definitions of a non-hazardous landfill and an inert landfill (see Figure 1). The individual tested waste into the inert landfill has to comply with the standards described in ANNEX II of the Landfill Directive. The aim of the Equifill landfill is compliance of the mixture of waste with ANNEX II. The Equifill performance has been explained by several references (Scharff et al, 2005); the results give an indication that after 15 years of operation the leachate of the mixture of waste will comply with the Landfill Directive. The critical components are chloride and sulphate.
The Landgraaf bioreactor test cell

The Landgraaf bioreactor (see Figures 2 and 3) test cell is subject of this paper. The aim of this project is enhancement of the (biological) stabilisation of the landfill body, flushing of salts and soluble stable organic material.

Figure 3. Cross section of the bioreactor.

During operation of the bioreactor the emissions to air and groundwater have to be negligible. The residue of the bioreactor has to meet the FSQ targets; these targets are also subject of the overall Sustainable Landfill study but the goal is compliance with the EU Landfill Directive ANNEX II standards for the inert landfill. The processes that influence the leachability of e.g. heavy metals in the mixture of waste have been summarised in Figure 4 (Van der Sloot et al, 2003).

Figure 4. Overview of processes that influence the actual leaching of heavy metals.
FIVE YEARS OPERATING THE BIOREACTOR TEST CELL

Materials and methods

The pilot was constructed with a rectangular footprint of 55x80 meter, with an average depth of approximately 6 meters (Figure 3). In order to optimise the infiltration and recirculation of leachate, two separate horizontal systems for leachate infiltration were constructed, one horizontal system is situated more or less at a depth in the middle of the waste and a supplementary system is located below the top-cover. Horizontal drains at the bottom collect the leachate. Landfill gas extraction is achieved by 6 vertical wells supplemented by 7 horizontal drains below the top-cover; these drains lay in a bed of wood chips and are covered with a geo-textile and a thin layer of loamy sand. In addition to the gas extraction the construction of the top cover is optimised with regard to the potential for methane oxidation. This semi-permeable top cover, existing of sandy soil enriched with coarse compost, allows the infiltration of rainwater (Brands-Van den Esschert et al, 2003). The organic waste is a mix of municipal solid waste and comparable industrial waste (Table 1).

<table>
<thead>
<tr>
<th>Waste-material</th>
<th>Weight-percentage (%)</th>
<th>Quantity (wet tons)</th>
<th>Quantity (dry solids)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Domestic waste</td>
<td>36</td>
<td>8.987</td>
<td>5.302</td>
</tr>
<tr>
<td>Industrial waste</td>
<td>12</td>
<td>2.988</td>
<td>1.404</td>
</tr>
<tr>
<td>Soil purification residue</td>
<td>19</td>
<td>4.718</td>
<td>3.762</td>
</tr>
<tr>
<td>Car shredding</td>
<td>19</td>
<td>4.825</td>
<td>3.699</td>
</tr>
<tr>
<td>Moulding sand</td>
<td>5</td>
<td>1.335</td>
<td>1.322</td>
</tr>
<tr>
<td>Screening residues</td>
<td>8</td>
<td>1.928</td>
<td>1.600</td>
</tr>
<tr>
<td>Biological sludge</td>
<td>2</td>
<td>379</td>
<td>258</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>100</td>
<td>25.160</td>
<td>17.348</td>
</tr>
</tbody>
</table>

Phasing and monitoring of the bioreactor

The operation of the bioreactor can roughly be distinguished in the following procedures:
- anaerobic degradation of the organic material enhanced by the recirculation of leachate
- flushing of salts and organic residues (DOC, humic and fulvinic acids)
- post degradation of microbial material and residual waste with special attention to removal of nitrogen by aeration of the landfill body and nitrification of the recirculated leachate
- stabilisation and humification of the residual waste with additional flushing to meet the Final Storage Quality standards.

In 2006 the third operational phase has been started: alternating aeration of the landfill body (via the water infiltration and discharge system) and supplementary nitrification (start up in 2005) of the recirculated leachate by a submerged rotating biological contacter (RBC system).

Summary of the phasing of the bioreactor landfill pilot:
- 2001-2002: construction and filling of the reactor
- 2002-2005: anaerobic stabilization; fill, recirculation and draw of leachate
- 2005-2006: semi-aerobic stabilization by ex situ nitrification
- 2006-2007: aeration of the bioreactor, ex-situ nitrification of leachate and recirculation of leachate
Gas management

The landfill gas extraction has been measured frequently; the quality during the first year was quite good (50% of methane) but deteriorated in 2005 to low levels (20% of methane). The calculated gas production (prognoses) seems to be an overestimation. This is probably caused by an overestimation of the biodegradable part of the organic waste and an aerobic composting phase during preparation of the waste (size reduction and homogenisation by the compactor), mixing (partly outside the bioreactor) of waste and the long period of filling in relative thin layers. The landfill gas production is presented in Figure 5.

Methane-emissions and methane oxidation have been measured (Woelders et al, 2005). The emission was approx. 2% of the extracted flow and the average oxidation measured by the 13C method could be calculated between 35 and 60%.

Figure 5. Landfill gas production (“adapted” is recalculated to 57% of methane)

Leachate management

The leachate management can be characterised by the “fill and draw” principle till 2005. During the “fill” period the leachate is also circulated to achieve a 3000 mm infiltration per year. In this period the anaerobic degradation is stimulated and soluble components increase in the water system. During the extended “draw” period the soluble components are drained off the landfill body. At first the leachate in the mobile zone is drained at the end of the draw period supplemented by leachate (and diffused soluble material) from the semi-stagnant zones in the landfill body. The rapid response of leachate generation and quality on changes in leachate infiltration lead to the conclusion that only limited parts of waste the test-cell are affected by leachate recirculation through convective transport. The size of this mobile zone is estimated to be 10% at maximum. The remaining 90% of the waste (stagnant and semi-stagnant zones) is most likely reached by slow convective transport or diffusion only.

Since 2005 a part of the recirculation flow has been treated by a rotating biological contactor. The average nitrification rate was 2.5 kg N/day. If the total recirculation flow (10 m3/day, 900 mm/a infiltration) had been treated the N conversion would be 1800 kg N/a. In chapter 6 the nitrogen removal will be discussed in more detail.
CRITICAL FACTORS TO ACHIEVE THE SUSTAINABLE LANDFILL

Table 2 indicates that the standards for inert landfill are not yet met for DOC, Chloride and for the metals Arsenic, Chrome and Nickel. Selenium is also indicated as too high. Beyond the criteria listed in Annex II, the values for nitrogen are of concern. A target value for the final concentration can be set at 11.3 mg N/l (corresponding with 50 mg NO3/l), either in the form of ammonia or nitrate (Groundwater Framework Directive). This indicates that the required reduction ratio for nitrogen would be 51 times, which is considerably higher then for the other elements in Table 2. To overcome this problem, an additional leachate treatment unit has been incorporated in the recycle loop for the Landgraaf pilot. This unit has been in operation since July 2005. Experience so far show that the conversion of ammonia to nitrate is achieved successfully at a conversion rate of 90%. The nitrified leachate is sent back to the landfill, where the denitirification under anaerobic conditions is expected to take place. The effect of denitirification cannot yet be verified, although the nitrate levels in the leachate have remained negligible. To quantify the long-term effect of this treatment modelling has been set up.

Table 2. Comparisons annex II criteria for inert waste and measured concentrations.

<table>
<thead>
<tr>
<th>Component</th>
<th>Symbol</th>
<th>Leachate Avg. concentrations 2005 (mg/l)</th>
<th>Leachate Concentrations April 2007 (mg/l)</th>
<th>Limit value C0 column test (mg/l)</th>
<th>Act. conc./ C0</th>
<th>Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arsenic</td>
<td>As</td>
<td>0.143</td>
<td>0.140</td>
<td>0.06</td>
<td></td>
<td>2.33</td>
</tr>
<tr>
<td>Barium</td>
<td>Ba</td>
<td>1</td>
<td>0.54</td>
<td>4</td>
<td>0.135</td>
<td></td>
</tr>
<tr>
<td>Cadmium</td>
<td>Cd</td>
<td>0.001</td>
<td>0.001</td>
<td>0.02</td>
<td>0.05</td>
<td></td>
</tr>
<tr>
<td>Chrome total</td>
<td>Cr</td>
<td>0.232</td>
<td>0.26</td>
<td>0.1</td>
<td>2.6</td>
<td></td>
</tr>
<tr>
<td>Copper</td>
<td>Cu</td>
<td>0.01</td>
<td>0.13</td>
<td>0.6</td>
<td>0.21</td>
<td></td>
</tr>
<tr>
<td>Mercury</td>
<td>Hg</td>
<td>0.0001</td>
<td>-</td>
<td>0.002</td>
<td>0.05</td>
<td></td>
</tr>
<tr>
<td>Molybdenum</td>
<td>Mo</td>
<td>0.014</td>
<td>0.026</td>
<td>0.2</td>
<td>0.13</td>
<td></td>
</tr>
<tr>
<td>Nickel</td>
<td>Ni</td>
<td>0.18</td>
<td>0.19</td>
<td>0.12</td>
<td>1.58</td>
<td></td>
</tr>
<tr>
<td>Lead</td>
<td>Pb</td>
<td>0.015</td>
<td>0.035</td>
<td>0.15</td>
<td>0.23</td>
<td></td>
</tr>
<tr>
<td>Antimony</td>
<td>Sb</td>
<td>0.01</td>
<td>0.01</td>
<td>0.1</td>
<td>10</td>
<td></td>
</tr>
<tr>
<td>Selenium</td>
<td>Se</td>
<td>0.01</td>
<td>0.01</td>
<td>0.004</td>
<td>2.5</td>
<td></td>
</tr>
<tr>
<td>Zinc</td>
<td>Zn</td>
<td>0.075</td>
<td>0.12</td>
<td>1.2</td>
<td>0.1</td>
<td></td>
</tr>
<tr>
<td>Chloride</td>
<td>Cl</td>
<td>1400</td>
<td>1200</td>
<td>460</td>
<td>2.6</td>
<td></td>
</tr>
<tr>
<td>Fluoride</td>
<td>F</td>
<td>-</td>
<td>-</td>
<td>2.5</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>Sulfate</td>
<td>SO4</td>
<td>479</td>
<td>130</td>
<td>1500</td>
<td>0.09</td>
<td></td>
</tr>
<tr>
<td>Fenolindex</td>
<td></td>
<td>-</td>
<td>-</td>
<td>0.3</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>DOC</td>
<td>DOC</td>
<td>707</td>
<td>440</td>
<td>160</td>
<td>2.75</td>
<td></td>
</tr>
<tr>
<td>Nitrogen</td>
<td>NKj</td>
<td>700</td>
<td>580</td>
<td>(11.3)</td>
<td>(51)</td>
<td></td>
</tr>
</tbody>
</table>

For the other elements mentioned in Table 2 flushing will be an important measure to reduce the concentrations to the required level. For the metals involved it cannot be directly assumed that their concentration will reduce with flushing as their concentrations may also be determined by solubility. Also the occurrence of complexation with DOC could have a great influence on concentrations. For the pilot in Landgraaf the correlation between the relevant metal concentrations and DOC concentrations has been investigated. The trends suggest that for the metals in question a correlation exists between the metal concentrations and DOC-values. If this correlation proves valid, the metal concentrations with the possible exception of arsenic could be reduced sufficiently by flushing of DOC (Van der Sloot et al, 2003 and Scharff et al, 2005).
MODELLING THE BIOREACTOR

Bioreactor processes are modelled combining relative simple descriptions of both biochemistry and mass transport. The overall model is depicted in Figure 6. The model itself and the model-parameters used are described in much more detail in Mathlener et al. (2006). Hydrological behaviour of the waste is described as a cascade of ideally stirred cells. Part of the cells represent the preferential channels, another part represents the stagnant phase. In the preferential channels (mobile zone), mass transfer takes place through convection; mass-transfer between mobile zone and stagnant bulk largely takes place through diffusion.

Within each individual cell, microbiological reactions take place. However in the model, the set of reactions is based on the work of McDougal et al (2001) and is significantly simplified compared to biochemical reactions schemes as being used in other modelling attempts.

One process that is incorporated is the accumulation of Nkj in the biologically active biomass formed during conversion on organic material, and only released slowly after decay of this biomass. Since the C/N-ratio in the biologically active biomass is lower than the original C/N-ratio of the waste, release of significant part of the nitrogen in the waste is delayed, making nitrogen a longer term problem compared to components that are more readily available.

For these calculations the following scenario was used for the hydrological parameters:

**Period 1:** from 0 – 5 years, recirculation flow equivalent to infiltration at 750 mm/yr;

**Period 2:** from 5 – 20 years, infiltration rate at 750 mm/yr, completely consisting of supplention of clean water. This period is characterised by flushing, no recirculation;

**Period 3:** from 20 – 30 years, infiltration rate at 300 mm/yr, also clean water, no recirculation.

The course of the main constituents of the leachate is presented in Figure 7 from which the main conclusions can be drawn:

- By applying this mode of operation the DOC concentration can be reduced to the level where the concentrations of the heavy metals should no longer constitute a problem;
- The reduction of concentrations achieved by flushing is much less pronounced for NKj as for the other components.

In order to investigate the possibilities of a further reduction of the long term nitrogen concentrations additional model calculations have been performed.

The scenarios that have been evaluated have the same hydrological characteristics as mentioned above. Changes have been made with regard to the possibility of treatment of the recirculation flow by biological treatment (nitrification/denitrification) and with respect to the life expectancy of the anaerobic biomass, which is capable of temporarily storing a certain amount of Nitrogen.
The scenario’s can be described as follows:

Scenario A: During the first 5 years the recirculation and treatment is increased to 1500 mm/yr. After 5 years the suppletion is increased to 750 mm/yr, recirculation and treatment is set at 900 mm/yr. After 20 years recirculation and treatment is stopped and suppletion is reduced to 300 mm/yr. Scenario B is identical to Scenario A except for the fact that the half-life of the anaerobic biomass has been reduced from 4 years to 1 year. This could possibly be achieved through aeration.

The results presented in Figure 8 indicate that the treatment of recirculated leachate is quite effective in reducing the NKj concentrations in the operational phase of the landfill. In that way it can contribute significantly in reducing the costs associated with the discharge of leachate. In the long run the effect of treatment reduces and the transfer of NKj from stagnant zones and from decaying biomass becomes dominant in the same way as for the reference case. Reducing the lifetime of the biomass can be an important step in achieving a more favourable course for the NKj-concentrations in the long term.

Figure 7. Course of chloride, Kjeldahl Nitrogen (NKj) and Dissolved Organic Carbon.

Figure 8. Effect of leachate treatment and biomass life-time on the NKj concentrations.
One way to get an impression of moisture distribution is from geo-electrical sounding measurements. In this method, a large number of electrodes are put in a line in the waste, and the resistivity between all combinations of electrodes is registered. Reverse modelling of the results give a 2D map of apparent resistivity in the waste. This method determines the apparent resistivity throughout the waste. Resistivity however is a product of conductivity of the waste (e.g. metal content), moisture content and concentrations of dissolved salts within the water. As a result geo-electrical sounding does not directly result in distributions in moisture content in the waste.

In Landgraaf test-cell five 2D cross-sections are being made, parallel to each other with several examples being shown in Figure 9. The measurements performed here seem to give useful insights, also because the Landgraaf test-cell, waste is relative homogeneous in composition, and after leachate infiltration also relatively wet (except for the upper part). In Figure 9, areas of very low (< 7 $\Omega$ m, dark blue), medium (10-30$\Omega$ m, yellow) and high (> 20 à 50 $\Omega$ m, redbrown) apparent resistivity can be distinguished, which are interpreted as resp. saturated zones with high salt contents (stagnant bulk); zones with low salt contents (mobile zones) and unsaturated zones.

The results of the geophysical measurements are compared with results of leachate samples from test-wells, yielding quite good results, test-wells being in the dark-blue low-resistivity area generally having higher COD and salt-contents and having a higher conductivity compared to test-wells in the lighter blue areas.

Concluding, the geophysical measurements seem to locate stagnant areas in the waste, and the pattern in Figure 9 is consistent with the other 2D cross-sections obtained:

1. a larger stagnant zone to the left, possibly as a result of a not optimal design of the system for leachate infiltration (This system consisted of horizontal perforated drains)
2. a zone with increased resistivity from the 30.0 m point downwards to the right, possibly being a preferential channel
3. some more heterogeneity at the right hand-side, with several smaller stagnant zones.
ENHANCEMENT OF NITROGEN REMOVAL BY AERATION

To enhance the stabilization of the landfill aeration started in June 2006 into the “middle” infiltration drains with 200 Nm3/hr; landfill gas extraction had been stopped and the off-gas passes through the oxidation top cover. Average off-gas quality is measured under the top cover CH4 1.8 v%, CO2 9 v%, O2 8.6 v% and N2 80.6 v%. The carbon release of the bioreactor is approx. 12 kg/hr, the NH3 emission 11 g/hr and the extra N2 release (denitrification of nitrified leachate) approx. 0.1 to 0.2 kg/hr. The enhancement of the stabilization is shown in Figure 10.

A mass balance of 5 years operation, from which 1 year aerated, is given in Table 3. About half of the bioreactor volume is aerated with a 100 ton C release per year. Enhancement is possible to over 150 ton C/a aerating the total bioreactor; release of residual biodegradable C in the waste and C stored in microbial biomass (528 ton C) will take several years to be converted aerobically (5 years?). The hydrolysed nitrogen however has been stored in the bioreactor for over 50% (in microbial biomass and leachate). Recirculation of leachate with treatment (nitrification) and drainage of the leachate will release the nitrogen (most NH4-N), but it will take a long time because of the reducing hydraulic conductivity (<900 mm/a) and the high volume of stagnant zones. An enhancement could be achieved by enlargement of the aeration without leachate recirculation and establishment of nitrification zones in the bioreactor. Full scale experience and model calculations have to prove this change of operation.

Table 3. Carbon and Nitrogen mass balance after five years operation of the bioreactor.

<table>
<thead>
<tr>
<th>Remarks</th>
<th>Carbon (C)</th>
<th>Nitrogen (N)</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Input : biodegradable waste input (1)</td>
<td>1200</td>
<td>100</td>
<td>32</td>
</tr>
<tr>
<td>Released (2): by leachate treatment</td>
<td>1</td>
<td>0.1</td>
<td>2.4</td>
</tr>
<tr>
<td>by flushing</td>
<td>2</td>
<td>0.2</td>
<td>2.0</td>
</tr>
<tr>
<td>by (landfill) gas (3)</td>
<td>664</td>
<td>55.3</td>
<td>0.1</td>
</tr>
<tr>
<td>stored in microbial biomass</td>
<td>133</td>
<td>11.1</td>
<td>8.0</td>
</tr>
<tr>
<td>stored in leachate bioreactor</td>
<td>5</td>
<td>0.4</td>
<td>9.0</td>
</tr>
<tr>
<td>Totally:</td>
<td>805</td>
<td>67.1</td>
<td>21.5</td>
</tr>
<tr>
<td>Residual: waste in bioreactor</td>
<td>395</td>
<td>32.9</td>
<td>10.5</td>
</tr>
</tbody>
</table>

(1) Overall C content analysed approx. 2100 ton; N not analysed properly and therefor estimated and correlated with C conversi
(2) The released C and N are measured values.
(3) N release by nitrification/denitrification in the aerated landfill not taken into account.
CONCLUSIONS

From the experimental data and model calculations the critical parameters in relation to FSQ (EU Landfill Directive Annex II for an inert landfill) can be selected: DOC, NKj, Cl, and heavy metals (As, Cr, Ni and Se). Flushing is the only pathway to reduce chloride and therefore considering and to overcome the negative influence of stagnant zones will be the target. Reduction of DOC to the limit of Annex II (160 mg/l) will reduce also the leaching of heavy metals within the criteria; DOC reduction by biological conversion and flushing is feasible.

Reduction of NKj in the leachate will be a combined effort of waste and microbial biomass degradation (“stabilization of the landfill”), in-situ conversion of organic N and recirculation with ex-situ nitrification. Enhancement by an improved aeration will be the effort for the next years.

ACKNOWLEDGEMENT

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In the Foundation the main Dutch landfill companies combine financial funds and technological know how together with Dutch research institutes and consultants. For information about the Dutch foundation and downloading research reports visit the website: [www.sustainablelandfilling.com](http://www.sustainablelandfilling.com)

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