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Evaluation of Existing and Evolving Techniques for Landfill Gas Upgrading in Comparison with Conventional Mitigation Systems

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Abstract
A new technique for upgrading of biogas or landfill gas (LFG) is introduced. With the new technique, biogas and/or LFG is upgraded by pumping it through weathered MSWI bottom ash (BABIU process). For evaluation of existing upgrading systems and the BABIU process, the greenhouse gas emissions of scenarios with and without utilization of LFG are calculated (in total: nine scenarios). The results indicate that an engineered bio-cover is the most important equipment for reducing GHG emissions in the aftercare. A maximum of CO_2 sequestration can be achieved by a combination of engineered bio-cover and application of the BABIU process.

1 Introduction
Methane emissions from landfills are a significant portion of the anthropogenic CH_4 emissions. It is clear that zero mitigation scenarios ("leave as it is") are acceptable only for small or older sites that contain mostly degraded material. For sites containing (potentially) degradable material, it has to be said that the question is not "*passive or active mitigation?*" but every landfill site needs a tailored solution in the aftercare phase. Passive mitigation systems, e.g., bio-covers for CH_4 oxidation, can be combined with active gas collection and subsequent utilization or with flaring of the collected gas. In combination with existing or optional biogas plants (or wastewater treatment plants) close to the landfill site, presently a large number of concepts are available or carried out at pilot scale. The focus of the paper presented here is an evaluation of scenarios where LFG is upgraded (enriched in CH_4) and of other scenarios applied frequently for mitigation of LFG in the aftercare.

2 Upgrading of LFG
2.1 Desired gas quality
The primary requirements for feeding upgraded biogas to the gas grid are: 1) Sufficiently high heating value, 2) low hydrogen sulfide content and 3) low moisture

Tab. 1: Desired gas quality according to Austrian guidelines

Parameter	**Limits (ÖVGW G31)**	**Parameter**	**Limits (ÖVGW G31)**
Wobbe index	13.3 – 15.7 kWh/m³	S total	30 mg S/m³
Heating value	10.7 – 12.8 kWh/m³	Mercaptane sulfur	6 mg/m³
Density	0.55 – 0.65	H_2S	5 mg/m³
Oxygen	0.5 Vol.%	COS	5 mg/m³
Carbon dioxide	2 Vol.%	Halogene compounds	"0 mg/m³"
Nitrogen	5 Vol.%	Ammonia	"technically none"
Hydrogen	4 Vol.%	Solids, liquids	"technically none"
Condensation point of hydrocarbons	< 0°C at applied pressure	Condensation point of water	< -8°C at 40 bar
Parameter	Limits (ÖVGW G33)	Parameter	**Limits (ÖVGW G33)**
Methane	> 96 Vol.% "or heating value > 10.7"	Si total	10 mg/m³

content. To meet these requirements, most of the CO_2 and H_2S have to be removed from the biogas. In Austria, the desired gas quality is defined by two guidelines (ÖVGW G31, ÖVGW G33).

The table values of the ÖVGW guidelines are oriented towards controlled mixing of gases from chemical processes with natural gas, and not towards utilization of biogas. In fact, the high limits for CH_4 and minimum heating value are an obstacle against biogas/LFG utilization projects. Furthermore, the limit for halogene compounds ("zero") is not executable. Therefore, at present only one upgrading plant for biogas is in operation in Austria. In none of the ≈ 55 existing landfills, LFG is upgraded or fed to the gas grid.

2.2 Available upgrading techniques

Several proven upgrading techniques are available for separation of CO_2 and CH_4. Besides that, a wide assortment of techniques for removal of H_2S or Si exists. The most abundantly applied processes for separation of CO_2 and CH_4 are:

1) Pressure swing adsorption (PSA),
2) Absorption of CO_2 in water under pressure (WA) and
3) Membrane separation (MS)

For each of the other techniques, e.g. cryogenic separation (KRYO) of CO_2 and CH_4, for chemical absorption (CHEM) and for membrane absorption (MA), only pilot plants exist or have been in operation in Europe for a few years. Cryogenic separation of LFG and MA are not state-of-the art. On the other hand, the cryogenic technique is frequently applied with success for special and rare natural gases, e.g. He-containing natural gas.

The principles of these upgrading techniques, and the number of existing

Tab. 2: Existing upgrading techniques

Technique	Very short description	Number of plants in EU
PSA	Adsorption and desorption on fine solids, e.g. zeolithes, carbon	> 32
WA	Removal of CO_2 by absorption in water at 6 to 12 bar	> 15
MS	Separation by permeable gas membranes (gas permeation)	few
KRYO	Cryogenic principle: "Freezing out" of CO_2	1
CHEM	Absorption of CO_2 in aqueous solutions of chemicals, e.g. amines	1
MA	Gas permeation combined with absorption in liquids	1

plants in Europe, as reported by JÖNSSON et al. (2001), TRETTER (2003) and SCHOLWIN et al. (2007), are shown in Table 2.

The high investment and maintenance costs of all existing upgrading techniques are a serious obstacle for a broad application of upgrading for LFG. Therefore, a new upgrading technique, the BABIU process, has been developed at our institute.

2.3 BABIU – a new upgrading technique

The BABIU process (Figure 1) has been developed at our institute in the year 2006. The principle of the BABIU technique is the absorption of CO_2 by hydrated (=weathered) MSWI bottom ash. BABIU means "**B**ottom **a**sh treatment and **bi**ogas **u**pgrading", because the process can be regarded as a treatment process for MSWI bottom ash as well. The subject now is primarily upgrading of LFG, and therefore, no comparison with other treatment techniques for ashes is made here.

Thirteen pilot scale experiments for optimization of the BABIU process were conducted in our laboratory during 2006. Samples (250 kg each) from two incineration plants from Vienna were used for the experiments. In each of the test runs, 70 to 90 kg of sieved MSWI bottom ash (fraction < 20 mm) have been treated in lab-scale static reactors with synthetic LFG (CH_4 + CO_2). To account for long-term scenarios on landfills and potential overdrawing, a low CH_4 content was used during the experiments. The most important results are:

- Hydration of the bottom ash by intermediate storage ("weathering") is beneficial. During intermediate storage, CaO reacts with water, and $Ca(OH)_2$ is formed. This way, the uptake of CO_2 is facilitated and "reactive" mineral phases of the bottom ash are stabilised. The optimum of duration of the intermediate storage is 14 to 28 days, and the bottom ash (or bottom ash fraction) should be moist during storage.
- To avoid formation of explosive gases, the bottom ash fraction has to be

flushed with an inert gas, e.g. N_2.

- Recirculation of the process gas (upgraded gas) does not enhance the CO_2 uptake. In addition, discontinuous (intermittent) recirculation may cause underpressure, which is not desirable. It is concluded that the process can be conducted without gas recirculation.
- As the experiments were conducted in thermally isolated reactors, the temperatures in laboratory scale were close to those expected in an industrial scale. The observed temperature maxima demonstrate that no excessive temperature stresses for the material of the static reactors is expected. The static reactors can be constructed similar to "tunnels" used for biological treatment of MSW or composting tunnels.
- To achieve good results, the operator of the plant has to keep to specified ranges of initial water content during storage of MSWI bottom ash, to specified pumping velocities for LFG and to a recommended range for volumes used for flushing.

The uptake of CO_2 leads to considerable enrichment of CH_4 in the offgas. Maximum CH_4 contents of > 90 Vol.% have been observed frequently in the upgraded gas. The mean uptake of the bottom ash fraction < 20 mm is 24.8 kg CO_2 per kg bottom ash (wet substance). The investigations are part of an ongoing scientific project (Mostbauer et al., 2006-2008) and only preliminary results are presented here. More experiments (at least four) are in preparation to quantify the H_2S uptake of the bottom ash and to measure H_2 in the upgraded gas.

In the context of potential utilization of bottom ash as a construction material, a substantial reduction of the leachability of Al and Pb has been observed. Details on metal leachability and pH reduction are already available, but will be published later. In total, the BABIU process reduces metal leachability of MSWI bottom ash and removes CO_2 and H_2S from biogas or LFG. The upgraded gas is very rich in CH_4 and may be used for local energy supply, fed to the gas grid or used as a vehicle fuel. At the moment, the patent approval for a national patent (Austria) is in progress.

Tab. 3: Preliminary results of the BABIU project (January 2007)

Experimental Run No.	CH_4 Input range (Vol.% CH_4)	CH_4 Output Medium (Vol.%CH_4)	CH_4 Output Maximum (Vol.%CH_4)	CO_2 Uptake per t bottom ash (kg/t)	Maximum of temperature (°C)
1 / 2	16.5 – 25.6	62 / 65	85 / 84	32.3 / 36.7	50.5 / 55.3
3	16.6 – 22.6	53	75	26.4	57.5
4	17.8 – 21.4	69	85	30.1	49.8
6 / 7	24.4 – 34.8	68 / 68	86 / 83	21.6 / 23.0	62.5 / 55.9

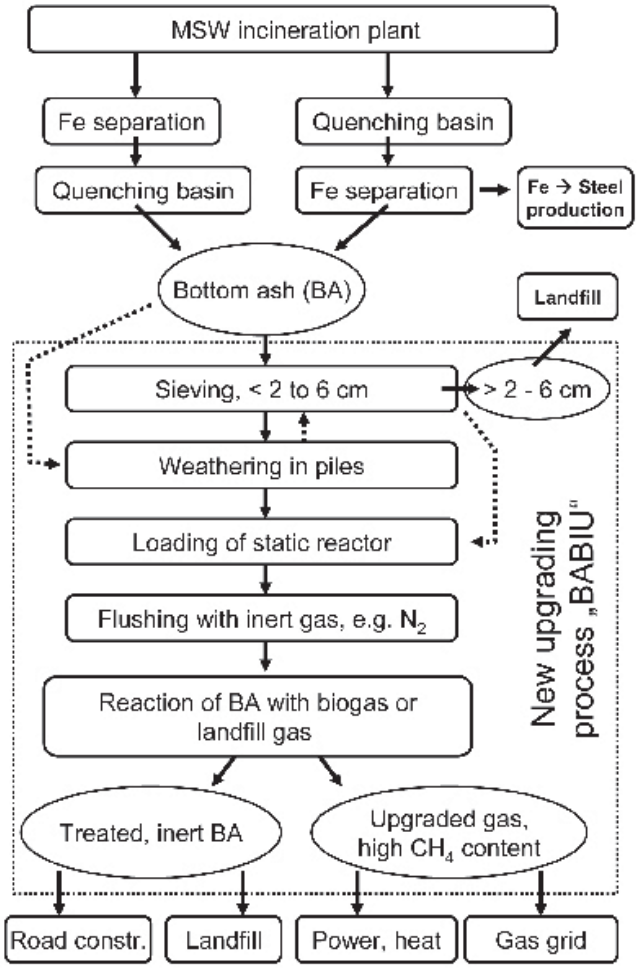


Fig. 1: The BABIU process.

8	17.9 – 40.4	61	83	22.7	59.4
9	17.4 – 23.0	84	92	22.5	54.6
10	26.7 – 30.1	≈ 70	98	15.5	43.7
11	32.4 – 37.9	77	99	17.9	49.1
12 / 13	16.0 – 18.9	82 / 73	95 / 84	24.1 /22.7	57.4 / 53.5

3 Impact assessment – scope

Within the framework of life cycle assessment (LCA), the definition of scopes and goals is always the fundamental step. The scope of the impact assessment as reported in section 4 of this paper is a comparison of emissions of greenhouse gases (CO_2 credits or emissions).

We are aware that shifts in environmental impacts to the offgas may occur when chloride or sulphide content of the landfill gas is extraordinary high, but in a first step these impacts were neglected, because hydrogen sulphide is removed during upgrading or percolation through bio-covers, and LFG with high chloride content is rare in middle Europe. Other potential impacts of upgrading and/or LFG utilization processes are: noise, odour, wastewater. Again, these impacts are of minor importance, with the exception of wastewaters coming from chemical upgrading processes. Because of the use of some toxic chemicals, for example methyl ethanolamine (MEA), chemical upgrading processes were not taken into account. The CO_2 sequestration for the MEA upgrading process is not shown here, but obviously the number for CO_2 credits will be similar to scenario C3 (bio-cover combined with membrane separation upgrading process, see section 4).

4 Definition of scenarios

The usefulness of application of passive or active mitigation systems depends on the size and history of the landfill. For the definition of scenarios, a standardization of the landfill size and assumed time span of active gas collection is useful. A landfill size typical for Austria (but not for Vienna, capital of Austria) was chosen as standard scenario, assuming an annual amount of 100,000 t of degradable waste and non-degradable waste each and an operation time of 20 years (total: 2.0 + 2.0 Mio t). To avoid complexity and in order to provide sample data for a preliminary environmental impact assessment, further assumptions were made:

- Start of aftercare period and active gas collection: immediately with closure
- Operation time for active gas collection and the utilization unit (or flare): 10 years
- Gas collection rate: constantly 60%

Scenarios without upgrading or utilization (A1, A2) can be regarded as reference scenarios, and scenario A1 serves as a "zero aftercare" or "worst case" scenario (Fig. 2). B-scenarios (B1, B2, B3) are conventional utilization scenarios, where LFG is captured and converted into power on-site or in vicinity to the landfill site (Fig. 3). The typical equipment for utilization in B-scenarios is: Gas collection system, dewatering, blower for compression, reciprocating engine or turbine, cooler, generator.

It was assumed that within the projected 10 years of operation, the CH_4 content of the landfill gas would not decrease significantly and that the gas fluxes are adjusted to the decreasing gas production of the landfill. Therefore, no engines/turbines constructed specifically for combustion of lean landfill gas (gas

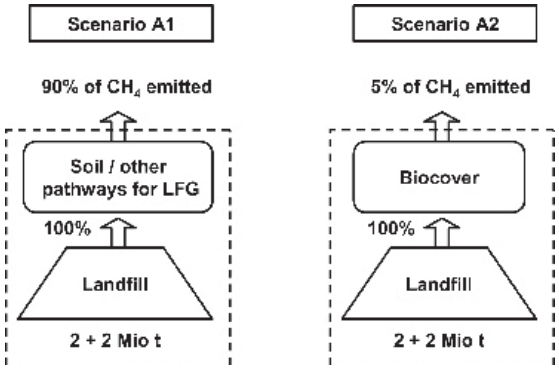


Fig. 2: Scenarios A1, A2

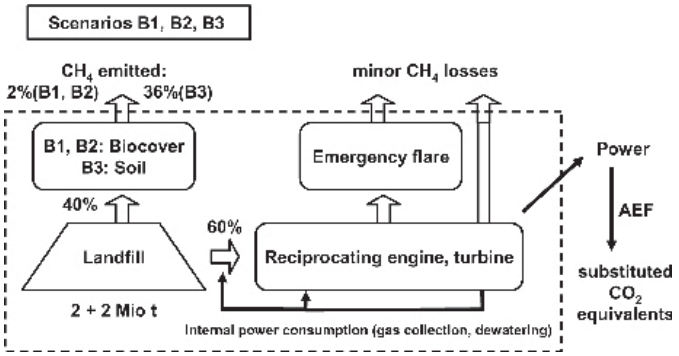


Fig. 3: Scenarios B1, B2, B3

with low CH_4 content) were taken into account. Also, the evaluation of the Haase VOCSI-box is out of the scope of this contribution. Further LCA studies have to be conducted in future to include also lean LFG scenarios and active aerobisation of landfills.

Scenarios B1, B2 and B3 represent the presently most frequently applied utilization system, which is power generation without utilization of the heat (Non-CHP scenarios). Combined heat & power (CHP) concepts were less frequently translated into reality and have a wide range of applications for the heat, like drying of sludge, evaporation of leachate, heating greenhouses for tomatoes, heating schools, artist studios, fish farms (aquaculture) and hydroponics (Voell, 2005, Goldstein, 2006). Because of this large variety of CHP projects, the CHP systems cannot be represented by just one standard scenario. Non-CHP power generation from LFG makes up about two-thirds of the currently operational projects in the USA (Goldstein, 2006) and three-fourths of operational projects in Austria (Rolland & Oliva, 2004). Scenarios B1 and B2 differ only slightly in that the assumed

electric efficiency of power conversion is approximately one fourth lower in scenario B1 (B2: η_{EL} = 33%, B1: η_{EL} = 25%).

For all scenarios where LFG is captured, a constant gas collection rate of 60% was assumed, and in our model 40% of the LFG reaches the top of the landfill, where either 95% of the methane mass (bio-cover scenarios; A2, B1h, B2, C1, C2, C3, C4) or only a marginal percentage (model: 10%, scenario B3) is oxidized biologically. For bio-cover scenarios, the emitted CH_4 amount, related to the CH_4-production of the deposited organic waste, is 0.05 x 40% = 2% or (only for A2) 0.05 x 100% = 5%. The estimation of the methane oxidation rate (95%) is conservative. The internationally increasing number of remediation projects, where optimized bio-covers are applied, points towards further reduction of emissions in future. With high performance bio-covers, methane oxidation rates of over 98% (annual mean) have been observed already in practice (Humer-Huber & Lechner, 2006).

Scenarios C1, C2, C3 and C4 (Fig. 4): Very frequently, the scope of upgrading of LFG or biogas in practice is to sell the high-calorific methane, to feed it to the regional gas grid or to transport it locally to a consumer. In case that the upgraded gas is fed to the gas grid, combined heat and power utilization (CHP) at the final consumer is assumed in the C-scenarios. Differences within the B-scenarios arise from the technique of utilization:

C1.....Pressure swing adsorption (PSA)
C2……Absorption of CO_2 in water (WA)
C3…..Membrane separation (MS)
C4……BABIU upgrading (see section 2.3)

For all utilization scenarios (B, C) except scenario C4 it was assumed that the availability of the system over time is 335 days per year (91.8%). During maintenance (8.2%), the LFG is combusted in an emergency flare. For scenario C4 (BABIU), an

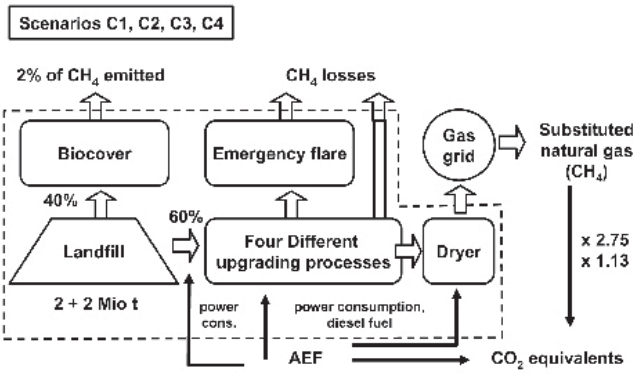


Fig. 4: Scenarios C1, C2, C3, C4

availability estimate of 97% was used, because it is expected that the systems will include redundant modules, and in most cases operation and maintenance can be conducted simultaneously.

Highly methane-enriched gases generated by the upgrading plants may be used for methanol synthesis, for feeding it to the gas grid or for combined heat-and-power generation. In Austria, CHP plants driven by natural gas play an important role for heat and power supply in winter. To account for the increasing importance of CHP, the substituted CO_2-equivalents of the scenarios C1, C2, C3 and C4 were calculated from the CO_2 amount formed during combustion of landfill-derived CH_4 in CHP plants.

5 Methods and input data for calculation of CO_2 equivalents

The method for calculation of the CO_2 emissions from the landfill is briefly described in Figure 5. A projection of the annual gas production of the deposited wastes is obtained using a recent gas production model (IPCC model 2006) and with the use of the default value for the delay time proposed therein by the IPCC (Time before start of the gas generation, 6 months). The rate constant for first-order decay of the organic substance was set to k = 0.1155, which is equivalent to a half-decay time of 6 years. The value for the initial organic matter content of MSW, DDOC(0) = 120 g/kg, was derived from DDOC-, OM- and GBX data (OM: organic matter, GBX: gas formation in laboratory over a period of X days, X = 90 to 375) on commercial and household waste from the Netherlands (Scharff

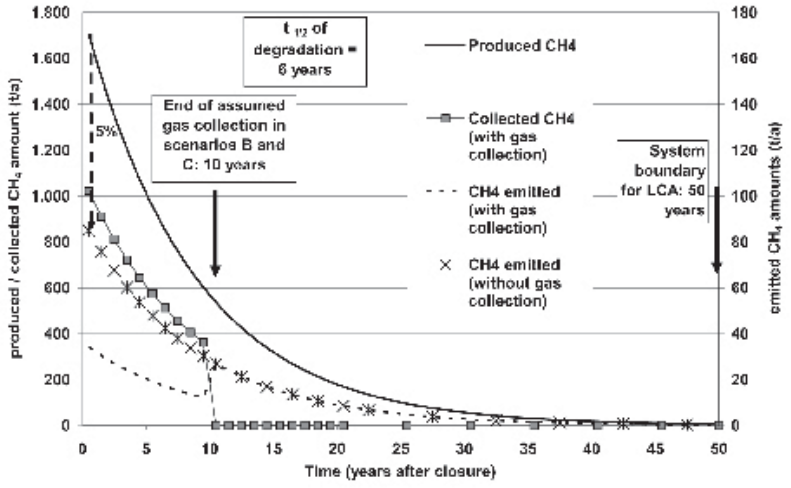


Fig. 5: Projected CH_4 emissions from the landfill

& Jacobs, 2006) and Austria. The dissimilation factor f = 0.75 expresses the fraction of degradable organic substance which is really converted into landfill gas. The value for f was taken from Scharff & Jacobs, 2006. The value for the methane correction factor was set to MCF = 1 which means that the chemical environment within the landfill remains anaerobic.

The CO_2 credits gained by substitution of the electric power supply from fossil resources (coal, oil, natural gases) were calculated with the average Austrian power supply emission factor (AEF) for the year 2004. The AEF2004 was estimated by dividing the national CO_2 emission associated with power and heat production for the public (12,800,000 t CO_2-equiv.) through the national primary power production (64,285 GWh). The resulting number of AEF2004 (0.199 kg/kWh) is quite low, since in Austria more than 50% of the electrical energy is produced by hydroelectric stations.

An equivalence factor of 22 (22 = 23 – 1, GWP_{100} = 23) was used to convert the mass of CH_4 (t) into CO_2-equivalents (t). Data on CH_4 losses and energy consumption of upgrading techniques and utilization processes were collected from the literature. Table 4 contains the input data and the most relevant references. The internal power consumption of the systems with active gas collection (B, C) consists of the energy demand for the gas collection and the power consumption for upgrading (including H_2S removal as a pre-cleaning step). For the specific power consumption of gas collection, an estimation of 0.29 kWh/m^3 collected gas was derived from the data provided by Hornbachner et al. (2005). The electric efficiency of reciprocating engines (33%) was taken from Bove & Lunghi (2006), which is in good agreement with producer specifications, as reviewed by the BHKW Infozentrum Rastatt (2001) and Tallner (2004). An electric efficiency η_{EL} of energy conversion of 33% was applied for scenarios B2 and B3. With decreasing gas amounts of the modelled landfill, the electric efficiency may decrease somewhat. To account for this and for potential use of engines (turbines, reciprocating engines etc.) of lower conversion efficiency, a 25% lower electric efficiency was assumed for scenario B1. Virtually, scenario B1 is the "low conversion efficiency" scenario.

To calculate the CO_2 equivalents from the amount of substituted natural CH_4 in scenario C1, C2, C3 and C4, the mass of CH_4 was multiplied with the stoichiometric factor 2.75 (CO_2/CH_4) and a second factor to account for losses coming from extraction and distribution of natural gas (13%).

The power consumption of the three existing upgrading processes (PSA, WA, MS) depends strongly on the capacity of the upgrading plant. The IPCC model predicts a decrease of the collected gas amount from 196 Nm^3/h (mean in first year after closure) to 69 Nm^3/h (10 years after closure). Therefore, the specific power consumption for upgrading to gas grid quality (per m^3 gas) for a capacity of 200 Nm^3/h to 250 Nm^3/h – as published by Tretter (2003) - was taken into account. The energy demand of the BABIU process will consist of diesel supply for operation of the front-end loader and electricity for blowers and refrigeration coolers.

Tab. 4: Input data for calculation of the CO_2 equivalents (Remark: Additional input data in text of chapter 4 and 5)

Process / Scenario	Type of LCA input	Dimension	Best estimate	References / data sources / comments
Degradation in landfill, all scenarios	CH_4 mass	t	14,005	CH_4 produced during 10 a, IPCC model calculation
	Landfill gas volume	Mio Nm^3	17.94	Gas amount produced during 10 a, IPCC model
B1, B2, B3, C1, C2, C3, C4	Energy consumption	kWh / m^3	0.29	Gas collection, Hornbachner et al. (2006)
Conversion to power, B2, B3	Electric efficiency	%	33	Bove & Lunghi (2006) and producers of engines
Conversion to power, B1	Electric efficiency	%	25	25% = approximately 0.75 * 33%
Conversion to power B1, B2, B3	H_u	GJ / t	7	Heating value of pure CH_4
Upgrading process C1	Energy consumption	kWh / Nm^3	0.3	Tretter (2003)
Upgrading process C2	Energy consumption	kWh / Nm^3	0.3	Tretter (2003)
Upgrading process C3	Energy consumption	kWh / Nm^3	0.08	Tretter (2003)
Upgrading process + drying C4	Energy consumption	kWh / a	77,200	Own estimate
Upgrading process C4	CO_2 sequestration	$kgCO_2$ / t BA	24.5	Mean of pilot scale experiments, per t bottom ash (BA)
Upgrading process C4	Bottom ash mass	t	722,180	Mass needed during 10 years, own estimate
BA handling C4	Diesel consumption	l /t /operation	0.12	Producers of wheeled loaders
Upgrading process C1	CH_4 losses	%	3	Tretter (2003)
Upgrading process C2	CH_4 losses	%	2	Tretter (2003)
Upgrading process C3	CH_4 losses	%	0.1	Tretter (2003)
Upgrading process C4	CH_4 losses	%	1.5	Based on pilot scale experiments
Upgrading process C1, C2, C3	Plant availability	%	91.8	Own estimate, similar to Bove & Lunghi (2006)
Upgrading process C4	Plant availability	%	97	Own estimate

The diesel fuel consumption of front-end loader per t of material was taken from personal information from the producers, assuming that the energy consumption during transport of the bottom ash fines (in and out from a batch reactor) was similar to manipulation of loamy soil. The relative CH_4 loss for the BABIU process was derived from laboratory scale experiments, and it is expected that this loss is very similar or lower at a 1:1 scale, because the porosity of the bottom ash is the most important factor, and storage tanks for intermediate gas storage may be used to reduce losses. Annual CH_4 losses of reciprocating or CHP engines during maintenance are approximately 0.75% of the total annual gas volume (Ronchetti et al., 2002).

6 Expected GHG emissions and CH_4 savings - Results

Not surprisingly, the CH_4 emissions that occur directly at the landfill are the critical GHG emissions. Scenarios with bio-covers are strongly recommended, and scenarios without bio-cover (A1, B3) are the "loosers" of the GHG balance. For the scenarios assuming direct utilisation in combination with a bio-cover (B1, B2), the GHG savings coming from substitution of power generated with fossil fuels and hydroelectric stations is not sufficient to achieve zero GHG emissions in the total system, if the methane oxidation efficiency of the bio-cover is 95%. With this CH_4 oxidation rate, the total emissions of scenario B1 are +5,875 t CO_2 equiv. and that of B2 +4,573 t CO_2 equivalents. Assuming a methane oxidation efficiency of

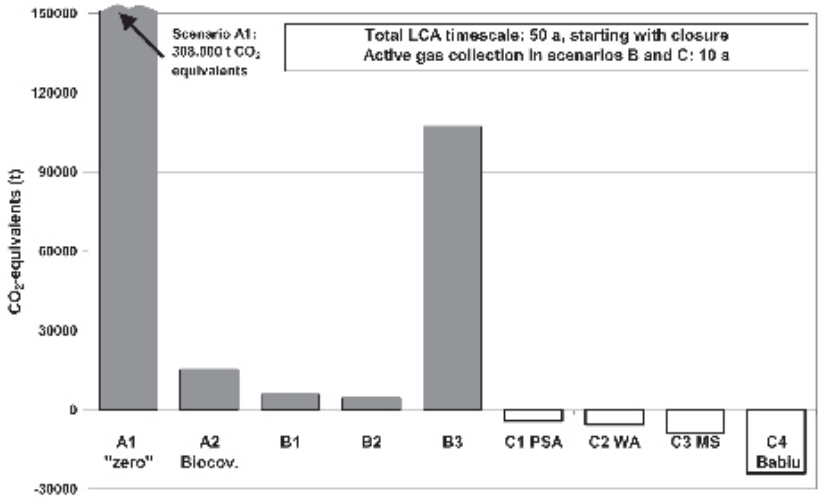


Fig. 6: Balance of GHG emissions

the bio-cover of 98%, the corresponding numbers for B1 were +865 t CO_2 equiv. and for B2 -437 t CO_2 equiv. (not shown in figure). This means that with optimized bio-covers the power conversion scenarios (B1, B2) result in a GHG emission/sequestration close to zero. With additional utilization of heat (CHP), a negative emission (sequestration) is expected. The GHG balance of the upgrading processes (C1 to C4) revealed a positive effect, and in total a CO_2 sequestration, which is attributed to the substitution of natural gas mainly.
In total, the BABIU process is the winner of the GHG balance. The uptake of CO_2 by the bottom ash and the substitution of natural gas are two large contributions to sequestration, and the total CO_2 balance is clearly negative (Figure 6). With the landfill scenario described in chapter 4 (Two Mio t of household waste, bio-cover applied, CH_4 oxidation efficiency 95%) -23,894 t of CO_2 are sequestered if the BABIU process is applied after closure for ten years.

7 Final remarks

Some of the proposed systems – especially C1, C2 and C3 – may require very high investment and maintenance costs. It can be assumed that the eco-efficiency of these scenarios is low compared to the construction of a bio-cover (A2, B1, B2, C4) and also in comparison with the BABIU process itself (C4).

The results presented here do not take into account overdraw scenarios, where a part of the landfill becomes aerobic, with the effect of decreasing CH_4 content in LFG. Of course, if overdraw happens or if it was applied to stabilize a landfill aerobically, the scenarios C (upgrading) or B1 / B2 (passive mitigation) should be favoured. Other promising options in case of overdraw or aerobisation are newer thermal systems, which provide the ability to utilize LFG with low CH_4 content for power of heat production. Additional studies are needed to include overdraw scenarios, but in any case it is clear from Table 3 that the BABIU process can be applied for upgrading of lean landfill gas as well.

According to the Austrian law, the fee for using the distribution network in most parts of Austria is always determined by the OMV, a company which provides and distributes fossil natural gas. As the motivation of the OMV to distribute also biogas in the gas grid is rather small, the corresponding distribution fees are quite high and a serious obstacle for biogas providers. In spite of this, it is very probable that with increasing prices for Russian fossil gas, biogas production will be more economic in future – with or without the use of the existing grid systems.

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