



# Carbon pools and flows during lab-scale degradation of old landfilled waste under different oxygen and water regimes



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## ABSTRACT

Landfill aeration has been proven to accelerate the degradation of organic matter in landfills in comparison to anaerobic decomposition. The present study aims to evaluate pools of organic matter decomposing under aerobic and anaerobic conditions using landfill simulation reactors (LSR) filled with 40 year old waste from a former MSW landfill. The LSR were operated for 27 months, whereby the waste in one pair was kept under anaerobic conditions and the four other LSRs were aerated. Two of the aerated LSR were run with leachate recirculation and water addition and two without. The organic carbon in the solid waste was characterized at the beginning and at the end of the experiments and major carbon flows (e.g. TOC in leachate, gaseous CO<sub>2</sub> and CH<sub>4</sub>) were monitored during operation. After the termination of the experiments, the waste from the anaerobic LSRs exhibited a long-term gas production potential of more than 20 NL kg<sup>-1</sup> dry waste, which corresponded to the mineralization of around 12% of the initial TOC (67 g kg<sup>-1</sup> dry waste). Compared to that, aeration led to threefold decrease in TOC (32–36% of the initial TOC were mineralized), without apparent differences in carbon discharge between the aerobic set ups with and without water addition. Based on the investigation of the carbon pools it could be demonstrated that a bit more than 10% of the initially present organic carbon was transformed into more recalcitrant forms, presumably due to the formation of humic substances. The source of anaerobic degradation could be identified mainly as cellulose which played a minor role during aerobic degradation in the experiment.

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## 1. Introduction

Landfilling still represents the dominant method for the disposal of municipal solid waste (MSW) on a global scale (Organisation for Economic Co-operation and Development, OECD, 2004). While the share of direct MSW landfilling is decreasing in many OECD countries, in particular in Europe due to the European Landfill Directive (Council of the European Union, 1999), there is a shift in non-OECD countries from open dumping or burning to waste disposal in controlled landfills (UNEP, 2010). Due to continuous

emissions of landfill gas and leachate from the deposited waste, MSW landfills still pose a threat to human health and the environment decades to centuries after landfill closure (Laner et al., 2012b). Therefore, old MSW landfills need to be managed also in countries such as Austria, Germany or the Netherlands, where the disposal of untreated MSW has been banned for several years already.

In order to shorten the potentially long periods of aftercare for MSW landfills (Laner et al., 2012a) and to reduce negative impacts associated with the release of landfill gas and leachate, in-situ aeration has been put forward as a measure to enhance waste degradation in landfills and thereby decrease the remaining emission potential (Ritzkowski and Stegmann, 2010). Low-pressure landfill in-situ aeration was used worldwide mainly for biological stabilization to reduce gaseous (CH<sub>4</sub>, odors) as well as leachate emissions (mainly NH<sub>4</sub>) and to decrease landfill volumes (Ritzkowski and Stegmann, 2012).

During landfill in-situ aeration the degradation of organic matter is accelerated by oxidative respiration. Aerating organic fractions of MSW shows various effects during the enhanced degradation caused by immediate oxygen availability. During in-situ aeration the formation of CH<sub>4</sub> was reported to be suppressed

*Abbreviations:* Bd, biodegradable fraction; BOD<sub>5</sub>, biological oxygen demand after 5 days; C, carbon; CI, confidence interval; COD, chemical oxygen demand; DW, dry weight; GP<sub>21</sub>, residual gas producing potential; LSR, landfill simulation reactor(s); MSW, municipal solid waste; NL liter, at standard temperature and pressure (0 °C and 1013 mbar); Nm<sup>3</sup> m<sup>3</sup>, at standard temperature and pressure (0 °C and 1013 mbar); RID, refractive index detector; RI<sub>4</sub>, respiration index after 4 days; RMSE, root mean square error; SD, standard deviation; TOC, total organic carbon; TOC<sub>init</sub>, initial total organic carbon; TW, total weight; WC, water content.

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(Ritzkowski and Stegmann, 2010), with an increased amount of carbon being released as CO<sub>2</sub> (Ritzkowski et al., 2006). In the leachate, NH<sub>4</sub> (He et al., 2011; Heyer et al., 1999) and also chemical as well as biological oxygen demand (Prantl et al., 2006; Ritzkowski et al., 2006) were reported to be reduced during in-situ aeration. Still, the main fraction of total nitrogen during aeration of MSW is considered to remain in the waste material (Fellner and Laner, 2011), even if the NH<sub>4</sub>-concentration in the leachate decreases during aeration.

Aeration measures are generally classified as successful if the waste material is considered biologically stabilized. This biological stabilization is measured in terms of aerobic (respiration index after 4 days, RI<sub>4</sub> – ON S 2027-1 (2002)) or anaerobic (residual gas potential after 21 days, GP<sub>21</sub> – ON S 2027-3 (2004)) biological reactivity. The reduction of these biological parameters was in addition to the differences in gaseous discharges used as a proof of concept for aeration measures, if possible (Hrad et al., 2013; Prantl et al., 2006; Ritzkowski et al., 2006). However, these biological tests only allow for assessing the readily degradable (short-term, 4 respective 21 days) biological available carbon pool of the waste material. These difficulties with the determination of the biodegradable fraction led to the development of a stabilization criterion for in-situ aeration measures: if in full scale more than 90% of the biodegradable fraction determined during lab-scale experiments was degraded, the landfill is considered stabilized (Ritzkowski et al., 2006).

For numerically assessing the performance of the applied aeration measures with respect to gaseous emissions, the aerobic and anaerobic cases are usually directly compared in terms of C-discharge. The mathematical difference between the carbon released during the aerobic and the anaerobic experiments is subsequently used to describe an acceleration effect of in-situ aeration measures in comparison to the non-aerated cases (Hrad et al., 2013; Prantl et al., 2006; Ritzkowski et al., 2006; Ritzkowski and Stegmann, 2010). This direct comparison of the anaerobic and the aerobic case would require that the degraded C-pools for both cases corresponded to each other, which has not been investigated so far.

Moreover, despite the fact that numerous lab scale aeration experiments have been conducted so far, no entire balance of all carbon flows and pools has yet been accomplished. Hence, the goal of this study is to investigate the extent of carbon mobilization during in-situ aeration of landfilled MSW in comparison to anaerobic landfill conditions. Thereto all flows (gaseous and leachate emissions) and pools of carbon are recorded for altogether six landfill simulation reactors, which are operated differently: two under anaerobic conditions, two under aerobic conditions without water addition and two under aerobic conditions with water addition. For all reactors cross referenced carbon balances are presented, including a discussion about carbon quality change for the respective settings as well as providing insight into the influence of water addition on aerobic degradation.

The latter was investigated for two reasons: firstly, the excavated waste from the landfill was very dry due to the conditions at the site (climate, low-permeability soil cover) and therefore it was of interest, whether the low water content (WC) could have an inhibitory effect on aerobic waste degradation during the full-scale project. And secondly, higher moisture content is generally considered to accelerate aerobic degradation of MSW (Pommier et al., 2008) and thereby might have an effect on carbon mobilization.

## 2. Material and methods

During the experiment, waste material from an old landfill was placed into six LSR and then incubated under different conditions: aerobic wet, aerobic dry, and anaerobic. Organic carbon flows and

pools were monitored by means of solid waste analysis, gas and leachate flow as well as concentration measurements. Mass balances were established considering the initial and final TOC (total organic carbon) content of the waste as well as for the carbon losses via leachate and gas. Gaseous C-discharge was modeled using a first order degradation model for the anaerobic and aerobic treatments. Changes of the carbon pool were observed via biological testing and lignin and cellulose analysis.

### 2.1. Experimental setup

#### 2.1.1. Waste material origin

The waste material was gathered from a landfill containing approximately 220,000 m<sup>3</sup> (fresh matter) of waste with an average deposition height of 3.7 m. The landfilled waste mainly consisted of MSW (66% moist mass), excavated soil (18% moist mass) and construction and demolition waste (16% moist mass, Brandstätter et al., 2014a). Landfilling took place at the site from 1965 to 1974. The excavated material was sieved with a mesh width of 20 mm. Before placing the material into the respective LSR, the total amount of waste material (~430 kg) was pooled and thoroughly mixed with shovels. The material was selected from landfill areas comparatively rich in organics, since one aim of the experiment was to determine the maximum degradation potential (Landfill A, Brandstätter et al., 2014b). Upon LSR installation, the material showed a WC of 23.2% and a dry bulk density of 846 (kg m<sup>-3</sup>).

#### 2.1.2. Landfill simulation reactor experiments

The experimental vessels consisted of stainless steel and they have already been used in a similar manner (Ritzkowski et al., 2006). The total volume of each vessel was 121 L. Analogous to Ritzkowski et al. (2006) the LSR were operated in duplicates, meaning that two LSR (A and B) were operated under the same conditions.

Three distinct experimental conditions (from here on named treatments) were applied: under the first treatment the waste material was aerated, water was added and leachate was recirculated (aerated wet). In the second treatment, only aeration took place, without leachate recirculation and no water addition (aerated dry). And in the third treatment, leachate was recirculated, but the material was kept anaerobic (anaerobic). The decision to not add water during the second treatment was made to investigate the impact of water addition and leachate recirculation on the acceleration of aerobic waste degradation.

The common design elements of all six LSR were as follows: To prevent water accumulation at the bottom of the waste columns, 8 cm from the LSR bottom a fine meshed grid was emplaced. This measure insured that excess water could drain out by gravity. The discharged leachate was captured in a closed container just before sampling. The temperature inside the waste columns was controlled by coating the LSR with insulation wool (2 cm thickness) and placing a temperature sensor in the center of the waste material. This sensor was connected to a thermostat, controlling a heating wire, which had been placed between the LSR wall and the insulation wool (see Fig. 1). After placing the waste material in the LSR it was slightly compacted with a closed steel pipe (Ø ~10 cm).

For the aeration treatments, air was introduced into the respective LSR via gas flasks (synthetic air, LINDE, Germany). One flask provided air for each treatment duplicate. From the gas flask the air flowed through a rotameter for pressure reduction, then through a splitter followed by non-return valves for each LSR and consequently by additional pressure reducing valves. All the tubes consisted of polyvinyl chloride (PVC). Afterward the air was streamed through digital mass flow meters (EM1NH, Sensirion

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