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Brandstätter, Ch.; Laner, D.; Fellner, J. (2015) “C- and N-Balances from a Landfill Aeration Experiment” In: CD-Proceedings Sardinia 2015, 15th International Waste Management and Landfill Symposium, 5-9 Oct. 2015, S. Margherita di Pula (Cagliari), Sardinia, Ed.: Cossu, R., Pinjing, H.; Kjeldsen, P. Matsufuji, Y.; Reinhart, D.; Stegmann, R., CISA Publisher, Sardina, paper 131, p 1-7, ISSN 2282-0027

C- AND N- BALANCES FROM A LANDFILL AERATION EXPERIMENT

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SUMMARY: In-situ aeration is increasingly applied to stabilize old MSW-landfills. However, a reliable assessment of the method´s overall influence on the long-term emission potential is still largely missing. In a first attempt, overall Carbon and Nitrogen balances during the in-situ aeration process are provided, leading to an increased process understanding. We could show a significant decrease in the biodegradable Carbon pool during the degradation experiment and the impact of water addition on in-situ aeration was investigated. While the overall Carbon balance was not affected by the addition of water, the Nitrogen removal could be enhanced with the addition of water. The main lessons learned from this experiment are firstly that leachate recirculation (using leachate with significant organic strength), may enhance denitrification during in-situ aeration. Thus, this could represent an option to enhance denitrification for full-scale implementations of in-situ aeration. Secondly, the formation of laughing gas during dry in-situ aeration (in the range of 200 mg N_2O/kg waste) significantly reduces the overall reduction in greenhouse gas emissions expected by in-situ aeration. In summary, during in situ aeration of old MSW 2/3 of the initial TOC remained in the solid materials, as did 75-90 % of the initial Nitrogen. The Nitrogen removal was strongly affected by the experimental setup.

1. INTRODUCTION

Landfilling of waste remains a crucial part of the global waste management. Even in the European Union, where landfilling of untreated MSW-waste is decreasing, many old landfill sites prevail. Landfills generally pose a threat to human health and environment, mainly in terms of gaseous and leachate emissions. A potential method of reducing the emission potential and thus the environmental threat is the method of in-situ aeration. This method, mainly applied as low pressure aeration (Ritzkowski and Stegmann, 2012), is increasingly applied on landfills all around the world.

Nonetheless, the assessment of the outcome of the aeration process until now lacks information of long-term emission potential and a profound comparison with a status quo bioreactor landfill. Many benefits of the in-situ aeration process were discussed in the literature (Hrad et al., 2013; Prantl et al., 2006; Ritzkowski et al., 2006), including the reduction of NH_4-N in the leachate and a decrease of the remaining gas producing potential of the landfilled waste.

What was missing so far, are complete and reliable Carbon and Nitrogen balances, respective transfer coefficients, giving a first hint to the long-term emission potential after the application of in-situ aeration of old landfilled waste. This was done for a laboratory experiment and will be presented in this article. In classical bioreactor landfills, Carbon is emitted mainly via the gas-phase

Proceedings Sardinia 2015, Fifteenth International Waste Management and Landfill Symposium
S. Margherita di Pula, Cagliari, Italy; 5 – 9 October 2015

and Nitrogen via the landfill leachate under mostly anaerobic conditions.

During in-situ aeration the gaseous Carbon emissions are even increased (Brandstätter et al., 2015a; Ritzkowski et al., 2006), while for Nitrogen the release patterns are less uniform. Typically, a decrease of the NH_4-N concentration in the leachate is well acknowledged during in-situ aeration (Berge et al., 2006), which was consequently termed as Nitrogen removal efficiency. However, a decrease in the leachate NH_4-N could also only denote a temporary decrease of the leachate concentration and an accumulation in microbial biomass or in the solid matrix of the MSW. Thus an overall mass balance for N may help highlighting the future fate of Nitrogen after waste stabilization via in-situ aeration. More specific details about these respective balances can be found for Carbon in Brandstätter et al. (2015a) and for Nitrogen in Brandstätter et al. (2015b, submitted).

2. MATERIAL AND METHODS

2.1 Laboratory Setup

The conducted laboratory experiment lasted in total for 823 days with an aerobic phase of 731 days for the aerated treatments. The experimental setup was unique, as in one aerated setup no water was added or recirculated (Fig. 1). In total three distinct setups (treatments) were applied with two reactors each: first an aerated wet setup, with water addition, leachate recirculation and sampling as well as aeration, secondly aerated dry, a treatment without water addition and thirdly an anaerobic case as a reference situation for a classical bioreactor approach.

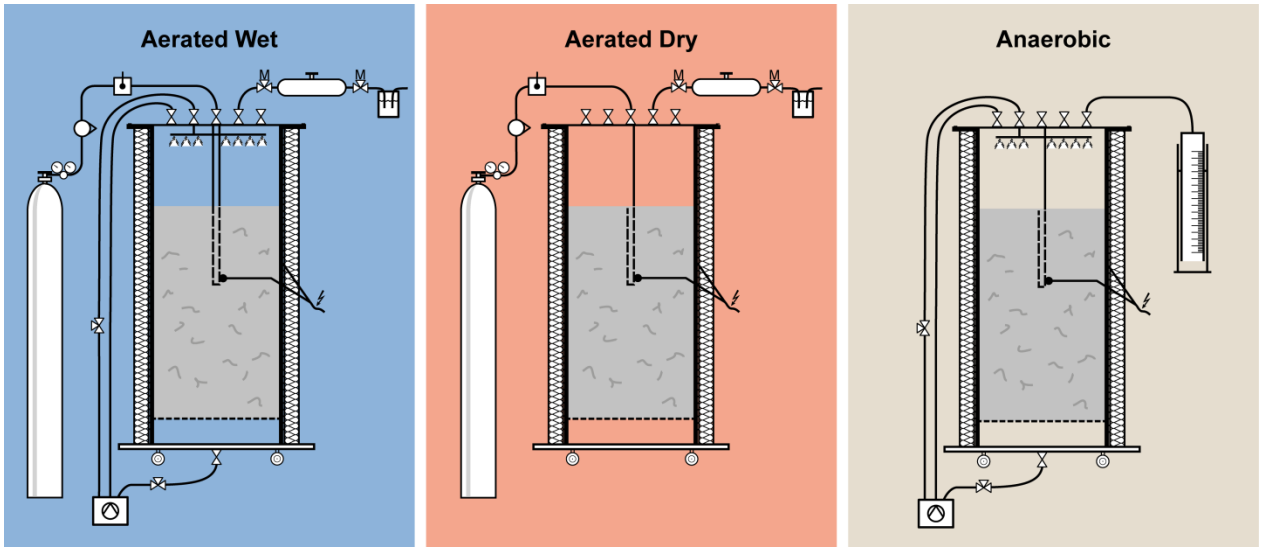

Figure 1. Experimental setup for three differing treatments (cf. Brandstätter et al. 2015a)

The air was introduced via gas flasks, the amount of the initially emplaced waste mass was roughly 90 kg for each reactor and during aeration about 40 Nm^3 of synthetic air was introduced. The material was collected from an old landfill (closed in 1974), and during sampling the waste was sieved (20 mm screen size) and vigorously mixed before emplacement.

For the treatments with water addition, roughly 50 L of water was added over time and continuously removed for sampling (in total 27 leachate samples were taken per reactor).

2.2 Analytical methods

The air flow for the aerated treatments was continuously measured at the gas inlet with a mass flow meter (EM1NH, Sensirion AG, Switzerland), while at the outflow the gas volume was mathematically corrected for water uptake with the assumption of full saturation. The amount of gas produced for the anaerobic case was assessed with eudiometers, plastic cylinders turned upside down in an acidic solution. The solution has to be sour to prevent CO_2 from dissolving.

The respective gas concentrations (CO_2, CH_4 and O_2) were assessed with a mobile device (GFM 400, Gas Data Limited, Coventry, UK). The concentrations of N_2O were determined (52 samples per reactor) via a GC-system in two different laboratories for cross-evaluation. The amount on NH_3 was determined via stripping in an acidic solution, emplaced at the off-gas stream of the aerated reactors and with Dräger-tubes in a monthly interval for the anaerobic case.

The solid TOC concentrations were assessed with a MACRO CHNS analyzer (Elementar Analysensysteme GmbH, Hanau, Germany) as a calculated difference between TC and TIC. The total Nitrogen was assessed using the same analyzer. The methods for additional parameters for the Carbon balance like lignin and cellulose are described in detail by Brandstätter et al. (2015a).

For the leachate losses of Carbon and Nitrogen, the species TOC (DIN EN 1484), respective NH_4-N, NO_3-N and TN were determined (DIN EN ISO 11732, DIN EN ISO 13395 and DIN EN ISO 11905-1, respectively).

2.3. Mass balancing

For the gaseous losses of C via CH_4 and CO_2 the respective amount (aerobic or anaerobic) was calculated by applying the general gas equation. For the aerobic case the mass flows of Carbon were determined using measurements of gas concentration (CO_2 and CH_4) and the measured volumetric flow rates of air injection. For the latter continuous records were available, whereas the gas concentrations were analyzed only once per weekday. Thus, gas concentrations had to be linearly interpolated between the single measurements. The gaseous losses of N_2 were derived via mass balancing. For leachate losses the amount of withdrawn sample were multiplied with the respective concentrations. And solid losses were calculated as the difference between the initial and final amount of the respective species.

3. RESULTS AND DISCUSSION

3.1. Carbon

During the experimental degradation of old landfilled MSW, roughly 1/3 of the initial TOC was released via gas phase (Fig. 2) during aeration, while only half of it during degradation in the anaerobic setup. Most surprising was the finding, that water addition did not affect the Carbon degradation for the aerated treatments.

We could observe an alteration of the Carbon quality during the degradation experiment (Table 1), as cellulose was completely exhausted for all treatments. For the aerated treatments, a certain amount of lignification was observed, indicating increased biological stabilization.

As was expected, the contribution of leachate losses was only of minor significance, even for the anaerobic treatment.

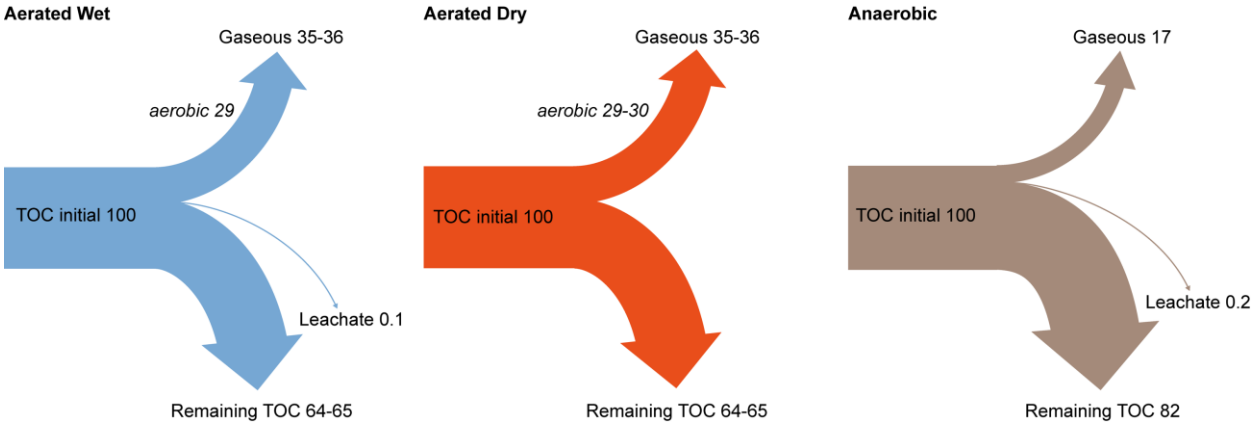

Figure 2. Transfer coefficients Carbon – overview (simplified from Brandstätter et al. (2015a))

Table 1. Transfer coefficients Carbon – detailed

Main category	Sub category	Aerated wet [% TOC_{init}]	Aerated dry [% TOC_{init}]	Anaerobic [% TOC_{init}]
Initial solid material	Cellulose	5.4	4.1	3.3
	TOC_{init}	100	100	100
Gaseous losses	Aerobic phase	29	30	-
	Anaerobic phase	6.4	5.9	17
Leachate losses		0.1		0.2
Remaining solid material	$Lignin_s$	10	12	
	$Non\text{-}Lignin_s$	54	52	82

Notes: Calculations based on Brandstätter et al. 2015a.

3.2. Nitrogen

For the Nitrogen balance, water addition played a significant role (Fig. 3). Denitrification was favoured under aerated wet conditions. Nonetheless, even under relatively favourable conditions for denitrification only 16 % of the initial Nitrogen could be removed via this pathway (Table 2). The leachate losses were relatively higher for the anaerobic case and for the aerated treatments an accumulation of nitrate was observed, while this was not the case for the anaerobic treatment (Table 2).

The lack of leachate recirculation led to a more pronounced laughing gas formation, which might counteract the overall generation of CO_2-credits (Ritzkowski and Stegmann, 2010) through in-situ aeration.

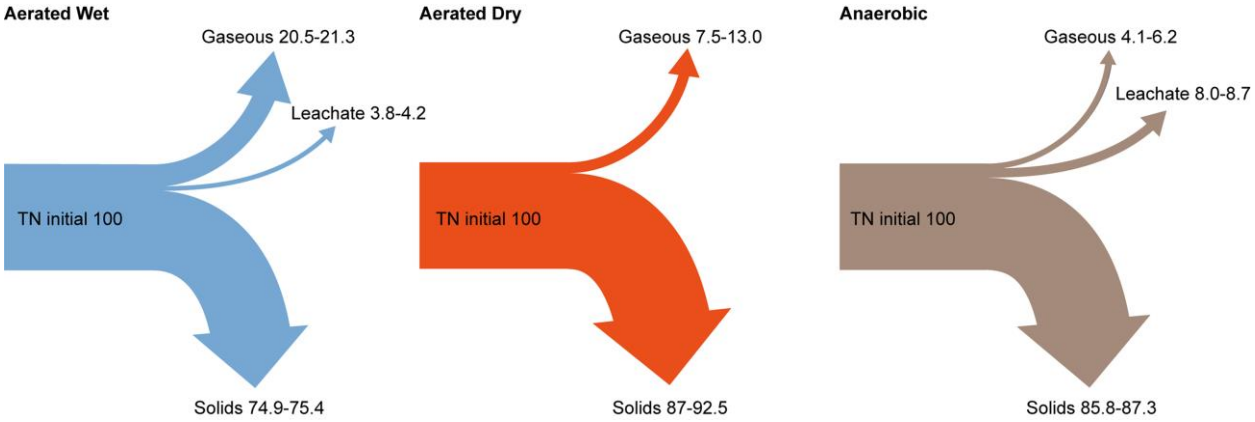

Figure 3. Transfer coefficients Nitrogen – overview (simplified from Brandstätter et al. (2015b))

Table 2. Transfer coefficients Nitrogen – detailed

Main category	Sub category	Aerated wet [% TN_{init}]	Aerated dry [% TN_{init}]	Anaerobic [% TN_{init}]
Initial solid material	NH_4-N	10.0	10.0	10.4
	NO_3-N	0.0	0.0	0.0
	TN_{init}	100.0	100.0	100.0
Gaseous losses	N_2O	1.3	3.9	0.0
	NH_3	0.3	0.7	0.0
	N_2 (indirectly)	16.0	5.7	5.1
Leachate losses	NH_4	2.1		7.7
	NO_3	1.5		0.0
	Total	4.0		8.3
Remaining solid material	NH_4	0.1	0.1	9.7
	NO_3	3.3	15.5	0.1
	TN	78.4	89.7	86.5

Notes: Calculations based on Brandstätter et al. 2015b.

The mechanism of denitrification was strongly affected by the experimental setup; the leachate was recirculated and then left for at least 16 hours in the reservoir prior to sampling. This also led to an accumulation of available Carbon sources, which would be needed for a complete denitrification. In comparison, the system of the aerated dry treatment was most probably faster Carbon depleted, leading to an increased formation of laughing gas.

4. CONCLUSIONS

The main lessons learned from this laboratory experiment are that

(1) in-situ aeration may degrade and transform almost 50% of the organic Carbon present in old landfills. Around one third of the initial Carbon pool is released (mainly via CO_2) independent whether water/leachate is added or not. The other 15% of Carbon are transformed (lignification) into more reluctant compounds, indicating increased biological stabilization.

(2) with respect to Nitrogen release and transformation in-situ aeration is strongly affected by the recirculation of leachate. A significantly smaller share of the initial N pool (in comparison to Carbon) is mineralized. Results indicate that only about 25% of the initial N pool is mineralized irrespective the treatment applied. From this 25% of mineralized N pool different shares are finally released. Whereas for the aerated wet treatment almost all Nitrogen mineralized has been emitted (mainly as N_2), more than half of the mineralized N remained in the aerated dry reactors. These reactors also showed the highest amount of N_2O emissions; about 40% of N release occurred via this path, thereby counteracting the aim of mitigating greenhouse gas emissions from a landfill through in-situ aeration.

ACKNOWLEDGEMENTS

This research was funded by the Kommunalkredit Public Consulting with the support of the Austrian Federal Ministry of Agriculture, Forestry, Environment and Water Management. Many thanks to our project partners WGM (Wiener Gewässer Managment), namely to Michael Enna. Many thanks also to our laboratory staff, namely to Manuel Hahn and Philipp Aschenbrenner for the analyses and great assistance during the sampling.

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