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Gebert, Julia; Knoblauch, C.

**Publication date**  
2017

**Published in**  
Proceedings Sardinia 2017. Sixteenth International Waste Management and Landfill Symposium, 2 - 6 October 2017, S. Margherita di Pula, Cagliari, Italy

**Citation (APA)**  
Gebert, J., & Knoblauch, C. (2017). Long-term gas generation from landfilled dredged sediment. In *Proceedings Sardinia 2017. Sixteenth International Waste Management and Landfill Symposium, 2 - 6 October 2017, S. Margherita di Pula, Cagliari, Italy* CISA Publisher.

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# LONG-TERM GAS GENERATION FROM LANDFILLED DREDGED SEDIMENT

J. GEBERT\*, C. KNOBLAUCH\*\*

\* *Hamburg Port Authority (HPA), Neuer Wandrahm 4, 20457 Hamburg, Germany, and Delft University of Technology, Department of Geoscience & Engineering, Stevinweg 1, 2628 CN Delft, The Netherlands*

\*\* *University of Hamburg, Institute of Soil Science, Allende-Platz 2, 20146 Hamburg, Germany*

**SUMMARY:** A share of the sediments dredged from the port of Hamburg is disposed of on mono-landfills. The anaerobic degradation of sediment organic matter leads to considerable gas production and subsequently necessitates gas treatment. However, little is known about the gas potential of dredged material, hence validated input parameters for gas production modelling are missing. On the occasion of drillings performed for the installation of inclinometers, the waste body of one of the two landfills operated by Hamburg Port Authority (HPA) was sampled, intercepting individual waste layers of known age. Complementing field measurements of gas production, these samples were analysed for gas generation in a long-term laboratory incubation experiment carried out for 695 days. It was found that the total residual gas potential of the deposited dredged material ranged between 2 and 12 m<sup>3</sup>/Mg, relating to 3 to 11% of degraded organic matter. Correlation analyses with material properties suggest a strong role of nitrogen, with the gas potential increasing with total nitrogen content and the share of degradable carbon decreasing with increasing TOC/TN ratio. The total gas potential as revealed from the long-term test was well-correlated to short-term values, but outreached the commonly applied potential after 21 days by the factor of four. The data substantiate the state of knowledge on gas production from the large mineral waste stream of dredged material and serve to improve gas production modelling for these kinds of wastes. The strong correlation of gas potential to TN suggests that TN may serve as a proxy to estimate total gas potential. Future investigations will focus on density fractionation of organic matter in order to shed light on the role nitrogen in the light, easily degradable organic matter fractions.

## 1. INTRODUCTION

In order to maintain the navigable depth in Germany's largest port, Hamburg Port Authority (HPA) annually contracts several million cubic meters of dredging of Elbe river sediment. While most of the material is relocated into the river downstream of Hamburg, treatment and disposal on land is required for the share of sediment with an increased level of contamination. Following classification and dewatering in the METHA plant (Detzner et al., 1997), the contaminant-bearing fine fraction is disposed of in one of the two mono-landfills (landfills Francop and Feldhöfe) operated by Hamburg Port Authority. Alternatively, the dredged material is flushed

into dewatering fields. Here, the natural processes of settlement, gravitational dewatering and evapotranspiration, aided by turning of the resulting heaps, also deliver an earth-like material fit for disposal in the landfills or re-use in construction of mineral liners (Detzner and Knies, 2004; LAGA, 2008), profiling layers, backfilling of harbour basins (Gutbrod, 2016) or, possibly, dike construction (Gröngröft et al., 2014; Saathoff et al., 2013). As can be expected from the natural content of organic matter, especially in the more finely-textured sediments, gas generation is observed on the two landfills (Gebert et al., 2015). Currently, biological as well as thermal gas treatment technologies are employed. However, little is known about the long-term gas potential of the material and its relationship to sediment properties. In 2015, drillings for the placement of inclinometers were carried out on Feldhöfe landfill, intercepting the most recent waste layers down to the oldest material at the base, providing the possibility to sample and analyse the disposed dredged material between 2 and 14 years of age. All layers consisted of material previously treated in dewatering fields.

The purpose of the investigation was to generate information on the total gas potential of dredged sediments in order to improve gas production modelling required for the choice of the appropriate gas treatment technology.

## 2. METHODS

The waste samples were taken directly from the open screw of the drill and packed on site into gas-tight containers. In the laboratory, 100 g of dredged material were placed in triplicate into 500 ml glass bottles and 30 g of anaerobic water added. The bottle headspace was flushed with 100% N<sub>2</sub> and samples incubated at 36 °C in the dark. Total gas generation was measured using a pressure gauge, additionally the evolution of CH<sub>4</sub> and CO<sub>2</sub> was monitored by gas-chromatographic analyses of the headspace.

Total generation of CH<sub>4</sub> and CO<sub>2</sub> was calculated from the concentration measurements in combination with the pressure readings. Further, the amount of produced CO<sub>2</sub> was corrected for the share dissolved in water and precipitated, based on the pH value of the solid sample. As to date, gas production was monitored over a period of 695 days.

The analysis of solids properties included particle size distribution, TOC (total organic carbon), TIC (total inorganic carbon), TN (total nitrogen), pH-value, electric conductivity, water content, and respiration activity (AT<sub>4</sub> test).

## 3. RESULTS AND DISCUSSION

### 3.1 Standard material properties

Selected properties of the investigated materials are summarized in Table 1. From the span between minimum and maximum values for each parameter, marked in bold font, it is apparent that the nature of the disposed dredged material varied quite significantly. The sample collective comprised highly sandy (#10) to very silty (#1) materials, TOC and TN contents spanned a factor of about 3 to 4. Great differences were found for the water content, ranging between 13 and 52% DW. All samples contained calcium carbonate, as evidenced by the pH values between pH 7 and pH 8. Respiratory activity overall was well below the threshold of 5 mg/g DW stipulated by the German landfill order (DepV, 2009) for the disposal of mechanically-

biologically pretreated (MBT) wastes.

Table 1. Properties of the investigated dredged material samples. B.s. = below surface; TOC = total organic carbon; TIC = total inorganic carbon; TN = total nitrogen; WC = water content; DW = dry weight; AT<sub>4</sub> = respiration activity in 4 days. Bold = min. and max. values.

| Nr. | Depth b.s. (m) | Sand (%)    | Silt (%)    | Clay (%)    | TOC (%)     | TIC (%)     | TN (%)      | TOC/TN (%)  | WC (% DW)   | pH         | AT <sub>4</sub> (mg O <sub>2</sub> /g) |
|-----|----------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|-------------|------------|----------------------------------------|
| 1   | 1.4-2.8        | 31.5        | <b>60.5</b> | 8.0         | 3.51        | <b>0.92</b> | 0.37        | <b>9.5</b>  | 50.4        | 7.4        | 1.1                                    |
| 2   | 4.3-4.6        | 56.1        | 27.1        | 16.8        | 2.11        | 0.53        | 0.21        | 10.1        | 32.7        | 7.3        | 0.2                                    |
| 3   | 5.4-6.8        | 46.5        | 42.8        | 10.7        | 2.66        | 0.65        | 0.28        | 9.5         | 41.0        | 7.5        | 0.9                                    |
| 4   | 7.1-7.9        | 50.2        | 30.4        | 19.4        | 2.49        | 0.19        | 0.22        | 11.3        | 29.7        | 7.3        | 0.6                                    |
| 5   | 7.9-8.8        | 48.2        | 24.5        | <b>27.3</b> | 2.95        | 0.24        | 0.26        | 11.4        | 36.3        | <b>7.2</b> | 1.0                                    |
| 6   | 9.2-10.4       | 37.1        | 37.2        | 25.8        | <b>3.92</b> | 0.65        | 0.39        | 10.1        | 50.4        | 7.4        | <b>1.6</b>                             |
| 7   | 10.8-11.3      | <b>30.5</b> | 49.9        | 19.6        | 3.83        | 0.81        | <b>0.40</b> | 9.6         | <b>51.8</b> | 7.5        | 1.3                                    |
| 8   | 11.6-12.6      | 37.9        | 35.9        | 26.2        | 3.44        | 0.54        | 0.35        | 9.8         | 45.7        | 7.5        | 1.1                                    |
| 9   | 12.9-14.5      | 78.2        | 13.3        | 8.6         | <b>1.35</b> | 0.40        | 0.13        | 10.4        | 23.2        | 7.8        | 0.4                                    |
| 10  | 15.0-15.9      | <b>83.3</b> | <b>9.9</b>  | <b>6.8</b>  | 1.42        | <b>0.18</b> | <b>0.12</b> | 11.8        | <b>13.4</b> | <b>7.9</b> | <b>0.1</b>                             |
| 11  | 17.1-17.8      | 63.6        | 18.1        | 18.4        | 3.09        | 0.20        | 0.25        | <b>12.4</b> | 30.7        | 7.5        | 0.8                                    |

### 3.2 Gas generation

Similar to the results reported by Gebert et al. (2011) for old municipal solid waste (MSW) samples, gas production followed an asymptotic function (Figure 1, left). The total gas potential, derived from the parameter y<sub>0</sub> of the asymptotic curve fit, ranged between 2 and 12 m<sup>3</sup> Mg<sub>dw</sub><sup>-1</sup> (Figure 1, right) which is in the lower range of values found for aged MSW wastes (Gebert et al., 2011) and MBT wastes (Bockreis and Steinberg, 2004). When compared to the organic carbon content of the samples, the modelled gas potential accounts for a share of 3 to 11% of organic carbon that was degraded during the experiment or will still be degraded in the landfill (Figure 1, stars in right panel).

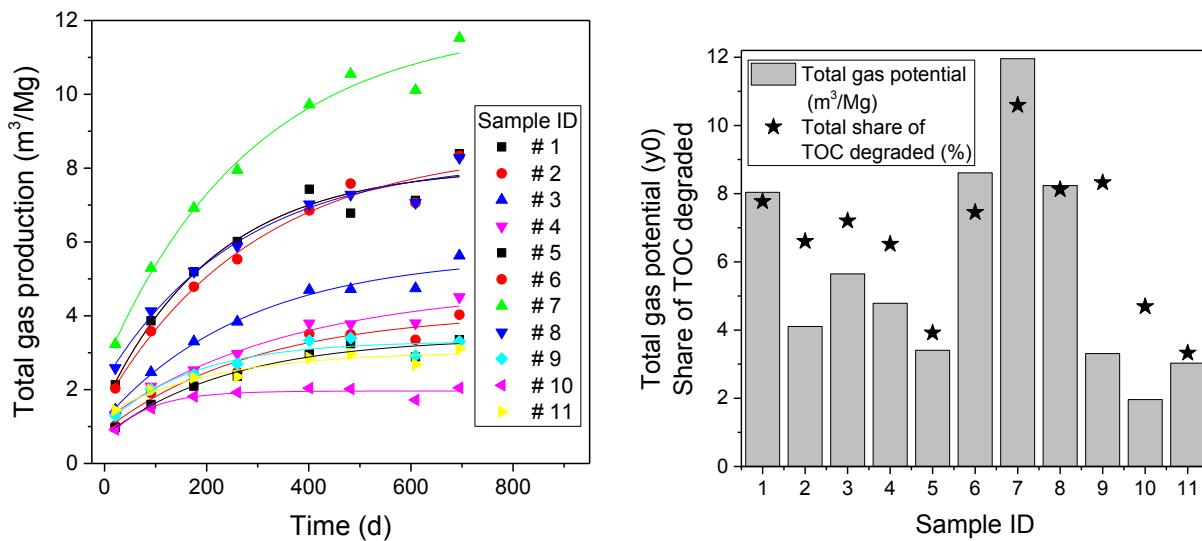


Figure 1: Course of total gas production over 695 days (left) and total gas production and share of degraded TOC as predicted from the asymptotic curve fits (right). Lines = asymptotic fits following the function  $y = A_1 \cdot \exp(-x/t_1) + y_0$ .  $p < 0.01$  for all fits.

Gas production after 21 days (chosen to reflect the GB<sub>21</sub>-value required in the German Landfill Ordinance, DepV, 2009) correlated very well to gas production after 91, 175, 401, 609 and 695 days (Figure 2). The decreasing difference in slopes of the individual regression functions (bold font) with time indicate the asymptotic nature of the time course of gas production already shown in Figure 1 (left). As also indicated by the magnitude of the slopes, the total gas potential as revealed from the long-term incubations and from the corresponding asymptotic fits is significantly higher than suggested by short-term tests such as the GB<sub>21</sub>, in this case by about a factor of four. For MSW samples drilled from three abandoned, non-sanitary landfills in Germany, long-term incubations were also by a factor of 3.5 higher than those obtained after 21 days (Gebert et al., 2011).

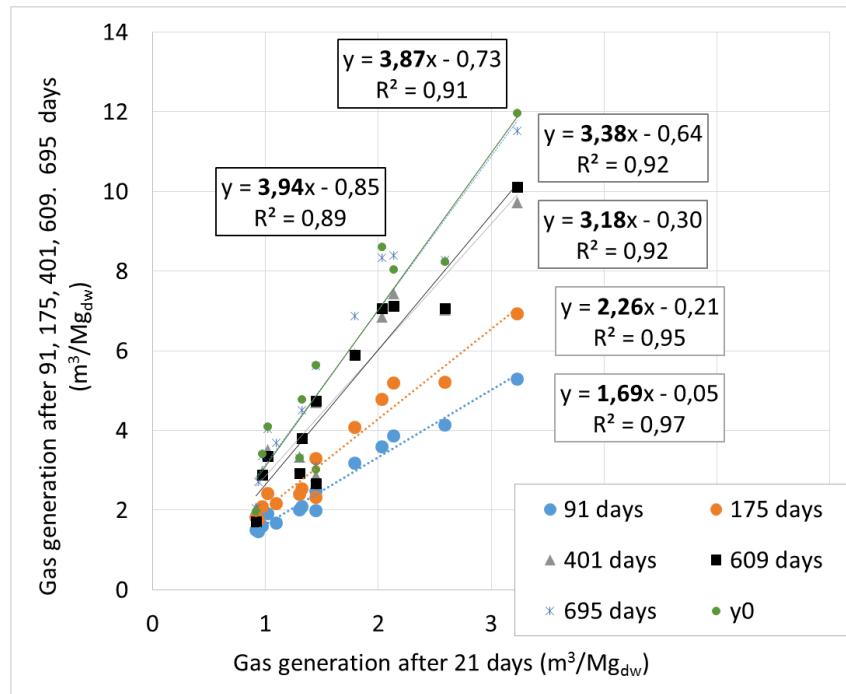


Figure 2: Relationship between short-term, long-term and modelled total gas potential.  $y_0$  = value derived from the asymptotic fits shown in Figure 1.

### 3.3 Relating gas generation to material properties

Relating gas production parameters to the materials' abiotic properties in a simple crosstabulation matrix revealed that the strongest relationship existed between the share of degraded organic carbon (measured) and the ratio of TOC to TN ( $r = -0.85$ , Figure 3 left) and for the relationship between total gas production and TN ( $r = 0.88$ , Figure 3 right). The ratio of TOC to TN is indicative of the degradability of organic matter, with high ratios usually related to less degradable, i.e. more stable organic matter and low ratios related to fresh, degradable organic matter. Indeed, the highest share of organic carbon degraded during the experiment were associated with the lowest TOC/TN ratios and vice versa. Similarly, total measured gas production increased with increasing nitrogen content of the samples. Also, the relationship between respiratory activity and total nitrogen was very close ( $r = 0.90$ ).

The results suggest a high relevance of available nitrogen for the microbial turnover of sediment-bound organic matter and confirm the results by Gebert et al. (2006) for freshly sampled riverine sediments, in which steady-state methanogenesis was correlated positively

with TN content. TN in that study was also positively linked to the share of organic matter in the light density fraction, suggesting that indeed nitrogen was indicative of the more easily degradable share not yet fixed in organomineral compounds.

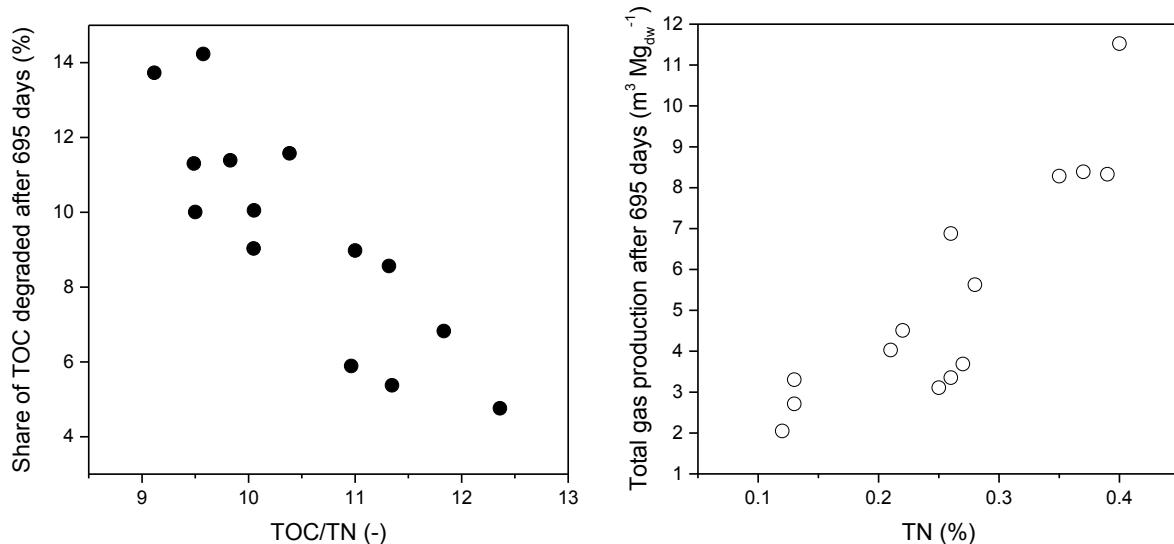


Figure 3: Correlation between TOC/TN and degraded TOC (left) and between TN and gas potential (right).

TOC and TN were correlated on a high level ( $r = 0.97$ ). With respect to the relationship between particle size distribution and organic matter parameters, the highest coefficients of correlation were observed for the share of silt and TN ( $r = 0.85$ ) and silt and initial water content ( $r = 0.89$ ) as well as between TOC and water content ( $r = 0.90$ ). This suggests that the organic matter, mainly responsible for the retention of moisture, is more tightly associated with the silt fraction than with the clay fraction, for which linear regression analysis yielded lower coefficients of correlation ( $r = 0.63$ ). As expected, not only the total amount of organic matter determines gas generation from dredged material but also its quality.

Contrary to expectations, the gas potential was not at all linked to the age of the samples (reflected by the depth of the layer). An inverse relationship would have been expected if the sediments disposed of over the years had been of comparable composition. As seen from Table 1, however, properties varied greatly, yielding different conditions for the microbial breakdown of organic matter which apparently overruled the relevance of age.

#### 4. CONCLUSIONS AND OUTLOOK

Long-term residual gas production of disposed dredged material depends on the nature of the organic matter. Available nitrogen appears to play a key role for degradability of organic matter and, subsequently, for microbial activity.

The gas potential derived from short-term incubations commonly applied in waste characterization, albeit strongly correlated to long-term data, underestimate the total gas potential significantly, here by a factor of four.

The data can serve to fine-tune gas production modelling for these types of wastes as grounds for decisions on appropriate gas collection and treatment options.

Future density fractionation of the investigated samples shall serve to investigate the hypothesis on the role of nitrogen as an indicator for the more degradable organic matter fraction.

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