

From waste to soil—carbon contents, respiration rates and ecotoxicological effects of an uncovered landfill site after 50 years

Andreas Keuter · Heinz-Christian Fründ · Michael Kerth

Received: 30 April 2010 / Accepted: 25 June 2010 / Published online: 15 July 2010
© Springer-Verlag 2010

Abstract

Purpose In this study, the ecotoxicological effects of the soil contaminations at an uncovered landfill site are assessed with two biological tests (earthworm avoidance test and luminous bacteria test). Furthermore, the state of rotting of the organic substance is estimated. Therefore, total organic carbon (TOC) contents and basal respiration rates are measured.

Materials and methods The study has been carried out with polluted samples originating from the old deposit I 27 in Bielefeld, North Rhine-Westphalia, Germany. To assess the ecotoxicological effects, heavy metal contents were determined and earthworm avoidance tests were conducted. Luminous bacteria tests with *Vibrio fischeri* were applied to the soil eluates. Furthermore, the TOC contents and the basal respiration rates were measured regarding to the stability of the organic substance.

Results and discussion Although the determined heavy metal contents showed high values, the results of the biotests do neither indicate an emission of contaminants with the seepage water nor a toxic disturbance of the soil function as a

biological habitat. Beyond that, the respiration rates turned out to be in a range that is typical for natural soils.

Conclusions Due to the aerobic decomposition of the organic matter and the associated development of humic substances, the contaminants contained in the material seem to be mainly immobile. The organic matter is stabilised to a large part. Altogether the results accord to the long-term perspective for the environmental behaviour of artificially aerated waste.

Keywords Aeration · Carbon dynamics · Earthworm avoidance test · Ecotoxicology · Landfill · Respiration · Waste

1 Introduction

In the context of the implementation of the first German waste disposal law (“Abfallbeseitigungsgesetz”; 1972), many landfills were closed and reclaimed. Part of the typical reclamation programme was covering of the wastes with a loam or clay layer to prevent access and to reduce seepage. As is known nowadays, this treatment led to a conservation of the organic substance in the wastes due to the slower and less complete degradation under anaerobic conditions. Recent assessments of former landfill sites which were closed in the 1970s show the necessity of maintenance for several generations (Stegmann et al. 2001; Kruse 1994).

In order to avoid future generations having to deal with today's wastes and to shorten maintenance, experiments aiming at the active aeration of those deposits are being conducted (e.g. biological in situ stabilisation via low-pressure aeration as well as the recirculation of seepage water) (Ritzkowski et al. 2008). Both the results of these experiments and experiments with waste samples carried out in the laboratory indicate a faster decomposi-

Responsible editor: Jaco Vangronsveld

A. Keuter (✉) · H.-C. Fründ
Fachhochschule Osnabrück-University of Applied Sciences,
1940, 49090 Osnabrück, Germany
e-mail: akeuter@gwdg.de

M. Kerth
Dr. Kerth + Lampe Geo-Infometric GmbH,
Walter-Bröker-Ring 17,
32756 Detmold, Germany

Present Address:
A. Keuter
Petrikirchstr. 12 a,
37077 Göttingen, Germany

tion of the organic substance almost avoiding methane emissions and reducing seepage water contamination. (Ritzkowski et al. 2008; Prantl et al. 2006; Östmann 2008). This can be explained by the aerobic decomposition of the organic substance which is not only respired by microbes to carbohydrate and water (mineralization) but also transformed to humic acids or similar substances (Heyer 2003).

The former landfill site I 27 studied in this paper has been covered with loam only in some parts. The investigated area of the former landfill site was not covered at all. This has led to a predominantly aerobic decomposition of the organic substance within the waste. Thus, the studied landfill is in an advanced state of aerobic decomposition—which took place without any active measures.

Waste that is deposited in a landfill becomes part of the pedosphere interacting with the hydrosphere, the atmosphere and the biosphere. Consequently, landfills may be classified as soils (e.g. Lehmann and Stahr 2007). With time, landfills may develop mollic properties (cf. Lehmann and Stahr 2007: soil profile #12) similar to natural terrestrial soils. The aim of the present study is to compare samples taken from the landfill to natural soils with respect to their suitability as a living space for soil organisms (toxic properties) and their intensity of carbon mineralization. This is of practical relevance for the further treatment of the landfill material: can it be excavated like soil or are waste properties prevailing that require special treatment?

2 Material and methods

2.1 The former landfill site I 27

The landfilling took place in a former limestone quarry. Commercial, domestic and industrial waste including building rubble was deposited since the 1950s. The former landfill site is located in North Rhine-Westphalia, Germany.

2.2 Pulse boring and sampling

In April 2009, six pulse borings with a steel tube of 50-mm diameter were carried out to a maximum depth of 6.5 m. Soil samples were taken from 0–20 cm depth, 20–50 cm depth and then downwards in 50-cm steps. The boreholes were equipped with PVC pipes and sealed against the atmosphere. Afterwards, the methane concentration in the soil air-drawn from the boreholes was measured with a handheld measuring device (Dräger Multiwarn).

For the ecotoxicological tests, larger amounts of soil were required. These were taken as additional soil samples from the upper 50 cm of the profile with a spade around the points of pulse boring.

2.3 Laboratory investigations

The pH values were measured according to DIN 19682-13 (2009) in 0.01 m CaCl₂ solution. Total carbon (TC) content was determined by elementary analysis according to DIN ISO 10694 (1995) with the Carbon-Sulfur-Determinator Eltra CS 500. The soil carbonate content (TIC) was measured with the Scheibler-gadget after DIN ISO 10693 (1995). The total organic carbon content (TOC) was calculated by subtracting the TIC from the TC.

Heavy metals were determined by microwave assisted digestion with nitrohydrochloric acid according to DIN ISO 11466 (1997). Weighted samples of 0.7 g (sieved to 2 mm, crushed and dried at 107°C) were used.

The basal respiration was determined with the Isermeyer-method (DIN ISO 16072 (2005)). Deviant from DIN ISO 16072 (2005), the soil samples had not been sieved to 2 mm since our investigations aimed at determining the “real-world” respiration rates of the landfill volume; 40 g of moist soil were filled into 1 l vessels. The mass of coarse material in the samples was weighed after the respiration measurement. From this, respiration rates of fine material could be calculated.

2.4 Ecotoxicological tests

The luminous bacteria test was carried out with the LUMISTOX 300 Luminometer following DIN ISO 11348-3 (2007). The test was applied to soil eluates made according to Römbke et al. (2006). Weighted samples equivalent to 100 g dry mass were shaken with 100 ml Aqua bidest and filtrated with Whatman Polydisc GW filters.

The Earthworm-Avoidance test was carried out according to DIN ISO 17512-1 (2005). Adult earthworms (*Eisenia fetida*) with fresh weights between 300 und 600 mg were used. The animals were from the breed of the University of Applied Sciences Osnabrück. As reference soil, loamy sand taken from an orchard of the University of Applied Sciences Osnabrück was used (pH 6.18, C_{org} 23.5 mg g⁻¹). Additionally, this reference soil was tested against the standard soil RefeSol 01-A (pH 5.67 C_{org} 9.3 mg g⁻¹) of the German Umweltbundesamt.

3 Results

3.1 pH values

The pH values of the soil samples range between 7 and 8. Only few samples show values between 6 and 7. The generally alkaline reaction is in accordance with the high amount of building rubble in the waste.

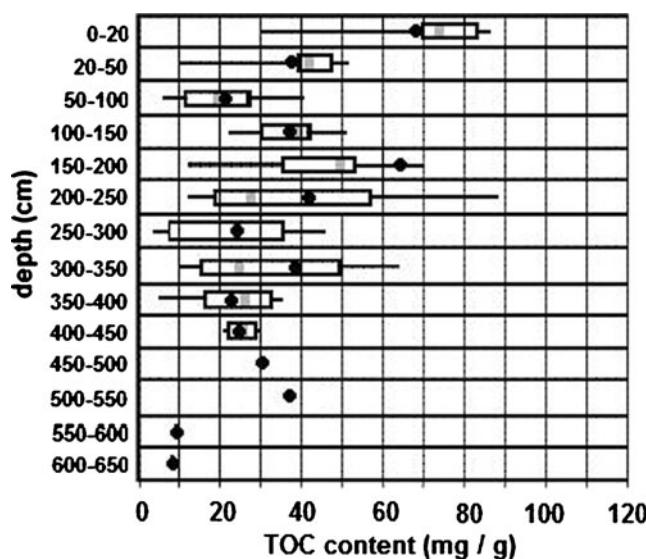


Fig. 1 TOC contents versus depths (the *black dots* represent the arithmetic mean; *grey bars* the median; *boxes* the 25th and the 75th percentile; *whiskers* the 5th and the 95th percentile)

3.2 Composition of the soil air

Only in two of the six measuring wells methane could be detected in the soil air with a maximum concentration of 0.42% v/v.

3.3 TOC contents

The TOC contents of the soil samples varied from 3 to 200 mg g⁻¹ with an arithmetic mean of 35 mg g⁻¹. The

highest average TOC content (75 mg g⁻¹) was found in the uppermost 20 cm below surface. Generally, the TOC contents decreased with increasing depth. However, there was a second maximum with contents of up to 60 mg g⁻¹ between 1 and 2 m below ground (Fig. 1).

3.4 Basal respiration

Respiration rates of the total samples including coarse material range from 0.1 to 9 mg CO₂ kg DM⁻¹ h⁻¹ (Table 1) with an arithmetic mean of 3.00 mg CO₂ kg DM⁻¹ h⁻¹. The highest respiration rates of about 4 mg CO₂ kg DM⁻¹ h⁻¹ on average were found in the upper 20 cm below ground. Between 20 and 550 cm, respiration was lower with rates up to 4 mg CO₂ kg DM⁻¹ h⁻¹; however, the statistical spread is very high in these depths. There were three extreme values measured in RKS-3 at 150–200 cm depth, and RKS-4 at 200–250 cm depth and 300–350 cm depth, respectively. A decrease of average respiration rates is observable in depths of more than 450 cm below ground, where the natural soil begins. Average respiration rates decrease to values of about 1.5 mg CO₂ kg DM⁻¹ h⁻¹.

In order to compare the measured respiration rates with literature data from 2-mm sieved natural soils the values have been recalculated to a fine soil basis taking 267 g kg⁻¹ as average content of coarse material in the landfill samples (see Table 1). The resulting frequency distribution of respiration rates per fine soil mass is shown in Fig. 2. Data on basal respiration of natural soils cover a

Table 1 Basal Respiration of samples taken from six pulse borings at the former landfill site

| Depth (cm) | Respiration rate (mg CO ₂ kg DM ⁻¹ h ⁻¹) | | | | | | Content of coarse material (g kg ⁻¹) | Mean respiration of fine material (mg CO ₂ kg ⁻¹ h ⁻¹) |
|------------|--|-------|-------|-------|-------|-------|--|--|
| | RKS-1 | RKS-2 | RKS-3 | RKS-4 | RKS-5 | RKS-6 | | |
| 0–20 | 4.48 | 6.88 | 1.60 | 5.76 | 5.02 | 2.19 | 4.32 | 5.78 |
| 20–50 | 4.97 | 3.08 | 1.70 | 1.80 | 4.70 | 2.53 | 3.13 | 3.70 |
| 50–100 | 2.10 | 0.63 | 0.68 | 2.40 | 2.31 | 0.18 | 1.38 | 1.71 |
| 100–150 | 3.05 | 4.55 | 2.99 | 2.76 | 2.00 | 2.27 | 2.94 | 4.24 |
| 150–200 | 2.85 | 2.19 | 8.73 | 2.74 | 1.12 | 1.89 | 3.25 | 4.49 |
| 200–250 | 1.91 | 0.89 | 3.83 | 8.84 | 2.26 | 5.71 | 3.91 | 4.97 |
| 250–300 | 0.55 | 1.60 | 2.78 | 3.41 | 4.70 | 4.75 | 2.97 | 4.15 |
| 300–350 | ND | 1.12 | 3.65 | 6.83 | 2.19 | 4.75 | 3.71 | 5.00 |
| 350–400 | ND | 0.10 | 4.63 | 2.39 | 3.53 | 1.49 | 2.43 | 2.97 |
| 400–450 | ND | ND | 3.46 | 3.99 | 3.97 | ND | 3.81 | 5.71 |
| 450–500 | ND | ND | 2.24 | 2.31 | ND | ND | 2.28 | 3.86 |
| 500–550 | ND | ND | 2.23 | 2.46 | ND | ND | 2.35 | 3.59 |
| 550–600 | ND | ND | 1.08 | 0.48 | ND | ND | 0.78 | 1.16 |
| 600–650 | ND | ND | ND | 1.49 | ND | ND | 1.49 | 1.87 |

ND not determined

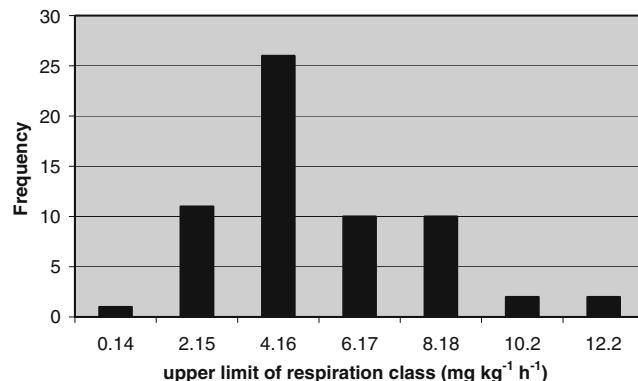


Fig. 2 Frequency distribution of respiration rates of fine material in the pulse-boring samples

wide range. Beck et al. (1997) report a basal respiration of arable soils in the range $0.15\text{--}0.70 \text{ mg CO}_2 \text{ kg DM}^{-1} \text{ h}^{-1}$. Characteristic rates of basal respiration in Swiss soils are given as $4.3 \text{ mg CO}_2 \text{ kg}^{-1} \text{ h}^{-1}$ for arable fields, and $8.5 \text{ mg CO}_2 \text{ kg}^{-1} \text{ h}^{-1}$ for permanent grassland, respectively (Rossier and Dessurault 2004). Sommer et al. (2002) measured the basal respiration in forest soils in southern Germany and found $0.62\text{--}3.72 \text{ mg CO}_2 \text{ kg DM}^{-1} \text{ h}^{-1}$ in A-horizons, and $10\text{--}200 \text{ mg CO}_2 \text{ kg DM}^{-1} \text{ h}^{-1}$ in humus layers of the forest floor. The samples from the investigated landfill are well within this range. There is no indication of an increased respiration in the landfill compared with natural soils.

3.5 Heavy metal contents

The spade samples and a selection of 13 samples from the pulse boring were analysed for heavy metal contents (Table 2). The range of metal concentrations found in the pulse-boring samples from depths below 50 cm was greater than that in the 0–50 cm spade samples. For an approximate conversion to toxic units, the measured concentrations of the different metals were divided by the respective precautionary values of the German Soil Protection Ordinance (Bundes-Bodenschutzverordnung BBodSchV 1999). Background for the derivation of the precautionary values are effects of contaminants on the natural soil functions. Thereby ecotoxicological threshold effect concentrations as well as long-term effects on the filtering and storage functions are considered. Furthermore, natural background values and—in the case of heavy metals—also increased background values due to the geological situation or human settlement are taken into account (König et al. 2003). Thus, heavy metal contents that exceed these precautionary values cannot be considered as a natural background contamination.

The sum of these relative concentrations was taken as an index of the combined heavy metal contamination in the samples. Table 3 shows, that copper is the main contaminant in the pulse-boring samples exceeding the precautionary values by a factor of 9.9 on average (range,

Table 2 Concentration of metals in microwave-HNO₃ extract in samples from spade sampling and pulse boring at the former landfill site (mg kg⁻¹)

| Sampling point | Depths (cm) | Cd | Cr | Cu | Ni | Pb | Zn |
|-----------------------|-------------|------|-----|-------|----|-------|-------|
| Spade sampling | | | | | | | |
| RKS-1 | 0–50 | 2.1 | 93 | 230 | 38 | 698 | 581 |
| RKS-2 | 0–50 | 3.0 | 143 | 169 | 43 | 393 | 803 |
| RKS-3 | 0–50 | 0.3 | 32 | 20 | 19 | 49 | 63 |
| RKS-4 | 0–50 | 2.1 | 54 | 122 | 33 | 208 | 550 |
| RKS-5 | 0–50 | 1.3 | 76 | 221 | 49 | 274 | 760 |
| RKS-6 | 0–50 | 0.4 | 28 | 39 | 22 | 74 | 118 |
| Pulse boring | | | | | | | |
| RKS-1 | 50–100 | 1.0 | 60 | 139 | 47 | 116 | 358 |
| RKS-1 | 150–200 | 1.7 | 726 | 175 | 72 | 176 | 383 |
| RKS-2 | 50–100 | 1.4 | 86 | 244 | 52 | 278 | 814 |
| RKS-2 | 200–250 | 2.2 | 465 | 197 | 66 | 848 | 359 |
| RKS-3 | 50–100 | 0.5 | 38 | 23 | 20 | 44 | 73 |
| RKS-3 | 200–250 | 1.2 | 58 | 1,195 | 22 | 383 | 2,786 |
| RKS-4 | 50–100 | 6.3 | 128 | 2,484 | 44 | 2,153 | 972 |
| RKS-4 | 150–200 | 1.4 | 103 | 295 | 58 | 181 | 565 |
| RKS-4 | 550–600 | 0.3 | 75 | 43 | 47 | 19 | 94 |
| RKS-5 | 50–100 | 45.5 | 58 | 57 | 34 | 36 | 428 |
| RKS-5 | 150–200 | 0.2 | 42 | 29 | 25 | 23 | 80 |
| RKS-6 | 100–150 | 11.1 | 62 | 180 | 38 | 102 | 321 |
| RKS-6 | 150–200 | 0.7 | 81 | 81 | 40 | 109 | 266 |

Table 3 Precautionary values for metal concentrations according to the German Soil Protection Ordinance (BBodSchV), metal concentrations in relation to precautionary values, and sum of relative concentrations as an index of combined metal contamination

| Sampling point | Depths (cm) | Cd | Cr | Cu | Ni | Pb | Zn | Sum (Cd...Zn) |
|--|-------------|------|------|------|-----|------|------|---------------|
| Precautionary values of German soil protection ordnance for loamy soils (mg kg ⁻¹) | | | | | | | | |
| 1 40 50 70 150 | | | | | | | | |
| Concentrations relative to precautionary values for spade samples 60 | | | | | | | | |
| RKS-1 | 0–50 | 2.1 | 1.5 | 5.8 | 0.8 | 10.0 | 3.9 | 24.0 |
| RKS-2 | 0–50 | 3.0 | 2.4 | 4.2 | 0.9 | 5.6 | 5.4 | 21.4 |
| RKS-3 | 0–50 | 0.3 | 0.5 | 0.5 | 0.4 | 0.7 | 0.4 | 2.8 |
| RKS-4 | 0–50 | 2.1 | 0.9 | 3.0 | 0.7 | 3.0 | 3.7 | 13.3 |
| RKS-5 | 0–50 | 1.3 | 1.3 | 5.5 | 1.0 | 3.9 | 5.1 | 18.0 |
| RKS-6 | 0–50 | 0.4 | 0.5 | 1.0 | 0.4 | 1.1 | 0.8 | 4.1 |
| Mean | | 1.5 | 1.2 | 3.3 | 0.7 | 4.0 | 3.2 | 13.9 |
| Concentrations relative to precautionary values for pulse-boring samples | | | | | | | | |
| RKS-1 | 50–100 | 1.0 | 1.0 | 3.5 | 0.9 | 1.7 | 2.4 | 10.4 |
| RKS-1 | 150–200 | 1.7 | 12.1 | 4.4 | 1.4 | 2.5 | 2.6 | 24.7 |
| RKS-2 | 50–100 | 1.4 | 1.4 | 6.1 | 1.0 | 4.0 | 5.4 | 19.4 |
| RKS-2 | 200–250 | 2.2 | 7.8 | 4.9 | 1.3 | 12.1 | 2.4 | 30.7 |
| RKS-3 | 50–100 | 0.5 | 0.6 | 0.6 | 0.4 | 0.6 | 0.5 | 3.2 |
| RKS-3 | 200–250 | 1.2 | 1.0 | 29.9 | 0.4 | 5.5 | 18.6 | 56.5 |
| RKS-4 | 50–100 | 6.3 | 2.1 | 62.1 | 0.9 | 30.8 | 6.5 | 108.7 |
| RKS-4 | 150–200 | 1.4 | 1.7 | 7.4 | 1.2 | 2.6 | 3.8 | 18.0 |
| RKS-4 | 550–600 | 0.3 | 1.3 | 1.1 | 0.9 | 0.3 | 0.6 | 4.4 |
| RKS-5 | 50–100 | 45.5 | 1.0 | 1.4 | 0.7 | 0.5 | 2.9 | 51.9 |
| RKS-5 | 150–200 | 0.2 | 0.7 | 0.7 | 0.5 | 0.3 | 0.5 | 3.0 |
| RKS-6 | 100–150 | 11.1 | 1.0 | 4.5 | 0.8 | 1.5 | 2.1 | 21.0 |
| RKS-6 | 150–200 | 0.7 | 1.3 | 2.0 | 0.8 | 1.6 | 1.8 | 8.2 |
| Mean | | 5.6 | 2.5 | 9.9 | 0.9 | 4.9 | 3.8 | 27.7 |

0.6–62). In the spade samples lead, copper, and zinc are rather similar in their exceedance of the precautionary values leading to a combined contamination that is about half of that in the pulse-boring samples.

A significant positive correlation was found between the heavy metal contamination and the TOC content of the pulse-boring samples from the former landfill (Spearman's rank correlation, Table 4). Rank correlation coefficients for the microbial respiration per gram TOC (Rbas/TOC) versus heavy metal contamination were not significantly different from zero.

Table 4 Correlation (Spearman's rho) between heavy metal contamination, TOC, respiration and respiration relative to TOC in 13 pulse-bored samples from depths below 50 cm

| Contaminant | TOC | Rbas | Rbas/TOC |
|---------------|----------|---------|----------|
| Cu | 0.8077** | 0.5384 | -0.4835 |
| Sum (Cd...Zn) | 0.6923* | 0.6209* | -0.1978 |

* $p<0.05$; ** $p<0.01$

3.6 Luminous bacteria tests

According to Römbke et al. (2006) a risk of toxicity release from soil contaminants into the seepage water is to be expected at LID values of ≥ 8 . Only sample RKS-4 has a LID value in this range while the eluates from the other samples have LID2 and LID3 clearly below this toxicity threshold (Table 5). The order of heavy metal contamination in the samples RKS-1 to RKS-6 (see Table 3) is not reflected by the results of the luminous bacteria test.

3.7 Earthworm avoidance tests

The results of the earthworm avoidance tests are shown in Table 6. None of the tested soil substrates from the landfill site was avoided by the earthworms. In three cases (RKS-2, -3 and -4) even a preference for the landfill soil was statistically significant (χ^2 test with null hypothesis 50%:50%). There was no correspondence between the amount heavy metal contamination in the soils and the distribution of *Eisenia fetida* between test soil and reference soil. A control test of the reference soil against the standard

Table 5 Least ineffective dilutions (LID) in luminous bacteria tests in soil eluates of spade samples (0–50 cm depth) from the former landfill site

| Sample | RKS-1 | RKS-2 | RKS-3 | RKS-4 | RKS-5 | RKS-6 |
|-----------|-------|-------|-------|-------|-------|-------|
| LID-value | LID3 | LID3 | LID2 | LID16 | LID2 | LID2 |

soil RefeSol 01-A resulted in 61% of the test worms being in the reference soil. Hence there is no reason to assume that the reference soil may be avoided by the earthworms.

4 Discussion

4.1 State of rotting and stability of the organic substance

The Deponieverordnung (2009)—the German landfill ordinance—sets limit values for the TOC-content in disposable material in order to minimise landfill gas production, emissions with seepage water, and subsidence of the landfill volume resulting from the decomposition of organic matter. The limit values are $\leq 1\%$ TOC for dump category I, and $\leq 3\%$ TOC for dump category II, respectively (Deponieverordnung 2009).

The TOC contents in the soils of the investigated former landfill site range between 0.3 and 20% with an arithmetic mean of 3.53% TOC. This exceeds the limit value for dump category II. About 50% of the determined TOC contents exceed the limit for dump category II. Only about 20% of the assayed samples could be assigned to dump category I.

However, it may be misleading to characterise the microbially available fraction of the organic matter by means of the TOC content (Department for Environment and comprehensive regional planning and Agriculture of North Rhine-Westphalia 1998). The TOC content covers all organic substances, including synthetic materials and/or so-called “black carbon” that may not be available to microbial consumption.

The Department for Environment and Comprehensive Regional Planning and Agriculture of North Rhine-Westphalia (1998) recommends the respiration activity

AT4 to characterise the microbially available fraction of the organic substance. The respiration activity AT4 is the amount of oxygen that is consumed by a sample in the Sapromat within 4 days (relating to 1 g dry mass). AT4 values of fresh, untreated waste usually average 20–60 mg O₂ g⁻¹ DM⁻¹ which corresponds to marginal decomposed litter originating from the L-horizons of forest soils (Soyez et al. 2000). The limit value in the Deponieverordnung for the parameter “AT4” is 5 mgO₂ gDM⁻¹. The measured respiration rates (CO₂ emission) were converted into AT4-values assuming a respiratory quotient of 1. Taking the 95th percentile of all measured respiration rates as a basis this conversion resulted in an AT4-value of 0.48 mgO₂ gTM⁻¹ which is much lower than the limit value of the Deponieverordnung although the TOC limit values are clearly exceeded. Furthermore, the calculated AT-4 values of the tested samples fall below the average respiratory activity of mechanically and biologically treated residual waste, which is about 3.5 mgO₂ gDM⁻¹ (Environmental Agency of North Rhine-Westphalia 2001). If the AT4 values are below the limit value of 5 mgO₂ gDM⁻¹, only marginal methane emission takes place and only tolerable contaminations of the seepage water are to be expected (Soyez et al. 2000). With respect to the state of degradation such material is similar to the organic substance in the A horizons of soils. The AT4 values for the samples in this study are only rough approximations since they were derived from CO₂-measurements according to the Isermeyer Method instead of a direct determination of O₂ consumption with a Sapromat (LAGA–Länderarbeitsgemeinschaft 1997). But even if the RQ is changed to 0.5 (which may be the case if hydrocarbons are mineralized and there is intense nitrification) this would only double the AT4 values which are by a factor of 10 below the limit value of 5 mg O₂ g⁻¹ DM (Deponieverordnung).

Although most measured TOC contents exceed the limit values of the Deponieverordnung, the level of the current mineralization is as low as in natural soils. The determined respiration rates indicate, that a relevant part of the TOC is of low microbial availability or even not available at all. This may be the result of an accumulation of recalcitrant organic compounds, such as synthetics, humic acids or other persistent substances like lignin. Organic matter can

Table 6 Results of the Earthworm Avoidance Tests: distribution of ten *Eisenia fetida* between a reference soil (loamy sand, pH 6.18) and soil substrate taken from sampling points at the former landfill site (spade samples 0–50 cm depth)

| Sample | Earthworms in reference soil (%) | Earthworms in tested soil (%) | χ^2 (critical value: 11.07) |
|--------|----------------------------------|-------------------------------|----------------------------------|
| RKS-1 | 31 | 69 | 8.5 |
| RKS-2 | 12 | 88 | 31.8 |
| RKS-3 | 33 | 67 | 11.9 |
| RKS-4 | 19 | 81 | 20.7 |
| RKS-5 | 51 | 49 | 4.00 |
| RKS-6 | 36 | 64 | 8.4 |

Entries in bold signifies $p < 0.05$

be stabilised by copper and other heavy metals in the soil (Sauvé 2006; Fründ et al. 2007). The significant correlation between TOC and copper found in this study may also be caused by the higher sorption capacity of organic matter compared to mineral sorbents (Lair et al. 2006). In general, the low rate of TOC mineralization can be explained by the advanced age of the old deposit. On the other hand the mainly aerobic conditions had an accelerating influence on the decomposition of the organic waste components (Environmental Agency of North rhine-Westphalia 2001).

4.2 Toxic potential

The results of the executed ecotoxicological tests do not indicate a toxic potential of the assayed material, neither regarding to a disturbance of the soil function as habitat for organisms nor regarding to a leakage of contaminants with the seepage water.

The earthworm avoidance test showed no avoidance of the landfill substrate although it contained considerable concentrations of heavy metals reaching 230 mg kg^{-1} of Cu and 803 mg kg^{-1} of Zn. There also was no correlation between the amount of heavy metal contamination and the degree of preference/avoidance of the earthworms. Other studies found an avoidance reaction of *E. fetida* with soils containing $4\ldots34 \text{ mg kg}^{-1}$ Cu (Van Zwieten et al. 2004). It is known that aged copper contaminations are less effective (van Zwieten et al. 2004, Lukkari and Haimi 2005, Fründ et al. 2005) but even than the contaminated soil is generally avoided by earthworms at Cu concentrations above 200 mg kg^{-1} .

Furthermore—according to the investigations by Kerth and van Straaten (1990)—elevated concentrations of organic contaminants, especially polychlorinated biphenyls, have also to be taken into account in the material originating from the former landfill site of this study.

5 Conclusions

The executed investigations relating to mineralization indicate that the bigger part of the former contained microbially available organic substance is already decomposed. Even so, the TOC contents exceed the limit value for dump category II of the Deponieverordnung, the respiration rates are in a range that is typical for natural soils. Therefore, the contained organic substance turns out to be stabilised or inert. The TOC depth profile is suggestive of the development to a natural ecosystem with high-average contents in the upper layers—obviously caused by the input of foliage biomass here.

While heavy metal contents are considerably high, the executed ecotoxicological tests do not indicate a toxic

potential. In terms of an emission of contaminants with the seepage water as well as in terms of a toxic disturbance of the soil function as a biological habitat the assayed material turns out to be ecotoxicologically harmless. According to this, it is assumed that the contained contaminants have formed mainly immobile species. This can both be due to the alkaline milieu and the formation of humic acids under aerobic conditions, which lead to an increase of the cation exchange capacity and have an influence on the mobility of pollutants. Yet in a long-term view a decrease of the pH values in the course of the pedogenesis and the leaching of carbonate leading to an increase of the heavy metal mobility cannot be excluded. Altogether the results of this paper indicate that due to rotting processes within the former landfill site of this study it has developed towards a natural ecosystem concerning the carbon dynamic as well as the toxic potential. In tendency this reflects the advantages of aerobically rotten waste concerning the environmental behaviour detected by Ritzkowski et al. (2008) and Östmann (2008) as well as the long-term abatement of the mobility of heavy metals prognosticated by Prantl et al. (2006) and Östmann (2008).

With regard to dump sites in developing countries where high-tech treatment of landfill gas and seepage water is not feasible, the results of this study should give food for thought, if treatment should aim at aerobic rotting processes rather than encapsulation. Probably, by doing so ecotoxicologically tolerable states would be achieved much quicker and the shifting of burden to following generations would be avoided.

References

- Beck T, Joergensen RG, Kandeler E, Makeschin F, Nuss E, Oberholzer HR, Scheu S (1997) An interlaboratory comparison of ten different procedures for measuring soil microbial biomass C. *Soil Biol Biochem* 29:1023–1032
- BBODSCHV (1999) Bundes-Bodenschutz- und Altlastenverordnung (BBODSCHV) vom 12. Juli 1999. BGBl. I, p 1554
- Department for Environment, Comprehensive Regional Planning and Agriculture of North Rhine-Westphalia (1998) Anforderungen und Kriterien für die Integration der mechanisch-biologischen Restabfallbehandlung in ein kommunales Abfallwirtschaftskonzept im Wege der Ausnahmeregelung der TA Siedlungsabfall. Available at: <http://www.abfallwirtschaftnrw.de/>. Accessed 17 July 2009
- Deponieverordnung (2009) Verordnung über Deponien und Langzeitlager vom 27. April 2008
- DIN ISO 10693 (1995) Soil quality—determination of carbonate content—volumetric method. International Organization for Standardization
- DIN ISO 10694 (1995) Soil quality—determination of organic and total carbon after dry combustion (elementary analysis). International Organization for Standardization
- DIN ISO 11466 (1997) Soil quality—extraction of trace elements soluble in aqua regia. International Organization for Standardization

- DIN ISO 16072 (2005) Soil quality—laboratory methods for determination of microbial soil respiration. International Organization for Standardization
- DIN ISO 17512-1 (2005) Soil quality—avoidance test for testing the quality of soils and the toxicity of chemicals—test with earthworms (*Eisenia fetida* and *Eisenia andrei*). International Organization for Standardization
- DIN ISO 11348-3 (2007) Water quality—determination of the inhibitory effect of water samples on the light emission of *Vibrio fischeri* (Luminescent bacteria test)—part 3: method using freeze-dried bacteria. International Organization for Standardization
- DIN 19682-13 (2009) Soil quality—field tests—part 13: determination of carbonate, sulfide, pH-value and iron(II)-ions. International Organization for Standardization
- Environmental Agency of North Rhine-Westphalia (2001) Langfristiges Emissionsverhalten von Altdeponien. Materialien des Landesumweltamtes Nordrhein-Westfalen Nr. 61. Essen
- Fründ HC, Frerichs C, Rück F (2005) Bewertung Schwermetall belasteter Böden mittels Regenwürmern—Siedlungsdichte und Vermeidungsverhalten im Fluchttest. Mitt Dtsch Bodenkundl Ges 107:191–192
- Fründ HC, Emmerling C, Bossung V, Vohland M, Tülp H, Hinsinger P (2007) Gibt es einen Zusammenhang zwischen Kupfergehalt und Humusgehalt im Boden? Mitt Dtsch Bodenkundl Ges 110:307–308
- Heyer KU (2003) Emissionsreduzierung in der Deponienachsorge. Hamburger Berichte 21. Verlag Abfall aktuell, Stuttgart
- Kerth M, van Straaten L (1990) Gutachten zur Gefährdungsabschätzung der Altablagerungen H 518, I 27 und B 154 in Bielefeld Hillegossen. Unveröffentlichtes Gutachten der Geo-Infometric Gesellschaft für Umwelt- und Geoservice mbH, 62S
- König W, Bachmann G, Kohl R (2003) Grundsätze des vorsorgenden Bodenschutzes zur Begrenzung von Stoffeinträgen in den Boden. In: Rosenkranz D, Bachmann G, Einsele G (eds) Bodenschutz – Ergänzbares Handbuch der Maßnahmen und Empfehlungen für Schutz, Pflege und Sanierung von Böden, Landschaft und Grundwasser. Erich Schmidt Verlag, Berlin, pp 400–479
- Kruse K (1994) Langfristiges Emissionsverhalten von Siedlungsabfalldeponien. Veröffentlichungen des Instituts für Siedlungswasserwirtschaft Technische Universität Braunschweig Heft 54, Braunschweig
- LAGA–Länderarbeitsgemeinschaft Abfall (1997): Anforderungen an die stoffliche Verwertung von mineralischen Abfällen—Technische Regeln. LAGA-Mitteilungen 20 vom 6. November 1997
- Lair GJ, Gerzabek MH, Haberhauer G (2006) Sorption of heavy metals on organic and inorganic soil constituents. Environ Chem Lett 5:23–27
- Lehmann A, Stahr K (2007) Nature and significance of anthropogenic urban soils. J Soils Sediments 7:247–260
- Lukkari T, Haimi J (2005) Avoidance of Cu- and Zn-contaminated soil by three ecologically different earthworm species. Ecotox Environ Saf 62:35–41
- Östmann M (2008) Ageing landfills—development and processes. Dissertation, Swedish University of Agricultural Sciences, Uppsala
- Prantl RM, Tesar M, Huber-Humer M, Heiß-Ziegler C (2006) In-situ Aerobisierung von Altablagerungen. Available at: <http://interland.arcs.ac.at/extranet/interland/documents/03%20LF%20InSitu%20Aerobisierung%202006-04%20.pdf>. Accessed 26 Nov 2009
- Ritzkowski M, Heyer KU, Stegmann R (2008) Biologische in-situ Stabilisierung der Altdeponie Kuhstedt/Niedersachsen. Müll und Abfall 7:354–361
- Römbke J, Eisenträger A, Hund-Rinke K, Jänsch S, Neumann-Hensel H, Weber G (2006) Handlungsempfehlung für die ökotoxikologische Beurteilung von Böden. Sidus, Limburg
- Rossier N, Dessurault J (2004) Die Entwicklung der bodenbiologischen Kenngrößen der freiburgischen Landwirtschaftsböden. VBB Bulletin 8/2004: 16–18. Available at: http://www.umweltschweiz.ch/buwal/de/fachgebiete/fg_boden/info/biologiesols. Accessed 01 April 2010
- Sauvé S (2006) Copper inhibition of soil organic matter decomposition in a seventy-year field exposure. Environ Toxicol Chem 25:854–857
- Sommer M, Ehrmann O, Friedel JK, Martin K, Vollmer T, Turian G (2002) Boden als Lebensraum für Organismen - Regenwürmer, Gehäuselandschnecken und Bodenmikroorganismen in Wäldern Baden-Württembergs, Hohenheimer Bodenkundliche Hefte, 63
- Stegmann R, Bidlingmayer W, Bilitewski B et al (2001) Strategie zum Umgang mit Altdeponien. Müll und Abfall 7:405–413
- Soyez K et al (2000) Verbundvorhaben “mechanisch-biologische Restabfallbehandlung”. Brandenburgische Umweltberichte (BUB) 8:289–297
- Van Zwieten L, Rust J, Kingston T, Merrington G, Morris S (2004) Influence of copper fungicide residues on occurrence of earthworms in avocado orchard soils. Sci Total Environ 329:29–41