Incineration Slag as Key Element within a Methane Oxidation Layer on a Closed Landfill - Assessment of Suitability: A Case Study -

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„You should make everything as simple as possible, but not simpler“

(Albert Einstein)

in memory of my beloved brother

Marcus Gruyters

dedicated to my dear children

Fabienne, Sandrine and Tom

Acknowledgements

Once upon a time, I believed that a dissertation was a completed work of a single person specializing in a distinct area. – Well, now that I have finished my own doctoral thesis, I have to revise this idea as this thesis is only one small piece within a superordinated mosaic and as this work could not have been realized without the help and support of numerous people, whom I wish to express my gratitude:

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Abstract

The bacterial oxidation of methane by means of semi-permeable landfill top covers has recently become an increasingly accepted tool to mitigate climate damaging landfill gas emissions. In this vein, different substrates and installation technologies have been investigated throughout the world, with respect to regional climatic conditions, as well as to local substrate availability and legal framework conditions. Thereby, methanotrophic bacteria turned out to be reliant on stable environmental conditions regarding soil temperature, moisture, and soil gas diffusivity; below or above certain variability ranges, the bacteria are actually capable of adapting to given environmental conditions, but only up to a certain extent, and only by reducing their methane oxidation activity. However, the conditions of these laboratory studies and/or simulations do not necessarily equate to given, local conditions and so far, there are only insufficient experiences about long-term studies on real landfills.

In this work, an interim coverage of a former landfill for municipal waste consisting of a ca. 20cm thick profiling layer of incineration slag, a ca. 80cm mighty layer of recultivation soil, and a ca. 20cm thick layer of compost as top soil was assessed regarding its suitability for bacterial methane oxidation. After a series of preliminary laboratory investigations, a testfield was installed at the oldest part of the landfill, where the gas production was most advanced, and where the active degassing system was switched off, to see if the interim coverage was able to cope with the given methane area loading during all seasons of the year. In an examination period of about three years, the soil temperature, soil moisture, and soil gas pressure were registered by means of data logs in different soil depths within the interim coverage to identify the environmental conditions. Periodical measurements of the gas concentrations of methane, carbon dioxide, and oxygen in various soil depths were performed to trace depth- and seasonal-related gas profiles, by which the depth of the methane oxidation layer was determined. At the same time, measurements of the stable carbon isotope composition of methane and carbon dioxide were performed to prove the bacterial methane oxidation. In order to examine the

impact of time and settlement on porosity and permeability of the constructed interim coverage, measurements of the hydraulic conductivity and of the intrinsic gas permeability, as well as micrographic examinations were performed.

The field experiments confirmed that a methane oxidation layer has actually established in a depth of about 120cm within, or just beneath the profiling layer, where the gas profiles of the methane and oxygen concentrations intersect. Furthermore, the stable carbon isotope composition indicates a bacterial methane oxidation as the methane in this horizon is isotopically slightly heavier than that in the anoxic zone, whereas the carbon dioxide is correspondingly lighter. The efficacy of the methane oxidation layer was substantiated, as during all seasons, no methane was detected in any soil depths < 100cm. The combination of applied layer thicknesses and substrates turned out to be well suited for methane oxidation purposes as it provides stable environmental conditions, well shielded against seasonal and short time atmospheric influences. Thereby, the incineration slag plays a key role within the methane oxidation layer as it was stabilized by carbonatic cementation, supporting resistance against settlement, and preserving porosity and permeability.

Because of decreasing gas quantities and methane concentrations, the passive phase, in which the methane loading can be mitigated by bacterial methane oxidation, actually overlaps with the active phase, in which the generated landfill gas can be utilized for electrification. Focussing solely on methane oxidation, the given, semi-permeable coverage should be left in the current state as it is actually capable of reliably mitigating the given methane loading, and as it provides the ongoing degradation of the organic matter within the anoxic zone, whereas a sealing of the landfill top would actually stop the degradation processes and mummify the landfill body.

With the combination of the respective examination tools, the chosen interdisciplinary approach is well suited to assess the suitability of the examined methane oxidation layer in the given landfill setting. However, there is still need for improvement, especially of the gas measuring and sampling devices and for the refinement of stable isotope analysis. Finally, further research regarding the emissions path of leachate is required.

Deutsche Zusammenfassung

Die biologische Methanoxidation in Deponieabdeckschichten ist eine wirksame Maßnahme zur Reduzierung von Treibhausgasemissionen, die in wissenschaftlich-technischen, als auch in politischen Gremien zunehmend Anerkennung findet. Im Zuge ihrer weltweiten Erforschung wurden – unter Berücksichtigung regionaler Gegebenheiten, wie etwa Klima, Materialverfügbarkeit und gesetzliche Rahmenbedingungen – verschiedenste Substrate und Konstruktionsarten untersucht. Dabei stellte sich eine deutliche Abhängigkeit der methanotrophen Bakterien von stabilen Umweltbedingungen hinsichtlich Bodentemperatur, -feuchte und -gasdiffusivität heraus; jenseits bestimmter Schwellenwerte sind die Bakterien zwar anpassungsfähig, jedoch nur innerhalb eines begrenzten Rahmens und bei verminderter Aktivität. Die Bedingungen dieser Laboruntersuchungen / Simulationen entsprechen jedoch nicht unbedingt den tatsächlichen Gegebenheiten vor Ort und bislang gibt es nur unzureichende Erfahrungen über entsprechende Langzeitstudien auf realen Deponien.

Die vorliegende Arbeit untersucht die Eignung einer Zwischenabdeckung aus einer ca. 20cm mächtigen Profilierungsschicht aus HMV-Schlacke, einer ca. 80cm mächtigen Schicht aus Rekultivierungsboden und einer ca. 20cm mächtigen Kompostauflage auf einer stillgelegten Siedlungsmülldeponie am Niederrhein. Nach einer Reihe von Voruntersuchungen wurde auf dem ältesten Bauabschnitt der Deponie, bei der die Gasbildung am weitesten fortgeschritten war, ein Testfeld eingerichtet und die aktive Gasabsaugung abgeschaltet, um zu untersuchen, in wie weit die aufgebrachte Zwischenabdeckung geeignet war, die in dem Bereich noch vorhandene Methanfracht abzubauen.

Während der dreijährigen Untersuchungskampagne wurden Bodentemperatur, -feuchte und -gasdruck in verschiedenen Tiefen mittels Datenloggern erfasst, um die herrschenden Umweltbedingungen zu beschreiben. Aus periodischen Messungen der Konzentrationen von Methan, Kohlendioxid und Sauerstoff in verschiedenen Bodentiefen wurden saisonale und tiefenabhängige Verlaufskurven erstellt; Messungen der Stabilen Kohlenstoffisotope von Methan und Kohlendioxid sollten die bakterielle Methanoxidation belegen. Zur Untersu-

chung des Einflusses von Setzungen auf die Porosität der Zwischenabdeckung wurde diese bodenphysikalisch und mikrographisch untersucht.

Die Untersuchungen ergaben, dass sich eine Methanoxidationsschicht in einer Tiefe von etwa 120cm im Bereich der Profilierungsschicht gebildet hat. Hier liegt zum einen der Schnittpunkt der Methan- und Sauerstoffkonzentrationskurven, zum anderen weisen die in diesem Bereich gemessenen Signaturen der stabilen Kohlenstoffisotope in Methan und Kohlendioxid auf eine entsprechende bakterielle Methanoxidation hin. Die gänzliche Abwesenheit von Methan in sämtlichen Bodentiefen oberhalb 100cm während der gesamten Versuchsdauer bestätigt die Wirksamkeit der vorhandenen Zwischenabdeckung hinsichtlich ihrer Methanoxidationsfähigkeit; dabei nimmt die Profilierungsschicht aus HMV-Schlacke eine gewisse Schlüsselrolle ein, da sie aufgrund ihrer karbonatischen Zementierung Setzungen widerstehen und so eine hohe Porosität und Permeabilität aufrecht erhalten konnte.

Durch die mit zunehmender Zeit weiter zurückgehende Deponiegasmenge und Methankonzentration überlappen sich auf der gegenständlichen Deponie die Aktivphase, in der noch eine Nutzung des Deponiegases durch Verstromung möglich ist, und die Passivphase, in der die vorhandene Methanfracht alleine biologisch abgebaut werden kann, um mehrere Jahre. Alleine aus Deponiegassicht betrachtet, sollte die vorhandene Zwischenabdeckung daher so lange wie möglich in dem aktuellen Zustand bleiben, da sie die vorhandene Methanfracht zuverlässig oxidiert und durch ihre Durchlässigkeit für Luft und Wasser den fortwährenden Abbau des organischen Materials im Deponieinnern aufrecht erhält, wohingegen eine Abdichtung die Abbauprozesse stoppen und die Deponie mumifizieren würde.

In ihrer Kombination sind die verwendeten Untersuchungsmethoden gut geeignet, die Wirksamkeit der Zwischenabdeckung hinsichtlich ihres Methanoxidationsvermögens zu untersuchen, wenngleich insbesondere das Auflösungsvermögen bei der Untersuchung der Stabilen Kohlenstoffisotope noch verfeinert werden muss, um auch quantitative Aussagen über die Methanoxidation ableiten zu können. Schließlich gibt es weiteren Forschungsbedarf hinsichtlich des Emissionspfades Sickerwasser.

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Index of Used Abbreviations and Symbols

Symbol	Meaning	(Unit)
a	year	
a_0 , a_1	soil type depending constants for conversion of soil moisture	–
A	area	m^2
AbfAblV	Abfallablagerungsverordnung = Ordinance on Environmentally Compatible Storage of Waste from Human Settlements and Biological Waste-Treatment Facilities	
AbfG	Abfallbeseitigungsgesetz = Federal Waste Law in Germany	
AC	air capacity	vol.-%
ADP	adenosine diphosphate	
ANMEs	anaerobic methanotrophs	
AOM	anaerobic oxidation of methane	
ATP	adenosine triphosphate	
AWC	available water capacity	vol.-%
BAM	Bundesanstalt für Materialforschung und –prüfung = Federal Institute for Materials Research and Testing in Germany	
BC	Before Christ	
BMU	Bundesministerium für Umwelt Naturschutz und Reaktorsicherheit = Federal Ministry for the Environment, Nature Conservation and Nuclear Safety in Germany	
BRD	Bundesrepublik Deutschland = Federal Republic of Germany	
C	carbon	
$C_6H_{10}O_4$	simplified molecular composition of organic matter in landfills with municipal solid waste	
c	concentration	$mol \cdot m^{-3}$, or $mol \cdot l^{-1}$
c	constant	
C_{org}	organic carbon	
Ca	calcium	
ca.	circa	
cbm	cubic meter	m^3
Cd	cadmium	
CH_3COOH	acetic acid	
CH_3OH	methanol	
CH_4	methane	
CHP	combined heat and power unit	
cm	centimetre	10^{-2} m
CMOS	complementary metal oxide semiconductor	
CO_2	carbon dioxide	
CO_2-eq	carbon dioxide-equivalents	
COP	Conference of Parties – annual meeting of UNFCCC	
Cu	copper	
Cyt C_{ox}	cytochrome complex oxidase	

Symbol	Meaning	(Unit)
Cyt C_{red}	cytochrome complex reductase	
C_{90}-value	time, when 90% of the biodegradable organic carbon is degraded	
D	diffusion coefficient, or diffusivity	$m^2 \cdot s^{-1}$
D_0	diffusion coefficient, or diffusivity in free air	$m^2 \cdot s^{-1}$
D_e	effective diffusion coefficient, or diffusivity in soil	$m^2 \cdot s^{-1}$
d_{eff}	effective grain diameter	mm
D-C GMP	density-corrected generalized macroporosity-dependant (model)	
DepV	Deponieverordnung = German landfill ordinance	
DepVerwV	Deponieverwertungsverordnung = German ordinance for the use of waste in landfills	
DGGT	Deutsche Gesellschaft für Geotechnik e.V. = German Society for Geotechnics	
DIN	Deutsches Institut für Normung = German Institute for Standardization	
DK II	Deponieklasse II = ~ landfill for municipal waste	
dyn.	dynamic	
e	Euler's number = 2.718…	
e	void ratio	–
e^-	electron	
EIT	Economies In Transition: Countries of the former Soviet bloc, which are transitioning to a market economy	
EC	European Community	
EPS	exopolymeric substances	
eq.	equation	
et al.	and others (et alii)	
etc.	and so on (et cetera)	
EU	European Union	
e.g.	for example (exempli gratia)	
e.V.	eingetragener Verein = registered association	
$f_0\%$	percentage of methane oxidized in transit through the cover soil	
FADH	formaldehyde dehydrogenase	
FC	field capacity	vol.-%
FDH	formate dehydrogenase	
FDR	frequency domain reflectometry	
Fe	iron	
FEM	finite element method	
fig.	figure	
FISH	fluorescence-in-situ-hybridization	
g	gram	10^{-3} kg
G_c	theoretically capturable gas volume	m^3
G_p	produced gas volume	m^3
G_t	total produced gas volume, up to a given instant in time	m^3

Symbol	Meaning	(Unit)
GC-IRMS	gas chromatography isotope ratio mass spectrometer	
GCS	gas collection station	
GDP	gross domestic product	
gen. nov.	new (found) genus	
GHG	greenhouse gas	
grav.	gravimetric	
GW	gas well	
GWP	global warming potential	
H, H_2	hydrogen	
h	hour	
h	height; difference of piezometric heads	m, or cm
h_W	difference of upper- and under water level	m, or cm
H^+	proton	
H_2O	water	
HCO_3^-	hydrogen carbonate ion	
HCOH	formaldehyde (methanal)	
$HCOO^-$	formate (methanoate)	
HCOOH	formic acid (methanoic acid)	
HFCs	hydrofluorocarbons	
HMV	Hausmüllverbrennungsschlacke = incineration slag (of municipal waste)	
hPa	hectopascal	10^2 Pa
HS^-	hydrogen sulfide ion	
i	hydraulic gradient	–
IPCC	International Panel on Climate Change	
J	Joule	$N \cdot m = kg \cdot m^2 \cdot s^{-2}$
J	diffusive flux	$mol \cdot s^{-1} \cdot m^{-2}$
K	Kelvin	
K	intrinsic permeability	m^2
K_{rel}	intrinsic permeability of a partly water-saturated soil	m^2
K_0	intrinsic permeability of a dry soil	m^2
k	decay constant	a^{-1}
k_f	(coefficient of) hydraulic conductivity	$m \cdot s^{-1}$
k_{fT}	hydraulic conductivity at ambient temperature T	$m \cdot s^{-1}$
$k_{f\,10\,(-calc)}$	(calculated) hydraulic conductivity for T = 10°C	$m \cdot s^{-1}$
$k_{f\,PSD}$	hydraulic conductivity, derived by particle size distribution	$m \cdot s^{-1}$
kg	kilogram	
kJ	kilojoule	10^3 J
km	kilometer	10^3 m
kPa	kilopascal	10^3 Pa

Symbol	Meaning	(Unit)
KrW-/AbfG	Kreislaufwirtschafts- und Abfallgesetz = Federal Law for Recycling and Waste Management	
KWA	Kreis Weseler Abfallwirtschaftszentrum	
kW_{el}	electric power	$W = kg \cdot m^2 \cdot s^{-3}$
l	liter	10^{-3} m^3
l	(perfused) length	m
l_0	height of test specimens	m
LAGA	Länderarbeitsgemeinschaft Abfall = task group “waste” of the German Conference of Environmental Ministers	
LFG	landfill gas	
LHV	lower heating value = net calorific value	$MJ * kg^{-1}$
LUA	Landesumweltamt = State Environmental Agency in Germany	
m	meter	
m	mass	kg
m_s	solid mass	kg
m_W	mass of water	kg
m_{Waste}	deposited mass of waste per year	$t \cdot a^{-1}$
mbar	millibar	10^{-3} bar = 10^2 Pa
MDH	methane-dehydrogenase	
Mg	magnesium	
mg	milligram	10^{-6} kg
Mio	million	10^6
min	minute	60 s
MJ	megajoule	10^6 J
mm	millimeter	10^{-3} m
MMO	methane monooxygenase	
MOL	methane oxidation layer	
MPa	megapascal	10^6 Pa
MSW	municipal solid waste	
MUNLV	Ministerium für Klimaschutz, Umwelt, Landwirtschaft, Natur- und Verbraucherschutz des Landes Nordrhein-Westfalen = The Ministry for Climate Protection, Environment, Agriculture, Nature Conservation and Consumer Protection of the German State of North Rhine-Westphalia	
MWh	megawatt hours	$3.6 \cdot 10^9$ J
m/z	mass number	
n	amount of substance	mol
n	porosity	vol.-%
n_A	air-filled porosity	vol.-%
N, N_2	nitrogen	
N_2O	nitrous oxide	
NaCl	sodium chloride	

Symbol	Meaning	(Unit)
NAD^+ / NADH	nicotinamide adenine dinucleotide: $NAD^+ + H^+ + 2e^- \Rightarrow NADH$	
NASA	National Aeronautics and Space Administration	
NH_4^+	ammonium ion	
NO_2^-	nitrite ion	
NO_3^-	nitrate ion	
NOAA	National Oceanic and Atmospheric Administration of the U.S.A.	
NRW	Nordrhein-Westfalen = North Rhine-Westphalia	
O, O_2	oxygen	
OECD	Organization for Economic Co-operation and Development: International economic organization of 34 countries, regarded as developed countries.	
ÖVA	Österreichischer Verein für Altlastenmanagement = Austrian Association for Management of Contaminated Sites	
p	pressure	Pa
Pa	pascal	$N \cdot m^{-2} = kg \cdot m^{-1} \cdot s^{-2}$
Pb	lead	
pF	negative common logarithm of suction tension in centimeter	–
PFCs	perfluorocarbons	
pH	negative common logarithm of effective hydrogen ion concentration	–
pMMO	particulate methane monooxygenase	
ppbv	parts per billion by volume	$1 \cdot 10^{-9}$
ppm	parts per million	$1 \cdot 10^{-6}$
ppmv	parts per million by volume	$1 \cdot 10^{-6}$
PWP	permanent wilting point	pF, or Pa
PSD	particle size distribution	
p.a.	per annum	
Q	laminary flowing discharge rate, or air current, resp.	$m^3 \cdot s^{-1}$
r	coefficient of roughness	–
R	ideal gas constant	$J \cdot K^{-1} \cdot mol^{-1}$
R^2	coefficient of determination	
R_A	ratio of substance A (versus substance B)	
R_B	ratio of substance B (versus substance A)	
R_{sample}	ratio of stable isotopes of ^{13}C / ^{12}C in a given sample	%
$R_{standard}$	ratio of stable isotopes of ^{13}C / ^{12}C in the standard (V-PDB)	%
resp.	respectively	
retr.	retrieved	
rRNA	ribosomal ribonucleic acid	
RuMP	ribulose monophosphate	
s	second	
SF_6	sulphur hexafluoride	
sMMO	soluble methane monooxygenase	

Symbol	Meaning	(Unit)
SO_4^{2-}	sulfate	
spec.	specific	
sp. nov.	new (found) species	
STP	standard temperature and pressure	
t	ton	10^3 kg
t	time	a
$t_{1/2}$	half-time	a
T	temperature	°C, or K
$T_{\text{min / max / opt}}$	minimum / maximum / optimum temperature	°C
T_{opt}	optimal growth temperature	°C
T_R	reaction temperature	°C
TA	Technische Anleitung Abfall = technical directive for hazardous waste	
tab.	table	
TASi	Technische Anleitung Siedlungsabfall = technical directive for municipal waste	
Tg	teragram = megaton	10^9 kg
TOC	total organic carbon	
TOC_d	content of degradable total organic carbon per ton (of waste)	$kg \cdot t^{-1}$
UGT	Umwelt-Geräte-Technik GmbH, Müncheberg	
UN	United Nations	
UNEP	United Nations Environment Programme	
UNFCCC	United Nations Framework Convention on Climate Change	
URL	uniform resource locator	
U.S.(A.)	United States (of America)	
USDA	United States Department of Agriculture	
V	Volt	
V	volume	m^3
V_A	air-filled volume	m^3
V_B	bulk volume	m^3
V_m	(ideal) molar volume at standard pressure / temperature conditions	22.4 l
V_s	solid volume	m^3
V_V	void-space volume	%
V_W	water-filled volume	%
V (DC)	Volt (direct current)	
V-PDB	Vienna–PeeDee-formation Belemnite (standard for stable isotopes)	
vol.-%	per cent in volume units	%
vs.	versus	
w/w	weight to weight	
x	space coordinate	m

Symbol	Meaning	(Unit)
ZfA	Zentralstelle für Abfallbeseitigung beim Bundesgesundheitsamt = (former) Central Department for Waste Management at the Federal Health Office in Germany	
Zn	zinc	
%	per cent	
‰	per mille	
°C	degree Celsius	
α_{ox}	isotope fractionation factor for bacterial oxidation	–
α_{trans}	isotope fractionation factor associated with gas transport	–
$\alpha_{\mathrm{A-B}}$	fractionation factor for conversions between substances A and B	–
γ	specific weight	$N \cdot m^{-2}$
γ_{W}	specific weight of water	$N \cdot m^{-2}$
$\delta^{13}\mathrm{C}$	ratio of stable isotopes of ^{13}C / ^{12}C in a sample vs. a standard	‰
δ_A	ratio ^{13}C / ^{12}C in the anoxic zone of a landfill	‰
δ_E	ratio ^{13}C / ^{12}C of emitted methane from a landfill	‰
Δ	difference; interval	–
ΔG^0	Gibbs energy	$kJ \cdot mol^{-1}$
∂	partial derivative	
ε	apparent permittivity	–
η	dynamic viscosity	$Pa \cdot s = kg \cdot m^{-1} \cdot s^{-1}$
η_{Air}	dynamic viscosity of air	Pa·s
η_T	dynamic viscosity at ambient temperature T	Pa·s
η_{W}	dynamic viscosity of water	Pa·s
η_{10}	dynamic viscosity at temperature T= 10°C	Pa·s
η_C	degree of gas capture for gas prognosis calculations	–
η_D	degradation factor for gas prognosis calculations	–
η_E	environmental factor for gas prognosis calculations	–
$\theta_{\mathrm{W}\ grav\ dry}$	gravimetric water content, related to dry matter	%
$\theta_{\mathrm{W}\ Vol}$	volumetric water content	%
µm	micrometer	10^{-6} m
ρ	density	$g \cdot cm^{-3}$, or $kg \cdot m^{-3}$
ρ_b	bulk density	$kg \cdot m^{-3}$
$\rho_{b\ \mathrm{dry}}$	dry bulk density	$kg \cdot m^{-3}$
$\rho_{b\ \mathrm{wet}}$	wet bulk density	$kg \cdot m^{-3}$
ρ_g	grain density	$kg \cdot m^{-3}$
ρ_r	raw density	$kg \cdot m^{-3}$
$\rho_{\mathrm{B\ Pro}}$	Proctor density = highest achievable density	$kg \cdot m^{-3}$
τ	ratio of kinematic viscosities of water at 10°C / in the aquifer	–
ψ	water potential	m water column, or hPa
ψ_m	matrix potential	m water column, or hPa

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