SUSTAINABLE EMISSION REDUCTION OF AMMONIUM FROM A LANDFILL BODY USING THE ANAMMOX-PROCESS AT THE VLAGHEIDE LANDFILL IN SCHIJNDEL (NL)

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SUMMARY: Within the framework of the ongoing sustainable landfill management programme (SLMP) at the Vlagheide landfill (started in 2005) the removal of the long term ammonium emission by means of the biological anaerobic ammonium oxidation process, the so-called 'Anammox-process' is being investigated. The objective is to examine the occurrence and activity of the Anammox-process in landfill leachate. The research program consists of five sequential steps. So far the first two steps have been completed. The results so far demonstrate a widespread presence of the potential of aerobic and anaerobic microbiological degradation of ammonium in the landfill body. At one specific location several types of Anammox-bacteria could be detected and quantified. This fact in combination with the low ammonium concentrations is the proof that at this location ammonium can biologically be degraded. This meant a 'go' for step 3 of the research program. Step 3 focuses on the kinetics and activity of the anammox-process as well as on the determination of the opportunities and risks of practical implementation of the anammox-process.

1. INTRODUCTION

The Urban District of 's-Hertogenbosch is the owner of the Vlagheide landfill in Schijndel (NL), which complies with the EU-standards with respect to aftercare measures. Within the framework of the ongoing sustainable landfill management programme (SLMP), at the Vlagheide landfill (started in 2005) the reduction or removal of long term ammonium emission by means of the anaerobic ammonium oxidation process, the so called 'Anammox-process', is being investigated. This paper presents the results obtained so far and microbial laboratory tests and the conclusions with respect to the feasibility of sustainable emission reduction of ammonium due to the anammox-process.

2. OBJECTIVES

The objective is to examine the occurrence of the Anammox-process in landfill leachate, and if so, to answer the following questions:

- to what extent does the Anammox-process contribute to a significant removal of ammonium in the leachate?
- what is the speed of the anammox-process with regard to the decrease of ammonium concentrations?
- will it result in admissible emission limit values?
- is it possible to stimulate the anammox-process by optimising the conditions, and if so, how?
- will it result in a significant reduction of the pollution rate and as a consequence in a reduction of the costs of leachate discharge?

3. BACKGROUND INFORMATION VLAGHEIDE LANDFILL

The Vlagheide landfill measures 40 hectares and 6 million m3 of municipal solid waste (MSW), stored over the last 35 years. The landfill closed in 2003. Half of the landfill comprises a newer part (exploited from 1990 to 2003) with a bottom liner, but still without a top liner. The other half is the older part (exploited from 1969 to 1985) without a bottom liner but partially covered by a top liner.

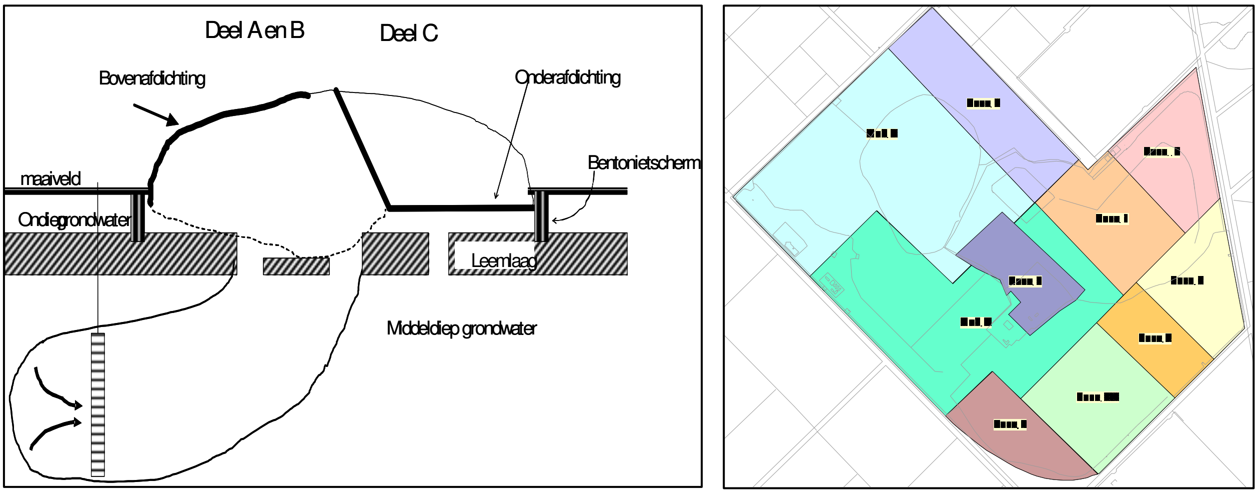

Figure 1. Cross-section and plan of the Vlagheide landfill

Leachate generated from landfills can be a great threat to the groundwater quality in case of leachate emission to the groundwater. Therefore the goal of the SLMP is to achieve a sustainable emission reduction in the landfill body in such a way that the leachate quality complies with the admissible limit values of emission to groundwater. This means that the landfill body is considered to be a bioreactor in which all kinds of natural processes occur, which are able to reduce concentrations of all kinds of compounds to admissible emission limit values.

The landfill leachate contains high concentrations of organic and inorganic compounds, of which ammonium is considered to be a long term emission problem. High concentrations of ammonium in the leachate, may become a hindrance to the effective functioning of sustainable landfill management (landfill stabilization). In table 1 the ammonium (N_{kj}) concentrations in the leachate, of the older and newer part of the landfill, are presented. The N_{kj}-concentrations in the older part are significantly lower than in the newer part. This was the reason to pose the question whether or not the lower concentrations might be attributed to the presence of the Anammox-process in the landfill body.

Table 1. Average ammonium concentrations in the Vlagheide landfill period 2008 - 2010

	Older landfill part (groundwater below waste)	*Newer landfill part (leachate)*
Average ammonium (N_{kj}) concentrations	150 – 250 mg/l	1000 – 1500 mg/l

4. FUNDAMENTALS OF THE ANAMMOX-PROCESS

The anammox-process is a part of the Nitrogen cycle (figure 2). The focus in this research is the removal of ammonium. In practice two methods are used to remove ammonium, which are:

- a combination of nitrification and denitrification;
- the anammox-process (figure 3)

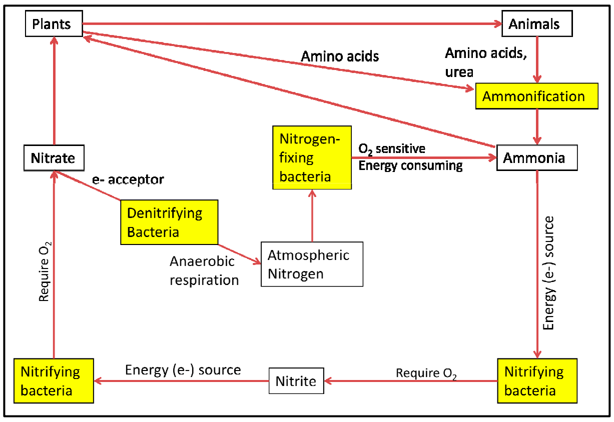

Figure 2. Nitrogen cycle

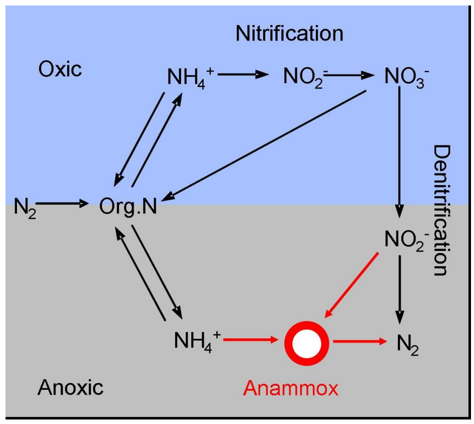

Figure 3. Anammox-process

During the <u>nitrification</u>-process ammonium is converted first into nitrite and finally into nitrate. This conversion process requires oxygen, but no source of organic carbon. Conversion of ammonium into nitrite as well as the conversion of nitrite into nitrate is executed by nitrifying bacteria. <u>Denitrification</u> is the process which converts nitrate into nitrogen by means of denitrifying bacteria. This process requires anaerobic conditions as well as a source of organic carbon (for instance methanol).

<u>Anaerobic ammonium oxidation (ANAMMOX)</u> is a process in which ammonium is converted into nitrogen by anaerobic denitrifying bacteria and in which nitrite is used as the electron acceptor. Different kinds of microbes have been identified, which are able to oxidize ammonium under anaerobic conditions. It concerns microbes named *Anammoxoglobus* spp., *Brocadia* spp., *Jettenia* spp., *Kuenenia* spp., *Scalindua* spp. The anammox-process offers great opportunities for the removal of ammonium in fully autotrophic systems with biomass retention. No organic carbon is needed in such nitrogen removal systems, since ammonium is used as electron donor for nitrite reduction and dissolved carbon is used as a source of carbon (autotrophic growth). The anammox-process is visualized in figure 2.

The anammox-process can only occur when certain process- and environmental conditions are present. The critical conditions are summarised in table 2.

Table 2. Process- and environmental conditions required for the Anammox-process

Critical parameters	*Anammox-process*
Organic carbon source	Bicarbonate (HCO_3^-)
Electron donor	Ammonium (NH_4^+)
Electron acceptor	Nitrite (NO_2^-), but inhibition at ± 100 mg/l
Nutrients	Nitrogen (N), phosphate (P)
End products	Nitrogen gas (N2), Nitrate (limited)
Temperature (^{0}C)	6 - 43
Acidity (pH)	6.7 – 8.3
Oxygen	Inhibition at ± 2 mg/l

5. RESEARCH PROGRAM

5.1 General approach

A stepwise approach has been chosen for a cost-effective execution of the research program. Only a positive result of an executed step leads to a 'go' for the execution of a next step, and so on. These sequential steps are:

1. A quick scan through the analysis of one sample of landfill leachate for the presence of anammox-bacteria.
2. A more extensive inventory through the analysis of 6 other samples of landfill leachate in order to test the structural appearance of the anammox-process in the landfill body.
3. Assessment of the potential and feasibility of sustainable emission reduction of ammonium by means of the anammox-process. Determination of the kinetics and activity of the anammox-process in the landfill body as well as of the opportunities and risks of implementation the anammox-process in practice.
4. Elaboration of the possibilities for optimization, stimulation and implementation of the anammox-process as a means for sustainable and cost-effective emission reduction of ammonium in the landfill body. Design of an implementation and monitoring program.
5. Implementation, supervision and monitoring at the Vlagheide landfill.

So far step 1 and 2 have been executed and at the time of writing step 3 is in progress.

5.2 Activities step 1 and 2

Six water samples were taken at the Vlagheide landfill for microbial analyses. Location, type and a short description of these samples is presented in table 3. All water samples were analyzed for specific microbial and chemical parameters.

Table 3. Sampling program step 1 and 2

Number	*Type*	*Location*
Comp 4	Leachate	Leachate from compartment 4 of the new part of the landfill
Comp 5/6	Leachate	Leachate from compartment 5/6 of the new part of the landfill; this leachate was sampled and analysedin step 1 and in step 2.
D2-1	Shallow groundwater	Groundwater from directly below the waste of the old part of the landfill without bottom liner and influenced by landfill leachate.
D4-D	Leachate	From the bottom of the waste body and in the middle of the old part of the landfill just outside the present top liner.
D5-D	Leachate	From the bottom of the waste body at the edge of the old part of the landfill and just at the edge of the present top liner.
A-04/3	Groundwater	Reference sample, natural groundwater upstream the landfill, which is not influenced by the landfill.

The microbial analyses, by means of the Q-PCR technique, give a clear insight into the number of micro-organisms that are present. This technique enables the accurate detection and quantification of micro-organisms based on distinguishing regions of their genetic material (DNA and RNA). Besides the aerobic nitrifying bacteria, the analyses included the quantification of the five Anammox-bacteria which have been identified up to now in literature (*Anammoxglobus*, *Brocadia*, *Jettenia*, *Kuenenia*, *Scalindua*).

The chemical analyses are related to the environmental conditions for an active anammox-process consisting of acidity (pH), bicarbonate, ammonium, nitrite, nitrate, nitrogen, oxygen, phosphorus and temperature (see table 2).

6. RESULTS

The results of the microbial and chemical analyses are presented in table 4 and 5.

Table 4. Results of chemical analyses in 2011 (n.a. = not analysed)

Chemical parameters	*Unit*	*Sample code*						
		A-04/3	Comp 4	Comp 5/6 March	Comp 5/6 April	D2-1	D4-D	D5-D
DOC (dissolved organic carbon)	mg/l	5.7	620		580	45	530	95
Bicarbonate (HCO_3^-)	mg/l	230	7,900		8,800	250	7,600	2,200
Kjeldahl-nitrogen (N)	mg/l	0.7	1,290		1,180	3.7	1,270	218
Ammonium (NH_4^+)	mg/l	0.2	1,300		1,100	0.8	1,200	220
Nitrate (NO_3^-)	mg/l	< 0.75	0.87		6.9	< 0.75	0.98	< 0.75
Nitrite (NO_2^-)	mg/l	< 0.3	< 0.3		13	< 0.3	< 0.3	< 0.3
Phosphorus (P)	µg/l	< 50	9,400		8,300	72	14,000	2,900

Table 5. Results microbial analyses in 2011 (n.p. = not present)

Type of micro-organism	*Unit*	*Sample codes*						
		A-04/3	Comp 4	Comp 5/6 March	Comp 5/6 April	D2-1	D4-D	D5-D
Total populations								
Total Bacteria	Cells/ml	8.4×10^{4}	1.4×10^{7}	5.3×10^{6}	2.1×10^{7}	1.3×10^{5}	5.5×10^{7}	4.8×10^{7}
Total Archaea	Cells/ml	5.5×10^{4}	1.2×10^{6}	9.9×10^{5}	8.4×10^{5}	2.7×10^{4}	1.8×10^{6}	7.4×10^{6}
Aerobic Ammonium Oxidation								
Bacterial ammonia monooxygenase	Cells/ml	37	140	680	311	n.p.	n.p.	248
Archaeal ammonia monooxygenase	Cells/ml	710	280	1,200	n.p.	n.p.	580	960
Anammox								
Anammoxoglobus	Cells/ml	present	present	120	n.p.	n.p.	present	185
Brocadia	Cells/ml	present	n.p.	n.p.	n.p.	n.p.		n.p.
Jettenia	Cells/ml	n.p.	n.p.	n.p.	n.p.	n.p.		present
Kuenenia	Cells/ml	n.p.	n.p.	n.p.	present	n.p.	n.p.	n.p.
Scalindua	Cells/ml	n.p.	n.p.	n.p.	n.p.	n.p.	n.p.	n.p.
Hydrazine oxidoreductase	Cells/ml	present	n.p.	n.p.	present	n.p.	n.p.	119
Additional information								
Number of ammonium degraders		747	420	2,000	311	n.p.	560	1,632
Detection limit		< 7	< 60	< 16	< 49	< 7	< 190	< 38

7. INTERPRETATION AND DISCUSSION

7.1 Environmental conditions

With reference to table 2 (process- and environmental conditions required for the anammox-process), the results of the critical chemical parameters in table 5 indicate that the environmental conditions can allow the anammox-process to be active.

7.2 Groundwater reference sample A-04/3

In comparison to the samples influenced by the landfill, the concentrations of the chemical analyses of the boundary conditions in this reference sample are far lower (by a factor 50 for DOC to a factor 3,000 for ammonium). The same is true for the concentrations of micro-organisms. The results indicate that the reference sample is clean for ammonium, but that a potential for the degradation of ammonium is present, in particular with respect to aerobic degradation of ammonium to nitrite (747 micro-organisms/ml). Furthermore anammox-bacteria were detected, although in very low numbers (< 7 cells/ml).

This means that the capacity for biological degradation of ammonium is not only found in situations with high ammonium concentrations. This corresponds to observations from literature, that biological decay of ammonium is a widespread phenomenon.

7.3 Leachate samples from the new part of the landfill (comp 4 and comp 5/6)

The concentrations of the chemical parameters analysed are significantly higher than in the reference sample. The same is true for the numbers of micro-organisms, which is probably caused by the degradation of organic matter in the landfill body. The numbers of micro-organisms for aerobic degradation of ammonium are higher than those for the anammox-process. The anammox bacteria were detected, but could not be analysed reliably owing to the low concentrations (< 60 cells/ml).

It should be pointed out that the numbers of bacteria in the latest sample of compartment 5/6 are significantly lower than in the previous sample (2,000 cells/ml versus 311 cells/ml). This might indicate that the anammox-process is not yet stable, possibly owing to the heterogeneity of the waste body.

Furthermore in the latest sample of compartment 5/6 nitrite was measured in low concentrations. As indicated before, nitrite is a prerequisite for the occurrence of the anammox-process under the condition that it may not exceed a concentration of 100 mg/l (inhibition). So far it is not yet clear if this is positive for the occurrence of the anammox-process, or if it is an indication that the anammox-process is not active resulting in an accumulation of nitrite.

7.4 Leachate samples from the old part of the landfill (D2-1, D4-D and D5-D)

It should be pointed out that sample D2-1 (shallow groundwater directly below the waste body) is chemically as well as biologically similar to the reference sample (upstream groundwater not influenced by the landfill). If the absence of ammonium was caused by biological degradation of ammonium, it is expected that high numbers of micro-organisms would have been detected as the responsible actor. As no micro-organisms were been detected, the water in this sample appears not to be influenced by landfill leachate.

The measured data of the sample D4-D are comparable to those of the samples from the new part of the landfill. This means high concentrations of ammonium and lower numbers of micro-organisms. The numbers of micro-organisms for aerobic degradation of ammonium are higher than those for anaerobic degradation of ammonium. The anammox bacteria were detected, but could not be quantified reliably owing to the relative high detection limit (< 190 cells/ml).

The results of sample D5-D show a relative low concentration of ammonium (220 mg/l) and simultaneously the highest number of micro-organisms for aerobic and anaerobic degradation of ammonium. This is an indication that this sample contains the highest capacity of decay for ammonium. It might therefore be concluded that the measured micro-organisms for aerobic and anaerobic degradation of ammonium are responsible for the low ammonium concentrations measured at this location.

8. CONCLUSIONS

The potential for aerobic and anaerobic degradation of ammonium was detected in all samples with the exception of sample D2-1. In sample D5-D several types of Anammox-bacteria were detected and quantified. This fact in combination with the low ammonium concentrations is the proof that at this location (D5-D) ammonium can be degraded biologically.

With the exception of sample comp 5/6, nitrite was not measured, even though the presence of nitrite is critical for the occurrence of the Anammox-process. It might be that the nitrite producing bacteria and the anammox-bacteria are living in symbiosis, which prevents the accumulation of nitrite. Another view could be that nitrite is the limiting factor for the anammox-process, so adding nitrite might accelerate the anammox-process.

It is a well known fact that the composition of the waste body is very heterogeneous. This also means that the conditions for the occurrence of the anammox-process can vary in time and place. This should be taken into account for the establishment of the sampling program and the interpretation of the results. This is also important to determine whether the anammox-process is stable or fluctuating.

The final conclusion is that the potential for biological degradation of ammonium is widespread in the Vlagheide landfill. This meant a ‘go’ for step 3 of the research program. Step 3 will focus on the kinetics and activity of the anammox-process as well as on the determination of the feasibility, opportunities and risks of practical implementation of the anammox-process.

9. ACKNOWLEDGEMENTS

The authors wish to thank Mr. Theo Folmer for assigning and financing this research program so far. He is the director of the Urban District of ‘s-Hertogenbosch, which is the owner of the Vlagheide landfill.