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Research Paper

# Application of the nitrogen-to-argon ratio to understand nitrogen transformation pathways in landfills under in-situ stabilization

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ABSTRACT

The ratio of nitrogen ($N_2$) to argon (Ar) in landfill gas was compared to the atmospheric gas ratio to quantify the balance between $N_2$ generating (anaerobic ammonium oxidation, denitrification) and $N_2$ consuming (nitrogen fixation) processes on three landfills undergoing in-situ stabilization. In the aerated landfills, as much as 22% of the extracted $N_2$ could be explained by net denitrification, with coexisting aerobic and anaerobic domains fostering nitrification-dependent denitrification. Nitrogen fixation was also occasionally observed. Removal of nitrogen via the gas phase exceeded nitrogen removed via the leachate by up to a factor of 33. Contrastingly, the anaerobic landfill under leachate recirculation showed a net reduction of $N_2$ in relation to Ar, indicating nitrogen fixation as the dominant mechanism, equivalent up to 28% of the nitrogen in the extracted landfill gas. The balance between denitrification and nitrogen fixation in the aerated sites varied seasonally, likely caused by increased evapotranspiration in the summer, allowing greater air intrusion through the cover soil, resulting in higher $NO_3^-$ and $NO_2^-$ availability for denitrification and anammox. No such variability was observed for the landfill under liquid recirculation. The nitrogen transforming microbial community comprised of species responsible for nitrification, ammonification, denitrification, and anammox, indicating all processes may coexist. The findings show aeration supports nitrogen removal through the gas phase, but also suggest that nitrogen fixation adds nitrogen to the waste body in anaerobic domains. This could delay reaching environmental compliance criteria for leachate nitrogen, both for in-situ treatment by aeration and by leachate recirculation.

## 1. Introduction

One of the primary goals of landfill in-situ stabilization in the Netherlands is to reduce the landfill management life-cycle by quickly reaching and maintaining the leachate quality criteria defined by local environmental protection regulations (Brand et al., 2016; Dijkstra et al., 2018). This is achievable by enhancing waste biodegradation, depleting waste reactivity, and reaching stability within a shorter timeframe (Laner et al., 2012; Scharff et al., 2010). Increasing biodegradation rates and quickly reducing the emission potential can be achieved by recirculating with leachate or water (Benson et al., 2007; Reinhart, 1996) or by promoting aerobic conditions using in-situ aeration systems (Ritzkowski and Stegmann, 2012).

Among a wide range of possible contaminants in the landfill leachate, ammonium ($NH_4^+$) is suggested to be most persistent (Barlaz et al., 2002). Seepage into the underlying soil and migration into the ground and surface waters from failing waste containment systems pose significant risks, as $NH_4^+$ can increase eutrophication, deplete dissolved oxygen, and increase toxicity in living organisms. Ammonium in the landfill leachate can be present in high concentrations (up to thousands of mg/L) as the main nitrogenous product of anaerobic decay in organic wastes (Li et al., 1999).

Leachate recirculation is intended to elevate anaerobic decay rates of organic matter and flush pollutants such as $NH_4^+$ out of the waste body over time. Landfill aeration, on the other hand, should stimulate the breakdown of organic matter under aerobic conditions, enhancing nitrification and promoting the formation of $NO_2^-$ and $NO_3^-$. However, waste heterogeneity and compaction often limit waste permeability, impeding spatial aeration efficiency. This leads to a multiplicity of anaerobic and aerobic domains alternating within the waste body, sometimes promoting denitrification and thereby enhancing the release of nitrous oxide ($N_2O$) and molecular nitrogen ($N_2$) through the gas phase (Berge et al., 2005; Dang et al., 2023). In both aeration and recirculation conditions, $NO_2^-$ can also be used to transform $NH_4^+$ into $N_2$ by anaerobic ammonium oxidation or anammox (Berge et al., 2005; Mulder et al., 1995).

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While $NH_4^+$, $NO_3^-$, and $NO_2^-$ concentrations are easier to monitor in the leachate, the fraction of nitrogen generated in the gaseous phase in the landfill is less understood. To faciliate mass balancing and more accurately assess the remaining nitrogen emission potential, quantifying nitrogen gas leaving the landfill is needed. $N_2O$ emissions as a product of nitrification–denitrification have been quantified by several researchers on full scale and in laboratory landfill simulation reactors (Chu et al., 2022; Harborth et al., 2013; Li et al., 2017, 2018; Sun et al., 2013; Zhang et al., 2009), but molecular nitrogen ($N_2$) as the final product of denitrification is less investigated. As the fraction of $N_2$ measured in the extracted or emitted gas also originates from the atmospheric air, especially in the case of actively aerated landfills, new approaches are needed to distinguish the atmospheric component from $N_2$ produced by denitrification within the waste body.

This study employs the noble gas argon (Ar), which can only originate from atmospheric air, as a tracer to differentiate the $N_2$ emerging from the waste biodegradation from the $N_2$ originating from atmospheric air. Hence, the difference between the $N_2$ to Ar ratio in the extracted landfill gas to the atmospheric ratio can be used to identify the net share of $N_2$ originating from microbially mediated nitrogen transformations (Nagamori et al., 2016; Shigemitsu et al., 2016). Enrichment of $N_2$ in relation to Ar (compared to their ratio in atmospheric air) indicates $N_2$ released from denitrification. In contrast, the depletion of $N_2$ in relation to Ar (compared to their ratio in atmospheric air) indicates nitrogen fixation. This study hypothesized that in the aeration pilots, enhanced nitrification would support denitrification in anaerobic domains, resulting in $N_2$ generation and, hence, a higher ratio of $N_2$ to Ar in the landfill gas compared to the atmospheric ratio. For the anaerobic recirculation pilot, this effect was assumed to be minimal.

## 2. Material and methods

### 2.1. Landfill description

The sustainable landfill management program in the Netherlands (iDS) investigates the viability of landfill in-situ stabilization in three landfill pilots: Braambergen (BRA), Wieringermeer (WIE), and De Kragge (KRA). More information on the characteristics of the three pilots is given in Supporting Information (Table S1 and in Fig. S1).

#### 2.1.1. Waste composition

Braambergen landfill is located in Almere, The Netherlands, and stores ~ $1.2 \times 10^6$ t of wastes distributed over four cells (11N, 11Z, 12W and 12O), landfilled between 1999 and 2008. The cells were filled mainly with contaminated soils and soil residues: 96 $\%_{mass}$ in cells 11Z and 12O, 73 $\%_{mass}$ in cell 11N, and 40 $\%_{mass}$ in cell 12 W. The remaining wastes were construction and demolition, commercial, and other household wastes. Cells 12O and 12W are presented as one compartment (12) herein, because the leachate and gas collection from 12O and 12W are combined.

Wieringermeer landfill located in Middenmeer, The Netherlands, contains ~ $2.8 \times 10^5$ t of mainly commercial waste (72 $\%_{mass}$), landfilled between 1992 and 2003 in compartment 6. The remaining wastes were sludge and composting materials (22 $\%_{mass}$), construction and demolition wastes (3 $\%_{mass}$), contaminated soils (2 $\%_{mass}$), domestic and household wastes (1 $\%_{mass}$), and shredded wastes (0.2 $\%_{mass}$).

Landfill De Kragge in Bergen op Zoom, The Netherlands, deposited ~ $1 \times 10^6$ t of mainly domestic wastes (29 $\%_{mass}$), construction and demolition wastes (21 $\%_{mass}$), and commercial wastes (21 $\%_{mass}$). The remaining wastes were sludge and composting wastes (15 $\%_{mass}$), bulky household wastes (13 $\%_{mass}$), soil residues and shredder wastes (1 $\%_{mass}$).

#### 2.1.2. Infrastructure and operation of landfill aeration

BRA and WIE started in-situ aeration in 2017. The aeration system consists of wells (230 in BRA and 109 in WIE) distributed over 10 ha and 2.6 ha, respectively. The distances between wells vary between 15 and 20 m in BRA and 12.5 to 13.5 m in WIE. In 2022, additional 98 wells were installed at BRA in compartments 11N and 11Z. Aeration wells were used to inject ambient air and extract landfill gas (*combi-aeration*) or to extract landfill gas at wells at higher rates than landfill gas production (*over-extraction*) to promote atmospheric air intrusion into the wastes through the cover soil (Cruz et al., 2021; Cruz and Lammen, 2023; Meza et al., 2022).

The BRA started two-years of *over-extraction* followed by another two years of *combi-aeration* before returning and maintaining *over-extraction*. The gases from the individual wells were transported to eight main pipes from the three compartments (two from 11N, four from 11Z, and two from 12) and combined at the blower station. From August 2022 to February 2024, the pressure and flow in BRA ranged −98 to 1 hPa and −17 to 1392 $m^3/h$, respectively.

In contrast, *over-extraction* was employed consistently at WIE since 2017. The proportion of the bulk gas in compartment 6 accounts for ~ 84.7 % of the total gas recovery since the aeration pipes extend ~ 20 % into the adjacent compartments to minimize boundary effects. Between August 2022 to February 2024, the mean pressure in both BRA and WIE was −35 hPa. The mean flow rate was lower in BRA (377 $m^3/h$) compared to WIE (437 $m^3/h$). WIE exhibited a pressure range of −69 to 1 hPa, and the flow ranged −10 to 1087 $m^3/h$.

#### 2.1.3. Infrastructure and operation of leachate recirculation

Leachate recirculation at KRA began in March 2018 over an area of 5.6 ha. Approximately 4.3 $m^3/h$ of leachate was produced between November 2022 to February 2024 and treated by the anammox water purification system. Approximately 81 $\%_{vol.}$ of treated leachate was then discharged into the sewage system, and the remainder ~ 19 $\%_{vol.}$ (0.8 $m^3/h$) was diluted with clean water (2.4 $m^3/h$) and then circulated to the KRA pilot at a rate of 3.2 $m^3/h$ during the course of the study. The infiltration rates varied between 1.8 and 7.2 $m^3/h$.

The gas extraction system at the KRA pilot landfill is integrated with other compartments. The landfill gases, extracted (approximately 19 $m^3/h$) via 13 gas wells from the recirculation landfill, are combined with gases from other compartments. The combined gases are converted into electricity and heat through a cogeneration unit used to power the plants and the surplus energy is fed into the electricity grid. Additionally, the heat is used to warm the anammox and nitritation reactors to temperatures of 25–30 °C.

### 2.2. Gas sampling

The gas samples were collected between August 2022 to February 2024 from pipes and wells fitted with gas sampling valves. The landfill gas (~10 ml) was injected into a 15 mL vial filled with HCl acidified distilled water (~pH 1.8) using a gas-tight syringe to inject the sample and displacing ~ 5 mL of water through a rubber-septum-fitted cap. The acidified water was used to minimize nitrogen transformation, and the vials were stored inverted to prevent gas leaks until further analysis. Table S2 (Supporting Information) lists the sample identification (ID) and the location where each gas sample was collected.

The gas samples were analyzed for $CH_4$, $CO_2$, $O_2$, $N_2$, Ar, and $N_2O$ using a gas chromatograph (Agilent 8860 GC System, Santa Clara, California, USA) installed with a MXT-Q-BOND 30 m × 0.53 mm × 20 μm coil column and a CP-Molesieve 5A 50 m × 0.53 mm × 50 μm column, equipped with a thermal conductivity detector (Agilent, Santa Clara, California, USA) using helium carrier gas at 10 mL/min, pressure at 137.27 kPa, and initial temperature set at 35 °C.

### 2.3. Composition of the microbial community

Leachate samples were collected in August and November 2022 from the leachate pump pits from the three compartments of Braambergen (11N, 11Z, and 12) and Wieringermeer. The leachate samples from

Kragge were collected in July and November 2022. The sample tap was opened and flushed at each location for approximately 10 minutes to remove sediments, precipitates, and sludge. Then 1–1.5 L of leachate samples were collected in a plastic bottle, stored at −80 °C and sent to Orvion B.V. (Stolwijk, Netherlands).

Approximately 1 L of the leachate sample was filtered and treated for DNA extraction using the Dneasy PowerSoil ProKit (Qiagen Group, Venlo, The Netherlands). The FastPrep-24 Classic Instrument (MP Biomedicals, Irvine, California, USA) was used for lysis. The lysate was purified with an equal volume of DNA binding solution and passed through a silica spin filter membrane that was washed in a two-step regime. The Silica-bound DNA was then eluted using a 10 mM Tris elution buffer to isolate the DNA and sequenced using the Oxford Nanopore Next Generation Sequencing (NGS). The sequencing data were submitted to the National Center for Biotechnology Information (NCBI) Database (National Library of Medicine, Bethesda, Maryland, USA) under BioProject number PRJNA1147207.

The identification of unassembled reads was then performed using a BLAST-based routine on a custom Bacteria-Archaea database containing a selection of accessions from the nucleotide database from the NCBI Database using the Whole Genome Shotgun (WGS) and the Reference Sequence (RefSeq) to identify species responsible for nitrification, nitrogen fixation, ammonification, anammox, and denitrification. The identified species were plotted by the % relative abundance of the hybridization product.

### 2.4. Aeration efficiency

The aeration efficiency (AE), expressed as the percentage of aerobic carbon ($CO_2$) in relation to total extracted carbon ($CH_4$ + aerobic and anaerobic $CO_2$), was estimated as follows (Gebert et al., 2023; Yazdani et al., 2010):

$$AE = \frac{[CO_{2,TOT}] - [CO_{2,AN}]}{[CO_{2,TOT}] + [CH_4]} \times 100 \qquad [1]$$

where AE is the aeration efficiency (%), $[CO_{2,TOT}]$ is the concentration of total $CO_2$ ($\%_{vol.}$), $[CO_{2,AN}]$ is the concentration of anaerobically produced $CO_2$ ($\%_{vol.}$) which is equal to the concentration of $CH_4$ ($\%_{vol.}$), $[CH_4]$ is the concentration of $CH_4$ ($\%_{vol.}$), with AE ≥ 0 %. The approach assumes that during anaerobic decay of waste organic matter, $CH_4$ and $CO_2$ are produced in a ratio of 1:1 (acetotrophic methanogenesis) (Ferry, 2010).

### 2.5. Ratio of nitrogen to argon and excess nitrogen

Net formation of $N_2$ by denitrification is assumed if the ratio of $N_2$ to Ar in the landfill gas ($[\frac{N_2}{Ar}]_{LFG}$) exceeds the atmospheric ratio ($[\frac{N_2}{Ar}]_{Atm}$), while the net consumption of $N_2$ by nitrogen fixation can be assumed if $[\frac{N_2}{Ar}]_{LFG}$ is lower than $[\frac{N_2}{Ar}]_{Atm}$ (Nagamori et al., 2016; Shigemitsu et al., 2016). For $[\frac{N_2}{Ar}]_{Atm}$, the atmospheric $N_2$ concentration was 78.08 $\%_{vol.}$, and the Ar was assumed as 0.93 $\%_{vol}$ (National Oceanic and Atmospheric Administration, 2023). $[\frac{N_2}{Ar}]_{LFG}$ reflects the measured $N_2$ ($N_{2\,LFG}$) and Ar ($Ar_{\,LFG}$) in the respective gas sample, thus $N_{2\,LFG}$ includes $N_2$ produced from waste degradation and $N_2$ from the atmosphere.

The $N_2$ in the landfill gas that is of atmospheric origin $N_{2\_Atm\_LFG}$ is calculated and expressed in unit $\%_{vol.}$ by:

$$N_{2\_Atm\_LFG} = Ar_{\_LFG}\left(\frac{N_{2\_Atm}}{Ar_{\_Atm}}\right) \qquad [2]$$

Nitrogen produced or consumed by the waste body is thus derived from the difference between $N_{2\,LFG}$ and the share of nitrogen that is of atmospheric origin based on Ar in the gas ($N_{2\_Atm\_LFG}$, Eq. 2). This difference is then expressed as a percentage of the total nitrogen measured in the landfill gas sample ($N_{2\,LFG}$) and the results are defined as $N_{2\_Excess}$ in this paper:

$$N_{2\_Excess} = \frac{N_{2\_LFG} - N_{2\_Atm\_LFG}}{N_{2\_LFG}} x\,100 \qquad [3]$$

with $N_{2\,LFG}$ and $N_{2\,Atm\_LFG}$ given in the unit $\%_{vol,}$ and $N_{2\_Excess}$ expressed as percentage (%).

The value of $N_{2\_Excess}$ represents the balance between all nitrogen producing and nitrogen consuming processes. Positive values of $N_{2\_Excess}$ indicate net generation of nitrogen within the landfill (denitrification), while negative values of $N_{2\_Excess}$ suggest that nitrogen fixation dominates the nitrogen balance, resulting in net nitrogen consumption.

## 3. Results

### 3.1. Composition of landfill gas

Fig. 1 illustrates the upper and lower quartiles, mean, and range of the concentration of six measured landfill gas components in samples from all locations and the expected atmospheric trace gas concentrations (shown in red dotted horizontal lines). The highest mean $CH_4$ concentration was found in KRA, the strictly anaerobically operated landfill (~51 $\%_{vol.}$), whereas the aerated pilots (BRA and WIE) showed reduced mean $CH_4$ concentrations of ~ 8 $\%_{vol.}$ and 6 $\%_{vol.}$, respectively (Fig. 1a).

The highest mean concentration of $CO_2$ (~28 $\%_{vol.}$) was in KRA, a liquid recirculation plant with a high share of domestic waste (Fig. 1b). The WIE landfill had the second highest mean $CO_2$ concentration (~17 $\%_{vol.}$), and BRA landfill had a mean $CO_2$ of 12 $\%_{vol.}$. However, the highest single measurement (52 $\%_{vol.}$) of $CO_2$ concentration was recorded in BRA (Fig. 1b).

The KRA showed the highest $CH_4$, $CO_2$, and the lowest $CO_2/CH_4$ ratio of < 1 (Fig. 2a), indicating a strictly anaerobic microbial activity evidenced by low $O_2$ (<0.5 $\%_{vol.}$) (Fig. 1c). The $CO_2/CH_4$ was the highest (mean of ~ 3.6) in WIE, and BRA had the following highest mean of ~ 2 resulting from each datapoint (Fig. 2a), indicating the highest share of aerobic processes coinciding with the highest $N_2O$ concentrations (Fig. 1f).

$N_2O$ was detected in concentrations as high as ~ 28 ppmv in WIE, with a mean concentration of ~ 2.4 ppmv. BRA produced $N_2O$, with a mean concentration of 0.6 ppmv $N_2O$ and a maximum of 23 ppmv measured on 11N compartment with the highest aeration efficiency (Figs. S2 and S4 in Supporting Information).

Argon concentrations were the lowest in KRA (~0.3 $\%_{vol.}$ Ar) due to the low share of atmospheric air present in the landfill. The argon concentration between the BRA compartments and WIE were similar (~0.8 $\%_{vol.}$), but low Argon (< 0.5 $\%_{vol.}$) concentrations were observed in individual gas wells. Higher Ar concentrations (near 0.9 $\%_{vol.}$) indicate the presence of atmospheric air in the landfill gas and low Ar indicates low shares or absence of atmospheric air in the respective sample.

Concentrations of $CH_4$, $N_2$, Ar, and $N_2O$ were similar between the two aerated pilots, BRA and WIE (Fig. 1). However, there were apparent differences between $CO_2$ and $O_2$. $O_2$ concentrations were higher in BRA than WIE, and WIE gas showed a higher $CO_2$ content than BRA, indicating higher rates of $O_2$ consuming processes and hence a higher AE at pilot WIE (Fig. 2).

### 3.2. Aeration efficiency

For Braambergen, ~38 % AE was observed in the investigation period, while WIE landfill exhibited the highest AE (~57 %). The lower efficiencies (AE ≈ 0 %) were also observed in individual wells in all seasons for BRA. Some individual wells can have poor air flow from clogged filters caused by accumulated water or from waste settlement that reduce permeability. Impermeable conditions can reduce $O_2$ available to the microorganisms, deterring aerobic conversion of waste

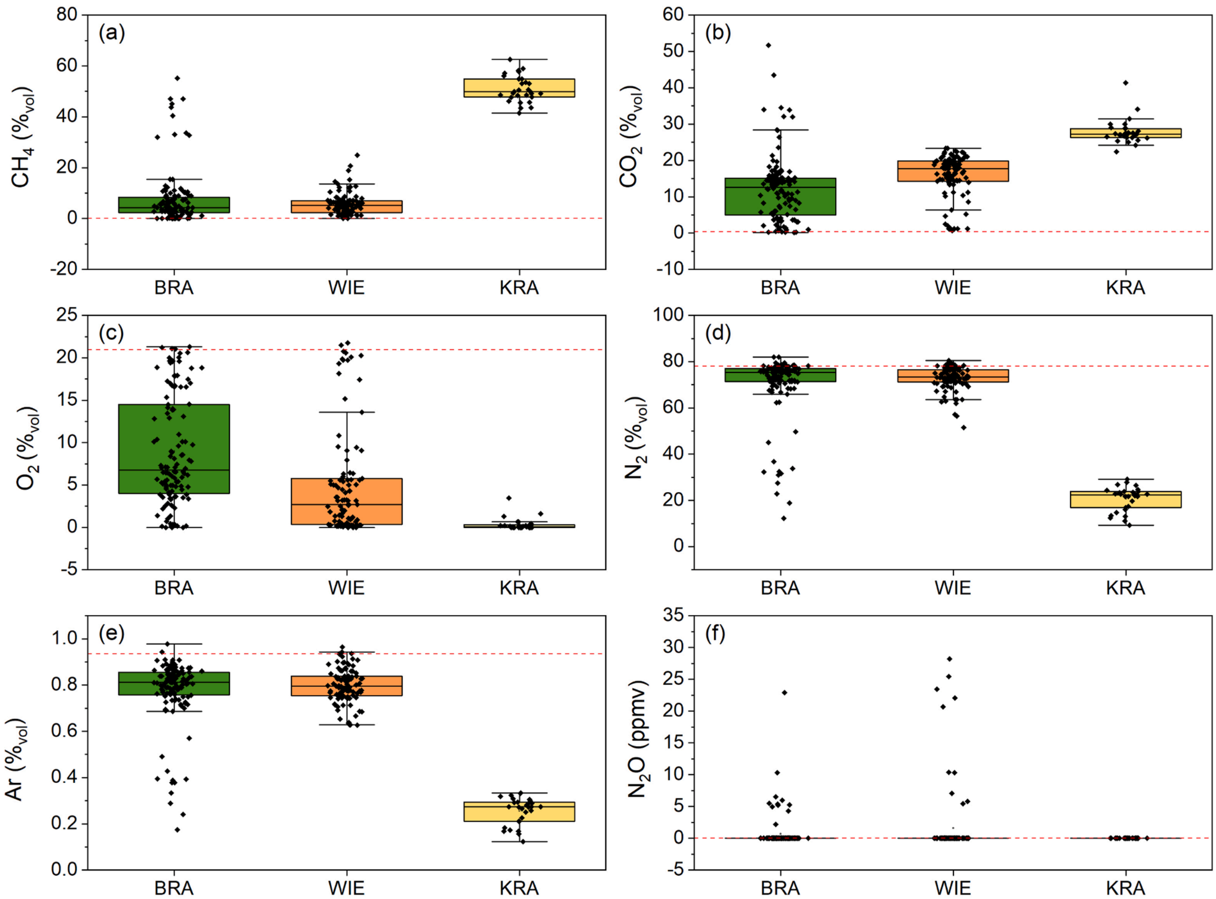


**Fig. 1.** Composition of landfill gas from pilots Braambergen (BRA), Wieringermeer (WIE) and Kragge (KRA). Box = 25th-75th percentile, line = mean, whiskers = 10th-90th percentile. The red dotted line presents expected atmospheric concentrations.

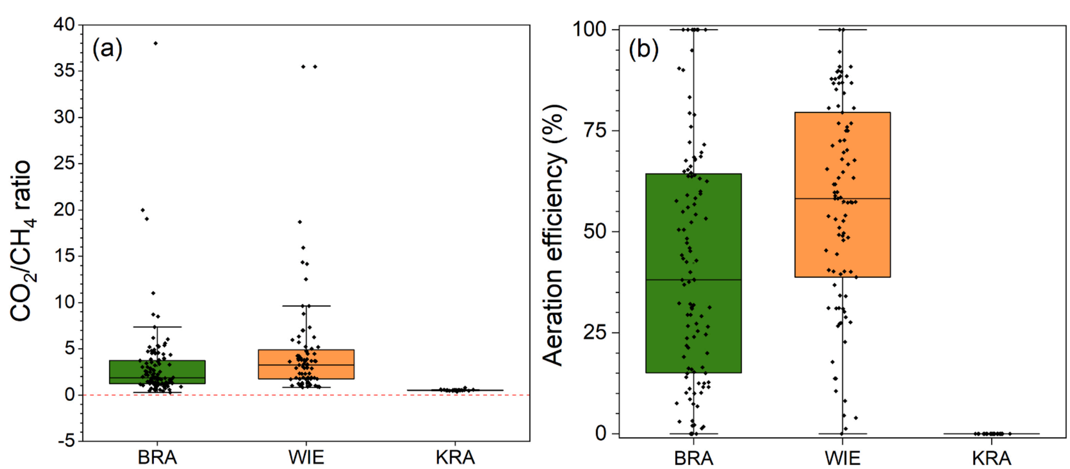


**Fig. 2.** (a) Ratio of $CO_2$ to $CH_4$; (b) aeration efficiency (Eq. 1) in landfill gas from pilots Braambergen (BRA), Wieringermeer (WIE) and Kragge (KRA). The red dotted line represents expected ambient concentrations.

organic matter and reducing $CO_2$ generation. Furthermore, the higher $CO_2/CH_4$ ratio at WIE (Fig. 2a) coincided with lower $O_2$ concentrations compared to BRA (Fig. 1c). The strictly anaerobic operational conditions for KRA under leachate recirculation reflects in the lowest $CO_2/CH_4$ ratio and corresponding zero or near-zero AE.

### 3.3. Nitrogen balance

Fig. 3a and 3b show the $N_2/Ar$ ratio measured in the landfill gas over the respective atmospheric ratio (Eq. 2) and the net nitrogen concentration leaving the landfill in the gaseous phase expressed as $N_{2\_Excess}$ (Eq. 3) assessed from the concentration of the Ar in % of the total $N_2$ in the landfill. Positive values of $N_{2\_Excess}$ indicate net nitrogen generation (denitrification) and negative values assume net nitrogen consumption (fixation). Fig. 3c shows the relationship between aeration efficiency and the $N_{2\_Excess}$ for all locations, whereas Fig. 3d demonstrates the same relationship only in the bulk gases of the three landfills. The release of $N_2$ from the net denitrification mainly occurred in landfills under

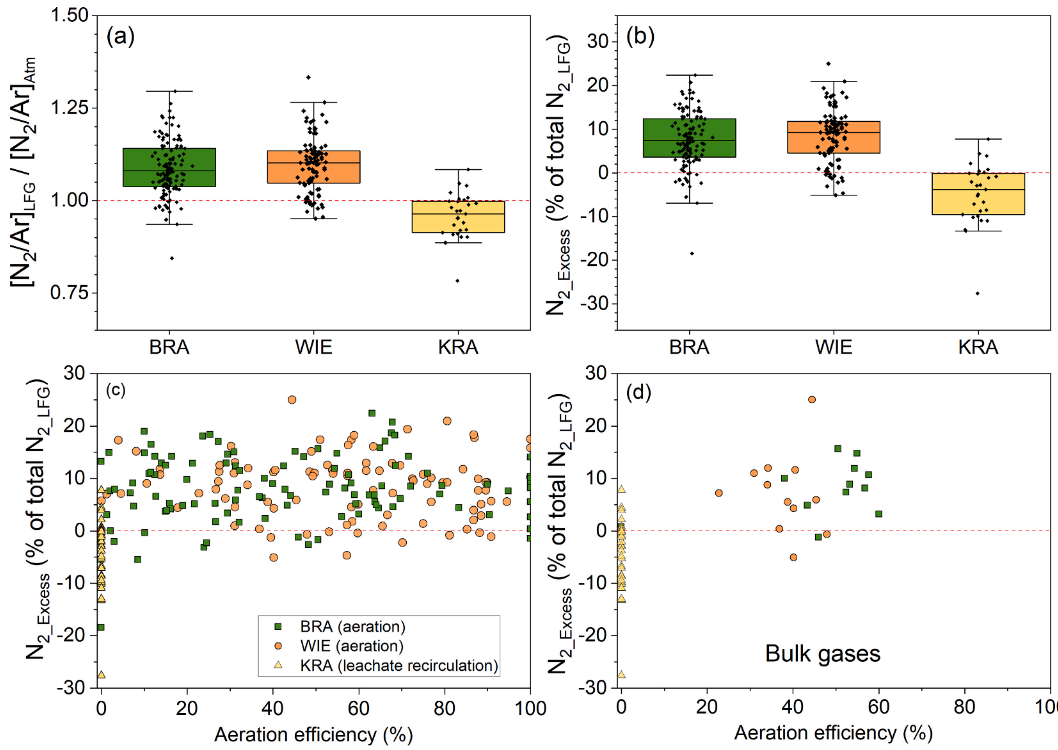


**Fig. 3.** (a) Ratio of $N_2$ /Ar in the landfill gas over the ratio of $N_2$ /Ar in the atmosphere; (b) the corresponding share of excess $N_2$ according to Eq. 3; (c) $N_2$ excess related to the aeration efficiency for individual wells; (d) $N_2$ excess vs. the aeration efficiency for the bulk extracted gas in pilots Braambergen (BRA), Wieringermeer (WIE), and Kragge (KRA). Box = 25th-75th percentile, line = mean, whiskers = 10th-90th percentile. The red dotted line presents the respective atmospheric condition.

aeration, with a mean 7.4 % of $N_{2\_Excess}$ in the BRA landfill and 8.5 % in landfill WIE. The balance under leachate recirculation (KRA) had a mean –4.9 % $N_{2\_Excess}$, indicating a net $N_2$ consumption (Fig. 3b).

Fig. 3c and 3d demonstrate the relationship between AE% as an indicator of the share of aerobic activity (Eq. 1) and the share of nitrogen production or nitrogen fixation (Eq. 3). In the absence of aerobic processes (values for AE% equal zero), primarily negative values, i.e., net nitrogen uptakes were observed, particularly visible if only data for the bulk extracted gas (Fig. 3d) were considered. For aerated conditions, a similar range of net nitrogen production was observed for a wide range of aeration efficiencies.

The data were plotted according to each monitoring location to illustrate spatial and seasonal differences in the $N_2$ balance (Figs. 4-6). Seasonal variation had a more significant impact on the balance between $N_2$ uptake and $N_2$ release than spatial variation or the differences in the waste inventory, particularly on aerated landfills. The peak of $N_{2\_Excess}$ was observed in spring (March-April) and in September in all three compartments in Braambergen (Fig. 4). The individual wells and the bulk gas reached the maximum in spring but remained mostly elevated with an $N_{2\_Excess}$ around or above 10 % throughout the summer.

In BRA compartment 11Z, sample 2P1 had a nitrogen fixing condition, indicated by a negative value for $N_{2\_Excess}$ in June 2023 (Fig. 4). Furthermore, compartments 11N (NP1 and NP2) and 12 (12P1 and 12P2) showed $N_{2\_Excess}$ ranging between −1 to −3%, suggesting a nitrogen fixating condition in the bulk gas in November 2022 (Fig. 4a to 4d), also a net negative balance (−5%) for sample 12P2 in February 2023 (Fig. 4c). The bulk gas from the WIE had nitrogen fixation conditions in October and December 2022, and in gas wells 12, 29, 62, 98, and 115 with $N_{2\_Excess}$ ranging from −0.3 % to −5% in the same two months, respectively (Fig. 5). The most negative $N_{2\_Excess}$, i.e., the highest share of nitrogen fixation was in KRA from well G26 (−28 %) from September 2023. During the investigation period, no consistent seasonal trend could be identified at de Kragge (Fig. 6).

Supporting Information presents the correlation between $N_{2\_Excess}$ and $O_2$, $CO_2$, $CH_4$, and the $CO_2/CH_4$ ratio (Figs. S7-S10). The anaerobic KRA under leachate recirculation showed low positive to negative $N_{2\_Excess}$ (nitrogen fixation) combined with the low $O_2$ (Fig. S7), high concentrations of $CO_2$ and $CH_4$ (Figs. S8 and S9), and a low $CO_2/CH_4$ ratio (Fig. S10). High $CO_2$ and $CH_4$ concentrations relate to more negative $N_{2\_Excess}$. On the aerated BRA and WIE landfills, in contrast, $O_2$ in the extracted landfill gas varied widely between near zero and atmospheric $O_2$ concentrations, and $CO_2$ and $CH_4$ concentrations were significantly lower than KRA, with most values below 25 %. $N_{2\_Excess}$ was mostly positive, indicating net denitrification, but a clear correlation to either gas component was not present.

Using the gas composition, flow rates, temperature, pressure from the bulk gases, along with the value of $N_{2\_Excess}$ (Eq. 3), the amount of nitrogen released by net denitrification or uptake by net nitrogen fixation through the gas phase was estimated for the investigation period (Table 1). The data were then compared to the nitrogen discharge load from the landfill via the leachate pathway, using the leachate flow rates and the average concentrations of $NH_4^+$, $NO_3^-$, and $NO_2^-$ (Table S3 in Supporting Information). It is estimated that on the aerated sites, nitrogen discharged in the gas phase exceeded leachate nitrogen discharge by a factor of ∼ 25 (BRA) to ∼ 33 (WIE), whereas nitrogen discharged on the anaerobically operated landfill KRA was dominated by the leachate pathway. Here, the net fixation of atmospheric nitrogen, as indicated by a decreased ratio of $N_2$ to Ar in the landfill gas compared to the respective atmospheric ratio (see also Fig. 3a) amounted to ∼ 3.5 t nitrogen per year, whereas a nine-fold higher amount is released with the landfill leachate.

### 3.4. Composition of the microbial community

Fig. 7 illustrates the microbial taxa at the genus level, identified in the leachate of the three landfills affiliated with nitrogen conversion

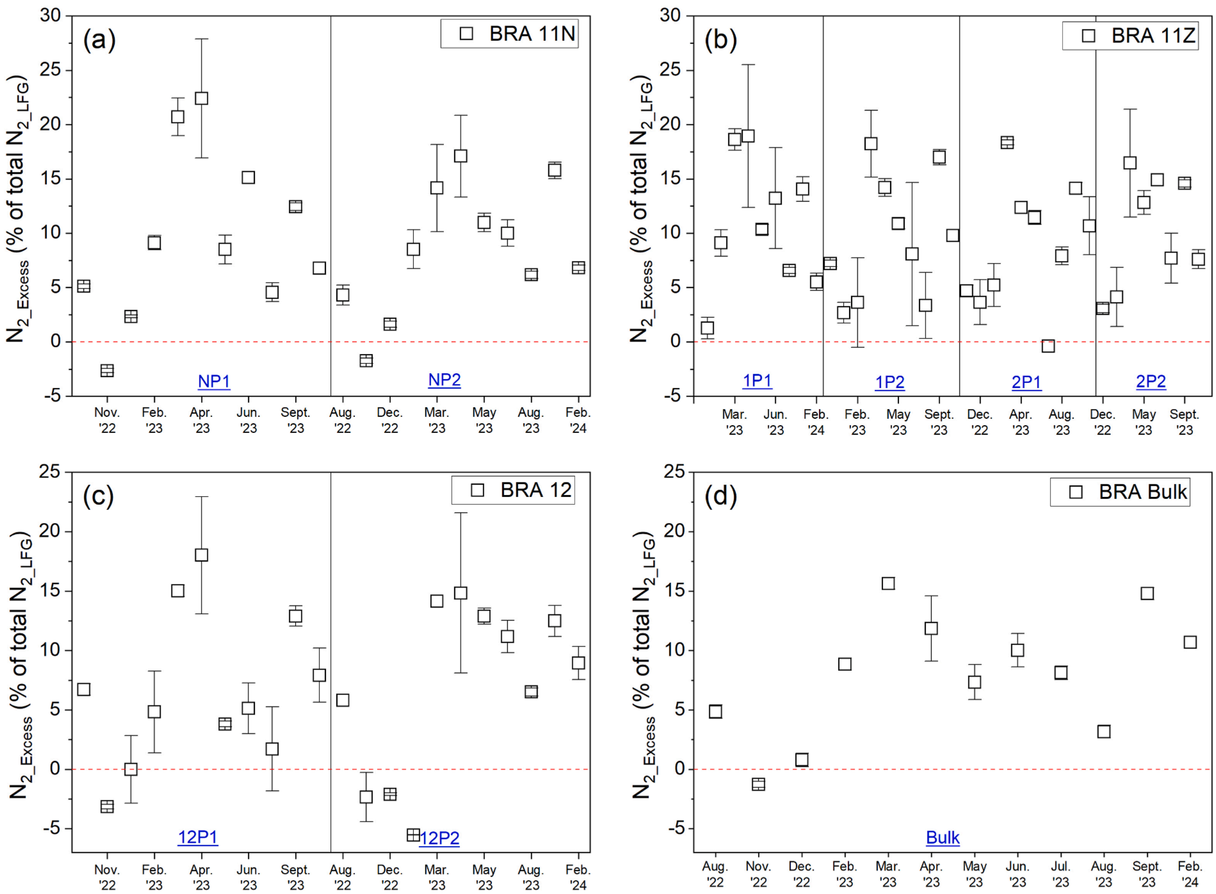


**Fig. 4.** Spatial and temporal variability of mean $N_{2\_Excess}$ (% of the total $N_{2\_LFG}$) in landfill gas (Eq. 3) from Braambergen (BRA) compartment 11Z, 11N, 12, and bulk gases. Blue underlined text = sample ID (for sampling location see Table S2 in Supporting Information). Error bars = standard errors.

pathways through: (a) nitrification; (b) nitrogen fixation; (c) anammox; (d) denitrification; and (e) ammonification from two sampling events in 2022. The % relative abundance reflects the relative share of each taxon within the total bacterial DNA in the sample. For all three sites, species with genes associated with denitrification, nitrogen fixation and ammonification were more abundant (>2 %) than species with genes that support nitrification (<1 %) and the anammox process (<1.5 %). The nitrogen affiliated microbial community composition was distinctly different at the three sites, with a striking presence of *Nitrospira sp.* at WIE, high abundance of *Pseudomonas* sp. at KRA, and high abundance of anammox related bacteria, *Candidatus Kuenenia*, at BRA. The abundance of the denitrifiers was lower in the BRA and WIE than for KRA (Fig. 7d).

In the BRA landfill, *Pseudomonas, Acinetobacter, Thiobacillus*, and *Paracoccus* were the four most abundant denitrifiers (~1 to 5 %). In August 2022, the population of *Acinetobacter* was more abundant than *Thiobacillus* in compartment 11N; however, in November 2022, *Acinetobacter* species decreased and the *Thiobacillus* species increased. *Streptomyces* species also increased in BRA compartments 11Z and 11N during November 2022. The abundance of *Pseudomonas* species remained relatively constant for both sampling events for all three compartments. There was a slightly higher abundance of nitrogen fixing bacteria than the denitrifiers in BRA in November than in August (~2 to 4 %). The largest abundance of nitrogen fixing bacteria in August 2022 were represented by *Pseudomonas, Thiobacillus*, *Desulfovibrio*, *Acidithiobacillus*, and *Halothiobacillus* species (Fig. 7b). In November 2022, *Acidithiobacillus, Thiobacillus,* and *Methylomonas* species increased for all three compartments in BRA. Also, the abundance of nitrogen fixing bacteria increased for all three compartments by 21 % (BRA 12) to 120 % (BRA 11Z).

The composition of the microbial community was similar in the WIE and BRA leachates, with a total abundance of denitrifiers increasing from August to November due to an increase in abundance of *Paracoccus* species. The denitrifiers consisted of *Acinetobacter* species with the highest 0.3 %, followed by *Pseudomonas* (0.3 %), *Paracoccus* (0.2 %), *Bacillus* (0.2 %), and *Thiobacillus* (0.2 %) in August. In November, *Acinetobacter* decreased (0.1 %), but *Paracoccus* increased (0.9 %). *Thiobacillus* also increased to 0.2 %. For the nitrifiers, the share of *Nitrosomonas* species increased from 0.04 to 0.1 % in WIE. WIE leachate was dominated by *Nitrosospira* (0.33 % and 0.49 %, respectively). *Pseudomonas* was maintained at ~ 0.3 % for both sampling events. The total nitrogen fixing organisms ranged from 1.5 to 1.9 % in WIE.

Compared to the aerated sites, the landfill under recirculation (KRA), with the highest share of biodegradable waste, showed the largest abundance of species affiliated with nitrogen fixation, ammonification, and denitrification. In contrast, the abundance of nitrifiers and anammox bacteria was in the same order of magnitude or considerably lower, respectively. The genus with the highest abundance of microorganisms associated with nitrogen transformation in KRA was *Pseudomonas* (6 %), collected during July 2022 sampling, which increased to ~ 28 % in November 2022. The genus *Pseudomonas* is categorized as both denitrifier and nitrogen fixer, along with *Thiobacillus*. *Alcaligenes* species was the third largest nitrogen group of nitrogen fixing bacteria. *Clostridium* was also identified, remaining at an equal share for both sampling events.

## 4. Discussion

The presented study addresses the knowledge gap regarding the occurrence and quantification of landfill $N_2$, as well as the balance between nitrogen formation and fixation in landfills under various

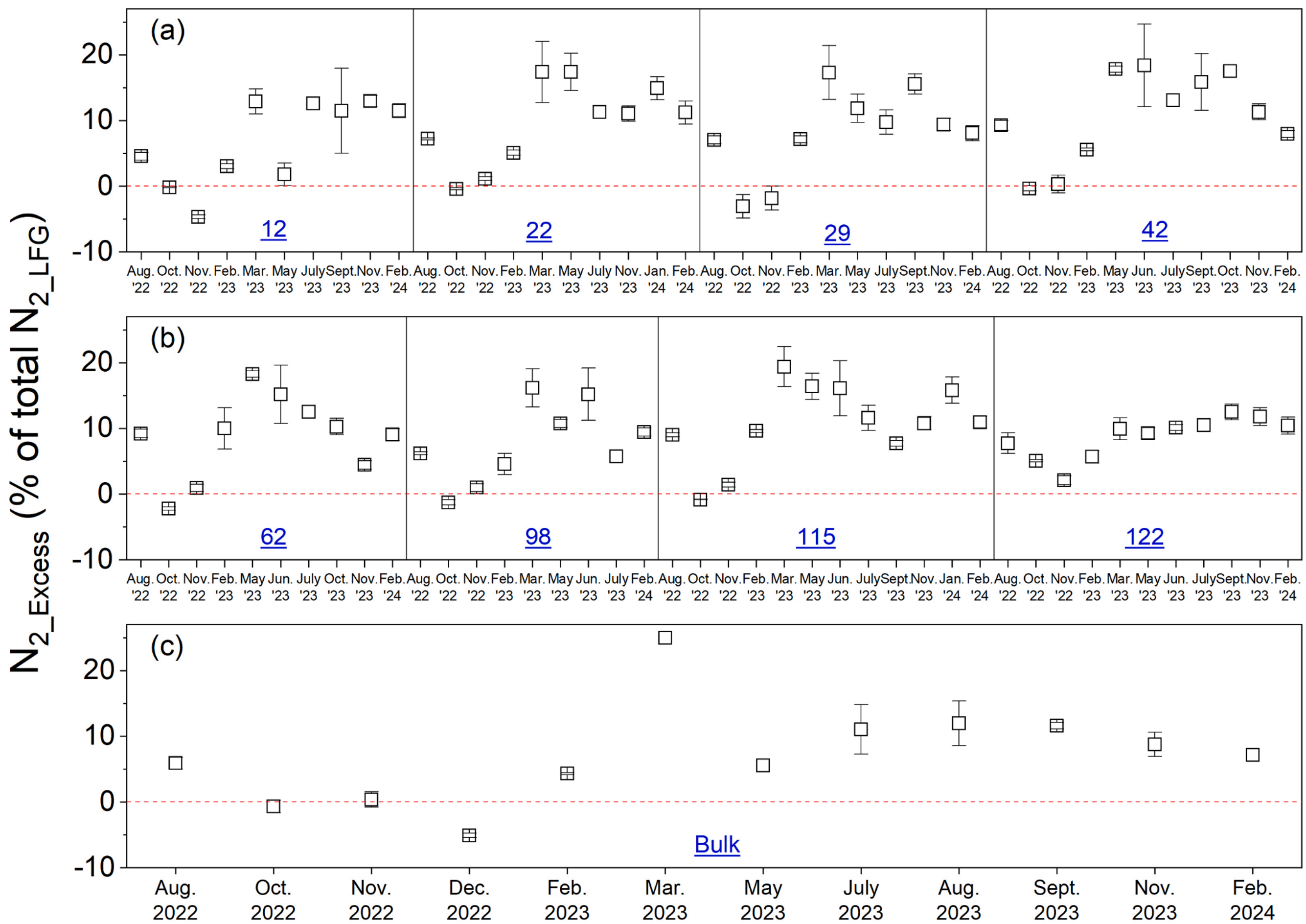


**Fig. 5.** Spatial and temporal differences of mean $N_{2\_Excess}$ (% of the total $N_{2\_LFG}$) in landfill gas (Eq. 3) from Wieringermeer (WIE). Blue underlined text = sample ID (for sampling location see Table S2 in Supporting Information). Error bars = standard errors.

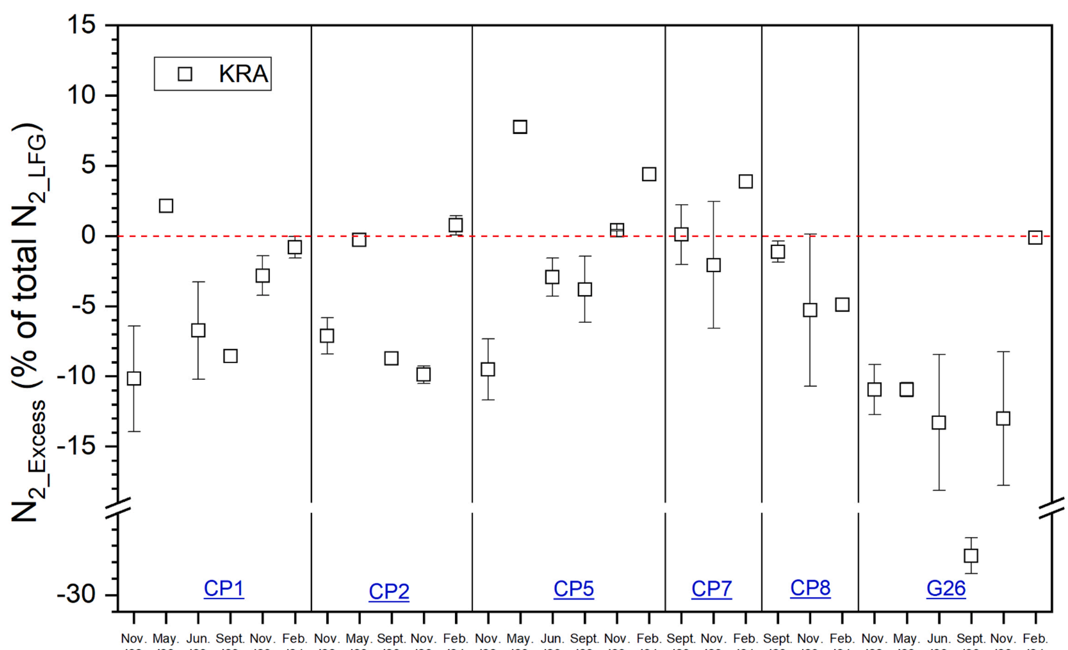


**Fig. 6.** Spatial and temporal differences of mean $N_{2\_Excess}$ (% of the total $N_{2\_LFG}$) in landfill gas (Eq. 3) from Kragge (KRA). Blue underlined text = sample ID (for sampling location see Table S2 in Supporting Information). Error bars = standard errors.

**Table 1**
Average annual balance between N release (denitrification, positive values) and N uptake (N fixation, negative values) at pilots Braambergen, Wieringermeer and de Kragge in the investigation period.

| Pilot | N release gas (t/a) | N release leachate (t/a) |
|---|---|---|
| Braambergen | 199 | 8 |
| Wieringermeer | 167 | 5 |
| De Kragge | −3.5 | 31.5 |

stabilization processes, by using the shift in the $N_2$/Ar ratio, thereby assisting with field-level nitrogen mass balancing. It was hypothesized that in-situ stabilization through aeration or liquid recirculation would promote conditions to modify nitrogen species and enhance $N_2$ formation. Aeration was expected to encourage denitrification due to the enhanced formation of $NO_3^-$ and $NO_2^-$. Recirculation of leachate from the nitritation unit of the wastewater treatment plant was expected to stimulate anammox under anaerobic conditions and produce $N_2$. Indeed, the shift in the $N_2$/Ar ratio of extracted landfill gas in relation to the atmospheric portion suggested a significant net $N_2$ release by denitrification for the aerated sites, whereas mostly net nitrogen uptake was detected for the anaerobic landfill undergoing leachate recirculation. Generally, the order of magnitude of the shares of $N_2$ generated by denitrification in the aerated landfills reported in this study (Fig. 3c and 3d) matches previous findings from a batch reactor study (Brandstätter et al., 2015), where the total nitrogen measured at the start of the study released 15.4 – 16.6 % $N_2$ from aerated-wet conditions, 2.6 – 8.8 % $N_2$ from aerated-dry process, and the anaerobic-wet conditions emitted 4 – 6.2 % $N_2$.

### 4.1. Landfills under aeration

The gas extracted from the three BRA compartments and from WIE exhibited elevated aeration efficiencies (Fig. 2b) and emitted excess $N_2$ in relation to the atmospheric concentration (Fig. 4a–d and Fig. 5a–c), suggesting that in combination with the desired aerobic decay of waste organic matter achieved net denitrification. The absolute amount of nitrogen leaving the waste body by net denitrification via the gas phase revealed that this pathway was more relevant to nitrogen discharge than the leachate pathway (Table 1). At both sites, $N_2O$ was detected, indicating partial heterotrophic denitrification (Berge et al., 2005; Harborth et al., 2013; Wrage et al., 2001; Zhang et al., 2009), also found by Brandstätter et al. (2015) for a bioreactor operated under aerated and dry conditions. These findings corroborate the presence of multiple parallel aerobic and anaerobic sites, which, regarding the desired high extent of aerobic carbon conversion processes, appears disadvantageous but is beneficial from the perspective of the demanding requirements on reduction of the nitrogen inventory through coupled nitrification–denitrification (Wrage et al., 2001). Net denitrification generated an $N_2$ excess of up to ~ 16 % of the total $N_2$ in BRA bulk gas and up to ~ 25 % of the total $N_2$ in WIE. However, there were samples with a lower $N_2$/Ar ratio in the gas compared to the atmospheric ratio at both sites,

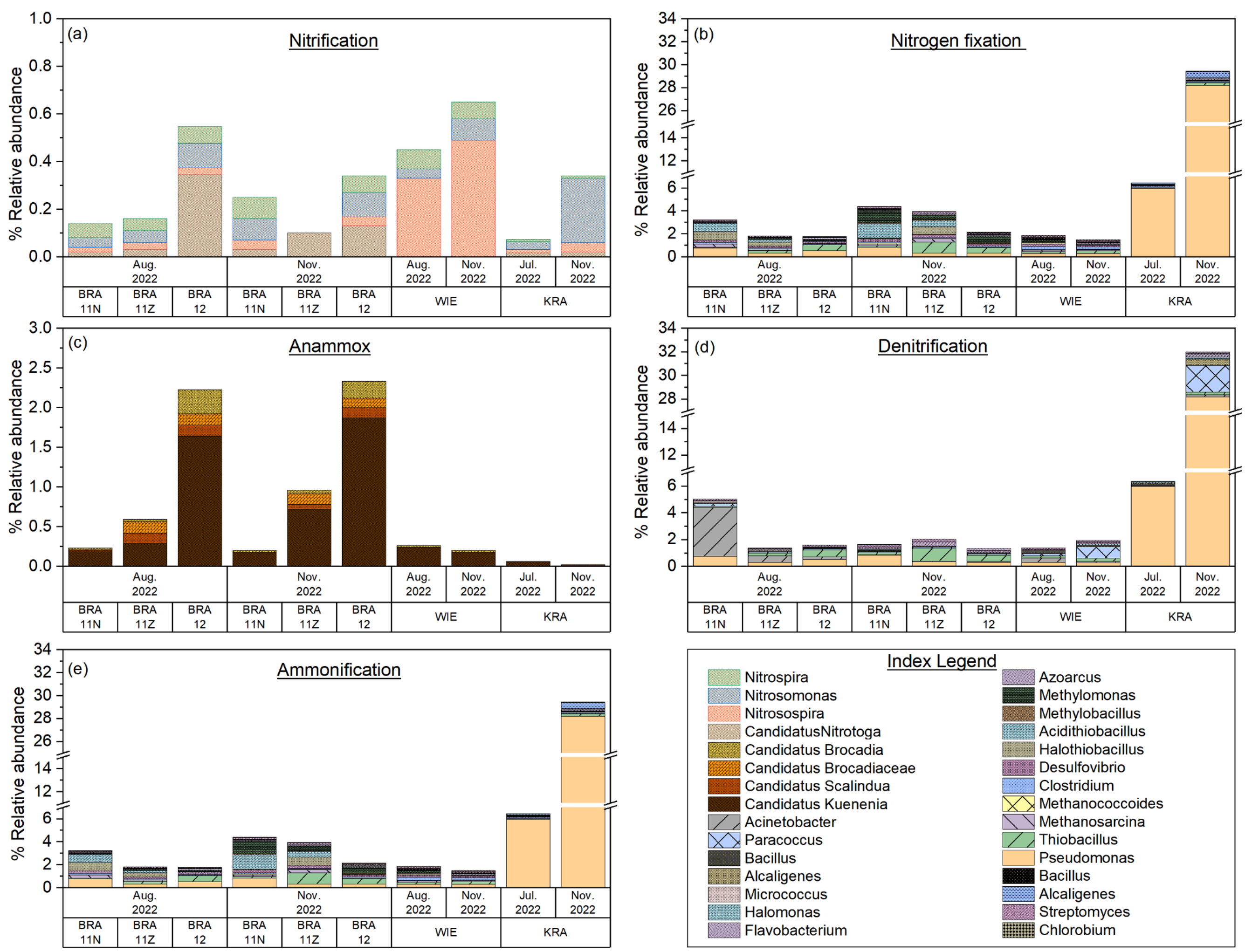


**Fig. 7.** Microbial species capable of: (a) nitrification; (b) nitrogen fixation; (c) anammox; (d) denitrification; and (e) ammonification detected in leachate samples from Braambergen (BRA) compartments 11Z, 11N, 12, Wieringermeer (WIE), and Kragge (KRA) landfills.

indicating an occurrence of net nitrogen fixation ($N_{2\_Excess} < 0$ %) around some gas wells or at some timeframe within the landfill. These samples mostly had high aeration efficiencies (Fig. 3d), around 45 to 50 %, suggesting that a higher aeration efficiency can conflict with conditions for denitrification, requiring the presence of anaerobic niches. At an aeration efficiency of 0 %, however, nitrogen fixation can occur, as shown in aeration well T3 from BRA compartment 12 that showed an extraordinarily high nitrogen fixation of ~ 18 % of the total $N_2$ (see Supporting Information Fig. S5).

Spatially varying gas composition indicated that the extent of aeration or the reactive processes affecting the balance of $CH_4$, $CO_2$, $O_2$, and $N_2$ varied within the waste body or both. In general, a higher aeration efficiency was observed in WIE than in BRA compartments (Fig. 2b). Spatial and temporal variability of intrinsic waste permeability governing fluid flow is due to heterogeneity (Xu et al., 2020), for example, from the level of compaction during waste placement, resulting in different permeability and water retention. Differences in waste permeability can also be the effect of previously enhanced biodegradation, varying in space and time relative to the differences in waste degradability and transport paths, leading to patterns in waste consolidation and subsequent changes in flow paths (Berge et al., 2005). In an earlier study in the BRA, it was shown that water levels indeed restricted gas flow in the wells (Meza et al., 2022), causing the aeration efficiencies to drop to ~ 0 %. Hence, a reduced extent of aeration or rapid $O_2$ consumption likely limited the nitrification-dependent denitrification in some parts of the waste body. However, this was mostly not the case, as shown in Fig. 3c and 3d. Low and high concentrations of $O_2$, $CO_2$, and $CH_4$, as well as low and high aeration efficiencies, coincided with both high and low values of excess $N_2$, respectively, for both BRA and WIE (Figs. S4, and S7 to S9 in Supporting Information). This is likely because the gas collected from any particular well reflects the mixture of all processes occurring within the sphere of influence and, hence, the multiplicity of concurrently existing aerobic and anaerobic pockets. Moreover, higher aeration efficiency does not necessarily coincide with higher flow. This means that the absolute amount of N intermediates attributed to aerobic decay of waste organic matter, such as $NO_2^-$ and $NO_3^-$, available for anammox or denitrification can be lower near a well of high aeration efficiency (but with lower flow). Similarly, high $O_2$ concentrations on a well could be indicative of bypass flow of atmospheric air or of flow along waste pathways that have already been stabilized and hence show minimal $O_2$ consumption, whereas low $O_2$ concentrations can indicate poor aeration flow or a high rate of $O_2$ consuming processes along the flow pathway. The concurrence of these processes in a heterogeneous waste body complicates the detection of clear correlations between individual parameters (Figs. S7 to S9).

Pronounced seasonal differences were observed at both aerated sites (Fig. 4a – 4d and 5a – 5c). It is assumed that the seasonal variability of cover soil moisture, resulting from seasonally changing balance between precipitation and evapotranspiration regulated air ingress, and subsequently, aeration efficiency (Gebert et al., 2023) and the nitrification-dependent nitrogen transformation. The net gaseous N balance and its variation over time were similar for all BRA compartments and WIE wells (Figs. 4 and 5), suggesting that the effect of aeration was more critical to the overall nitrogen balance than the differences in the waste inventory. This is likely the case since, on both sites, the strictly linear increase in cumulatively extracted carbon since 2017 for landfill WIE (Gebert et al., 2023) indicates that waste biodegradation is still limited by $O_2$ supply through the achieved air flow and not yet by possible differences in degradable organics.

The microbial community at landfill BRA related to N transformation pathways was characterized by the highest abundances of *Candidatus Nitrotoga* in the group of nitrifiers and *Candidatus Kuenenia* in the group of anammox bacteria. At WIE, the composition of the microbial community responsible for nitrogen transformation was different (Fig. 7a–d) with a notably higher abundance of *Nitrospira* species, absence of *Candidatus Nitrotoga*, lower relative abundance and species related to anammox, and different species associated with nitrogen fixation, ammonification, and denitrification. *Nitrospira* species achieve both ammonia and nitrite oxidation within a single cell (Daims et al., 2006; van Kessel et al., 2015) and have been considered key nitrite-oxidizing bacteria in municipal and industrial wastewater treatment plants (Daims et al., 2006; Spieck et al., 2021; Wu et al., 2019). The high share of net denitrification in both sites suggests that the activity of the denitrifying species fully consumed the products of nitrification, explaining the observed absence of $NO_2^-$ and $NO_3^-$ in the landfill leachate at both sites (Table S3 in Supporting Information), while the differences in composition of nitrogen transformation microbiota can likely be attributed to the differences in waste composition (see section 2.1).

Data on the microbial community composition are available from only two sampling dates (July/August and November 2022). While the data indicated that the abundance of taxa can vary over time, the number of sampling was too limited to derive any seasonal trend or relate the community composition to details of the landfill operation. The spatially complex patterns of gas and water flow give rise to the simultaneous existence of anaerobic and aerobic sites, as demonstrated by the variability in gas composition in this study, and other environmental conditions such as temperature, pH, and redox potential. In combination with the variable carbon sources available from the different waste types, these conditions influence the composition and activity of the microbial community and, hence, the nitrogen transformation pathways. The bulk biochemical signals detected in the landfill leachate combine all these effects. Thus, it is impossible to relate the abundance of individual microbial taxa to any single environmental condition present within the waste body.

### 4.2. Landfill under leachate recirculation (De Kragge)

In the KRA landfill, operating strictly under anaerobic conditions with high $CO_2$ and $CH_4$ concentrations (Fig. 1), the ratio of $N_2$ to Ar in the landfill sampled from November 2022 to February 2024 was mainly lower than the atmospheric ratio (Fig. 6), except for location CP1 and CP5 from May, also for CP2 and CP5 in February 2024. Nitrogen fixing processes consumed more nitrogen than was released by denitrification, resulting in a significantly different balance between N generation and N uptake (average net $N_{2\_Excess}$ = -4.2 $\%_{vol.}$ of total $N_{2\_LFG}$) than observed for the two aerobic landfills. Also, here, the temporal variability in the gas phase nitrogen balance was high. In contrast to the aerated sites, partial denitrification appears absent, as $N_2O$ was never detected.

Compared to the high extent of nitrogen discharge from denitrification on the aerated sites, net N uptake on the anaerobic landfill de Kragge was low (Table 1). Remarkably, the net nitrogen uptake occurred despite the markedly high average $NH_4^+$ concentrations of ~ 1161 mg/l in the leachate (Table S3 in Supporting Information). Compared to the aerated sites, the leachate from the KRA landfill was characterized by a low abundance of anammox-related species, and a very high abundance of bacteria related to nitrogen fixation, ammonification, and denitrification. The group of denitrifiers was markedly dominated by *Pseudomonas* species, suggesting that the primary driver of nitrogen fixation was *Pseudomonas* with the ability to also activate denitrification (Chen et al., 2013; Wei et al., 2021). *Pseudomonas* and the *Thiobacillus* belong to a group of diazotrophic microorganisms with a *nifH* gene that can fix atmospheric $N_2$ (Levy-Booth et al., 2014). Some *Pseudomonas* has the microbial functional genes of *napA*, *narG*, *norZ*, and *nirK* that assist in denitrification (Levy-Booth et al., 2014). Also, *Paracoccus* was the second largest genus of denitrifiers. *Paracoccus* and *Pseudomonas* are the most widely distributed bacteria in natural soil ecosystems and some species have denitrifying genes *narG*, *napA*, *nirS*, *norBC*, and *nosZ*, and have the ability to reduce nitrate to $N_2$ at different rates (Carlson and Ingraham, 1983).

Nitrogen can be fixed within the landfill cover soil, for example, by diazotrophic methanotrophs such as the *Methylocystis, Methylosinus, Methylocella* or *Methylomonas* genera (Cui et al., 2022; Ho and Bodelier, 2015), which were detected in all three landfills except for species of *Methylocella* (see Fig. S6 in Supporting Information). Aerobic $CH_4$ oxidation is likely a relevant process, as the landfill is not sealed and the gas extraction is not fully optimized for $CH_4$ recovery, allowing for $CH_4$-rich landfill gas to migrate through the cover soil. Interestingly, *Paracoccus denitrificans* was also detected in KRA leachate, an aerobic denitrifier that can reduce $N_2O$ to $N_2$ (Qu et al., 2016). Due to the predominantly anaerobic conditions within the waste body, this process is thought to be only possible in the landfill cover soil. Methanotrophic nitrogen fixation in the cover soil with ensuing ammonification and nitrification could enhance activity of *Paracoccus denitrificans* or leaching of ammonia and nitrate into the waste (Burton and Watson-Craik, 1998). In contrast to the aerated sites, the value of excess $N_2$ appeared to correlate with concentrations of $CO_2$ and $CH_4$ at KRA (Figs. S8 and S9), with more net N fixating conditions occurring with increased $CO_2$ and $CH_4$ concentrations. This suggests a causal relationship between $CH_4$ consuming and nitrogen fixing processes.

The growth of anammox bacteria, showing the lowest abundance at the KRA landfill, is slow, and competing with denitrifiers for nitrate and nitrite (van de Graaf et al., 1996). Their presence indicates that $N_2$ generation from anammox under anaerobic conditions likely adds to $N_2$ generated by denitrification (Burton and Watson-Craik, 1998), albeit possibly at a lower share. At the given aeration flows on landfills BRA and WIE, the coexistence of aerobic and anaerobic domains appeared to deliver more nitrate and nitrite to facilitate denitrification than anammox on anaerobic landfill KRA.

## 5. Conclusions

Denitrification and nitrogen fixation significantly affected the total landfill nitrogen balance in landfills treated by aeration and by leachate recirculation. The ratio of $N_2$ to Ar enabled the net balance between $N_2$ generating and $N_2$ consuming processes to be quantified, thereby serving as a tool for nitrogen mass balancing at full scale. However, the absolute magnitude of these processes could not be identified, and $N_2$ generation was not differentiated regarding anammox and coupled nitrification–denitrification. The results corroborated the assumption that landfill aeration resulted in $N_2$ removal via the gas phase, which was shown to represent the main pathway for nitrogen discharge, far exceeding the discharge via the leachate phase. This effect is due to anaerobic regions persisting or re-developing as evidenced by significant concentrations of methane in the gas and of ammonium in leachate. The condition of incomplete aeration, creating coupled aerobic-anaerobic domains within the waste body, can therefore be regarded favorable for maximizing nitrogen removal. Net nitrogen outflow through the gas phase was mostly absent for the site under leachate recirculation with a higher share of degradable organic matter, where nitrogen fixation dominated the nitrogen gaseous phase, thus adding nitrogen to the landfill. These findings suggest a delay in meeting environmental compliance criteria for leachate nitrogen as long as the degradable organic matter is present. Notably, net nitrogen fixing conditions can prevail despite high leachate nitrogen concentrations on both the sites under aeration and under leachate recirculation. Microbial taxa responsible for nitrogen transformation via nitrification, nitrogen fixation, anammox, and denitrification existed concurrently on all three sites, indicating the potential for all processes to be effective in the landfills and for organic matter degradation and nitrogen transformation to coexist under both aerobic and anaerobic conditions. It would be of interest to further explore computational models of electron transfer processes in the landfills to determine and characterize the biochemical reactions that predict the $N_2$ concentrations and the effects of liquid recirculation and aeration on nitrogen dynamics in the landfills.

## CRediT authorship contribution statement

**Susan Yi:** Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Data curation, Conceptualization. **Nathali Meza:** Methodology. **Julia Gebert:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.wasman.2024.12.042.

## Data availability

Data will be made available on request.

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