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Theoretical Analysis of Municipal Solid Waste Treatment by Leachate Recirculation under anaerobic and aerobic Conditions

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Abstract

Long-term emissions of Municipal Solid Waste (MSW) landfills are a burden for future generations because of the required long-term aftercare. To shorten aftercare, treatment methods have to be developed that reduce long-term emissions. A treatment method that reduces emissions at a lysimeter scale is re-circulation of leachate. However, its effectiveness at the field scale still needs to be demonstrated. Field scale design can be improved by theoretical understanding of the processes that control the effectiveness of leachate recirculation treatment. In this study, the simplest and most fundamental sets of processes are distilled that describe the emission data measured during aerobic and anaerobic leachate recirculation in lysimeters. A toolbox is used to select essential processes with objective performance criteria produced by Bayesian statistical analysis. The controlling processes indicate that treatment efficiency is mostly affected by how homogeneously important reactants are spread through the MSW during treatment. A more homogeneous spread of i.e. oxygen or methanogens increases the total amount of carbon degraded. Biodegradable carbon removal is highest under aerobic conditions, however, the hydrolysis rate constant is lower which indicates that hydrolysis is not enhanced intrinsically in aerobic conditions. Controlling processes also indicate that nitrogen removal via sequential nitrification and denitrification is plausible under aerobic conditions as long as sufficient biodegradable carbon is present in the MSW. Major removal pathways for carbon and nitrogen are indicated which are important for monitoring treatment effectiveness at a field scale. Optimization strategies for field scale application of treatments are discussed.

Keywords: Municipal Solid Waste treatment, leachate recirculation, aeration, fundamental biogeochemical model, optimisation

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1. Introduction

A major challenge for this human generation is to develop treatment methods that reduce long-term emissions from Municipal Solid Waste (MSW) landfills. Because landfills can emit gas and leachate for hundreds of years (Belevi and Baccini, 1989), they pose a risk for human health and the environment. Long-term aftercare is required which puts a burden on future generations.

Reduction of long-term emissions can be achieved by accelerating degradation within a waste-body (Scharff et al., 2011) which is normally slow due to inhibitions and transport limitations (Kjeldsen et al., 2002, Laner et al., 2011, Meima et al., 2008). Enhanced degradation leads to accelerated release of carbon and nitrogen containing compounds. In the case of methane, accelerated release is also economically attractive because this gas can be utilized as an energy source.

Treating MSW by recirculating leachate under anaerobic or aerobic conditions has shown to accelerate emissions in experiments at a lysimeter scale (Veeken et al., 2000, Kasam et al., 2013, Erses et al., 2008, Brandstätter et al., 2015a,b, Bilgili et al., 2007). By inducing (increased) water flow, leachate recirculation stimulates mixing of solutes and bacteria within the waste body which removes inhibitions and transport limitations for degradation (White et al., 2011). Dissolved compounds in the leachate are furthermore removed by bleeding of the leachate stream. Normally, the enhanced consumption of readily available electron acceptors leads to strictly anaerobic conditions.

In order to generate (partly) aerobic conditions, leachate recirculation can be combined with aeration (Ritzkowski and Stegmann, 2012). This has two main advantages: aerobic degradation is generally faster than anaerobic degradation (Heijnen and Kleerebezem, 1999) and ammonium can be removed by oxidation to nitrogen gas via subsequently nitrification and denitrification (Bolyard and Reinhart, 2016).

Although effective in enhancing biodegradation, full-scale application of leachate recirculation or aeration has not yet been proven to reduce long-term leached emissions (Benson et al., 2007, Hrad et al., 2013). Apparently, the understanding of processes that control the effectiveness of these treatments is insufficient and needs to be improved. We believe that a good point to start is to revisit data obtained in lysimeter experiments and to distill the simplest, most fundamental biogeochemical process networks that explain the measured emissions.

A fundamental model that is as 'simple' as possible highlights the controlling processes, reactions and factors that drive (measured) emissions. It allows to identify major and minor degradation and transport pathways and rate-limiting factors. Moreover, such a model provides insight in any specific mass balance which is not directly measured in the context of the processes, inhibitions and limitations. The challenge, however, is to identify which combination of fundamental processes are least ambiguous (or subjective) and contain minimal uncertainty. We use the toolbox developed by Van Turnhout et al. (2016) to find the most simplistic and best describing fundamental model; it allows to integrate several environmental frameworks, quickly build different models and compare performances objectively with a set of qualitative and quantitative criteria.

This paper presents the least ambiguous biogeochemical process networks that are responsible for the emissions measured in lysimeter experiments performed by Brandstätter et al. (Brandstätter et al., 2015a,b). With these networks, the processes and factors controlling the effectiveness of the applied anaerobic and aerobic leachate recirculation treatment are discussed. Major and minor removal pathways for carbon and nitrogen compounds are presented. Implications of the findings for full-scale treatment design are given together with suggestions for improvement.

2. Material & Methods

2.1. Characteristics of the lysimeter experiments

For this study we used the data measured in two lysimeter experiments of Brandstätter et al. (2015a,b). In both experiments leachate was recirculated and the temperatures in the lysimeters were kept constant ($\sim$ 309K). To allow for drainage of leachate by gravity, a fine meshed grid of 8cm was placed at the bottom of the reactors. Leachate removed during sampling was replaced with distilled water (Aqua dest) to maintain the degree of water saturation.

In one experiment the conditions were anaerobic and in the other (partly) aerobic due to continuous injection of air. The experiments were carried out in duplicate on MSW which was mined from a landfill where operations stopped 40 years ago. The MSW was sieved to a grain size of < 20 mm. In the reactors, the MSW had initially a water content of 23% and a dry bulk density of 846 $\frac{\text{kg}}{\text{m}^3}$. An illustration of both set-ups is presented in figure 1 and the initial and environmental conditions are listed in table 1.

From each experiment, the following time series are used which were measured over 2.25 years: cumulative produced biogas (CO_2 and CH_4), partial pressure of CO_2, CH_4 and O_2, pH, Biological Oxygen Demand (BOD), ${NH_4}^+$ and Cl^- concentrations. One time series of each experiment was used for calibration and the other for validation. The calibration dataset was used to find the least ambiguous process network with the toolbox developed by van Turnhout et al. (2016). Subsequently, the network was tested on the validation dataset without further parameter fitting.

Although the MSW was mined from a 40 year old landfill, it contained a significant percentage of readily degradable organic content such as cellulose (Brandstätter et al., 2015a). This can be explained by the heterogeneity of landfill bodies which can cause many types of local limitations and inhibitions on the degradation of its organic content.

In order to arrive at a generic modeling framework, average compositions of the biodegradable fraction of Solid Organic Matter (SOM) of $C_6H_{10}O_5N_{0.036}$ and $C_6H_{10}O_5N_{0.032}S_{0.03}$ are used for respectively the anaerobic experiment and the aerobic experiment following generic principles of anaerobic digestion modeling ((Batstone et al., 2002, Reichel et al., 2007)). The fractions of nitrogen and sulfur are deduced from the fractions measured in the MSW at the beginning and the end of the experiment. Because sulfur was not significantly detected under anaerobic conditions, it was not considered in the model. Most likely, the released sulfur under anaerobic conditions was immediately converted into H_2S. For generic purposes, BOD is assumed to consist of Acetic Acid which accumulates strongest when methanogenesis is rate-limiting. The elemental composition of bacteria of $CH_{1.4}O_{0.4}N_{0.2}$ is taken from Henze et al. (1995).

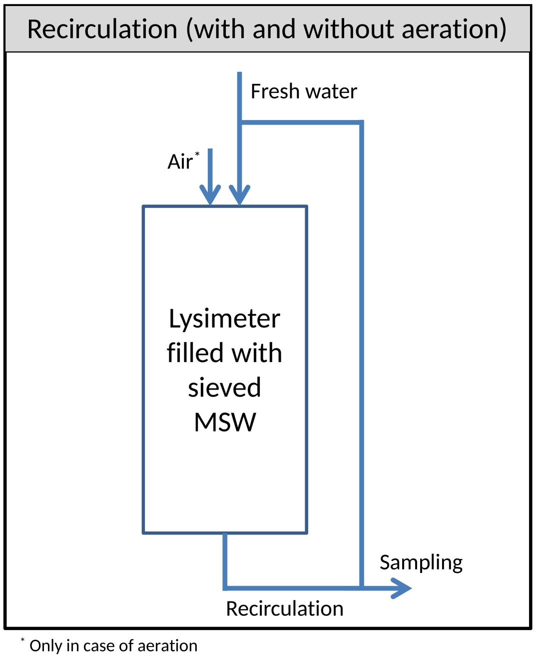


Figure 1: Illustration of the experimental set-ups for leachate recirculation under anaerobic and aerobic conditions (Brandstätter et al., 2015a,b). The lysimeters were made of stainless steel and had a cylindrical shape with a volume of 121 L.

Table 1: Initial and environmental conditions in the duplicate experiments.

	Initial conditions						Environmental conditions					
	p_{CO_2}	p_{O_2}/p_{CH_4}	NH_3	pH	Cl^-	V_{liquid}	$\phi^{in1)}_{waterflow}$	$\phi^{out1)}_{sampleflow}$	$\phi^{in}_{airflow}$	V_{gas}	p^{tot}_{gas}	T
Type of experiment	[atm]	[atm]	[M]	[-]	[M]	[L]	[L d^{-1}]	[L d^{-1}]	[L d^{-1}]	[L]	[atm]	[K]
Anaerobic (calibration)	0.10	0	0.03	8	0.025	19.5	0.03	0.03	-	41.9	1	308.5
Anaerobic (validation)	0.10	0	0.03	8	0.025	19.5	0.03	0.03	-	41.9	1	308.5
Aerated (calibration)	0.41	0	0.025	7.25	0.025	21.24	0.0524	0.0483	56.67	38.8	1	308.5
Aerated (validation)	0.41	0	0.025	7.25	0.025	20.95	0.0526	0.0491	56.67	38.82	1	308.5

1) The flows of water are averaged over the entire measurement period. After the first week, the MSW reached field capacity and flows were relatively constant.

2.2. *Selecting the least ambiguous biogeochemical process network to describe measured data*

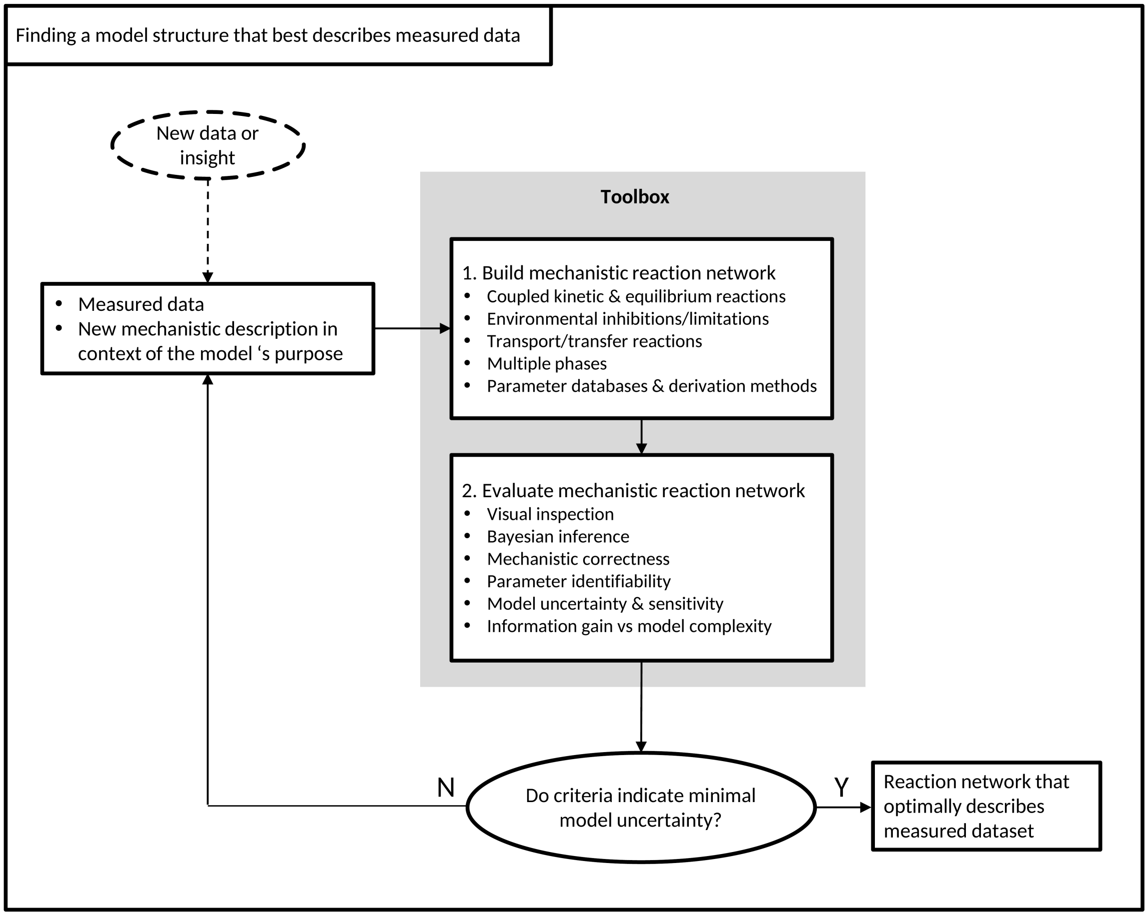


Figure 2: Iterative procedure for finding the biogeochemical process network that describes measured data with minimal ambiguity and uncertainty (van Turnhout et al., 2016). In step 1, the user manually builds the best network with the toolbox which is automatically optimized for the measured data in step 2. The user then decides if the network describes the measurements sufficiently accurate with the evaluation criteria generated in step 2. If not, the user starts another iteration and adapts the network.

Figure 2 shows the approach to select the least ambiguous biogeochemical process network describing the measured data (van Turnhout et al., 2016). First, a model structure is build to be evaluated. Relevant kinetic, equilibrium, transfer and transport reactions within a multiphase environment are selected together with appropriate environmental inhibitions and limitations. Fundamental parameters can be obtained from an extensive geochemical database and a method to derive biochemical parameters from thermodynamic principles (Kleerebezem and van Loosdrecht, 2010). Then, the probability distribution functions (pdf) of the most uncertain parameters are obtained by fitting modeled data to the measured data. Pdfs are obtained with a Bayesian statistical optimization method and allow to evaluate the performance of the network with an extensive set of qualitative and quantitative criteria. Finally, model performance is assessed with these criteria which reveals if model uncertainty and ambiguity is small enough to be acceptable. If this is not satisfactory, a next iteration can be started with an alternative biogeochemical process network selected by the user.

In this study, three criteria were used to evaluate model performance. The first is the visual fit between modeled data and measured data. The second is the practical identifiability of a parameter which is indicated by the $5\% - 95\%$ quantile range of its pdf. A smaller range means a better identifiability. The third is the agreement between the $5\% - 95\%$ quantile range of a parameter and its intrinsic value when measured under or estimated for non-limiting environmental conditions e.g. in perfectly mixed batch reactors. The closer (all) calibrated parameter ranges agree with intrinsic reference values, the stronger the evidence is that all controlling fundamental processes and limitations are included in the network. The model that performs best for all three criteria has minimal ambiguity and uncertainty.

3. Results & Discussion

3.1. A fundamental biogeochemical reaction network for leachate recirculation under anaerobic conditions

Figure 4 presents the least ambiguous biogeochemical process network that describes emissions during anaerobic leachate recirculation treatment. The network contains five kinetic reactions: 1) lumped hydrolysis and acidogenesis with hydrolysis as rate limiting step, 2) methanogenesis, 3-4) bacterial biomass decay with a maximum decay rate of 5% of the maximum growth rate (Angelidaki et al., 1999) and 5) lumped nitrification and denitrification with nitrification as rate limiting step. Although pure anaerobic conditions are compromised by including nitrification, it allowed to explain observations in the measured data. Kinetic mechanisms and stoichiometry are listed in table 2. Also a set of equilibrium reactions (i.e. speciation, complexation, precipitation and gas-liquid transfer reactions) is included in the network. Corresponding species are presented in the bottom part of figure 4.

Figure 3 shows the visual agreement between the best fit of the modeled data and the measured data. The uncertainty in the total model error is also indicated. Overall, the calibration and the validation data sets are well described. A good description of the pH was only possible by including the set of chemical/physical equilibria. The high alkalinity of leachate could be modeled with a sufficient amount of readily available Calcite.

Only the dynamics in ammonium concentrations are not accurately described by the network which indicates that fundamental processes are missing in the network. We could account for the total amount of removed nitrogen in MSW by including a lumped nitrification/denitrification reaction together with an intrusion flow of air. Evidence for air intrusion is given by the decrease in partial pressures of methane and carbon dioxide. However it must be noted that at field scale conditions, intrusion of air is most likely irrelevant. The measured data did not contain sufficient information to further fit the ammonium concentrations by including additional fundamental processes. Important processes could have been adsorption of ${NH_4}^+$ to the MSW as suggested by Bolyard and Reinhart (2016) and autotrophic denitrification with reduced sulfur compounds as suggested by Onay and Pohland (1998). Follow-up experiments should include measurements to calibrate these processes in order to identify their relevance.

In order to evaluate the ambiguity and uncertainty of the network, we calibrated the parameters with the highest uncertainty. Table 3 presents the $5\% - 95\%$ quantiles of the posterior pdfs, the prior ranges and the reference values of the optimized parameters for non-limiting environmental conditions. The small range of all posterior quantiles indicate that all parameters are identifiable from the information present in the measured data set. Even more interesting is that, although the process network is relatively simple, most calibrated bandwidths fall in the range of the reference values. We believe this indicates that the process network includes most fundamental reactions and processes controlling the emissions. Calibrated bandwidths of maximum rates are a bit lower than the reference values presumably due to mass transport limitations in the experiments which have not been included in the model.

The process network closely resembles the network identified for a similar type of lysimeter experiment by van Turnhout et al. (2016). This suggests that the selected processes describe biodegradation of MSW on a general level for lysimeter scale. The main difference between the networks is that we did not include environmental inhibition relations because concentrations of potential inhibitors remained low.

The calibration of the network allows to estimate the amount of organic carbon in the MSW that could be removed under the experimental conditions. The mean fitted value for the initial amount of biodegradable carbon ($C_{\mathrm{SOM}}^{\mathrm{ini}}$) of 2.3 $\frac{\mathrm{Cmol}}{\mathrm{L}}$ or 0.018 $\frac{\mathrm{kgSOM}}{\mathrm{kgdrywaste}}$ is comparable with the total removed carbon measured by MSW sampling. This indicates that all significant emission pathways are considered in the network. For further study, it would be even more interesting to be able to predict how much of SOM in the MSW can be removed under different experimental conditions. This, however, requires to extend the network with processes that describe the potential of different fractions of SOM present in the MSW to be hydrolyzed.

Because the model describes the emission controlling processes, it also indicates which phases are dominant in removing carbon and nitrogen from MSW. The mass percentages of organic carbon and nitrogen in the phases MSW, solutes, bacteria, sampled leachate, $\mathrm{CO_2}$(gas), $\mathrm{CH_4}$(gas), $\mathrm{N_2}$(gas) and $\mathrm{NH_3}$(gas) over time are presented in figure 5. It shows that most of the organic carbon and nitrogen ($85\% - 83\%$) in the MSW is unaffected by the treatment. Considering the controlling processes, the main limitation seems to have been the transport of reactants e.g. oxygen or methanogenic bacteria to the biodegradable fraction of MSW. The optimal network and parameter values do not indicate any other severe biochemical rate limitations or inhibitions. In addition, part of the remaining carbon could have been non-biodegradable. Figure 5 also shows that carbon is mainly removed via the gas phase and nitrogen is mainly removed via sampled leachate. Phases such as bacterial biomass and stripped ammonia contribute minimally as a final sink.

In order to successfully monitor the efficiency of the treatment methods at full scale, it is crucial to measure the dominant final sinks of carbon and nitrogen. Inconclusive field scale results so far, may (partially) be caused by incomplete monitoring. To optimize field scale application, important reactants such as oxygen and methanogens should be distributed more homogeneously throughout the waste-body. This could be achieved for instance by placing aeration wells or infiltration trenches closer together. Important topics for further research are therefore optimization of reactant distribution and monitoring of final sinks.

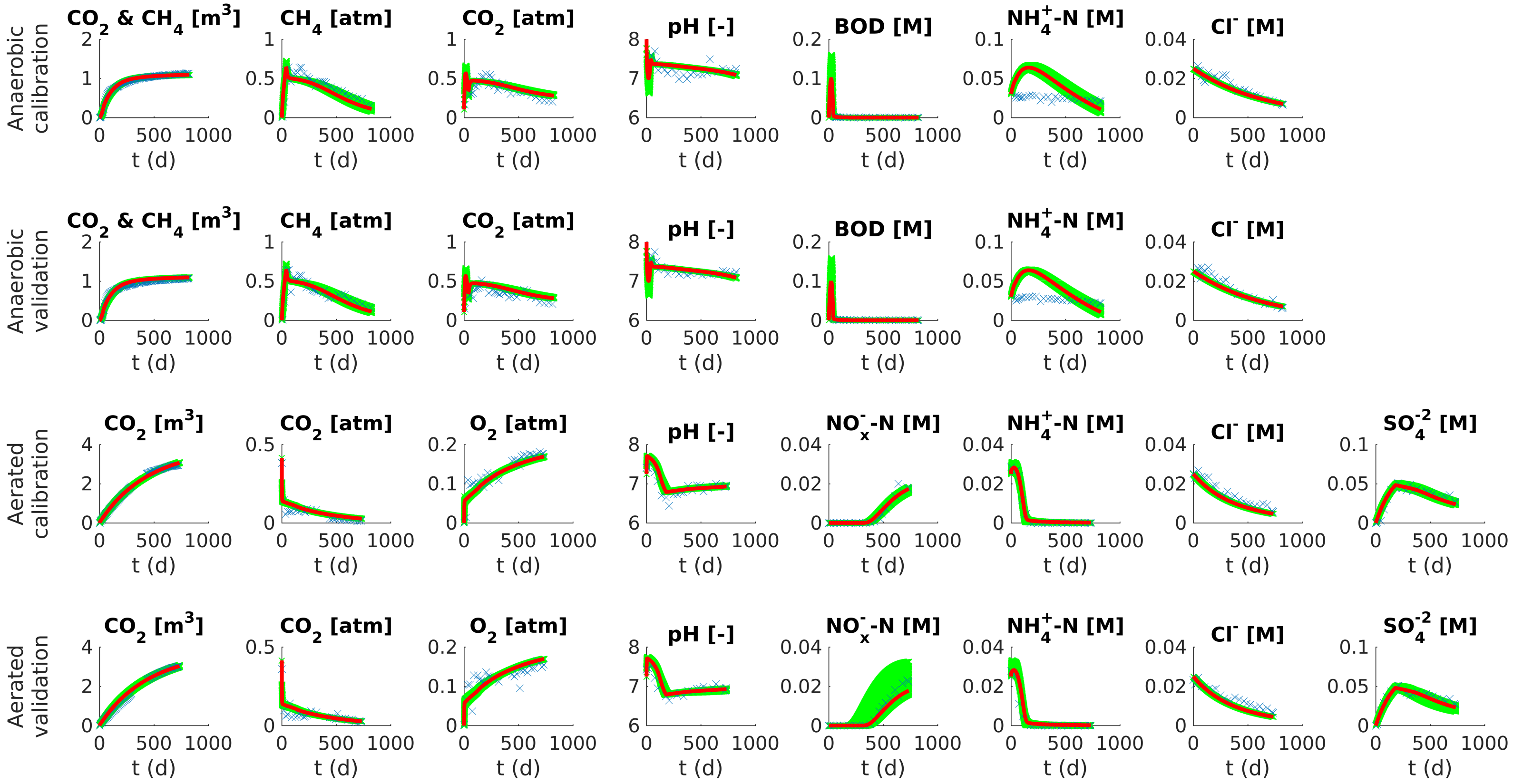
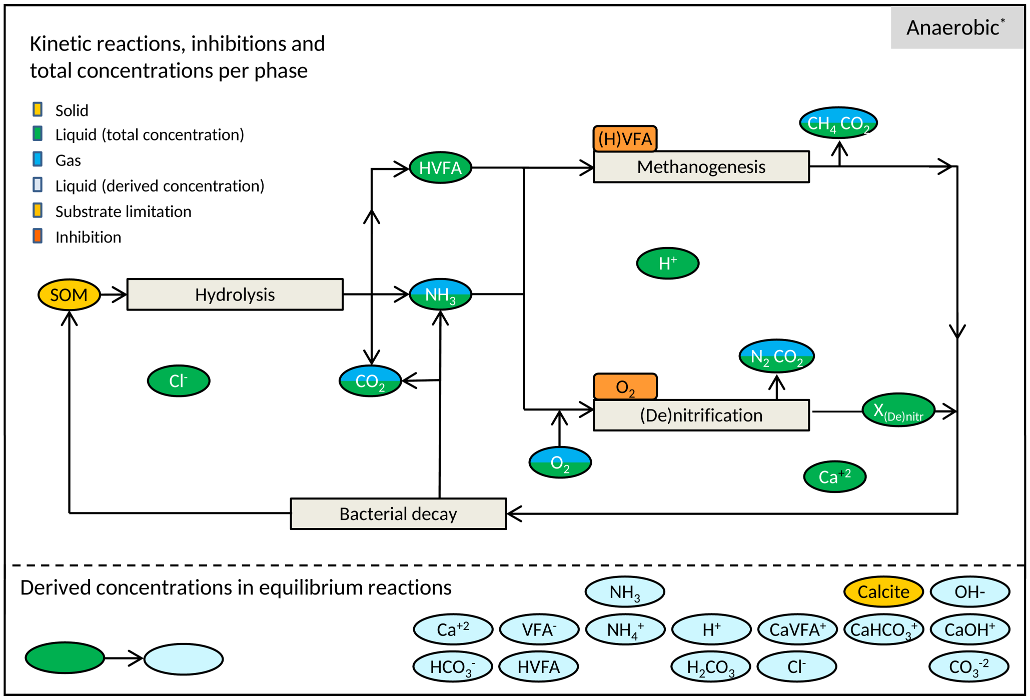
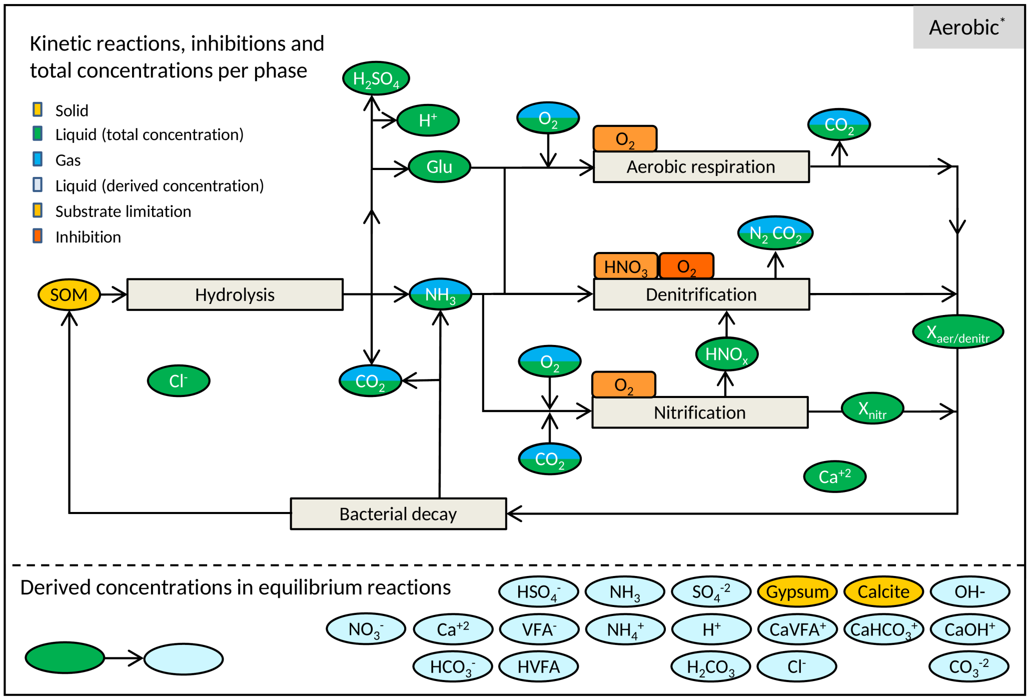


Figure 3: The visual agreement between the best fit of the modeled data (in red) and the measured data (in blue) for calibration and validation data measured during leachate recirculation under anaerobic and aerated conditions. The uncertainty in the total model error is indicated with green bandwidths surrounding the modeled data.



*with some oxygen present due to leakage



*with remaining anaerobic pockets because air is transported through preferential flow paths

Figure 4: The least ambiguous biogeochemical process networks for anaerobic (top) and aerobic (bottom) recirculation treatment on a lysimeter scale. In these schemes, gas and leachate flows that go into and out of the reactors are not included, but their values are listed in tables 1 and 3.

Table 2: Stoichiometry for the kinetic reactions in the biogeochemical process networks derived with the method from (Kleerebezem and van Loosdrecht, 2010) or deduced from the composition of MSW measured during sampling. Also the kinetic mechanisms are indicated. Parameters in bold were selected for calibration because they are highly uncertain and can significantly influence the model outcome.

Stoichiometry for the recirculation experiment under anaerobic conditions [mol]												Kinetics$^{c)}$
	SOM$^{a)}$	Acetic Acid	CO_2	NH_3	CH_4	H_2O	H^+	$X^{b)}_{\text{meth}}$	$X_{\text{(de)nitr}}$	O_2	N_2	
Hydrolysis$^{e)}$	-1	0.5	0	0.036	0	-0.172	0	0	0	0	0	$\mathbf{k}_{\text{hyd}} \cdot \mathbf{C}^{\text{T}}_{\text{SOM}}$
Methanogenesis	0	-12.91	12.41	-0.2	12.41	0.6	0	1	0	0	0	$\boldsymbol{\mu}^{\max}_{\text{meth}} \cdot \mathbf{C}_{\text{X}} \cdot \mathbf{f}^{\text{SL}}_{C^{\text{T}}_{\text{Ac}}} \cdot f^{\text{SL}}_{C^{\text{T}}_{\text{NH}_3}}$
Bacterial decay $^{d)}$	0	0.5	0	0.2	0	-0.6	0	-1	0	0	0	$0.05 \cdot \mu^{\max}_{\text{meth}} \cdot C_{\text{X}}$
(De)nitrification	0	-0.12	0.39	-0.54	0	1.5	0	0	1	-0.63	0.17	$\boldsymbol{\mu}^{\max}_{\text{(de)nitr}} \cdot \mathbf{C}_{\text{X}} \cdot f^{\text{SL}}_{C^{\text{T}}_{\text{O}_2}} \cdot f^{\text{SL}}_{C^{\text{T}}_{\text{NH}_3}}$

Stoichiometry for the recirculation experiment under aerobic conditions [mol]													Kinetics
	SOM	Glucose	CO_2	NH_3	H_2O	H^+	$X_{\text{aer-denitr}}$	O_2	SO_4^{-2}	NO_3^-	X_{nitr}	N_2	
Hydrolysis	-1	0.167	0	0.032	-0.172	$\boldsymbol{\nu}_{\text{H}^+}$	0	0	0.03	0	0	0	$\text{k}_{\textbf{hyd}} \cdot \mathbf{C}^{\text{T}}_{\text{SOM}}$
Aerobic respiration	0	-0.23	0.39	-0.2	0.99	0	1	-0.39	0	0	0	0	$\boldsymbol{\mu}^{\max}_{\text{aer}} \cdot \mathbf{C}_{\text{X}} \cdot f^{\text{SL}}_{C^{\text{T}}_{\text{Glu}}} \cdot f^{\text{SL}}_{C^{\text{T}}_{\text{O}_2}} \cdot f^{\text{SL}}_{C^{\text{T}}_{\text{NH}_3}}$
Nitrification	0	0	-1	-11.42	10.82	11.22	0	-21.44	0	11.22	1	0	$\boldsymbol{\mu}^{\max}_{\text{nitr}} \cdot \mathbf{C}_{\text{X}} \cdot \mathbf{f}^{\text{SL}}_{C^{\text{T}}_{\text{NH}_3}} \cdot f^{\text{SL}}_{C^{\text{T}}_{\text{CO}_2}}$
Denitrification	0	-0.24	0.42	-0.2	2.62	-0.33	1	0	0	-0.33	0	0.17	$\boldsymbol{\mu}^{\max}_{\text{denitr}} \cdot \mathbf{C}_{\text{X}} \cdot f^{\text{SL}}_{C^{\text{T}}_{\text{Glu}}} \cdot f^{\text{SL}}_{C^{\text{T}}_{\text{NO}_3^-}} \cdot f^{\text{SL}}_{C^{\text{T}}_{\text{NH}_3}} \cdot \mathbf{f}^{\text{NC}}_{C^{\text{T}}_{\text{O}_2}}$
Bacterial decay $^{d)}$	0	0.17	0	0.2	-0.6	0	(-1)	0	0	0	(-1)	0	$0.05 \cdot \mu^{\max} \cdot C_{\text{X}}$

a) SOM is the biodegradable fraction of Solid Organic Matter in MSW generically modeled as $C_6H_{10}O_5N_{0.036}$ and $C_6H_{10}O_5N_{0.032}S_{0.03}$ for respectively anaerobic and aerobic conditions. b) X are species of bacteria with the elemental composition $CH_{1.4}O_{0.4}N_{0.2}$ taken from Henze et al. (1995). c) Substrate limitation factors (f^{SL}) and inhibition factors (f^{NC}) range between 0 and 1 (van Turnhout et al., 2016). Half saturation constants which are not calibrated have low values and are primarily included as switch factors. d) Bacterial biomass decay is included as a separate process for each species. e) Hydrolysis under anaerobic conditions is lumped with acidogenesis with hydrolysis being rate limiting.

Table 3: Results of the optimization: $5\% - 95\%$ quantiles of the posterior ranges. They are listed together with the prior ranges and the reference ranges measured under or estimated for non-limiting environmental conditions.

Anaerobic recirculation experiment

	prior	posterior	reference		prior	posterior	reference
k_{hyd} [d^{-1}]	0.0005 – 0.15	0.0094 – 0.0104	0.09 – 0.26$^{1)}$	X_{meth} [mM]	0.005 – 15	0.59 – 11	0.27 – 19$^{3)}$
$\mu^{\mathrm{max}}_{\mathrm{meth}}$ [d^{-1}]	0.01 – 3	0.04 – 0.12	0.8$^{2)}$	$X_{\mathrm{(de)nitr}}$ [mM]	0.015 – 15	1.2 – 14	0.27 – 19$^{3)}$
$\mu^{\mathrm{max}}_{\mathrm{(de)nitr}}$ [d^{-1}]	0.001 – 1	0.11 – 0.96	1.4$^{2)}$	$K^{\mathrm{meth}}_{\mathrm{s},C^{\mathrm{T}}_{\mathrm{Ac}}}$ [mM]	1 – 1000	5.5 – 16.7	0.03 – 420$^{4)}$
$\phi_{\mathrm{in,air}}$ [Ld^{-1}]	0.005 – 1.5	0.12 – 0.19	–	$C^{\mathrm{ini}}_{\mathrm{SOM}}$ [M]	2 – 3	2.27 – 2.33	–

Aerated recirculation experiment

	prior	posterior	reference		prior	posterior	reference
k_{hyd} [d^{-1}]	0.00024 – 0.026	0.0021 – 0.0023	0.09 – 0.26$^{1)}$	X_{nitr} [mM]	0.09 – 110	1.1 – 2.7	0.27 – 19$^{3)}$
$\mu^{\mathrm{max}}_{\mathrm{aer}}$ [d^{-1}]	1.2 – 121	25.6 – 118	57$^{2)}$	$X_{\mathrm{aer-denitr}}$ [mM]	7 – 800	9.5 – 35.4	0.27 – 19$^{3)}$
$\mu^{\mathrm{max}}_{\mathrm{nitr}}$ [d^{-1}]	0.003 – 0.31	0.01 – 0.02	1.4$^{2)}$	$K^{\mathrm{nitr}}_{\mathrm{s},C^{\mathrm{T}}_{\mathrm{NH_3}}}$ [mM]	0.9 – 110	1.6 – 7.1	0.04$^{5)}$
$\mu^{\mathrm{max}}_{\mathrm{denitr}}$ [d^{-1}]	2 – 201	6.4 – 198	54$^{2)}$	$K^{\mathrm{denitr}}_{\mathrm{i},C^{\mathrm{T}}_{\mathrm{O_2}}}$ [mM]	0.005 – 0.6	0.047 – 0.093	–
$C^{\mathrm{ini}}_{\mathrm{SOM}}$ [M]	5 – 7	6.2 – 6.5	–	$\nu^{\mathrm{hyd}}_{\mathrm{H^+}}$ [mol]	0 – 0.06	0.037 – 0.041	–

1) Veeken and Hamelers (1999) 2) Kleerebezem and van Loosdrecht (2010) 3) Nopharatana et al. (2007) 4) Meima et al. (2008) 5) Kantartzi et al. (2006)

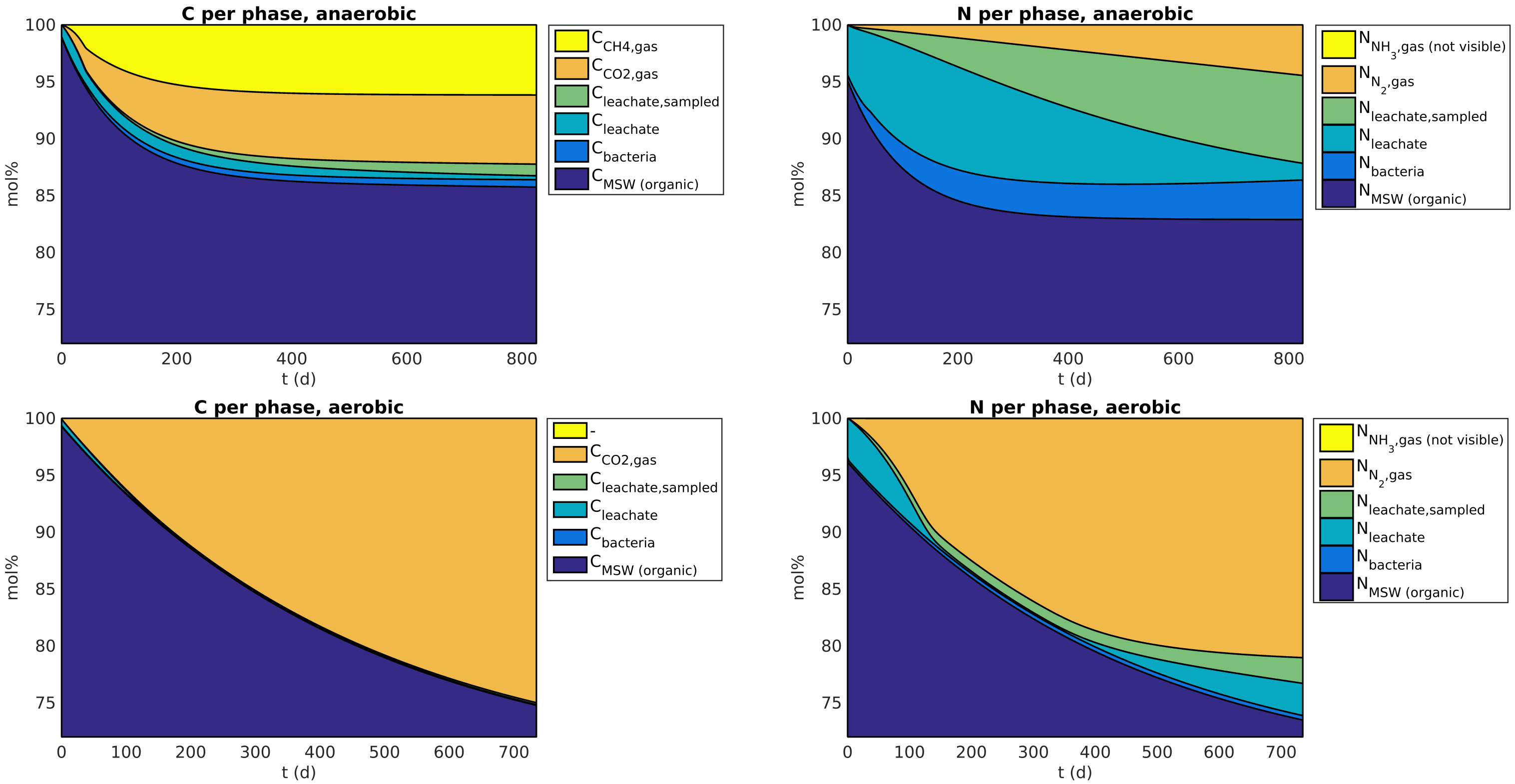


Figure 5: Modeled mass percentages of organic carbon and nitrogen in different phases during anaerobic and aerobic leachate recirculation. They indicate which phases are dominant in removing carbon and nitrogen from MSW for each treatment method.

3.2. A fundamental biogeochemical reaction network for leachate recirculation under aerobic conditions

Figure 4 also presents the least ambiguous process network that describes emissions during aerobic leachate recirculation treatment. This network contains six kinetic reactions: 1) hydrolysis of SOM, 2) aerobic respiration of glucose, 3) nitrification, 4) denitrification and 5-6) bacterial biomass decay. The set of equilibrium reactions is identical to that used for modeling the anaerobic experiment except that sulphate speciation and gypsum precipitation are included as well. Table 2 lists the stoichiometry and kinetic mechanisms.

The high sulfur/carbon ratio in SOM (table 2) suggests that gypsum is co-dissolved during hydrolysis because only protein hydrolysis would give a much lower ratio. The amount of protons released per sulfate (ν_{H^+}) is calibrated on the measured data and its mean calibrated value (table 3) indicates mainly release of $\mathrm{HSO_4}^-$. A more detailed process description of the release and conversion of sulfur was not possible because the measured data was too limited. When future studies do include additional measurements, it would be very interesting to extend the process description.

The best visual fit between modeled and measured data for aerobic treatment is presented in figure 3. Again all model results are in good agreement with the measured data for both the calibration dataset and the validation dataset. Dynamics in measured pH and dissolved sulphate could be reproduced because of inclusion of the set of chemical equilibrium reactions.

The optimization results (table 3) indicate that also this network contains minimal ambiguity and uncertainty. The quantile ranges of the posterior pdfs show that the identifiability of most calibrated parameters is good. Only the maximum rates of aerobic respiration and denitrification have a wide range with high values. This is, however, reasonable because these reactions are limited by the rate of oxygen or nitrate supply. Therefore, any high value for these maximum rates gives a satisfying model result. Most quantile ranges fall close to the reference values, but similar to the findings for the anaerobic case, values are slightly lower due to missing mass transport limitations in the model. The higher values of the initial concentration of aerobic respirators seem to compensate for a high initial growth rate which was not recorded because of too large measurement intervals.

The consumption of ammonium (figure 3) via sequential nitrification and denitrification suggests that anaerobic pockets were present in the MSW during air injection. This implies that air is transported through preferential flow paths which is reasonable given the strong heterogeneity of the MSW. Figure 6 schematically explains how preferential flow of air leads to simultaneous nitrification and denitrification. With sufficient biodegradable carbon available, the oxygen is readily consumed near the preferential channels which leaves the bulk of the waste anaerobic. Nitrate that is produced in the aerobic region by nitrification diffuses into the anaerobic bulk where it is utilized by denitrification. Interestingly, this mechanism only holds when sufficient biodegradable carbon is available. When carbon depletes, the oxygen penetrates deeper into the bulk of the waste violating the anaerobic conditions. This eventually leads to oxygen overtaking nitrate as oxidizer of the remaining carbon. This effect is noticeable in the measured data (figure 3) by the increase in nitrate/nitrite concentrations at the end of the experiment.

The inhibition of denitrification by oxygen is included in the process network with the factor:

$$f^{\mathrm{NC}}_{C^{\mathrm{T}}_{\mathrm{O}_2}} = \left(\frac{K_{\mathrm{inh}}}{K_{\mathrm{inh}} + C^{\mathrm{T}}_{\mathrm{O}_2}} \right)^6 \tag{1}$$

where K_{inh} is the inhibition constant and $C^{\mathrm{T}}_{\mathrm{O}_2}$ is the concentration of oxygen in the water phase. The inhibition factor $f^{\mathrm{NC}}_{\mathrm{denitr},C^{\mathrm{T}}_{\mathrm{O}_2}}$ decreases from 1 to 0 as the oxygen concentration rises which indirectly indicates that biodegradable SOM is depleting. The main consequence of this mechanism for ammonium removal by aeration is that sufficient biodegradable carbon should be present in the MSW. Otherwise aeration will only lead to increased concentrations of nitrate/nitrite in the leachate.

Our mechanism that facilitates denitrification generalizes the one proposed by Brandstätter et al. (2015) (Brandstätter et al., 2015b). They propose that denitrification mainly occurred at the bottom of the reactor where leachate remained anaerobic. Although this could have amplified the presence of denitrification, we believe it is not a prerequisite for denitrification. Anaerobic regions can exist everywhere near preferential flow-paths of air because of the limited penetration depth of oxygen in water.

Aeration also causes a rise in temperature in waste-bodies which subsequently affects biodegradation and leachate quality. Ritzkowski and Stegmann (2012) showed that temperature rise due to aeration can lead to enhanced ammonification, reduced nitrification and increased pH of the leachate. Our process networks for lysimeter scale do not consider the impact of temperature changes on biodegradation because temperatures were kept constant throughout the experiments. However, in order to predict field scale conditions, processes should be included that describe enthalpy changes and the effect of temperature on kinetics (e.g. Arrhenius equations).

Figure 5 shows the mass percentages of carbon and nitrogen in different phases during both aerobic and anaerobic leachate recirculation. Clearly, more carbon is removed under aerobic conditions ($\sim$ 25%) than under anaerobic conditions ($\sim$ 14%). This indicates that the electron acceptor (i.e. oxygen) was better mixed throughout the waste and/or the fraction of biodegradable carbon was higher. Also more nitrogen is removed under aerated conditions: $\sim$ 27% against $\sim$ 17%. Nevertheless, the amount of carbon and nitrogen removed is still quite small compared with the total amount in MSW. Therefore, we have the same conclusion for the aerobic recirculation treatment as for the anaerobic one: optimal electron acceptor distribution throughout the waste-body at field scale is a main issue to be tackled. Figure 5 furthermore indicates that carbon and nitrogen are mainly removed via CO_2 and N_2 during aeration. These phases are therefore important to measure for monitoring treatment effectiveness at a field scale.

Another interesting difference between anaerobic and aerated treatment is that the hydrolysis constant is lower during aeration. Stripping of water could have caused this by decreasing the amount of water per surface area of MSW. Although hydrolysis was not intrinsically enhanced, aeration still increased the rate of hydrolysis because the fraction of biodegradable carbon in MSW was increased. An optimal treatment strategy could be to alternate anaerobic and aerobic leachate recirculation which can generate both a high hydrolysis constant and a high fraction of biodegradable carbon.

Adopting a different perspective (and corresponding treatment goals) on how to achieve low long-term emissions can also increase the efficiency of treatment methods. Rather than aiming to completely remove all biodegradable carbon and nitrogen in the waste body, it could be sufficient to only remove the carbon and nitrogen in the proximity of the preferential flow-paths. In the latter case, slow diffusion from the bulk of the waste and the continuing degradation in the close vicinity of the preferential pathways jointly contribute to maintaining low emission levels. Follow up studies, however, should demonstrate that settlements do not significantly influence preferential flow-paths and therefore long-term emissions. Also, it should be investigated if emissions after treatment are not only temporarily low due to limitations such as drying of the preferential flow-paths.

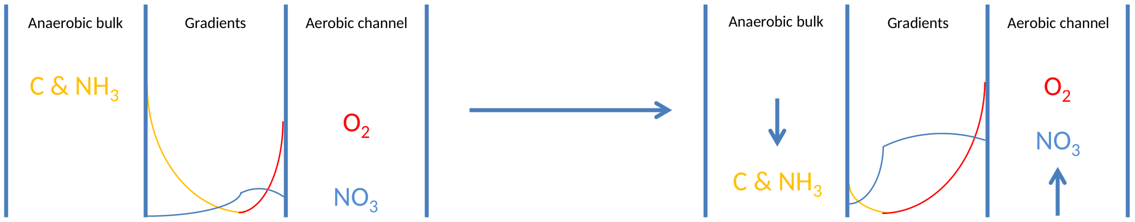


Figure 6: Illustration of the concentration gradients between aerobic and anaerobic regions in the MSW due to gas transport through preferential flow paths. When sufficient biodegradable carbon is present in the MSW, oxygen penetration is limited which leads to an anaerobic bulk where nitrate can be converted by denitrification (left). However, when the amount of biodegradable carbon becomes too small, oxygen penetrates deeper into the bulk which leads to failure of denitrification and increased nitrate concentrations (right).

4. Conclusion

This paper discusses factors and processes that control the effectiveness of leachate recirculation treatment under anaerobic and aerobic conditions. Fundamental process networks were found which closely reproduce the measured data from experiments at the lysimeter scale. Using qualitative and quantitative criteria, we indicate that fundamental uncertainty and ambiguity in these networks were small. The networks therefore represent factors and processes that control emissions of methane, carbon dioxide, pH, chloride and ammonium (under aerobic conditions) sufficiently accurate. Emission of ammonium under anaerobic conditions is only adequately described.

The effectiveness of both treatment methods is mainly controlled by how homogeneously important reactants such as oxygen and methanogens are distributed through-out the MSW. Under aerated conditions, a higher fraction of biodegradable carbon and nitrogen is reached and removed than under anaerobic conditions: 25% compared to 14% for carbon, and 27% compared to 17% for nitrogen. Interestingly, the rate constant of hydrolysis during aeration is lower than under anaerobic conditions. The rate of hydrolysis is nevertheless higher during aeration because the fraction of biodegradable carbon is higher.

Consumption of ammonium by sequential nitrification and denitrification suggests that anaerobic pockets were present in the MSW during aeration. This can be explained by transport of air through preferential flow-paths which is reasonable given the strong heterogeneity of the MSW. Ammonium removal via N_2 is only possible when sufficient biodegradable carbon is available. Otherwise, denitrification fails and the concentration of nitrate rises in the leachate.

To accurately monitor the effectiveness of treatments at field scale, the most important emitted phases to measure are CO_2, CH_4 and NH_4^+ under anaerobic conditions. For aerobic conditions, these phases are CO_2 and N_2. Gas phases, however, might be difficult to measure at field scale with current techniques. Phases such as bacterial biomass and stripped NH_3 contribute minimally to carbon and nitrogen removal.

To maximize degradation of organic matter in waste bodies at full-scale, the main challenge is to maximize distribution of the electron acceptors and other reactants throughout the waste-body. This could be achieved by optimizing geometry of gas wells and infiltration trenches and other operational parameters. It is, however, doubtful if removal rates of carbon and nitrogen can be improved beyond those observed in the lysimeter experiments where conditions were already quite optimal. A more realistic goal to achieve low long-term emissions might be to remove all biodegradable carbon and nitrogen in the proximity of preferential flow-paths; emissions should remain low afterwards because of slow diffusion from the bulk and continuing reactivity near the flow-paths. If this is indeed a feasible and stable mechanism, however, must be demonstrated in further studies.

5. Acknowledgements

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Tables and figures

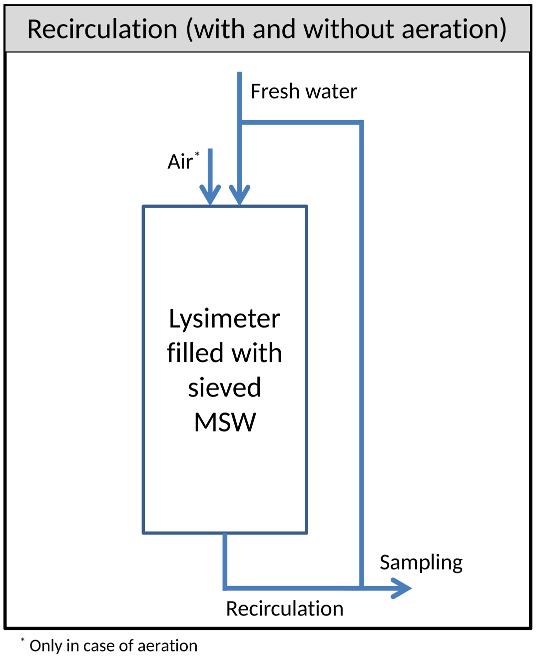


Figure 1: Illustration of the experimental set-ups for leachate recirculation under anaerobic and aerobic conditions (Brandstätter et al., 2015a,b). The lysimeters were made of stainless steel and had a cylindrical shape with a volume of 121 L. To allow for drainage of leachate by gravity, a fine meshed grid of 8cm was placed at the bottom of the reactors. Experiments were run for 2.25 years and reactor temperature was kept constant (~ 309K). MSW was sieved to a grain size < 20 mm and initially had a water content of 23% and a dry bulk density of 846 $\frac{\mathrm{kg}}{\mathrm{m}^3}$.

Table 1: Initial and environmental conditions in the duplicate experiments.

	Initial conditions						Environmental conditions					
	p_{CO_2}	p_{O_2}/p_{CH_4}	NH_3	pH	Cl^-	V_{liquid}	$\phi^{in1)}_{waterflow}$	$\phi^{out1)}_{sampleflow}$	$\phi^{in}_{airflow}$	V_{gas}	p^{tot}_{gas}	T
Type of experiment	[atm]	[atm]	[M]	[-]	[M]	[L]	[L d^{-1}]	[L d^{-1}]	[L d^{-1}]	[L]	[atm]	[K]
Anaerobic (calibration)	0.10	0	0.03	8	0.025	19.5	0.03	0.03	-	41.9	1	308.5
Anaerobic (validation)	0.10	0	0.03	8	0.025	19.5	0.03	0.03	-	41.9	1	308.5
Aerated (calibration)	0.41	0	0.025	7.25	0.025	21.24	0.0524	0.0483	56.67	38.8	1	308.5
Aerated (validation)	0.41	0	0.025	7.25	0.025	20.95	0.0526	0.0491	56.67	38.82	1	308.5

1) The flows of water are averaged over the entire measurement period. After the first week, the MSW reached field capacity and flows were relatively constant.

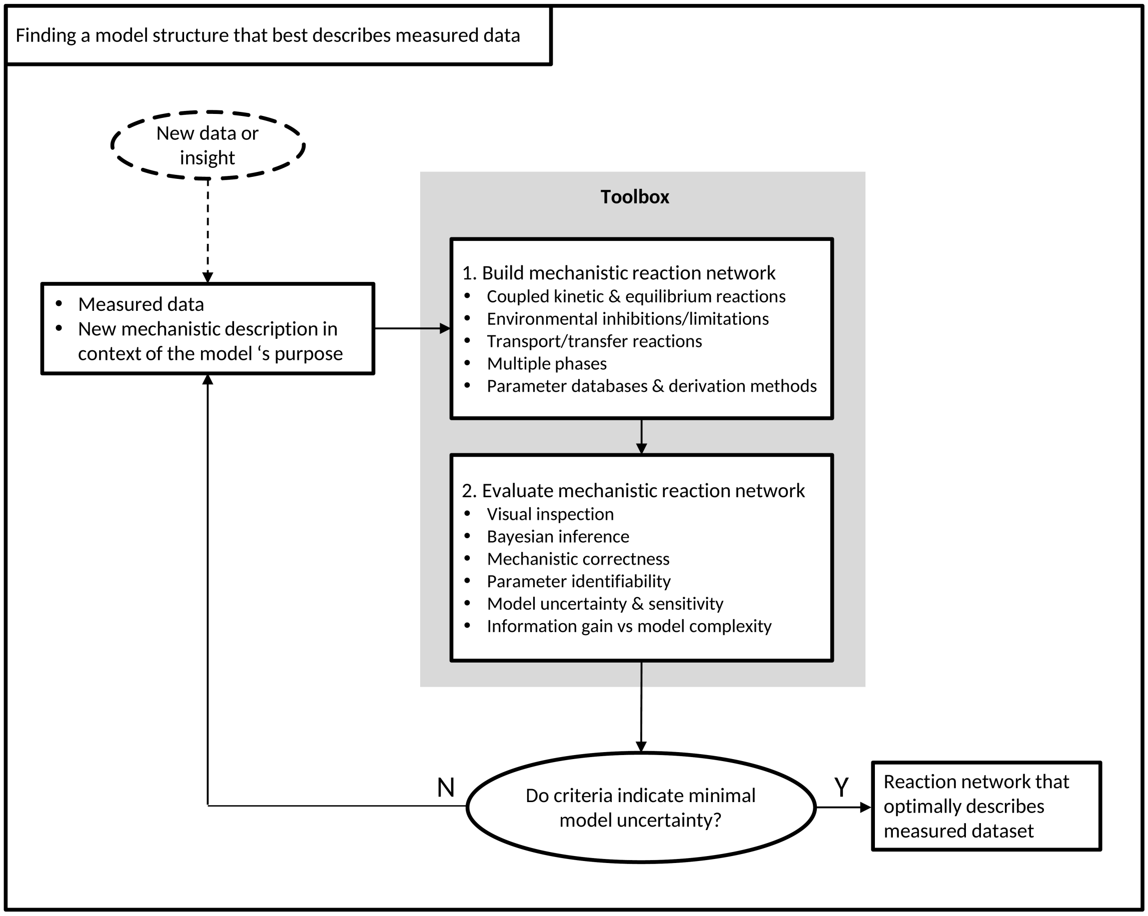


Figure 2: Iterative procedure for finding the biogeochemical process network that describes measured data with minimal ambiguity and uncertainty (van Turnhout et al., 2016). In step 1, the user manually builds the best network with the toolbox which is automatically optimized for the measured data in step 2. The user then decides if the network describes the measurements sufficiently accurate with the evaluation criteria generated in step 2. If not, the user starts another iteration and adapts the network.

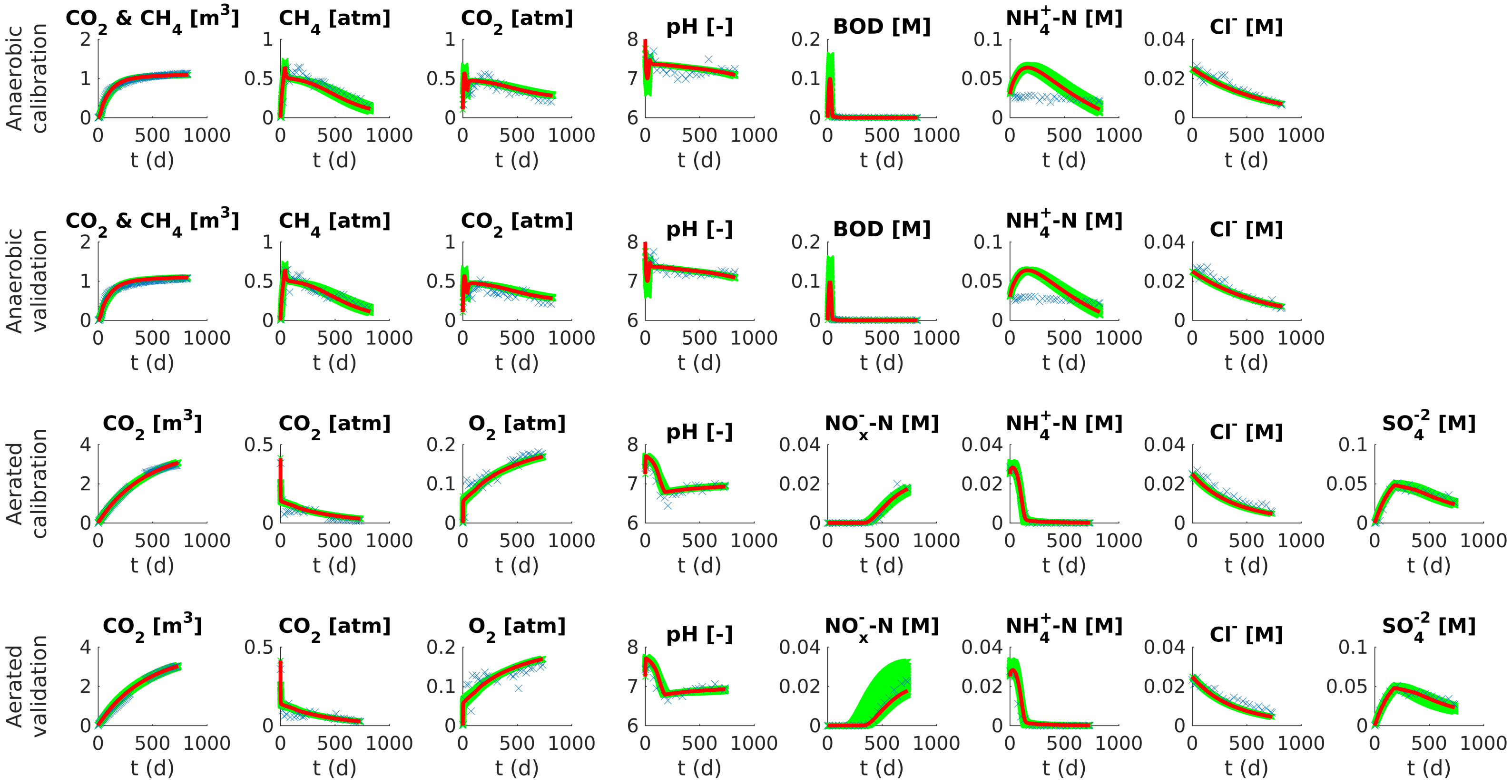
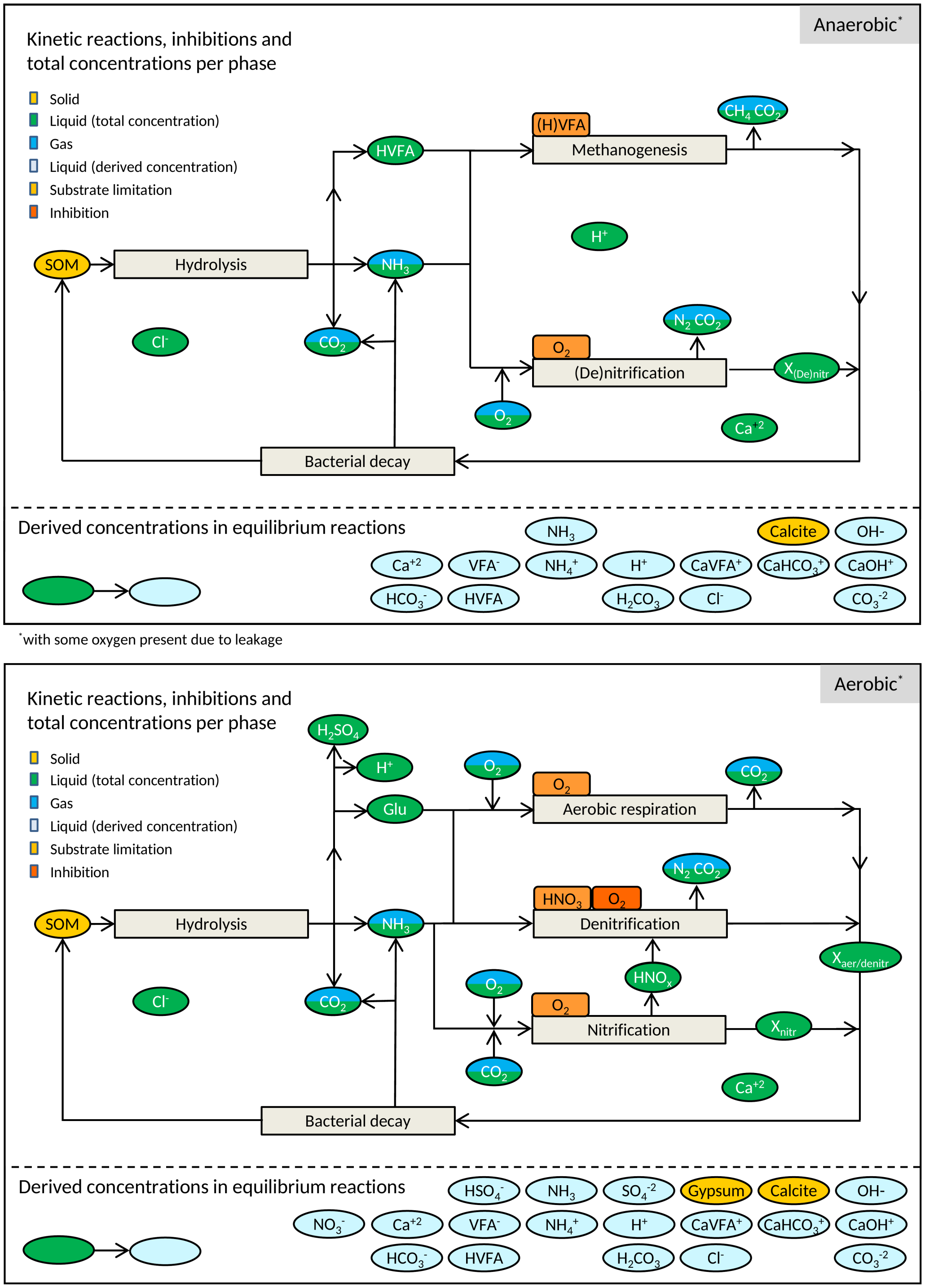


Figure 3: The visual agreement between the best fit of the modeled data (in red) and the measured data (in blue) for calibration and validation data measured during leachate recirculation under anaerobic and aerated conditions. The uncertainty in the total model error is indicated with green bandwidths surrounding the modeled data.



*with some oxygen present due to leakage

*with remaining anaerobic pockets because air is transported through preferential flow paths

Figure 4: The least ambiguous biogeochemical process networks for anaerobic (top) and aerobic (bottom) recirculation treatment on a lysimeter scale. In these schemes, gas and leachate flows that go into and out of the reactors are not included, but their values are listed in tables 1 and 3.

Table 2: Stoichiometry for the kinetic reactions in the biogeochemical process networks derived with the method from (Kleerebezem and van Loosdrecht, 2010) or deduced from the composition of MSW measured during sampling. Also the kinetic mechanisms are indicated. Parameters in bold were selected for calibration because they are highly uncertain and can significantly influence the model outcome.

Stoichiometry for the recirculation experiment under anaerobic conditions [mol]												Kinetics$^{c)}$
	SOM$^{a)}$	Acetic Acid	CO_2	NH_3	CH_4	H_2O	H^+	$X^{b)}_{meth}$	$X_{(de)nitr}$	O_2	N_2	
Hydrolysis$^{e)}$	−1	0.5	0	0.036	0	−0.172	0	0	0	0	0	$\mathbf{k}_{\mathrm{hyd}} \cdot \mathbf{C}^{\mathrm{T}}_{\mathrm{SOM}}$
Methanogenesis	0	−12.91	12.41	−0.2	12.41	0.6	0	1	0	0	0	$\boldsymbol{\mu}^{\max}_{\mathrm{meth}} \cdot \mathbf{C}_{\mathrm{X}} \cdot \mathbf{f}^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{Ac}}} \cdot f^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{NH_3}}}$
Bacterial decay $^{d)}$	0	0.5	0	0.2	0	−0.6	0	−1	0	0	0	$0.05 \cdot \mu^{\max}_{\mathrm{meth}} \cdot C_{\mathrm{X}}$
(De)nitrification	0	−0.12	0.39	−0.54	0	1.5	0	0	1	−0.63	0.17	$\boldsymbol{\mu}^{\max}_{\mathrm{(de)nitr}} \cdot \mathbf{C}_{\mathrm{X}} \cdot f^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{O_2}}} \cdot f^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{NH_3}}}$

Stoichiometry for the recirculation experiment under aerobic conditions [mol]													Kinetics
	SOM	Glucose	CO_2	NH_3	H_2O	H^+	$X_{aer\text{-}denitr}$	O_2	SO_4^{-2}	NO_3^-	X_{nitr}	N_2	
Hydrolysis	−1	0.167	0	0.032	−0.172	$\boldsymbol{\nu}_{\mathrm{H^+}}$	0	0	0.03	0	0	0	$\mathrm{k}_{\mathbf{hyd}} \cdot \mathbf{C}^{\mathrm{T}}_{\mathrm{SOM}}$
Aerobic respiration	0	−0.23	0.39	−0.2	0.99	0	1	−0.39	0	0	0	0	$\boldsymbol{\mu}^{\max}_{\mathrm{aer}} \cdot \mathbf{C}_{\mathrm{X}} \cdot f^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{Glu}}} \cdot f^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{O_2}}} \cdot f^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{NH_3}}}$
Nitrification	0	0	−1	−11.42	10.82	11.22	0	−21.44	0	11.22	1	0	$\boldsymbol{\mu}^{\max}_{\mathrm{nitr}} \cdot \mathbf{C}_{\mathrm{X}} \cdot \mathbf{f}^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{NH_3}}} \cdot f^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{CO_2}}}$
Denitrification	0	−0.24	0.42	−0.2	2.62	−0.33	1	0	0	−0.33	0	0.17	$\boldsymbol{\mu}^{\max}_{\mathrm{denitr}} \cdot \mathbf{C}_{\mathrm{X}} \cdot f^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{Glu}}} \cdot f^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{NO_3}^-}} \cdot f^{\mathrm{SL}}_{C^{\mathrm{T}}_{\mathrm{NH_3}}} \cdot \mathbf{f}^{\mathrm{NC}}_{C^{\mathrm{T}}_{\mathrm{O_2}}}$
Bacterial decay $^{d)}$	0	0.17	0	0.2	−0.6	0	(−1)	0	0	0	(−1)	0	$0.05 \cdot \mu^{\max} \cdot C_{\mathrm{X}}$

a) SOM is the biodegradable fraction of Solid Organic Matter in MSW generically modeled as $C_6H_{10}O_5N_{0.036}$ and $C_6H_{10}O_5N_{0.032}S_{0.03}$ for respectively anaerobic and aerobic conditions. b) X are species of bacteria with the elemental composition $CH_{1.4}O_{0.4}N_{0.2}$ taken from Henze et al. (1995). c) Substrate limitation factors (f^{SL}) and inhibition factors (f^{NC}) range between 0 and 1 (van Turnhout et al., 2016). Half saturation constants which are not calibrated have low values and are primarily included as switch factors. d) Bacterial biomass decay is included as a separate process for each species. e) Hydrolysis under anaerobic conditions is lumped with acidogenesis with hydrolysis being rate limiting.

Table 3: Results of the optimization: $5\% - 95\%$ quantiles of the posterior ranges. They are listed together with the prior ranges and the reference ranges measured under or estimated for non-limiting environmental conditions.

Anaerobic recirculation experiment

	prior	posterior	reference		prior	posterior	reference
k_{hyd} [d^{-1}]	0.0005 – 0.15	0.0094 – 0.0104	0.09 – 0.26$^{1)}$	X_{meth} [mM]	0.005 – 15	0.59 – 11	0.27 – 19$^{3)}$
$\mu^{\mathrm{max}}_{\mathrm{meth}}$ [d^{-1}]	0.01 – 3	0.04 – 0.12	0.8$^{2)}$	$X_{\mathrm{(de)nitr}}$ [mM]	0.015 – 15	1.2 – 14	0.27 – 19$^{3)}$
$\mu^{\mathrm{max}}_{\mathrm{(de)nitr}}$ [d^{-1}]	0.001 – 1	0.11 – 0.96	1.4$^{2)}$	$K^{\mathrm{meth}}_{\mathrm{s},C^{\mathrm{T}}_{\mathrm{Ac}}}$ [mM]	1 – 1000	5.5 – 16.7	0.03 – 420$^{4)}$
$\phi_{\mathrm{in,air}}$ [Ld^{-1}]	0.005 – 1.5	0.12 – 0.19	–	$C^{\mathrm{ini}}_{\mathrm{SOM}}$ [M]	2 – 3	2.27 – 2.33	–

Aerated recirculation experiment

	prior	posterior	reference		prior	posterior	reference
k_{hyd} [d^{-1}]	0.00024 – 0.026	0.0021 – 0.0023	0.09 – 0.26$^{1)}$	X_{nitr} [mM]	0.09 – 110	1.1 – 2.7	0.27 – 19$^{3)}$
$\mu^{\mathrm{max}}_{\mathrm{aer}}$ [d^{-1}]	1.2 – 121	25.6 – 118	57$^{2)}$	$X_{\mathrm{aer-denitr}}$ [mM]	7 – 800	9.5 – 35.4	0.27 – 19$^{3)}$
$\mu^{\mathrm{max}}_{\mathrm{nitr}}$ [d^{-1}]	0.003 – 0.31	0.01 – 0.02	1.4$^{2)}$	$K^{\mathrm{nitr}}_{\mathrm{s},C^{\mathrm{T}}_{\mathrm{NH_3}}}$ [mM]	0.9 – 110	1.6 – 7.1	0.04$^{5)}$
$\mu^{\mathrm{max}}_{\mathrm{denitr}}$ [d^{-1}]	2 – 201	6.4 – 198	54$^{2)}$	$K^{\mathrm{denitr}}_{\mathrm{i},C^{\mathrm{T}}_{\mathrm{O_2}}}$ [mM]	0.005 – 0.6	0.047 – 0.093	–
$C^{\mathrm{ini}}_{\mathrm{SOM}}$ [M]	5 – 7	6.2 – 6.5	–	$\nu^{\mathrm{hyd}}_{\mathrm{H^+}}$ [mol]	0 – 0.06	0.037 – 0.041	–

1) Veeken and Hamelers (1999) 2) Kleerebezem and van Loosdrecht (2010) 3) Nopharatana et al. (2007) 4) Meima et al. (2008) 5) Kantartzi et al. (2006)

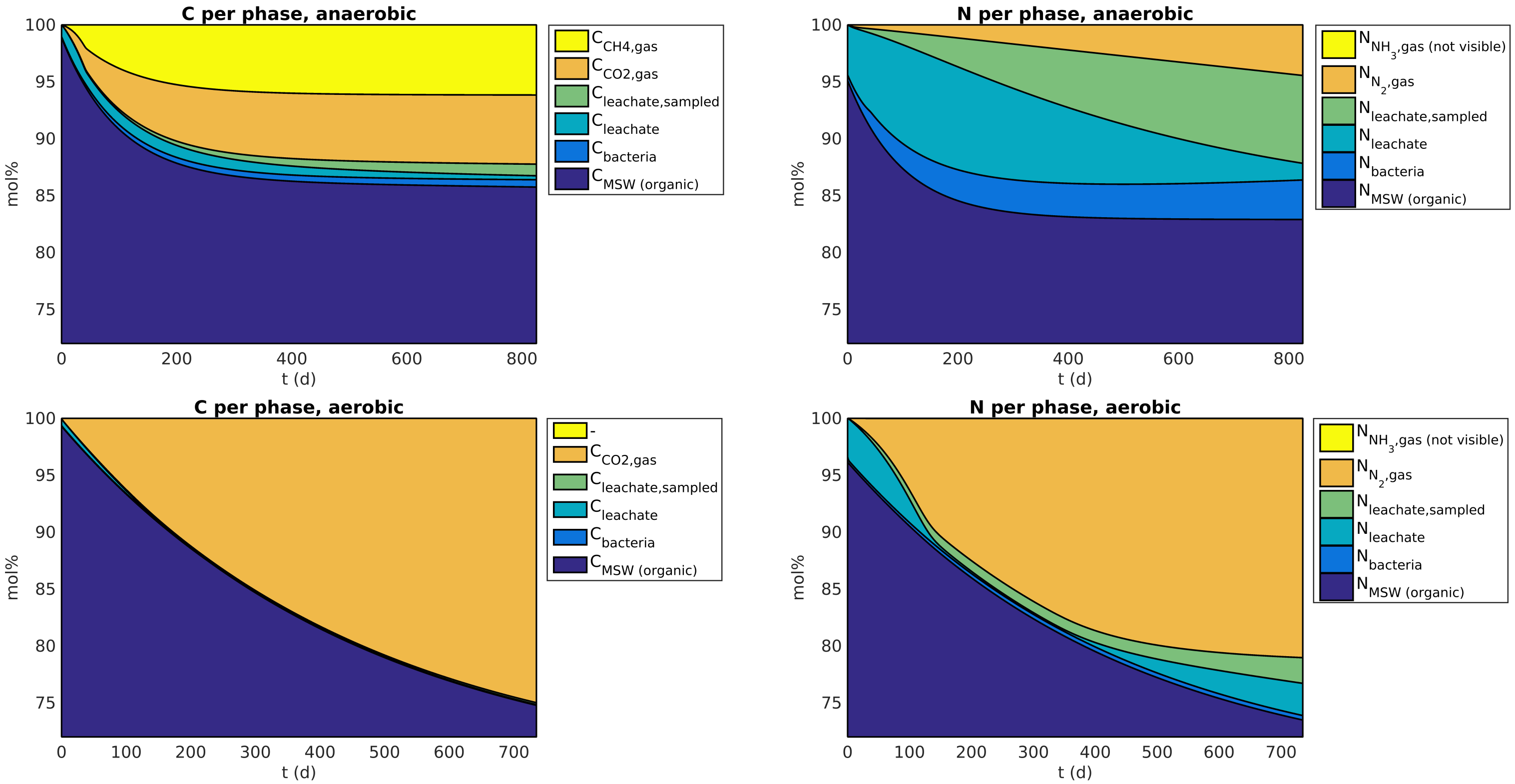


Figure 5: Modeled mass percentages of organic carbon and nitrogen in different phases during anaerobic and aerobic leachate recirculation. They indicate which phases are dominant in removing carbon and nitrogen from MSW for each treatment method.

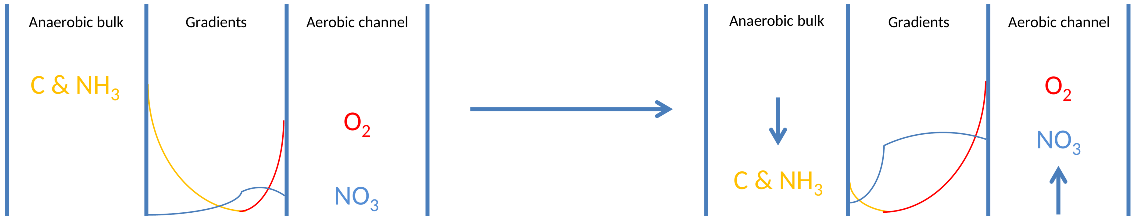


Figure 6: Illustration of the concentration gradients between aerobic and anaerobic regions in the MSW due to gas transport through preferential flow paths. When sufficient biodegradable carbon is present in the MSW, oxygen penetration is limited which leads to an anaerobic bulk where nitrate can be converted by denitrification (left). However, when the amount of biodegradable carbon becomes too small, oxygen penetrates deeper into the bulk which leads to failure of denitrification and increased nitrate concentrations (right).