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MECHANISMS GOVERNING CARBON AND NITROGEN PATHWAYS DURING ENHANCED WASTE DEGRADATION IN LANDFILL SIMULATOR REACTORS

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ABSTRACT: Nitrogen undergoes multiple biogeochemical transformations during waste degradation, which depend on speciation, prevailing geochemical boundary conditions, and waste surface properties. This study developed a waste biodegradation model with high flexibility in accommodating reaction pathways to assess different process dynamics. The model was applied to landfill simulator reactors operating anaerobically. Model results show that dilution with adsorption matches the experimental dissolved NH_4^+ concentration (C/N=25) at the early experimental stages. Also, NH_4^+ binding decreases due to competition with Ca^{2+}, and the model better captures the dissolved NH_4^+ behavior when $CaSO_4$ is present in solution. Mass removal due to sampling and posterior dilution are the main mechanisms to reduce NH_4^+ concentration in the leachate. The model highlights the role of nitrogen sorption as the main mechanism for nitrogen accumulation in the solid phase of municipal solid waste.

Keywords: Nitrogen, sorption, municipal solid waste, inhibitions, boundary conditions.

1. INTRODUCTION

Next to carbon, nitrogenous compounds are released from the microbial degradation of organic matter in landfilled waste. As a result, landfill gas may contain N2O as potent nitrogenous greenhouse gas (Berge et al., 2007). The leachate composition is typically dominated by high concentrations of ammonium (NH4+), which next to other contaminants, needs to be reduced before discharge into the environment is possible (Kjeldsen et al., 2002). In light of efforts to reduce the intensity of aftercare, it is of interest to understand and predict landfill nitrogen transformations.

Experimental results with landfill simulator reactors (LSRs) are useful to identify nitrogen transformations at the different solid, liquid, and gas phases (Brandstätter et al., 2015; Fricko et al., 2021). However, up to this date, results have not been conclusive regarding governing nitrogen conversion mechanisms and pathways (Lubberding et al., 2012). For instance, N2 generation from denitrification is difficult to quantify with sufficient accuracy, impacting the nitrogen mass balance estimation in the gas phase. In addition, there is a lack of waste degradation models applied to nitrogen, which prevents understanding the system dynamics and potentially predicting nitrogen turnover in a landfill.

Nitrogen can undergo multiple biogeochemical transformations (Oudart et al., 2015) and physical (de-)sorption processes, depending on its speciation, and hence on the prevailing geochemical boundary conditions and waste surface properties (He et al., 2017; Liao et al., 2013). Boundary conditions such as

O2 availability and in-out water flow in landfills (or LSRs) determine the rate of organic matter degradation, where O2 is usually a limiting factor.

This research developed a mechanistic biodegradation model based on an existing toolbox (van Turnhout et al., 2016). The biodegradation model is a powerful tool for testing hypotheses and identifying process dynamics. It has great flexibility to include and evaluate various bio-physico-chemical mechanisms under different environmental conditions. The model tracks chemical speciation at the solid, liquid, and gas phases. Therefore, gas and leachate quantity/quality are obtained as a function of time. In addition, the inclusion of boundary conditions allows to estimate mass outflows and assess the impact of external measures such as water or O2 inclusion.

The biodegradation model is applied to experimental results from LSRs operated under anaerobic conditions (Brandstätter et al., 2015), and follows a similar modeling approach as presented by Reichel et al., 2007 and van Turnhout et al., 2018 to advance on an explanation for measured NH4+ concentration in the leachate. Previous modeling efforts have proven unsuccessful to explain NH4+ concentrations in the leachate under anaerobic conditions. The model presented here contributes to an explanation by including sorption mechanisms and rigorous handling of boundary conditions.

2. NITROGEN TRANSFORMATION PATHWAYS

Landfill waste degradation accompanies the release of nitrogen compounds. Ammonification, which results from hydrolysis, releases NH4+ as well as volatile fatty acids (VFA). The extent of NH4+ released depends on the waste C/N ratio, which varies depending on regions and waste age (Campuzano & González-Martínez, 2016). VFA and NH4+ accumulation might hinder hydrolysis due to toxicity (Oudart et al., 2015). In addition, NH4+ volatilization to NH3 is significant at high temperatures, that are achievable at aerobic conditions.

NH4+ adsorption to municipal solid waste (MSW) is a nitrogen sink that is often overlooked. Experimental studies have shown that MSW has a high sorption capacity and a high affinity for NH4+ adsorption (He et al., 2017; Liao et al., 2013). On the other hand, desorption is weak, pointing out a strong NH4+ binding to MSW. This physicochemical mechanism depends on pH, organic matter content, other cations in solution, and the availability of reactive sites, which increase with MSW age (He et al., 2017; Liao et al., 2013).

In addition, biochemical mechanisms such as nitrification, denitrification, anaerobic ammonia oxidation (anammox), or nitrogen fixation, can occur if the required substrates and electron acceptors are available (Aasfar et al., 2021; Berge et al., 2007; Meyer-Dombard et al., 2020; Strous et al., 1997). Microbial growth assimilates NH4+, reducing its release into the leachate, and dilution, for example by infiltrating precipitation, further decreases NH4+ concentration in the leachate. All the above-mentioned mechanisms can occur simultaneously.

3. MODEL DESCRIPTION

A bespoke bio-geochemical model has been developed based on van Turnhout et al. (2016). The model allows great flexibility in accommodating reaction pathways to assess different process dynamics that represent the degradation of MSW. The biodegradation model calculates out-of-equilibrium conditions or process kinetics in Python and, as a result, determines the change in concentration of different species within the system. For instance, the growth of microorganisms can be tracked, or it is possible to identify the production or consumption of byproducts from waste degrading in time.

Thermodynamic principles allow the creation of biochemical metabolic pathways. In addition, energetic differences between autotrophic and heterotrophic organisms are considered when calculating metabolic yields (Kleerebezem & Loosdrecht, 2010). Furthermore, the model is capable to handle physicochemical equilibrium conditions through a direct coupling between the

ORCHESTRA software (An Object Oriented Framework for Composing Chemical Speciation and Mass Transport Models, Meeussen, 2003) and the kinetics part of the biodegradation model built in Python. Specific concentrations of chemical compounds from the kinetic solver can be passed at every time step to ORCHESTRA. Afterward, the chemical equilibrium solver can calculate chemical speciation at equilibrium in the solid, liquid, and gas phases. The model integrates a wrapper for ORCHESTRA so that its capabilities can be accessed from Python. A C++ version of ORCHESTRA was used with pybind11 to develop a package (PyORCHESTRA) that is imported to the model so that data can be passed as NumPy or Pandas data-structures.

The equilibrium state and speciation of certain state variables calculated in ORCHESTRA, such as pH or NH_4^+ dissolved in the leachate can be passed back to the kinetic solver where certain inhibitions to the rates of the kinetic pathways take place. For instance, a high pH might cause specific reactions to stop, or high concentrations of dissolved species can be toxic to certain microorganisms also causing reactions to halt. The model includes a broad range of environmental conditions that act as inhibitions to the system, as well as other enzymatic inhibitions, substrate limitations, and toxicities to represent how certain processes affect the reaction rates (El-Fadel et al., 2009; Monod, 1949; Reichel et al., 2007; van Turnhout et al., 2016; White et al., 2004).

For every biochemical reaction, the model has a substrate limitation inhibition that accounts for conditions when the reactants are limited. Toxicity in the model accounts for conditions in which the presence of certain chemical compounds within a threshold affects reaction rates. An example of this inhibition is O_2 limiting methanogenesis (Jarrell, 1985). The model can also accommodate competitive and non-competitive inhibitions to reflect cases in which enzymes are blocked due to the presence of other compounds in solution. In the model, every biochemical reaction has an optimal pH range and temperature. Therefore, pH and temperature inhibitions account for deviations from the optimal range. The model includes mechanisms such as dilution or O_2 inclusion into the system as boundary conditions. These types of mass exchange allow for quantifying mass loss and better simulating operating conditions of landfill simulator reactors. Furthermore, the model can assess interactions between the liquid and the solid phases. For instance, different adsorption models in ORCHESTRA allow to quantify the adsorption of specific cations in solution to the surface of MSW.

The model coding philosophy tries to maximize performance. The Phyton scripts include classes and vectorized calculations, and the kinetic solver uses an imaginary derivative approach implementation. These arrangements lead to a fast integration time for the assessed reaction network. The methods in the classes were coded following a generic approach so that it could calculate the rates for different numbers of degradation pathways and state parameters. A main script contains a class with all the functions to calculate the kinetic rates, obtain the equilibrium calculations from ORCHESTRA, and manage boundary conditions. This script connects to a stoichiometry module, and a non-equilibrium correction function obtains a new metabolic stoichiometry at every time step. The model uses a calling script in which the input information is loaded, the class gets initialized, the integration instruction occurs, and the figure plotting takes place.

In summary, the user can quickly build and test a reaction network and assess the relevance of the tested mechanisms. Model complexity can be added in a stepwise approach. Also, the incremental change in model results aids the understanding of system dynamics. Particularly, model inhibitions can indicate the cause(s) for system stabilization in time, or trace back the resulting concentrations to a fundamental bio-physico-chemical mechanism.

4. EXPERIMENT DESCRIPTION

This study focuses on the simulation and analysis of LSRs presented by Brandstätter et al. (2015). The LSRs have a capacity of 121 L and were operated under anaerobic conditions. LSRs were filled with old MSW, and leachate was recirculated every time gas was sampled. A comprehensive experimental description can be found in (Brandstätter et al., 2015; Turnhout et al., 2018) as well as model parameters.

The experiments lasted for 823 days, with a constant temperature of 308.15 ºK, leachate sampling removed approximately 2 L of leachate at irregular intervals with a total of 27 samplings. Similarly, deionized (DI) water replaced the volume removed. In addition, the gaseous C-discharge (as CH_4-C and CO_2-C) was measured.

5. MODEL IMPLEMENTATION

Table 1 shows the resultant reactions this study considers from the catabolic and anabolic pairings**.** This study followed a similar approach from van Turnhout et al. (2018), in most cases model parameters were obtained from that research. In this study, there is a clear distinction between non-metabolic and metabolic pathways. Metabolic pathways follow a coupling between catabolic and anabolic reactions, as well, yield determination is corrected by the activities of the compounds present in solution.

Table 1. System's resultant metabolic reactions from the catabolic and anabolic pairings. SOM = Solid organic matter, AnaX = Biomass generation.

Reaction_ID	Reaction name	Reaction pairing	Resultant reaction	Maximum rate (μ_{max}) (d^{-1})
C1	Hydrolysis of SOM	Non-metabolic	Hydrolysis of SOM	0.0101*
C2	Propionate to acetate	C2-A1	Acetogenesis	0.035
C3	Acetate to CO2 & CH4	C3-A1	Methanogenesis	0.013
C4	Ammonium oxidation	C6-A2	Nitrification	0.2669*
C5	Acetate-_NO3	C7-A1	Denitrification	0.2669*
C6	Microbial decay	Non-metabolic	Microbial decay	0.05· μ_{max} 0.003 (Methanogens)
A1	AnaX with acetate			
A2	AnaX with HCO3-			

***** Maximum rates ($\boldsymbol{\mu_{max}}$) from van Turnhout et al. (2018)

Figure 1 shows the conceptual model applied to the LSRs. Environmental conditions influence biochemical and physico-chemical processes. Gas venting and leachate sampling are the governing mass outflows from the system. Leachate sampling removes mass of dissolved compounds at the sampling times. On the other hand, DI water replenishes the sampled volume and carries dissolved O_2 with it. Dissolved O_2 is relatively toxic to methanogens, but is also an electron acceptor for nitrification/denitrification.

This model considers hydrolysis as a non-metabolic reaction, while acetogenesis, methanogenesis, and nitrification/denitrification are metabolic pathways. The selection of metabolic and non-metabolic pathways falls on the model user. For every metabolic pathway involved in the reaction network, the model considers microbial growth and decay. In this implementation, substrate limitations apply to every substance involved in a reaction. In the same way, non-ideal pH and temperature modify reaction rates. VFA and NH_4^+ accumulation reduces hydrolysis (Oudart et al., 2015), as well as dissolved O_2 coming from the DI water to methanogens. The model considers those effects as toxicities.

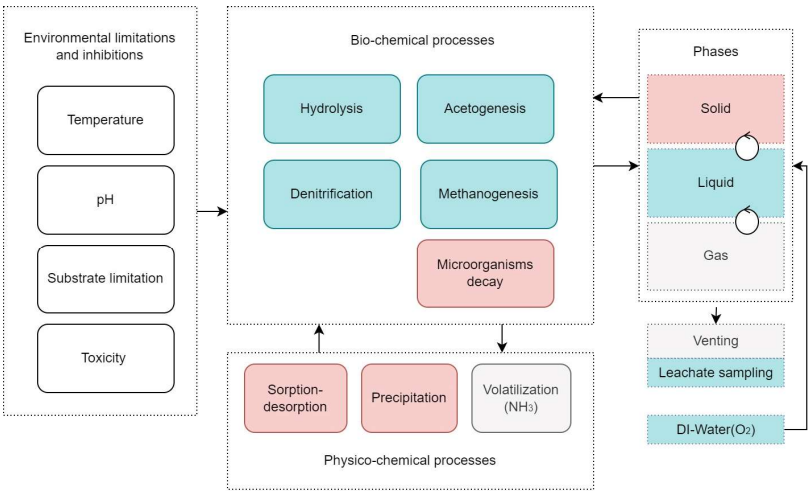

Figure 1. The conceptual model applied to the LSRs at anaerobic conditions.

Table 2 presents the stoichiometry for every catabolic and anabolic mechanism, as well as the state parameters considered at non-equilibrium conditions. Further, the initial compound concentrations are shown. Gibbs formation energies of every compound needed to obtain metabolic yields were obtained from Kleerebezem & Loosdrecht (2010) and Tro (2017). The ideal pH for all reactions lies between 7 and 9, and the optimal temperature for all reactions was assumed to be 315.15 ºK (Nie et al., 2021). The model includes a NICA-Donnan model from ORCHESTRA to capture the adsorption of cations in solution to the MSW. Default parameters are used in the model, but a specific reaction for NH4+ adsorption with H+ parameters was added. Simulations consider the presence of Ca2+ and SO42- in solution, so that gypsum and calcite can precipitate. This mechanism buffers pH.
This study considers the effect of different C/N ratios (C/N = 166, C/N = 25) on the amount of dissolved NH4+; the selection was based on van Turnhout et al. (2018) and Fricko et al. (2021). The initial carbon content was kept constant while the nitrogen concentration varies. The elemental waste compositions the model used are as follows: C6H10O4N0.036 for the high and C6H10O4N0.24 for the lower ratio.

Table 2. Reactions stoichiometry and initial compound concentrations of state parameters at non-equilibrium conditions.

Reaction name\initial concentrations [mol/L]	SOM	H_2O	VFA[1*]	VFA^{2*}	NH_4^+	HCO_3^-	H_2	H^+	X_{bio}	O_2	CO_2	CH_4	NO_3^-	N_2
	2.3	55.6	0	0.5	0.027	0.4	0.01	0.0204	(0.001 -0.01)	0	0	0	0	0
SOM_Hydrolysis	-1	-7	1	0	0.036	3	4.5	6.856	0	0	0	0	0	0
Propionate_Acetate	0	1	-1	2	0	-1	-1	0	0	0	0	0	0	0
Acetate_CO2_CH4	0	0	0	-1	0	0	0	-1	0	0	1	1	0	0
Ammonium_ox	0	1	0	0	-1	0	2	0	0	-2	0	0	1	0
Acetate-_NO3	0	0.8	0	-1	0	2	0	-0.6	0	0	0	0	-1.6	0.8
Microbial_decay	0	-0.6	0	0.5	0.2	0	0	0	-1	0	0	0	0	0
AnaX_Acetate	0	0.4	0	-0.525	-0.2	0.05	0	-0.275	1	0	0	0	0	0
AnaX_HCO3-	0	0.925	0	0	-0.725	-1	0	0.25	1	0	0	0	0.53	0

SOM (hydrolysable solid organic matter → $C_6H_{10}O_4N_{0.036}$), Xbio/biomass → $CH_{1.8}O_{0.5}N_{0.2}$, VFA^{1*}/ Propionate → $C_3H_5O_2^-$, and VFA^{2*}/Acetate- → $C_2H_3O_2^-$

6. RESULTS

Figure 2 (top left) shows that degradable solid organic matter (SOM) hydrolyzes relatively fast during the first 200 days of the simulation. On the other hand, degradation occurs at a slower pace between days 200 and 400, but degradable SOM nearly depletes by day 500. Propionate generation peaks by day 70. From that time on, concentration decreases at different rates, and propionate nearly depletes by day 150. Acetate concentration in the simulation sharply rises in the first 200 days, then the concentration decreases, until depleting at day 800. Abrupt changes in concentrations correspond to leachate sampling events (2 L per sampling event). Mass removal due to leachate sampling and the addition of DI water strongly affect the concentration of microorganisms, as Figure 2 (top right) shows. Dissolved O2 concentration is small and follows an off-and-on trend, similar to NO3- and N2, as depicted in Figure 2 (bottom left). Finally, Figure 2 (bottom right) shows measured and modeled bio-gas. Modeled results do not capture the sharp rise at the early experimental stages, but reach the cumulative experimental gas production.

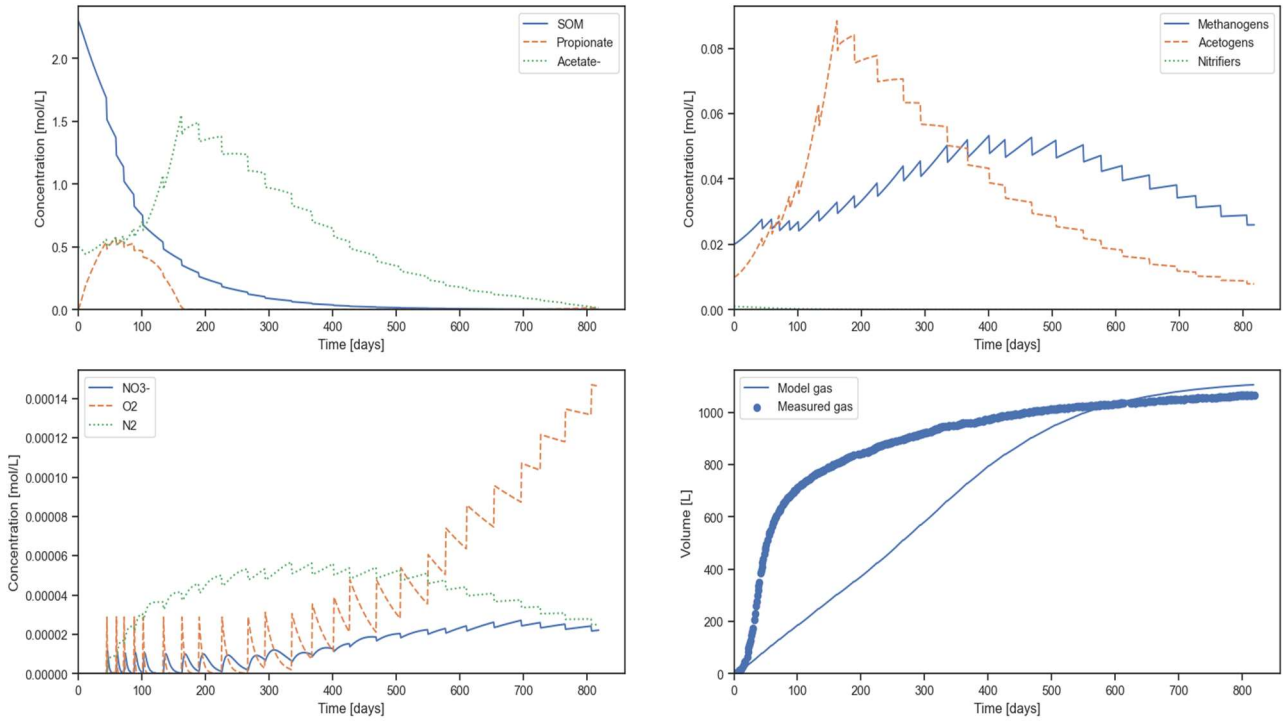

Figure 2. Modeled state parameters at out-of-equilibrium conditions in a system with all bio-physical-chemical mechanisms considered in the conceptual model at high C/N. Top left: solid organic matter and VFAs, top right: microorganisms in the system, bottom left: dissolved O_2 and nitrification/denitrification byproducts, bottom right: modeled and measured biogas.

Figure 3 shows substrate limitations inside the system. NH4+ is a limiting substance for acetogenesis, while degradable SOM impacts hydrolysis. Those reaction rates are highly impacted after day 500 when substrate limitations reach a factor below 0.6. Furthermore, a small O2 concentration strongly substrate inhibits nitrification and, in a similar manner, NO3- to denitrification. pH and temperature inhibitions are not strong in the system, due to a stable pH and a constant temperature close to optimal conditions as stated above.

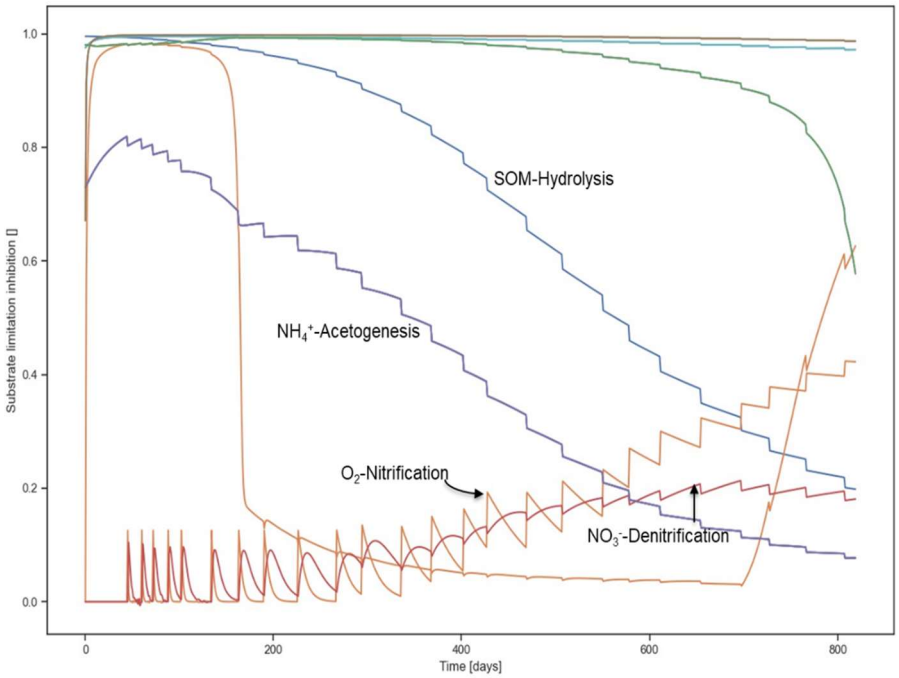

Figure 3. Overview of the main substrate limitations in the system.

It is possible to identify the dilution effect in all the state parameters within the system. Concentrations sharply decline in a staircase shape. Figure 4 shows the experimental and model chloride concentration in time. Figure 4 presents the model data with the experimental dilution volume of 2 L. However, the model obtains a closer match with a dilution volume of 1 L (Figure 4, right).

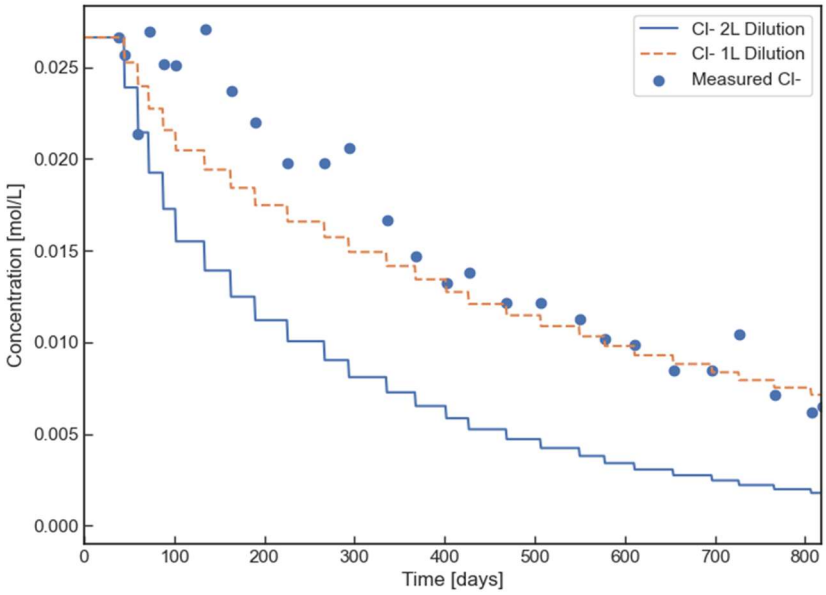

Figure 4. Experimental (dotted) and modeled leachate chloride concentration in time at two dilution conditions.

Figure 5 presents dissolved NH4+ in five different scenarios. The model considers two different C/N ratios; one that was used by van Turnhout et al. (2018), C/N = 166 (so-called high C/N), and another one with C/N = 25. Model mechanisms superimpose each other. For instance, simulations considered only a dilution effect, then with sorption, and ultimately with Ca2+ and SO42- present in solution.

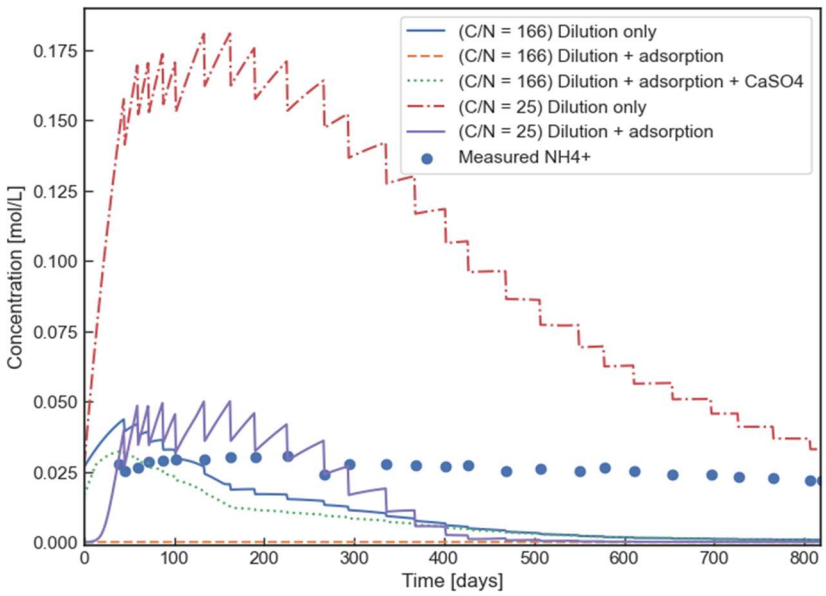

Figure 5. Experimental (dotted) and modeled NH_4^+ present in the leachate. Dilution due to replenishment of sampling volume, adsorption, and adsorption with enough Ca^{2+} and SO_4^{2-} (gypsum) present in solution are considered in the simulation at two different C/N ratios (C/N = 166, C/N = 25).

Results show that at a high C/N ratio dilution matches the experimental NH4+ concentrations trend during half of the experiment. Afterward, modeled NH4+ falls below experimental values. On the contrary, when adding a sorption mechanism, the dissolved NH4+ goes to 0, but when Ca2+ and SO42- are present in solution the model obtains a better match for dissolved NH4+. At C/N = 25, dilution is not able to capture the experimental dissolved NH4+ behavior. However, by adding a sorption mechanism, the model better captures dissolved NH4+ behavior during the first 300 days.

7. DISCUSSION

In the investigated LSR, O2 inclusion, which is little due to its low solubility in water (0.00028 mol/L, Montgomery et al., 1964), comes from the injection of DI water. Nitrification is limited by O2 availability but appears to be enough to produce NO3- in small quantities, which is then consumed by denitrification to further produce N2. This condition explains the appearance of NO3- in LSR that operate without an apparent O2 source. Concentrations are in the order of ppm, but without O2 limitations, these mechanisms may become dominant due to high reaction rates. NH4+ volatilization as NH3 is minor.
Mass removal due to leachate sampling and posterior dilution with DI water constitutes the dominant removal pathway of NH4+ in the studied LSR. The dilution factor is around 10% in the experimental case. Furthermore, at a high C/N ratio and without other excess competing cations in solution, sorption can bind all NH4+ present in solution. This strong effect is in agreement with the literature (He et al., 2017; Liao et al., 2013), but does not explain the experimental case results. On the contrary, by adding excess cations in solution (Ca2+), NH4+ binding decreases due to competition with Ca2+, and the model better captures the dissolved NH4+ behavior. At C/N = 25, dilution continues to play a significant role in decreasing dissolved NH4+ concentrations. In this scenario, a better fit for the experimental data is obtained with the sorption mechanism. In all scenarios, the model underestimates dissolved NH4+ concentrations at later experimental stages, this could be related to a missing mechanism in the model. Furthermore, a better selection of reaction rates might improve the fitting with experimental gas production.

8. CONCLUSION

The model shows that the systems considered in this study reach equilibrium or exhaust their reactivity by substrate limitations. Furthermore, reaction rates are highly regulated by specific environmental conditions, which makes the system progress toward equilibrium slowly. Mass removal due to sampling and posterior dilution is the main mechanism to reduce NH4+ concentration in the leachate. Finally, the model highlights the role of nitrogen sorption as the main mechanism for nitrogen accumulation in the solid phase of MSW which is more than 40 years old.

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