JEE Journal of Ecological Engineering

Journal of Ecological Engineering 2021, 22(5), 205–213
https://doi.org/10.12911/22998993/135314
ISSN 2299–8993, License CC-BY 4.0

Received: 2021.02.16
Accepted: 2021.04.14
Published: 2021.05.01

Performance of Reactive Nitrogen in Leachate Treatment in Constructed Wetlands

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ABSTRACT

Reactive Nitrogen (Nr) is produced from natural and human activity, the use of fuel, the activities of industry, and agriculture. The Nr from agriculture is used to produce food crops, but excess Nr has an impact on the surrounding land. Landfills also generate Nr from the decomposition of waste which then releases the leachate containing Nr. This study aimed to determine the value of Nr generated by landfills, the effect of Nr on the environment, and the performance of Nr when used in Constructed Wetlands (CW). Review papers were collected from several studies and publications. Nr commonly found in leachate landfills include: NH_4, NH_3, NO_2, and NO_3. The Nr present in landfill leachate at CW can be used for proper plant development and growth, which significantly increases and enhances its quality and yield by playing an important role in the biochemical and physiological functions of plants. In addition, the content of hazardous substances in landfill leachate can also be processed using CW. This review paper discusses the effects of Nr from human activities ending up in landfills. The landfill leachate with Nr content can be used in CW for plant growth.

Keywords: Reactive Nitrogen, performance, effects, landfill leachate, constructed wetland

INTRODUCTION

All nitrogen compounds except N_2, are Reactive Nitrogen (Nr). Naturally available, Nr is generated primarily by and Biological N_2 Fixation (BNF), lightning, and forest fires. Nitrogen is an essential constituent for all life, and is part of protein and many other compounds (Bach et al., 2020). In order to increase agricultural production, humans have developed the technology to produce synthetic Nr fertilizers (Schlesinger, 2009). In addition, humans also generate Nr by accident. Gases such as CO_2 and sulfur dioxide, but also NOx produced from fossil fuels combustion by industry, transportation and the energy sector emit not only emissions (Erisman et al., 2011). Reactive nitrogen is an indispensable nutrient for agricultural production. The largest contributor to Nr pollution which causes severe damage to human health and ecosystem is the agricultural sector. Globally, about 425 TgNr are generated through natural processes in the oceans, land, and by human activities (Sutton et al., 2011). The outcomes of all human activities finally enter the landfill which produces Nr in leachate. The resulting landfill leachate contains NH_3 about 6.83 mg/L (Kahar, 2017), NH_4 166.07 mg/L (Yalçuk and Ugurlu, 2020), NO_2 about 2,497 mg/L, and NO_3 with a value 12.5 mg/L (Silvestrini et al., 2019). From year to year, the human activities that produce Nr are increasing 15 $TgNyr^{-1}$ to 140 $TgNyr^{-1}$ from 1890–1990 (Raza et al., 2018).

The excess Nr can cause environmental pollution problems, such as radiation emission, air and water quality pollution (Ibrahim et al., 2020; Ravishankara et al., 2009). The reactive nitrogen generated can enter the groundwater, rivers, and estuaries, causing eutrophication (Braun, 2007). Eutrophication can negatively impact biodiversity and fish stocks (United Nations Environment Programme (UNEP), 2014). The increased release of Nr into the environment has a negative impact on living things and ecosystems (Lassaletta et al., 2014; Leach et al., 2012). When released from soil, reactive nitrogen can make surface water and groundwater unfit for human consumption. After the Nr molecules are produced, they can remain in the environment for quite a long time because they are highly mobile and can contribute to several environmental effects as they flow through soil and water (Erisman et al., 2011). Excessive Nr release to the environment has led to many problems, including the loss of marine and terrestrial biodiversity, the formation of greenhouse gases, air pollution, and increased levels of nitrates in groundwater and marine ecosystems. The limit for nitrogen has been exceeded so that reducing the nitrogen emissions should be a key objective of environmental policy (Bach et al., 2020).

The mechanisms involved in Nr removal in Constructed Wetland (CW) include evaporation and adsorption (Kadlec and Wallace, 2009). The performance of Nr mechanism in CW depends on the microbial and vegetation processes (Gupta et al., 2016; Kadlec and Wallace, 2009). These organisms contribute to Nr uses such as microbial interactions and uptake by vegetation. Abiotic mechanisms include sedimentation, filtration, chemical precipitation, and adsorption (Dordio et al., 2009). Organic matter is excreted in SSF built up in wetlands by aerobic bacteria that adhere to porous media and plant roots. Plant roots not only provide the necessary surface for bacterial growth, but also oxygen. The performance of Nr in CW is basically achieved by microbial interaction and adsorption (Gupta et al., 2016; Mangkoedihardjo & Samudro, 2014).

Reactive nitrogen is an important plant nutrient and some can fertilize agricultural areas (Sutton et al., 2011). As it passes through the agricultural supply chain, the Nr from these sources is then lost as waste (Bodirsky et al., 2014). The average total nitrogen removal efficiency was 98.5%, 68.9% and 79.6% using CW (Wojciechowska, 2017). Biological assimilation of CW can recover Nr in 83–87% (Sengupta et al., 2015). The P increased deposition of N improves plant growth (Y. Liu et al., 2013; Wang et al., 2019; Xu et al., 2014). Some data show that Nr in plants has a function in overcoming aging in plant seeds, as a factor that has the potential to improve seed vitality, thereby increasing the rate of germination (Ciacka et al., 2020). Nitrogen can contribute to improved vegetation restoration and soil fertility (Samudro and Mangkoedihardjo, 2020; Wang et al., 2010, 2011). The objectives of this literature review were to determine the origin of Nr generated from landfills, to analyze the effect of leachates containing Nr, and to analyze the reactive performance of nitrogen in landfill leachate by plants in CW.

SOURCE OF REACTIVE NITROGEN

The natural sources Nr include, for example, the process of nitrification while anthropogenic sources, comprise, for example, the application of nitrogen fertilizers and the burning of fossil fuels (Wang et al., 2019). Reactive nitrogen is an essential nutrient for important crops. Primarily, it can fertilize agricultural areas; however, this can cause environmental pollution (Bodirsky et al., 2014; Callesen et al., 2011; Sutton et al., 2011). The Haber-Bosch process can determine the amount of Nr produced. In 2010, the Nr value was twice that of natural sources with 120 $TgNyr^{-1}$ (Fowler et al., 2013; Raza et al., 2018).

In order to reduce agricultural Nr pollution, mitigation measures including fertilizer, livestock management, waste recycling, and community consumption, are implemented. However, these measures have not been assessed quantitatively as to how much they reduce the Nr pollution on a global scale. The previous analyses of the global agricultural nitrogen cycle focused largely on the estimates of current and past Nr flows, references to future Nr flows, and nitrogen fixation (X. Liu et al., 2013; McCabe and Arnold, 2016; Szporak-Wasilewska et al., 2015; Zhang et al., 2017). The mitigation program for Nr production has not yet been able to contribute to reducing Nr (Conley et al., 2009; Cui et al., 2016; Li et al., 2008; McCabe and Arnold, 2016; Rice et al., 2017; Rousseau et al., 2004).

The increase in agricultural yields is based on the presence of N in the soil (Massignam et al., 2009). The wise use of N ensures the largest yields of better quality (Leghari et al., 2016). The Nr from human activities eventually enters the landfill and produces leachate. The Nr value of the leachate landfill can be seen in Table 1.

Landfill leachate is defined as liquid waste resulting from the percolation of rainwater through solid waste discharged into landfills, as well as moisture present in waste and waste degradation products (Costa et al., 2019; Arliyani et al., 2021). The amount of leachate produced is mainly

Table 1. The Nr value of the leachate landfill

Cimate	Country	Landfill	Landfill age	Parameter	High concentration	Unit	Reference
Tropical	Malaysia	Jeram	Old (> 5 Years)	TN	0.75	mg/L	(Moktar and Mohd Tajuddin, 2019)
Tropical	Singapura	Lorong Halus	Old (> 5 Years)	NH_4-N	5.6–75.1	mg/L	(Sim et al., 2013)
Tropical	Indonesia	Sambutan, Samarinda	Medium (1–5 Years)	NH_3-N	6.83	mg/L	(Kahar, 2017)
				Nitrite	1.85	mg/L	
				Nitrate	22.13	mg/L	
Sub-tropical	Turki	Mamak, Ankara	Old (> 5 Years)	NH_4-N	166.07	mg/L	(Yalçuk and Ugurlu, 2020)
				NO_3-N	3.8	mg/L	
Sub-tropical	Argentina	Villa Domínico Buenos Aires	Old (> 5 Years)	NH_4-N	2,484	mg/L	(Silvestrini et al., 2019)
				NO_3-N	12.5	mg/L	
				NO_2-N	2,497	mg/L	
Cold	Canada	Brady, Winnipeg	Old (> 5 Years)	NH_4-N	800–1000	mg/L	(Xu et al., 2019)

influenced by rainfall, evapotranspiration, surface runoff, groundwater infiltration, and the level of compaction in the landfill (Miao et al., 2019). Various techniques are used to control the ingress of water into the landfill, including the installation of watertight coatings and covers to minimize leachate (Dajić et al., 2016).

REACTIVE NITROGEN EFFECTS

The anthropogenic intervention into the biogeochemical cycles of nitrogen has substantially been increasing the global levels of Nr, thus causing damage to the environment on a local, regional and global level. Such increases could exacerbate the impact on climate balance either through changes in atmospheric constituents, or feedback in terrestrial ecosystems. Reactive nitrogen has an indirect or direct effect on the GHG sources and removers and on climate (Erisman et al., 2011). The direct effects of Nr on climate include:

a) Formation of N_2O, which is a potent greenhouse gas produced by industrial processes, combustion, or microbial conversion of nitrogen-containing substrates (Davidson, 2009),
b) O_3 formation on ground surface from NO_x. O_3 can cause immediate health damage such as cancer and asthma (Sutton et al., 2011; Sutton MA et al., 2013).

The most important indirect relationships between climate and Nr include:

a) Excessive Nr deposition which can increase or decrease ecosystem productivity as well as C sequestration (Diaz and Rosenberg, 2008),
b) Production CH_4. Deposition of Nr to CW which can trigger vascular crop production, thereby increasing the supply of C substrate to the system and triggering the CH_4 production in the absence of sufficient oxygen.

The absorption of Nr in the agricultural sector is low so that the excess Nr from that sector causes Nr pollution (De Vries et al., 2013; Rockström et al., 2013). Reactive nitrogen can be converted into a non-reactive form (N_2) by a denitrification process. The form of Nr which is released from the environment by leaching or evaporation can create various environmental effects (Sutton et al., 2011). The productivity of aquatic ecosystems is one of the effects of the Nr presence in the ecosystem (Breitburg et al., 2009). Global gross domestic product is estimated to be damaged by 0.3–3% in monetary terms (Bodirsky et al., 2014).

Reactive nitrogen is highly soluble in water, so that if it is released into the environment it can contaminate surface water and groundwater. As a result, the Nr that enters the surface of the water will flow and affect the aquatic ecosystem. The presence of Nr in aquatic ecosystems encourages algal growth. Algal blooms produce eutrophication that blocks sunlight from entering the water. The absence of sunlight entering the waters will disrupt the productivity of the aquatic ecosystem. The drinking water treatment process can also produce Nr. The content of Nr in drinking water can cause methemoglobinemia. Nitrates in the body are converted into nitrites in the digestive system by binding to oxygen which can irritate the respiratory system (Holmes et al., 2019).

MECHANISM AND PERFORMANCE NR FROM LANDFILL LEACHATE IN CW

In CWs, the mechanism of ammonia nitrogen utilization is caused by adsorption, nitrification, plant uptake and volatilization (Cui et al., 2016; Gupta et al., 2016; Marsili-Libelli, 2010; Spokas et al., 2012; Taghizadeh-Toosi et al., 2012). The mechanisms for utilizing Nr in CW include NH_3 evaporation, microbial uptake, ammonification nitrification, denitrification, nitrogen fixation, plant uptake, fragmentation, absorption (Soares et al., 2021), desorption, washing, and burial (Vymazal, 2007). The main process of converting Nr to N which is not reactive is nitrification-denitrification (Dong and Sun, 2007; Kadlec and Wallace, 2009; Noor et al., 2010).

Urea is the dominant form of nitrogen (N) fertilizer used globally (Souza et al., 2019). The presence of Nr has been shown to be an important nutrient input from the external environment to ecosystems (Wang et al., 2019). The performance of Nr in biochar used in CW media can increase the soil microbial activity (Cayuela et al., 2013; Mangkoedihardjo & Triastuti, 2011). The use of Nr on CW through the denitrification process can reach 82.8–92.08% (Gupta et al., 2016). NH_3 which is adsorbed by biochar on the medium in CW can provide nutritional needs for plants (Taghizadeh-Toosi et al., 2012). The biochar used at CW showed a reduction in ammonia 58.3–50.01% at CW. The growth of rhizobacteria communities in CW was supported by NH_3 which was adsorbed by biochar in the pores (Verhamme et al., 2011).

Landfill leachate contains various pollutants (Tangahu et al., 2021). The removal of BOD, ammonium, and nitrate is dependent on the oxygen concentration affected by the plants' transport and consumption of nutrients in the constructed wetland (Fuchs, 2011). The study by (Lavrnić et al., 2018) used a flow meter system on the constructed wetland on influent and effluent calculated by Kadlec and Wallace, (2009):

$$Q_{\mathrm{inf}} + (P \times A) - Q_{\mathrm{eff}} - I - (ET \times A) = dV/dt \quad (1)$$

where: $Qinf$ = Influent discharge (m^3/s),
P = Precipitation rate (m/s),
A = Upper surface area of the constructed wetland (m^2),
$Qeff$ = Effluent discharge (m^3/s),
I = Infiltration rate (m^3/s),
ET = Evaporation rate (m/s),
V = Water volume in the constructed wetland (m^3),
t = time (s).

Changes in time (Δt), changes in water volume (ΔV) can be neglected (Kadlec and Wallace, 2009; Lavrnić et al., 2018). The measurement of infiltration and evapotranspiration rates cannot be carried out separately, so that the water balance for hydrological flow equilibrium is calculated as:

$$Q_{\mathrm{inf}} + (P \times A) - Q_{\mathrm{eff}} = I + (ET \times A) \quad (2)$$

In equation (2) ($I + (ET \times A)$) as overall water loss in the constructed wetland (Lavrnić et al., 2018). The effect of capacity and discharge vary depending on time differences (Johannesson et al., 2017; Reinhardt et al., 2005). Water is stored in the constructed wetland system and is partially lost due to infiltration and evapotranspiration (Lavrnić et al., 2018). The influent will enter the constructed wetland system; microbes in the soil convert organic nitrogen into ammonium, which is then available for adsorption and absorption by plants to be reduced to nitrate (Samudro & Mangkoedihardjo, 2020). The available of nitrate to plants is reduced to nitrogen gas (N_2). The input of oxygen from root transport and gas is vital for the BOD and ammonium removal. Anoxic conditions are needed to reduce nitrates. The carbon source for denitrification in Carbon BOD (CBOD) is needed to increase the BOD and nitrogen removal (Fuchs, 2011). The mechanism and performance Nr in CW can be seen in Figure 1.

The process of using Nr is 3549 g with an average load of 36 g TKN/m^2/d in CW which is adsorbed by 32%, the nitrification-denitrification process is 59%, and the rest is released into the environment. During the rest period, the amount of adsorbed ammonium is drastically reduced to 186 g (5%) and converted to nitrate, increasing the mass of nitrified nitrogen to 2,543 g (72%) ammonium and involving high concentrations of nitrate in the waste (Morvannou et al., 2014). The performance of Nr in plants in CW can be seen in Table 2.

The form of Nr that is absorbed by plants at CW is different. For example, the NH_4 preference is common in macrophytes that live in limited nitrifying environments, where NH_4 is abundant (Garnett et al., 2001). The rate of absorption and storage of nutrients by plants depends on the concentration of nutrients in their tissues. The desirable traits of a plant used for storage and nutrient assimilation include rapid growth, the ability to obtain an upright plant, and high tissue nutrient content. Conversely, plants that have large

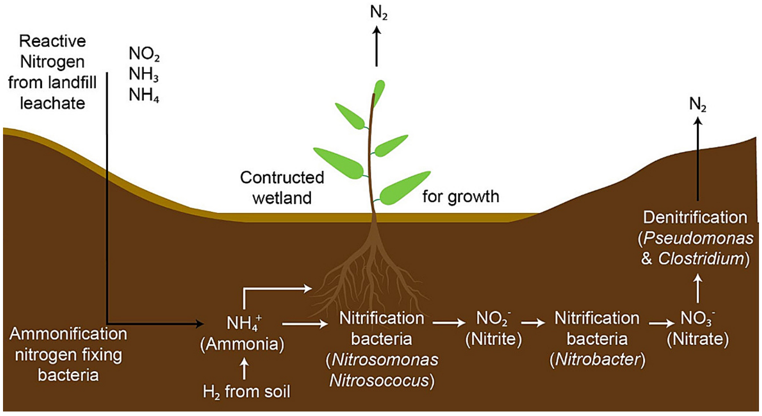

Figure 1. The mechanism and performance Nr in CW (Modified [Lee et al., 2009])

Table 2. The performance of Nr in plants in CW

No.	Parameters	Performance	Refrences
1.	NO_3	Chloride (Cl^-) and nitrate (NO^-) ions play a capable role in osmoregulation can have a positive effect on plant organs (Dorais et al., 2010)	(Anjana et al., 2007)
2.	NO_2	Plants exposed to soil nitrogen with NO_2 gas can increase nutrient uptake, photosynthesis, and nutrient metabolism so that shoot biomass, total leaf area, and content of C, N, P, K, Ca, Mg, per shoot, and S (or Fe), free amino acids and crude protein were roughly double that of the control plants.	(Takahashi and Morikawa, 2014)
3.	NH_3	The uptake of ammonia and nitrate by macrofites changes the form of inorganic nitrogen to organic compounds, as building blocks for cells and tissues (Lewin, 1999).	(Lee et al., 2009)
4.	NH_4	All plants utilize Nr in the form of NO_3 and NH_4. These are the most important element for proper growth and development of a plant which significantly increases and enhances its yield and quality by playing an important role in the biochemical and physiological functions of plants.	(Leghari et al., 2016)

biomass accumulations during fall and winter can release a lot of nitrogen accumulated back into the water during winter (Lee et al., 2009; Vymazal, 2007).

Reactive nitrogen is a molecule that regulates various physiological processes in plants, including germination, seed formation, dormancy, and maturation. Aging causes a decline in seed quality, which limits not only agricultural production, but also the preservation of global biodiversity. NO and other compounds belonging to the Nr family appear to reduce the negative effects of seed aging (Ciacka et al., 2020). Nitrogen occupies a prominent place in the metabolic system of plants. All important processes in plants are related to protein, of which nitrogen is an essential constituent. As a result, to achieve greater crop production, nitrogen application is indispensable and unavoidable (Massignam et al., 2009). Nitrogen plays a key role in agriculture by not only increasing yields, but also improves the food quality (Ullah et al., 2010). The optimal amount of N can increase the photosynthesis process, the production of leaf area, the duration of the leaf area and the net assimilation rate (Ahmad et al., 2009). Maximum leaf area and total plant leaf biomass are determinants of higher yields (Rafiq et al., 2010). All crops, including cereals, vegetable oils, fiber, and sugar and horticulture, require a balanced amount of nitrogen for a vigorous plant growth and development processes (Leghari et al., 2016).

CONCLUSIONS

Human activities can produce by-products, some of which can be harmful to the environment and humans themselves. Activities such as industry, agriculture, transportation, and other daily activities produce waste. The waste that cannot be reused is placed in landfills, some of which are separated, such as landfills for organic, non-organic and medical waste. Some landfill sites are not separated, so that they contain many hazardous materials, one of which is the presence of

Nr. The Nr of landfill leachate found in the literature review was NH_4, NH_3, NO_2, and NO_3. The Nr content in landfill leachate which is directly removed to the environment will cause negative effects. However, Nr is needed by plants as nutrition. Therefore, it was found that Nr had a good performance when used in CW for the growth of plant metabolism and photosynthesis, thereby releasing a non-reactive form of N. N which is not reactive in the form of N_2 can be accepted by the environment. Besides changing Nr to be unreactive, CW can also change other hazardous content in the leachate landfill.

Acknowledgements

This research receives the financial support from the Ministry of Research and Higher Education of the Republic of Indonesia with Master leading to Ph.D. scholarship (PMDSU).

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