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Infiltration Mechanism Controls Nitrification and Denitrification Processes under Dairy Waste Lagoon

S. Baram,* S. Arnon, Z. Ronen, D. Kurtzman, and O. Dahan

Earthen waste lagoons are commonly used to store liquid wastes from concentrated animal feeding operations. The fate of ammonium (NH_4^+) and nitrate (NO_3^-) was studied in the vadose zone below earthen-clay dairy farm waste lagoons using three independent vadose zone monitoring systems. The vadose zone was monitored from 0.5 to 30 m below land surface through direct sampling of the sediment porewater and continuous measurement of the sediment profile's water content variations. Four years of monitoring revealed that wastewater infiltration from the lagoon is controlled by two mechanisms: slow (mm d^{-1}), constant infiltration from the lagoon bed; and rapid (m h^{-1}) infiltration of wastewater and rainwater via preferential flow in desiccation cracks formed in the unsaturated clay sediment surrounding the lagoon banks. The preferential flow mechanism is active mainly during wastewater-level fluctuations and intensive rain events. The vadose zone below the waste sources remained unsaturated throughout the monitoring period, and all infiltrating NH_4^+ was oxidized in the upper 0.5 m. The NH_4^+ oxidation (nitrification) was coupled with NO_3^- reduction (denitrification) and depended on the sediment water content, which was controlled by the infiltration mechanism. Coupled nitrification–denitrification (CND) resulted in 90 to 100% reduction in the total nitrogen mass in the vadose zone, with higher removal under high water content (~0.55 m^3 m^{-3}). Mass balance of nitrogen and isotopic composition of NO_3^- indicated that CND, rather than cation exchange capacity, is the key factor regulating nitrogen's fate in the vadose zone underlying earthen waste lagoons.

Earthen waste lagoons are commonly used to store liquid wastes from concentrated animal feeding operations (CAFOs) such as dairy farms. The liquid waste stored in the lagoons contains high concentrations of nitrogen (N), phosphorus (P), salts, organic compounds, and other nutrients. For economic reasons, most dairy farms store the liquid waste on-site in earthen (soil-lined or unlined) lagoons rather than in plastic-lined lagoons, steel tanks, or concrete tanks (Ham and DeSutter, 2000). However, this practice fails to prevent the substantial leaching of organic-nitrogen (organic-N) and ammonium nitrogen (NH_4^+–N) into the subsurface (Gooddy et al., 1998, 2002; Ham and DeSutter, 2000; Ham, 2002; Harter et al., 2002; DeSutter et al., 2005).

To minimize downward or lateral seepage of the wastewater from earthen waste lagoons and prevent groundwater contamination, it is recommended that the saturated hydraulic conductivity coefficient (K_{sat}) of the earthen lining would be less than a prescribed level ($K_{sat} < 1 \times 10^{-6}$ cm s^{-1}, the level in many U.S. states) (SCS, 1997). Hence, during the construction of the waste lagoon, clay is typically mixed with local sediment and compacted to form an earth liner along its bottom and banks. Following the introduction of wastewater into the lagoon, the hydraulic conductivity of the earth lining will most likely be reduced by at least an order of magnitude due to physical, chemical, and biological processes, commonly termed *seal formation* (SCS, 1997; Cihan et al., 2006; Tyner et al., 2006).

Laboratory-scale experiments on dairy waste infiltration into clay, loam, and sand sediments have shown that all sediment types have similar infiltration fluxes (4.6 to 6.9 × 10^{-7} cm s^{-1}) within 10 d of manure application (Culley and Phillips, 1982). The effect of the hydraulic parameters of the organic seal and the underlying sediment on the infiltration rate from animal waste lagoons was analyzed by Tyner and Lee (2004) using a steady-state two-layer model. Their model predicted that sediment thickness has no effect on the infiltration rate, and sensitivity analysis predicted that infiltration rate is highly dependent on seal hydraulic conductivity and minimally dependent on

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Abbreviations: BLS, below land surface; CAFO, concentrated animal feeding operation; CEC, cation exchange capacity; CND, coupled nitrification–denitrification; FTDR, flexible time domain reflectometry; USS, ultraviolet spectrophotometric screening; VMS, vadose zone monitoring system; VSP, vadose zone sampling port.

sediment hydraulic conductivity. Cihan et al. (2006) developed a model for describing the sealing process in dairy and swine waste lagoons with time. After a stable seal develops, the model predicts that the seal properties, and not the sediment properties, are responsible for limiting infiltration. Furthermore, Tyner et al. (2006) showed, in a laboratory experiment, that within 85 d of manure application to undisturbed columns of silt loam, the sediment profile becomes unsaturated. They indicated that unsaturated conditions appeared immediately below the sediment–liquid interface. "In situ" field measurements showed that seepage rates from earthen lagoons are typically on the order of several millimeters per day or less (Miller et al., 1985; Parker et al., 1999c; Ham and Baum, 2009). These seepage rates were observed in various sediment types (sand to clay) and validated the findings of Cihan et al. (2006) and Tyner et al. (2006) that the hydraulic conductivity of the seal, rather than that of the sediment, controls the infiltration rate. While the potential seepage loss from the bottom of waste lagoons has been well studied, that from the lagoon banks, which are subjected to wetting/drying cycles during frequent water-level fluctuations, warrants further research (Parker et al., 1999c).

Clayey earth lining is very attractive for use in waste lagoons due to its low hydraulic conductivity. However, clay sediments are highly sensitive to water content and tend to form desiccation cracks under unsaturated conditions (Chertkov and Ravina, 1998; Parker et al., 2001). These desiccation cracks serve as preferential flow paths for water and contaminants. Quick rises in sediment water content following rain events and temporal wastewater overflows were observed across a clay vadose zone exceeding 10-m depth (Baram et al., 2012). The infiltration velocities calculated from these wetting events (0.4 to 23.6 m h^{-1}) indicated that the desiccation-crack network in the unsaturated clay sediment remains open and serves as a preferential flow pathway year-round, even at high sediment water contents (~0.50 m^3 m^{-3}). Kurtzman and Scanlon (2011) concluded from the chemical composition of the vadose zone sediment and groundwater that preferential flow is the major mechanism dominating the recharge processes in the noncultivated vertisol of the region in which this research was performed.

The main concern with seepage from earthen waste lagoons is the risk to groundwater quality. Organic-N and NH_4^+ are the dominant N species in CAFO anaerobic waste lagoons, whereas in their subsurface, nitrate (NO_3^-) is the most common contaminant. High NO_3^- levels in drinking water have been associated with risks for methemoglobinemia (blue baby syndrome) in infants and diarrheal and respiratory diseases, and have even been suggested to be a risk factor for specific cancers (Ward et al., 2005). Accordingly, many countries and organizations around the world have provided guideline values for maximum NO_3^- levels in drinking water (for example: World Health Organization—50 mg L^{-1}, United States—45 mg L^{-1}, Israel—70 mg L^{-1} [Ward et al., 2005; WHO, 2007; Israel Internal Affairs and Environment Committee, 2010]). Microbial oxidation of ammonia (NH_3) and NH_4^+ into NO_3^- (nitrification) in porous medium requires the presence of molecular oxygen (O_2) (Prosser, 1989). Nitrate can be further reduced by microorganisms into N gas (N_2) through denitrification. In sediments where favorable conditions for both nitrification and denitrification are present in neighboring microhabitats, nitrite (NO_2^-) or NO_3^- produced during nitrification can be utilized by denitrifiers. This process is termed *coupled nitrification–denitrification* (CND). Coupled nitrification–denitrification plays an important role in the removal of nitrogenous compounds in sediments (Nielsen and Revsbech, 1998; Kremen et al., 2005).

Studies on the fate of N species in the vadose zone and groundwater in dairy earthen waste lagoon environments have shown that their fate varies from place to place. Monitoring of NO_3^- concentration in the porewater of the vadose zone under and around earthen lagoons constructed in sand to clay loam sediments demonstrated that during lagoon operation, in some cases NO_3^- concentrations remained similar to background concentrations (Meyer et al., 1972; Parker et al., 1999c), and in others they reach very high concentrations (>900 mg L^{-1}) (Oliver and Meyer, 1974; Korom and Jeppson, 1994). Groundwater monitoring for NO_3^- contamination from earthen lagoons has demonstrated that the impact of the lagoon varies with time from its initial operation (Sewell, 1978). While in some studies, elevated NO_3^- concentrations in groundwater were attributed to lagoon seepage (Nordstedt et al., 1971; Withers et al., 1998), in others, elevated NO_3^- could not be distinguished against the background levels from manure application in surrounding fields (Harter et al., 2002). A recent study has even shown that the NO_3^- concentration in the saturated zone varies with depth due to denitrification processes in the groundwater column (Singleton et al., 2007).

The objective of this research was to study the fate of NO_3^- in the clay vadose zone underlying dairy earthen waste lagoons and their margins, by implementing a unique array of vadose zone monitoring systems (VMSs). More specifically, this study evaluates the impact of infiltration mechanism and sediment water content on NH_4^+ oxidation and denitrification in the subsurface.

Materials and Methods

Study Area

The study area was located in the Beer Tuvia region (40 km^2), above the southern part of the Coastal Aquifer in Israel. The phreatic aquifer in the area is composed of calcareous sandstone (Kurkar Group) overlaid by Pleistocene-age clays (Issar, 1968; Weinberger, 2007). The land surface area is covered by a clay layer, dominated by smectite minerals, with a typical thickness spanning 3 to 12 m. The climate is characterized as Mediterranean with average summer and winter temperatures of 24.3°C and 14.2°C, respectively. The average annual precipitation is ~450 mm, falling during the winter season (November to March) mostly in five to eight rainy episodes. The main recharge to the aquifer is related to percolation of seasonal rainwater and agricultural return flow from irrigated and rain-fed fields that have been intensively cultivated in the area for >60 yr. Over the past four decades, the quality of the groundwater in the Beer Tuvia region has been continuously deteriorating, as reflected by a gradual increase in chloride concentrations (>600 mg L^{-1}) and local spots of high NO_3^- concentration (Weinberger, 2007).

The Beer Tuvia region currently hosts ~10,000 cows in 140 dairy farms. The dairy farm selected for this study has 60 dairy cows and 30 heifers and calves, which is representative of the region. Wastewater and liquid manure from the farm (7 m^3 d^{-1}) is constantly discharged into a single-stage earthen unlined lagoon (also commonly termed a *waste storage pond*) without treatment and remains untreated. The lagoon is roughly 20 m long, 10 m wide, and 1 m deep. Wastewater overflow is constantly drained into a waste channel roughly 200 m long, 2 m wide, and 1 m deep. The dairy farm has been operating in the same way for the past 50 yr and no specific maintenance procedures, such as solids removal, have been used at the site. Both the waste lagoon and the channel are flooded with liquid manure year-round. Natural annual shallow-rooted plants (mainly *Malva sylvestris* L.) grow on the waste source banks from November to April.

The vadose zone underlying the waste lagoon and drainage channel is composed of clay sediments extending from land surface to depths of 6 m and 12 m, respectively. The clay overlies sand and calcareous sandstone down to the water table at ~46 m below land surface (BLS). Particle size distribution of the clay sediment in the area is composed of 26% sand, 22% silt, and 52% clay, 90% of which consists of illite–smectite minerals. Desiccation cracks form regularly in the clay sediment and create polygons with average dimensions of 1.0 × 0.70 m^2, separated by cracks with an average aperture of 0.055 ± 0.017 m. The cracks extend to a depth of 0.65 ± 0.19 m (cracks are defined when the aperture is >0.006 m). The desiccation-crack network remains active year-round, even after rain events and under high water contents (>0.50 m^3 m^{-3}). A detailed discussion of the role of the desiccation cracks in the water-infiltration process through the clay sediment at the studied site can be found in Baram et al. (2012).

Monitoring Approach and Field Instrumentation

The hydraulic and chemical properties of the sediment in the vadose zone underlying the waste lagoon and its margins were evaluated by VMS. The VMS is designed to collect continuous real-time measurements of the sediment water content, and to allow frequent sampling of the sediment porewater across the vadose zone (Dahan et al., 2007, 2008; Rimon et al., 2007, 2011). These latter publications present a detailed description of the monitoring system and its installation procedure. Therefore, in this article, only a brief description is provided of the monitoring concept and the technical aspects that were important for evaluating the presented data. The VMS is composed of a flexible sleeve made of a thin, flexible liner hosting a set of flexible time domain reflectometry (FTDR) probes and vadose zone sampling ports (VSPs). The VMS is installed in a 140-mm-diameter, uncased, slanted borehole (30–35° from the vertical). During the installation, the sleeve is inserted into the borehole and filled with two-component high-density liquid urethane (~1.65 kg L^{-1}) that solidifies shortly after its application. The hydrostatic pressure generated by the liquid urethane inside the sleeve expands the sleeve and seals the borehole void to prevent generation of preferential pathways along the borehole axis, while ensuring proper attachment of the probes to the sediment facing the upper side walls of the borehole. Assuming that the general flow direction in the vadose zone is rather vertical, each point on the upper side of a slanted borehole faces an undisturbed sediment column (Fig. 1).

The volumetric water content of the sediment was measured using the FTDR probes operated with a TDR100 instrument (Campbell Scientific, Logan, UT). Time domain reflectometry measurements were calibrated by accounting for the specific structure of the FTDR probe, instrumentation setup, variety of soil types (including the clay at the site), and salinities (Baram et al., 2012). The overall accuracy of water content measurements under field conditions using the VMS is ±5% of the measured value (Rimon et al., 2011).

The sediment porewater from the unsaturated zone was sampled using VSPs. The VSPs are essentially suction lysimeters that have been modified for operation with the VMS in deep sections of the vadose zone. Porewater sampling from unsaturated sediments is achieved by creating hydraulic continuity between the sediment and the sampling cell using a flexible porous interface (Dahan, 2005, 2008, 2009a,b; Dahan et al., 2009).

The vadose zone at the study site was monitored using three independent VMSs that were installed under the waste lagoon and its drainage channel. One VMS with five monitoring units was installed in a 35-m-long borehole extending from the lagoon margins (1.5 m from the lagoon bank) toward the center of the waste lagoon to a vertical depth of 30.5 m (I in Fig. 1a and Table 1). Two additional VMSs (II and III in Fig. 1b), with seven monitoring units each, were installed ~150 m downstream of the waste lagoon in 24-m-long boreholes under the center of the waste channel and 2 m away from the channel bank to a vertical depth of 21.3 m (Table 1). In addition to the monitoring units

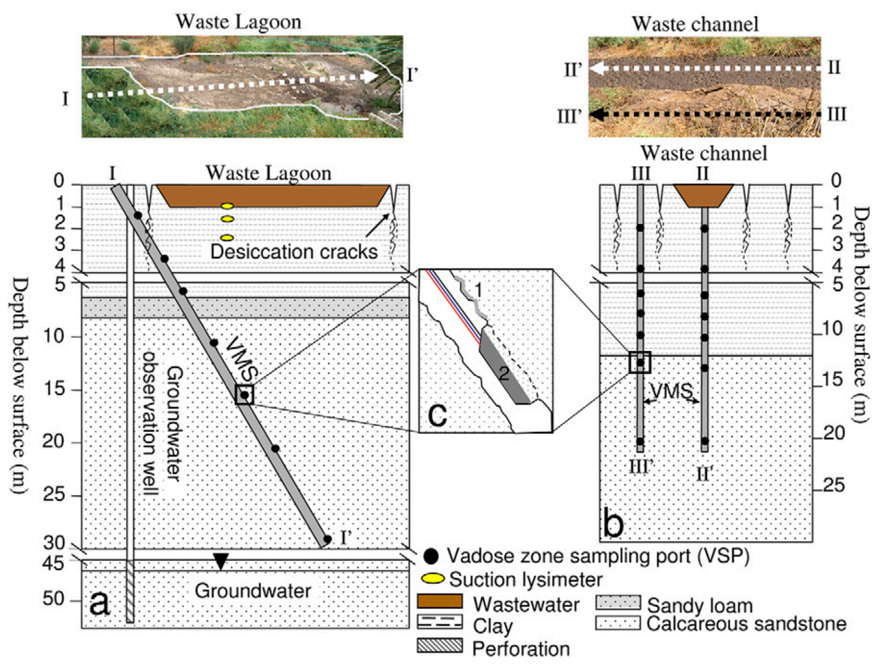

Fig. 1. Schematic illustration of the vadose zone monitoring systems (VMS) and observation well installed in the sediment profile under (a) the waste lagoon and its margins, and (b) the waste channel and its margins. The monitoring units (c) include (1) a flexible time domain reflectometry (FTDR) probe, and (2) a vadose zone porewater sampling port (VSP). (a) shows a side view of the waste lagoon while (b) shows a front view of the waste channel, and thus VMSs II and III appear as vertical lines in (b) although they were installed in slanted boreholes.

that were installed via the VMS, three conventional suction lysimeters (0.06 m long, 0.02 m in diameter; A.M.I., Ashdod, Israel) were installed in the sediments underlying the central part of the waste lagoon (0.5 and 1.4 m below the lagoon bottom), and at the interface between the bottom of the lagoon and the underlying sediments (Fig. 1a and Table 1). The suction lysimeters were installed in 0.05-m holes that were backfilled with a few centimeters of fine quartz sand (<75 μm) and sealed to the surface with bentonite clay. A groundwater observation well was drilled a few meters away from the lagoon bank to a depth of 54 m with a screen interval from the water table at 46 m BLS to the bottom of the well.

The volumetric water content of the sediment in the vadose zone was measured every 60 min during the dry season (April–October) and every 15 min during the wet season (November–March). Water samples from the lagoon, vadose zone, and groundwater were collected every 6 to 12 wk. The wastewater samples were collected using suction lysimeters. Groundwater was sampled from the observation well using a submersible pump (Model MP1, Grundfos, Denmark). All water samples were stored in polypropylene bottles and kept on ice until they reached the laboratory (<12 h), where they were filtered (45-μm glass fiber filter) and kept at 4°C until analysis (<2 wk).

Table 1. The monitoring setup.

Monitoring unit depth (m)†		Sediment type	No. of porewater samples
VSP‡	FTDR		
0		Lagoon sludge	14
Vadose zone underlying the waste lagoon			
1.5		Clay	16
2.4		Clay	14
6.5	6	Clay	15
10.5	10	Calcareous sandstone	8
15.5	15	Calcareous sandstone	18
20.5	20	Calcareous sandstone	
30.5		Calcareous sandstone	15
Vadose zone underlying the waste lagoon margins			
2.5	2	Clay	14
3.5	3	Clay	14
Vadose zone underlying the waste channel			
2.4	1.7	Clay	8
4.5	3.9	Clay	9
6.5		Clay	8
8.7	8.1	Clay	7
10.7	10.2	Clay	8
14.0	13.5	Calcareous sandstone	3
21.3	19.8	Calcareous sandstone	2
Vadose zone 2 m away from the overflow waste channel bank			
2.3	1.6	Clay	4
4.3	3.7	Clay	7
6.5	6.0	Clay	8
8.4	7.9	Clay	4
10.5	10.0	Clay	4
13.8	13.3	Calcareous sandstone	4
21.1	19.8	Calcareous sandstone	
Groundwater			12

† Vertical depth measured relative to land surface at each site.

‡ VSP, vadose zone sampling port; FTDR, flexible time domain reflectometry probe.

Chemical Analysis

Concentrations of N species (NO_3^-–N, NO_2^-–N, NH_4^+–N, and total N) were determined within 48 h after sampling. The NO_3^-–N concentration was measured using two methods: (i) ultraviolet spectrophotometric screening (USS) (APHA 4500-NO3 B), and (ii) ion chromatography (Dionex-4500i, Sunnyvale, CA). Ion chromatography was also used to measure NO_2^-–N, and the phenate method was used for NH_4^+–N (APHA 4500-NH3 F). Total N was determined using the persulfate digestion method (APHA 4500-N C) (APHA, 1998). Accuracy of the measured concentrations was validated using 10% duplicates and spike recovery tests. Matrix interference was not observed in the USS method, and high recovery (91–98%) was observed in all cases. During the first 2 yr (2007 and 2008), the USS method was used to determine the NO_3^-–N concentration; later, ion chromatography was used.

$\delta^{18}O$ and $\delta^{15}N$ in NO_3^- of porewater samples from the vadose zone were measured at the Laboratory of the Israel Geological Survey, using an isotope ratio mass spectrometer (Delta Plus XP Thermo Scientific, Braunschweig, Germany), after chemical conversion of NO_3^- to nitrous oxide (McIlvin and Altabet, 2005). Values are given as mean ± standard deviation, with total number of samples (*n*) in parentheses.

Infiltration Rates

Conductance ($K/\Delta x$) of the waste lagoon bed and waste channel bed was measured "in situ" using pipe infiltrometers that were hammered into the top ~15 cm of the clay soil (where K [L t^{-1}] is the bulk hydraulic conductivity of the unknown thickness Δx [L] of the soil through which infiltration takes place, under positive pressures, before reaching the point of drainage in unsaturated conditions [L]). The infiltrometers (2 and 1 m long with diameters of 0.35 and 0.2 m, respectively) were filled with wastewater filtered to remove coarse floating particles (using sieve ASTM No. 40; Ari. J. Levi, Petach Tikva, Israel) and covered at the top to eliminate water loss due to evaporation. The flux was evaluated by measuring the drop-in water level inside the infiltrometer at nine different locations around the lagoon and channel. Infiltration was measured under a range of hydraulic heads between 0.1 m below and 1.2 m above the wastewater level. In each experiment, the water level was allowed to drop by up to 5% of the initial level to measure infiltration under relatively constant water head.

Sediment Sampling

Detailed sampling of the water content and organic matter content in the upper 0.1 m of the sediment under the waste lagoon and channel was performed by exposing small sections of the lagoon and channel bottom in the following manner. A metal cylinder (0.9 m long and 0.6 m in diameter) was first pushed through the wastewater into the waste lagoon and channel bottom. Then, before sampling, the sediment, wastewater, and 0.1 to 0.2 m of fine organic matter that had settled on the bottom were carefully removed from the cylinder, exposing the bottom. Sediment cores were then collected from the exposed surface using plastic cylinders (0.11 m long and 0.028 m in diameter) and kept on ice until they were brought to the laboratory. The core samples were immediately dissected to 0.005-m-thick slices that were analyzed for water content (oven-dried at 105°C for 72 h) and total organic matter content through combustion (450°C for 4 h) (Nelson and Sommers, 1996). Altogether, six different locations were sampled under the center of the channel and the center of the lagoon (three each).

Sampling of the water content and NH_4^+–N distribution in the upper 0.5 m of the sediment under the waste lagoon was performed once a year from three random locations by hammering a metal pipe (0.05 m in diameter and 0.5 m long) into the lagoon bed. Two cores were taken from each location. Sections of 0.1 m from one core were used to measure the volumetric water content of the sediment, while 0.1-m sections from the other core were extracted for NH_4^+–N using 2 M KCl (Maynard et al., 2008).

Saturation of the clay sediment (θ_{sat}) was evaluated using 10 undisturbed core samples (0.02 m long and 0.01 m in diameter) taken from 10 to 20 cm BLS. The samples were oven-dried (at 105°C for 72 h), flushed with CO_2 gas (5 min), and then placed in degassed water. After 10 h the samples were weighed and their volume was remeasured to obtain their volumetric and gravimetric saturated water contents.

Table 2. Seepage rates under dairy and cattle waste lagoons.

Reference	Sediment type	Seepage rate: Range	Seepage rate: Avg.
		—— mm d^{-1} ——	
Full-scale seepage studies			
Meyer et al. (1972)	Sand–clay loam	NA†	~1
Parker et al. (1999c) and references therein	Sandy loam–clay loam	0.26–0.87	0.48
Ham (2002)	18–30% clay	0.2–2.4	1.1
This study	Clay	0.64–7.6	2.4
Small-scale laboratory studies			
Parker et al. (1999c) and references therein	Sand–clay	0.24–36	3.12
Cihan et al. (2006)	Sand–clay	NA	~1
Tyner et al. (2006)	Silt loam	NA	0.7

† NA, data not available.

Results and Discussion

While measurements of the infiltration pattern from the lagoon and channel bottom enabled assessment of water losses through the sediment matrix, changes in sediment water content enabled detection of water losses following preferential flow from the lagoon and channel banks. Combining results of both methods showed that infiltration of polluted wastewater from the dairy farms to the subsurface can occur via four major regions: (i) the area under the waste lagoon bottom, which is permanently flooded; (ii) the area along the waste lagoon banks, which is subjected to fluctuations in wastewater level; (iii) the area under the waste channel, which is permanently flooded; and (iv) the area underlying the waste channel margins, which is subjected to occasional flooding by wastewater overflows and rain events. Monitoring of the waste channel area, a narrow waste source proximal to two hydraulically active banks, represents the process occurring at the lagoon banks, while monitoring of the lagoon represents the whole lagoon area including the area far from the banks (Fig. 1).

Infiltration from the Bottom of the Waste Sources and Seal Formation

Average infiltration fluxes from the waste lagoon and channel bottom, estimated from the pipe infiltrometers, was 2.4 mm d^{-1} (n = 20) (Table 2) with an average conductance value of 4.4 × 10^{-8} s^{-1}. No significant differences were observed between the infiltration fluxes measured in the lagoon and channel (p = 0.22). The infiltration rates measured in this study were very similar to measurements conducted previously in earthen lagoons and in laboratory studies with various sediment types (sand to clay) (Table 2). The similarities between the infiltration rates through different types of sediment further support conclusions of Tyner et al. (2006) and Cihan et al. (2006) that the hydraulic conductivity of the organic seal rather than the hydraulic conductivity of the underlying sediment matrix controls the infiltration rate from the bottom of waste lagoons.

The organic matter content and the water content in the clay sediment below the lagoon and channel rapidly decreased from the surface down to a depth of 2 cm, while they remained relatively constant below 2-cm depth (Fig. 2). The water content in the upper 1.5 cm resembled the laboratory-measured water contents at saturation (θ_{sat} 0.64 ± 0.06 g g^{-1}), indicating that unsaturated conditions appeared immediately below that depth (1.5 cm). This water content profile indicates that the bulk hydraulic conductivity of the upper few centimeters of the clay layer (matrix and desiccation cracks under unsaturated conditions) is lower than that of the rest of the clay profile to depths of 6 to 12 m. Reduction of the hydraulic conductivity in the top section of sediments below the wastewater lagoon has been attributed to seal formation (Rowsell et al., 1985; SCS, 1997; Cihan et al., 2006; Tyner et al., 2006). The linear correlation between the water content and organic matter content in the upper 6 cm (R^2 = 0.66 for the lagoon, R^2 = 0.99 for the channel) suggests that seal formation is driven by clogging of the clay sediment pores by

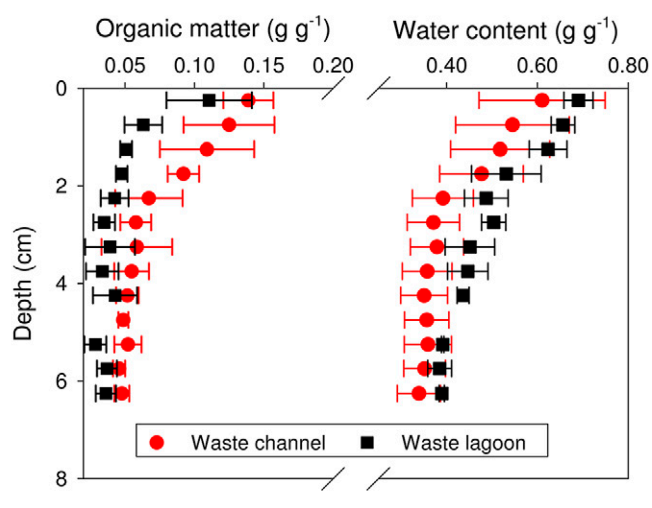

Fig. 2. Organic matter content and water content in the sediments under the waste channel and the waste lagoon. Results are presented as averages, and the horizontal bars are the standard deviation.

organic matter. This organic seal is produced by either biological activity (SCS, 1997; Parker et al., 1999c) or physical settling of fine organic particles from the wastewater (SCS, 1997; Parker et al., 1999c; Tyner et al., 2006).

Deep Percolation

Percolation of water through the vadose zone was evaluated by the spatial and temporal variations in the sediment water content across the unsaturated zone beneath the waste sources and their margins (Fig. 3). Water content of the clay sediment underlying the central parts of the lagoon was significantly lower (~0.40 m^3 m^{-3}) than that under the channel (0.55–0.62 m^3 m^{-3}). Temporal variations in sediment water content across the unsaturated profile at both sites showed dramatic variations throughout the year: although these variations were more pronounced during the rainy season (November–April) following intensive rain events, they were also observed throughout the dry summer season (May–October) when no rain was recorded (see vertical dashed arrows on Fig. 3). Baram et al. (2012) attributed these rapid changes in water content to quick infiltration and drainage of percolating water through preferential flow paths formed by desiccation-crack networks in the unsaturated clay sediment. They showed that the desiccation-crack network remains open and hydraulically active year-round, and serves as a fast conduit for water movement, even under conditions of near saturation (laboratory-measured water contents at saturation θ_{sat} were 0.63 ± 0.04 m^3 m^{-3}). The repeated observations of rapid increase in sediment water content during the dry season indicated that preferential infiltration events occur regularly from the lagoon and channel banks during fluctuations in the wastewater level and overflows: the wastewater flows through the desiccation-crack network into the clay matrix and redistributes in the subsurface, sustaining elevated water contents in the clayey vadose zone around the lagoon and channel banks.

This mechanism suggests that the water content in the bulk sediment is highly dependent on the intensity of the crack network and on proximity to the cracks. Crack intensity and connectivity increase as the sediment dries (Chertkov and Ravina, 1998), increasing the likelihood of interconnected preferential flow paths being generated. The crack intensity in clayey sediments, such as in our study site, is more pronounced at the waste source (lagoon or channel) margins and banks. As a result, at the lagoon banks there are interfaces between the water source, the wet sediments (under the source), and the dry, cracked hydraulically active sediments. Therefore, the dimensions of the waste source on the surface have a strong influence on water content distribution in the subsurface, dependent on the extent of the wet–dry interfaces at the banks. For example, the vadose zone under a narrow lagoon is near two hydraulically active banks. Preferential water infiltration from the banks and redistribution into the sediment matrix over such a narrow area sustains high water content in the vadose zone (i.e., the channel, with water content of 0.50–0.60 m^3 m^{-3}; Fig. 3). On the other hand, the sediment under the central part of a wide lagoon is relatively more distant from the hydraulically active banks. This area will be less affected by the infiltration processes at the banks and will sustain a lower water content (i.e., the lagoon, ~0.40 m^3 m^{-3}; Fig. 3). The described mechanism explains the elevated water contents observed under the lagoon banks (0.40–0.60 m^3 m^{-3}) (Baram et al., 2012). Similar observations have been made by Parker et al. (1999a, 1999c), who suggested that drying/freezing processes enhance seepage from the lagoon banks, and by Gooddy et al. (2002), who suggested that a large component of the wastewater migrates through the lagoon banks.

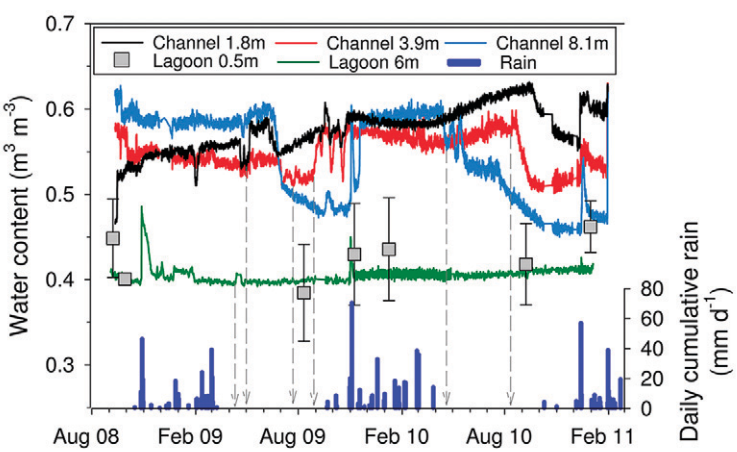

Fig. 3. Temporal changes in sediment water content measured by flexible time domain reflectometry probes under the waste channel and waste lagoon (continuous lines) and by gravimetric methods on sediment cores (squares), along with daily cumulative rain events during 2008–2011 (bars). Dashed arrows indicate events in which sharp increases in sediment water content were not associated with rainstorms.

Infiltration Mechanism and Nitrogen Transformations

The dominant N species in the dairy farm wastewater was NH_4^+–N, with an average concentration of 2012 ± 482 mg L^{-1} (n = 14). The NO_3^-–N concentration in the wastewater was always below 6.8 mg L^{-1} (n = 14), and NO_2^-–N was below detection (n = 14). The NH_4^+–N concentrations in the sediment (KCl extract) under the lagoon and the channel decreased dramatically in the upper shallow cross section, from 2700 to 4200 mg kg^{-1} dry sediment at 0.05 m to ~10 mg kg^{-1} dry sediment at a depth of 0.45 m (Fig. 4). Moreover, NH_4^+–N concentration in porewater samples collected from deeper parts of the vadose zone (>0.5 m) remained very low (<5 mg L^{-1}) throughout the entire sampling period (over 200 samples in 26 sampling campaigns from January 2007 and January 2011).

The disappearance of NH_4^+–N from the vadose zone was accompanied by NO_3^-–N formation (Fig. 5), suggesting intensive microbial oxidation of NH_4^+–N to NO_3^-–N (Fig. 4 and 5), which requires molecular O_2 (Francis et al., 2007). Gooddy et al. (1998) showed a clear inverse relationship between NH_4^+ and NO_3^-, which was demonstrated to be driven by oxygen availability. We therefore assume that aerobic conditions exist within the clay vadose zone under the flooded waste lagoon and channel. The development of aerobic conditions might be explained by the development of unsaturated conditions in the vadose zone (Fig. 2), and the well-developed desiccation-crack networks at the banks which further enhance aeration of the vadose zone. It is likely that aerobic conditions exist in regions close to the desiccation cracks, while anaerobic conditions prevail in sediment microstructures within the clay matrix which are not well aerated by these cracks. The constant supply of organic-N and NH_4^+–N and the proximity of aerobic and anaerobic zones suggest that the NO_3^-–N derived from nitrification is immediately available for denitrification, resulting in CND. Coupled nitrification–denitrification can occur in all sediment types where favorable conditions for both nitrification and denitrification are present in neighboring microhabitats (Kremen et al., 2005). The abundance of both nitrifying and denitrifying bacteria and the nitrification and denitrification potentials were determined in sediment samples collected from the lagoon bed down to a depth of 0.5 m. A detailed description of the methods used and the different microbial groups can be found in Sher et al. (2012). The high number of nitrifying and denitrifying bacteria (~10^8 gene copies g dry sediment^{-1} of each) and the high nitrification and denitrification potentials found in the uppermost sediment suggest that CND is likely to occur under the lagoon and channel.

Examinations of NH_4^+–N profiles from other studies on seepage from earthen waste lagoons have shown similar rapid attenuation of NH_4^+–N with depth (Ham and DeSutter, 2000; Ham, 2002; DeSutter et al., 2005). These studies were conducted in soils with clay contents between 18 and 46%. In those studies, the rapid attenuation of NH_4^+–N was attributed to the cation exchange capacity (CEC) of the sediment under the lagoon. To compare our results with those of Ham and DeSutter (2000), Ham (2002), and DeSutter et al. (2005), we calculated the total N mass that had infiltrated into the subsurface by multiplying the influx of wastewater by the operation period and by the concentrations of organic-N, NH_4^+–N, and NO_3^-–N in it. The total N mass infiltrated into the subsurface was then compared to the total N mass stored in the subsurface (integrating the N-species concentration profiles reported in those papers). Our calculations showed that in all of those studies, regardless of the clay content, >90% of the N mass was removed during transport in the subsurface. In most cases, the removal occurred within the upper 2 m of the sediment profile. The results from the current study showed that 85 to 100% of the N mass is removed from the subsurface (above 1.5 m) under the lagoon and channel through CND (Fig. 4 and 5). Therefore, while high CEC values can explain some attenuation of NH_4^+–N concentration in the sediment profile, it cannot explain the extent of N removal from the subsurface environments under waste lagoons.

Clear differences between NO_3^-–N concentrations in the propagating porewater underlying the waste lagoon and waste channel were observed. Whereas NO_3^-–N concentration in the clay sediments underlying the lagoon was very high (183–524 mg L^{-1}), the concentrations under the waste channel were very low (0–29 mg L^{-1}) (Fig. 5). The isotopic composition of NO_3^- (δ^{15}N and δ^{18}O) in the propagating porewater had a distinct denitrification signature (Kendall, 1998) (Fig. 6), further supporting the assumption of CND reactions occurring in the vadose zone under the waste lagoon and channel. We were unable to evaluate the NO_3^- isotopic composition in the shallow clay profile under the waste channel, since the NO_3^-–N concentrations in that area were mostly close to zero (Fig. 5 and 6). Nevertheless, the isotopic composition in one of the porewater samples from 4 m below the channel (a sample with relatively high NO_3^-–N concentration), along with the low NO_3^-–N concentrations in that area, suggest that denitrification was more prominent in locations with relatively high water contents (0.50–0.60 m^3 m^{-3}). This observation is in agreement with other studies, for example, Arah and Smith

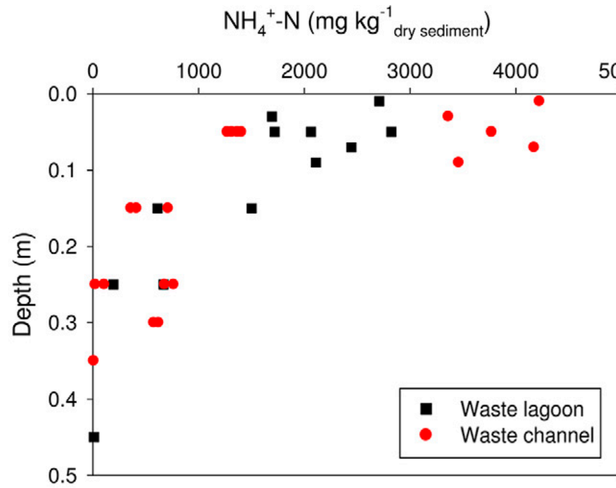

Fig. 4. Vertical distribution of NH_4^+–N concentrations in the sediment at three different locations underneath the waste lagoon and three different locations underneath the waste channel.

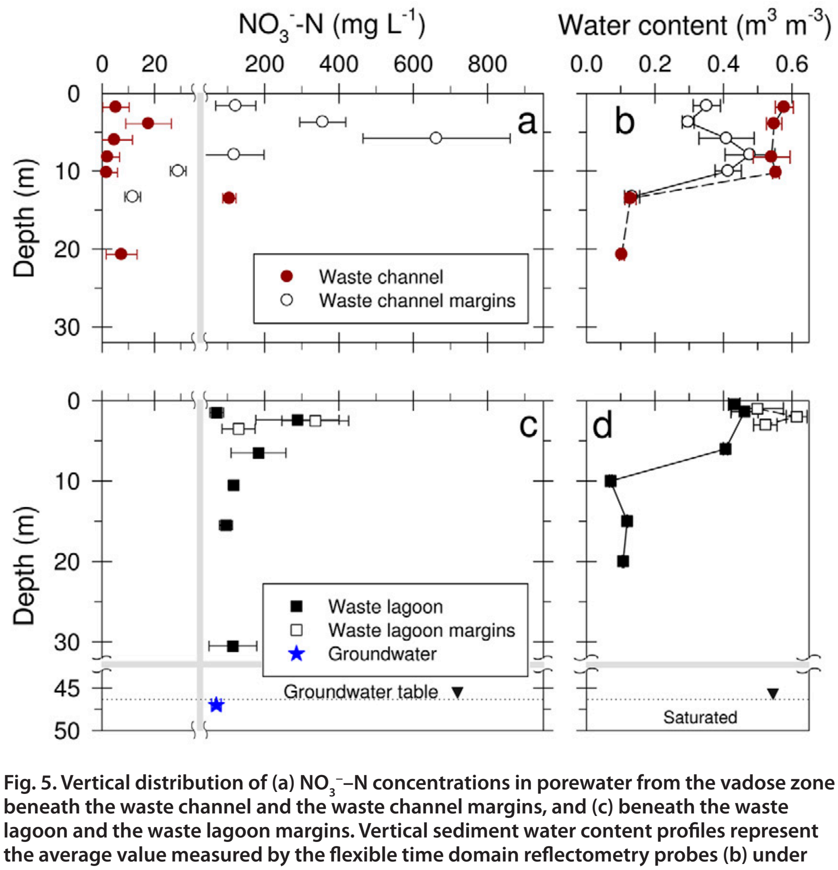

Fig. 5. Vertical distribution of (a) NO_3^-–N concentrations in porewater from the vadose zone beneath the waste channel and the waste channel margins, and (c) beneath the waste lagoon and the waste lagoon margins. Vertical sediment water content profiles represent the average value measured by the flexible time domain reflectometry probes (b) under the channel and its margins and (d) under the lagoon and its margins. Concentrations in the groundwater presented at 47 m. Results are presented as averages and the horizontal bars are the standard deviation. Transition from clay to sand/loam occurs at 6 m below land surface under the lagoon and at 12 m under the channel (Fig. 1).

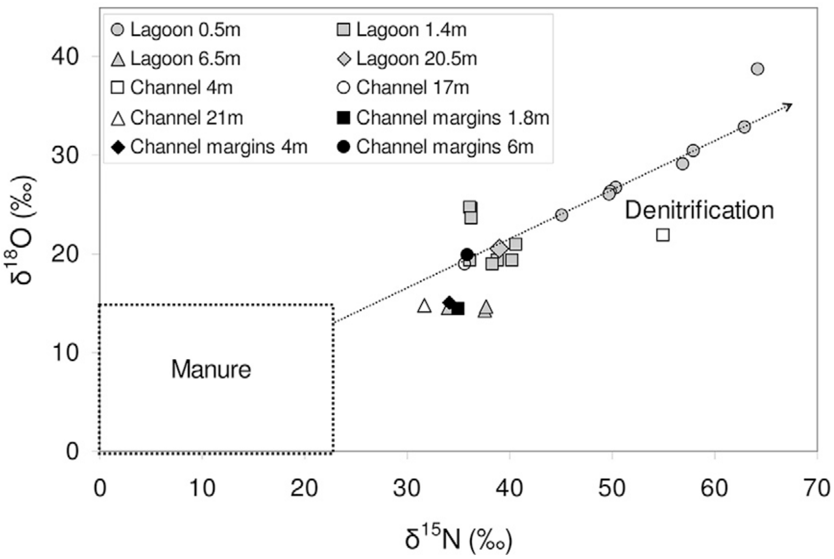

Fig. 6. Isotopic composition of NO_3^- ($\delta^{15}N$ and $\delta^{18}O$) in the propagating porewater from the vadose zone beneath the waste lagoon, waste channel, and their margins. The isotopic composition of dairy manure is based on Kendall (1998).

(1989) and Schurgers et al. (2006). Bakken and Dörsch (2006) further suggested that sediment water content acts almost like an on/off switch with respect to denitrification. Because sediment aeration is dependent on water content, it is reasonable to assume that as the conditions in the vadose zone approach saturation, more anaerobic niches will be generated and denitrification will become more prominent. Despite the importance of water content on the fate of N under CAFOs, a number of key studies on seepage from waste lagoons did not report the values of water content in the sediments (Oliver and Meyer, 1974; Korom and Jeppson, 1994; Ham and DeSutter, 2000; Gooddy et al., 2001; Harter et al., 2002; DeSutter et al., 2005). On the other hand, the work of Parker et al. (1999b) revealed that NO_3^-–N is not found beneath the lagoon bottom, but is found in isolated areas beneath the lagoon side-slopes where higher water contents are observed in the sediment.

The results of the study by Parker et al. (1999b) and those reported here strongly suggest that the water content and consequent fate of N in the vadose zone are impacted by proximity to the waste source banks. The mechanism governing N-species transport under the center of large waste lagoons may differ significantly from that at the banks of the flooded area. While minor fluctuations in water level in the waste lagoon or channel have a negligible effect on the infiltration rate from the lagoon bed, these fluctuations might cause seepage of wastewater through desiccation cracks formed at and near the flooded area's banks, leading to wetting and drying cycles in the subsurface. When preferential infiltration occurs regularly, the water content of the sediment will be higher and closer to saturation. Consequently, NO_3^-–N removal from the vadose zone will be much more effective, as observed here under the waste channel (Fig. 5c and 5d). On the other hand, overflows and isolated preferential infiltration events from the banks will not sustain high water contents and will lead to very high NO_3^-–N concentrations, as observed under the waste channel margins (Fig. 5c and 5d).

Gooddy et al. (1998) summarized the primary physical and chemical processes beneath an unlined dairy waste lagoon where no seal has formed. They suggested that wastewater infiltration would generate saturated anaerobic conditions in the sediment underlying the lagoon, while the lagoon banks would remain unsaturated and aerobic. Accordingly, NH_4^+–N would be the dominant N species in the anaerobic saturated environment under the lagoon, while NO_3^-–N would be the dominant N species in the aerobic unsaturated environment under the lagoon banks. In this work, the influence of the percolation mechanisms on the sediment water content and the consequent fate of the N species in the vadose zone beneath an unlined dairy waste lagoon where a seal has formed are illustrated in Fig. 7. Three different zones should be considered when studying the fate of redox-related compounds in seepage from clayey earthen waste lagoons. The first zone is the vadose zone under the center of the waste lagoon (away from the banks). Water in this zone continuously infiltrates from the bottom flooded area at relatively low fluxes (mm d^{-1}). Clogging of the clay matrix at the bottom of the flooded area by fine organic matter (seal formation) leads to the development of unsaturated conditions (70% saturation) in the underlying sediment of the vadose zone. The desiccation-crack network developing in the unsaturated clay allows air penetration and formation of aerobic conditions with anaerobic niches where CND occurs. Coupled nitrification–denitrification leads to substantial reduction in N mass.

The second zone consists of the banks of the waste lagoon. This zone is subjected to continuous slow infiltration under its flooded section as well as to repeated, rapid (m h^{-1}) preferential infiltration via the desiccation-crack network formed in the dryer bank areas during fluctuations in wastewater level. Redistribution of the preferentially infiltrating wastewater generates higher sediment water content (90% saturation) and reduces its aeration, such that aerobic conditions exist in niches. In this section of the vadose zone, CND leads to nearly complete removal of the propagating N mass.

The third zone is the area underlying the waste lagoon margins. This area is subjected to rare flooding events from wastewater overflow and to deep percolation during intensive rain events. The vadose zone in this section is characterized by low water content (45% saturation) and contains a well-developed desiccation-crack network. Aerobic conditions prevail in the vadose zone and wastewater overflow preferentially infiltrates into its deep sections, where NH_4^+–N is oxidized to NO_3^-–N. The NO_3^-–N propagates deeper into the vadose zone with minor transformation.

Nitrate-N was the only N form found in the groundwater under the lagoon (71 ± 19 mg L^{-1} [n = 12]) (Fig. 5). The average concentration under the lagoon was 3.5 times higher than the average concentration in the regional groundwater (~20.2 mg L^{-1}; Weinberger, 2007). Moreover, NO_3^-–N concentration in porewater sampled across the entire (~40 m) vadose zone under the waste lagoon was similar to concentrations measured in the upper groundwater, indicating that leachates from the waste lagoon have reached the groundwater (Fig. 5c). On the other hand, NO_3^-–N concentrations in the deep section

(>10 m) of the vadose zone below the channel and its margins were lower than the concentrations in the groundwater under the lagoon (Fig. 5). Under the channel margins, preferential infiltration during rainstorms transports substantial amounts of water with low NO_3^-–N concentration (22 ± 25 mg L^{-1} [n = 55]; Baram, unpublished data, 2012) into the deep parts of the vadose zone (>10 m) (Baram et al., 2012). Subsurface mixing between the channel leachates and the preferentially infiltrating rainwater could lead to such a decrease in NO_3^-–N concentrations.

This study strengthens the generally accepted idea that the hydraulic conductivity of the organic seal formed at the bottom of waste lagoons controls the infiltration rate from the lagoon bed. The results indicate that the formation of an organic seal in the bed of a waste lagoon leads to the development of unsaturated conditions under the lagoon. Consequently, the unsaturated sediment under the lagoon is sufficiently aerated to support the microbial oxidation of NH_4^+–N to NO_3^-–N. An examination of the published literature has shown that this process is likely to occur in lagoons where a seal has formed, regardless of the sediment in which the lagoon was constructed. For example, in the work of Gooddy et al. (2002) on lagoons constructed in chalk, high NH_4^+–N concentrations were observed under lagoons where no seal had formed, while almost complete oxidation of NH_4^+–N to NO_3^-–N was observed within the upper vadose zone under lagoons where a seal had formed. In the works of Ham and DeSutter (2000), Ham (2002), and DeSutter et al. (2005) on lagoons constructed in sediment with clay contents of 18 to 46% where a seal had formed, almost complete oxidation of NH_4^+–N to NO_3^-–N was observed within the upper vadose zone.

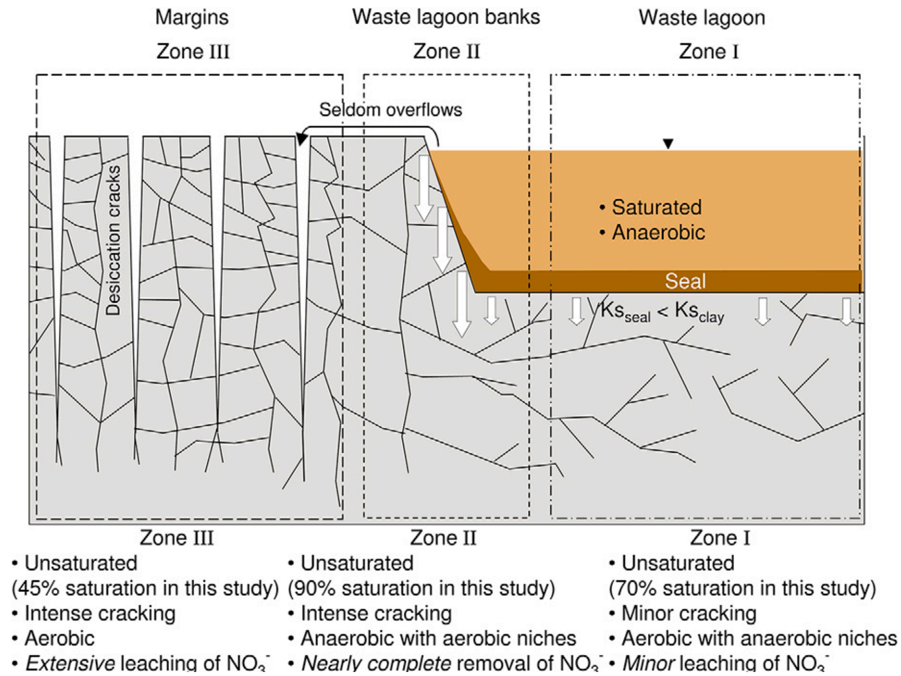

Fig. 7. Conceptual model (not to scale) of the three zones of a waste lagoon constructed in clay sediment, the governing seepage mechanisms, and the resulting fate of the N species in the subsurface. Zone I represents the vadose zone under the center of the waste lagoon (away from the banks); Zone II represents the banks of the waste lagoon; and Zone III represents the lagoon margin area, which is subjected to rare flooding events by wastewater overflow and rain events. Small arrows represent continuous slow (mm d^{-1}) infiltration flux and long arrows represent preferential infiltration via the desiccation-crack network. Relative saturation (%) is the ratio between the measured water content and the water content at saturation.

Conclusions

The subsurface under a dairy waste lagoon, waste channel, and their margins can be divided into two main zones: (i) that under the permanently flooded area and (ii) that under the banks; this division is based on the infiltration mechanism, the sediment water content, and their impact on N transformations. The sediment water content under the permanently flooded area was below saturation (~0.40 m^3 m^{-3}) due to formation of an organic seal at the bottom and continuous slow (mm d^{-1}) infiltration through it. At the same time, the sediment water content under the banks was closer to saturation (~0.50 m^3 m^{-3}) due to rapid (m h^{-1}) and deep (<8 m) preferential infiltration and redistribution of wastewater through the desiccation-crack network to the sediment matrix, which occurs regularly during lagoon-level fluctuations and rainstorms. The formation of desiccation cracks enhanced vadose zone aeration and hence aerobic processes. Consequently, organic-N and NH_4^+–N were completely oxidized in the upper 0.5 m of the sediment below the lagoon, channel, and their banks. Ammonium-N oxidation was coupled with NO_3^-–N reduction, removing >90% of the leached N, with up to 100% N removal under regions with higher water contents. Nitrogen removal suggests that neither NH_4^+–N nor NO_3^-–N can serve alone as indicators of lagoon leakage and that CND, rather than the CEC of the sediment, regulates the fate of N in the vadose zone under permanently flooded waste lagoons.

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