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Key Points:

- Nitrate removal in river-dominated deltas is limited by lateral exchange with wetlands
- Nitrate removal efficiency increases with delta topset slope and channel complexity
- Diverting coarse-grained sediment for coastal reclamation projects may increase retention capacity of newly formed deltas

Supporting Information:

Supporting Information may be found in the online version of this article.

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The Relationship Between Delta Form and Nitrate Retention Revealed by Numerical Modeling Experiments

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Abstract River deltas display a wide range of morphologic patterns that influence how nutrients interact in channels and wetlands on their way to the coast. To quantify the role of delta morphology on nitrate fate, we simulated reactive nitrate transport over steady base flow conditions for six synthetic, morphologically unique river-dominated deltas created in Delft3D by varying incoming grain size distributions. We parameterized nitrate removal kinetics using an observed relationship with elevation from Wax Lake Delta (Louisiana, USA). Total nitrate retention across the six synthetic deltas and Wax Lake Delta ranged from 1.3%–13%, suggesting that these river-dominated deltas have limited ability to remove nitrate from incoming river water. Nitrate removal is constrained by limited hydrologic connectivity with the areas of greatest nitrate demand, which are found at higher elevation. In these synthetic numerical experiments, the efficiency of nitrate removal is greatest for deltas with more topologic complexity and greater proportions of the delta plain at higher elevation, which are both common characteristics of coarser-grained deltas. The positive relationship between grain size and nitrate removal may help guide land reclamation projects if project goals include minimizing nitrate export to the sea.

1. Introduction

River deltas often receive large amounts of nitrate from land-based human activities. Excess nitrate accelerates the development of harmful algal blooms and reduces water quality of the coastal ocean (Diaz & Rosenberg, 2008; Rabalais & Turner, 2001; Rabalais et al., 2017). For example, the Gulf of Mexico receives over 1.6 million metric tons of nitrogen annually through the Mississippi River Delta (Goolsby et al., 1999), contributing to an approximate 18,000 km² seasonal dead zone (Rabalais et al., 1996; Rabalais & Turner, 2001). Similarly, the Pearl River Delta experiences annual summertime episodes of hypoxia that span approximately 200 km² due to abundant inorganic nitrogen input (Dai et al., 2006; Wang et al., 2018), and the eastern Niger Delta is experiencing a declining capacity to support fish and aquatic grass habitats due to high levels of nitrate sourced from sewage discharge (Ezekwe & Edoghotu, 2015). Seeing that most global freshwater passes through deltas before discharging to the coast (Caldwell et al., 2019; Nienhuis et al., 2020; Syvitski & Saito, 2007), it is important to understand how deltas impact water chemistry.

Deltas modify nitrate concentrations of water by physical and biochemical processes acting in distributary channels and islands (DeLaune et al., 2005; Friedrich et al., 2003; Hiatt et al., 2018; Lane et al., 2003). Denitrification, an anaerobic, microbially mediated process that converts nitrate to elemental nitrogen (or nitrous oxide), is the dominant pathway of permanent nitrogen removal in coastal settings (Lane et al., 2003; Whitney et al., 1981). Other common and temporary removal pathways include nitrate assimilation by plants (Matheson et al., 2002; Kreiling et al., 2011), dissimilatory reduction of nitrate to ammonium (Burgin & Hamilton, 2007; Tiedje, 1988), and anaerobic ammonium oxidation (Jetten et al., 1998; Rysgaard et al., 2004).

Evidence suggests that most nitrate removal in deltas occurs in densely vegetated wetlands (DeLaune et al., 2005; Lane et al., 2003; Leopold, 1970), which are associated with shallowly submerged delta islands as opposed to deeper, open-water embayments and channels (Carle et al., 2014, 2015; Ma et al., 2018). For example, in Wax Lake Delta, 73% of the nitrate that is removed from surface water is associated with shallowly inundated island areas (Hiatt et al., 2018). Knights et al. (2020) showed that submerged island areas with denser vegetation have greater nitrate processing rates, and Henry and Twilley (2014) demonstrated a positive correlation between delta

island age and nitrate removal. Both these studies demonstrate the potential for more developed soils and vegetation communities on older deltaic islands to remove nitrate, given adequate flow of water and nutrients between areas of fast transport (channels) and high reactivity (submerged islands) (Powers et al., 2012). However, the connectivity of biogeochemically “hot” wetlands to nutrient sources entering at the delta head is not well constrained (DeLaune et al., 2005; Hiatt et al., 2018) and this connectivity will vary with delta morphology. Connectivity on Wax Lake Delta strongly influence sediment transport pathways and deposition (Olliver & Edmonds, 2021) and likely impacts nitrate retention. To improve understanding of nitrate retention in deltas, it is necessary to understand how the extent of hydrologic connectivity—as controlled by delta geomorphology—influences removal in deltas.

In general, the transport of water and nitrate within deltas depend on morphologic characteristics such as topset slope, river bifurcation geometry, channel planform and cross-sectional geometry, and the structure of the distributary network (Bolla Pittaluga et al., 2003; Carlson et al., 2018; Edmonds and Slingerland, 2008; Tejedor et al., 2016), though flow also varies with vegetation, tides, and wind (Nepf, 2012; Buschman et al., 2010; Salles et al., 2015). The structure of the distributary network is of particular importance to nitrate removal because this sets the size of islands within the delta and their inundation dynamics. The morphology of deltas has been studied for several decades (Galloway, 1975), but only recent advances in both remote sensing and data analysis have led to new morphometrics for characterizing delta shape and the organization of their channel networks that may hold insight into the potential for deltas to retain nutrients. For instance, shoreline characteristics such as shoreline length and delta front rugosity (shoreline smoothness or sinuosity) are important descriptors of deltaic complexity, influenced in part by grain size distribution (Caldwell & Edmonds, 2014; Wolinsky et al., 2010; Yu et al., 2011). Other shoreline metrics such as discontinuity associated with channel presence have been proposed to distinguish tidal, wave, and river-dominated deltas (Geleynse et al., 2012). Quantitative metrics, such as the number of channels and channel width distribution, describe transport patterns such as deltaic channel networks and subnetworks (Nardin & Fagherazzi, 2012; Piliouras & Rowland, 2020; Syvitski & Saito, 2007) while commonly used metrics such as island size distribution and nearest edge distance (proximity of points on islands to the nearest channel) (Edmonds et al., 2011; Piliouras and Rowland, 2020) may hold predictive power for a delta's nutrient removal efficiency. Recently, graph-theoretic frameworks quantify delta morphological complexity resulting from varying grain sizes in terms of entropy rates, the number of alternative pathways, and channel leakage indices (Tejedor et al., 2016, 2017, 2019) allowing for quantitative morphological comparison across different deltas.

In this study, we use numerical modeling and a suite of morphometrics to understand the fundamental processes controlling nitrate retention and removal potential across morphologically diverse river-dominated deltas. The delta topset area, delta topset slope, non-local entropy (nER , Tejedor et al., 2017), the number of alternative pathways for discharging water at channel mouths (N_{ap} , Tejedor et al., 2016), and leakage indices between channel subnetworks (LI , Tejedor et al., 2016) are the morphometrics examined. We hypothesize that nitrate removal efficiency increases with increasing delta topset area, a measure of the area available for wetland establishment, and delta slope, a proxy for the proportion of biological active wetlands present. We further hypothesize that nitrate removal efficiency increases with nER , N_{ap} , and LI as the increasingly diverse flow will improve the chances of nitrate being exposed to biologically active wetlands. We test the hypotheses with a numerical experiment consisting of six synthetically generated deltas with distinct morphologies (Caldwell & Edmonds, 2014). We also run one experiment where we examine changes in nitrate removal across various stages of a single delta's growth. We end by using our findings to speculate on potential management practices that may improve removal efficiency in newly constructed river-dominated deltas.

2. Methods

2.1. Fluid Flow and Reactive Transport Framework

Model development was performed using Delft3D, a morphodynamic modeling suite with fluid flow, sediment transport, and water quality modules (Deltares 2014, 2016). Delft3D has been widely used to simulate flow and sediment transport in coastal rivers, estuaries, and deltas (Caldwell & Edmonds, 2014; Edmonds & Slingerland, 2007; Lesser et al., 2004; Olliver et al., 2020; Sawyer et al., 2015; Van Maren et al., 2009). The fluid flow component solves the 2-D depth-averaged, shallow-water equations for incompressible free-surface flow. The

equations are appropriate for describing flow in deltas where vertical momentum is relatively small and negligible (Lesser et al., 2004).

Reactive transport was solved using the unsteady two-dimensional advection-dispersion reaction equation:

$$\frac{\partial C}{\partial t} = -v_x \frac{\partial C}{\partial x} + D_x \frac{\partial^2 C}{\partial x^2} - v_y \frac{\partial C}{\partial y} + D_y \frac{\partial^2 C}{\partial y^2} - kC \quad (1)$$

where C is nitrate concentration in water [M L^{-3}], t is time, and v_x and v_y are flow velocities in the x and y directions, respectively [L T^{-1}]. D_x and D_y are the hydrodynamic dispersion coefficients in the x and y directions, respectively [$\text{L}^2 \text{T}^{-1}$]. D_x and D_y were chosen as $1 \text{ m}^2 \text{s}^{-1}$ based on validation efforts for a Wax Lake Delta model (Supplemental Material). k [T^{-1}] is the first-order nitrate removal rate, equivalent to V_f/h , where V_f is the nitrate mass transfer velocity [L T^{-1}], and h is water depth. Equation 1 assumes that nitrate removal proceeds as a lumped first-order reaction occurring at the bed due to the combined effects of reactions that remove nitrate from the water column (i.e., denitrification, assimilation, and dissimilatory nitrate reduction to ammonium) and processes that regenerate it (i.e., nitrification). As removal processes often outweigh regeneration in estuarine environments (Delaune et al., 2005; Seitzinger et al., 2006), including Wax Lake Delta (Knights et al., 2020), the lumped first-order reaction here is represented by a negative term.

Processes that remove nitrate such as autotrophic and heterotrophic uptake, and denitrification tend to occur within biofilms along short flow paths through sediments and on vegetation (Böhlke et al., 2009; Boano et al., 2010; Harvey et al., 2013). These inherently three-dimensional processes have commonly been represented as a quasi-two-dimensional, thin-film process that occurs at the bed in nutrient transport models for rivers and wetlands (Ensign & Doyle, 2006; Wollheim et al., 2006, 2008), and we adopt the same widely used approach. In relatively fine-grained deltas where groundwater residence times are long (Shaw et al., 2016 reasonable approximation, perhaps even more so than in the coarse-grained stream systems where similar nutrient spiraling models have traditionally been applied (Ensign & Doyle, 2006; Wollheim et al., 2006, 2008).

The challenging part of Equation 1 is specifying nitrogen removal rate because removal kinetics vary spatially across deltas (Henry & Twilley, 2014; Knights et al., 2020; Li et al., 2020). Knights et al. (2020) showed that summertime nitrate removal rates on Wax Lake Delta positively correlate with NDVI (a vegetation index), elevation, and, to a lesser degree, nitrate concentration. Because vegetation distribution and topography are interrelated in deltaic environments (Cahoon et al., 2011; Ma et al., 2018; Olliver & Edmonds, 2017), we parametrize spatially distributed nitrate removal kinetics across the synthetic deltas as a function of elevation instead of NDVI, based on removal rate estimates from 23 field stations across one island on Wax Lake Delta (Knights et al., 2020):

$$V_f = ae^{bz} \quad (2)$$

where a is 8.41 m s^{-1} , b is 2.01 m^{-1} , and z is bed elevation in meters referenced to mean lower low sea level (MLLW). The coefficient of determination (R^2) between V_f and z is 0.63.

Linking nitrate removal rates to elevation is a simplification; removal rates depend on many biogeochemical processes controlled by a variety of factors, including sediment grain size, organic matter content, temperature, and pH (Dawson & Murphy, 1972; Glass & Silverstein, 1998; Seitzinger, 1994). However, this simplification is a reasonable first approximation in river-dominated, sub-tropical deltas, where field data show that elevation correlates with nitrate removal rates (see Knights et al. (2020) for a detailed discussion). Given the uncertainties of this simplification, and to develop more robust interpretations, we ran replicate versions of all models using upper and lower limits of coefficient a (0.5 and 2 times) resulting in average V_f across the six deltas ranging from 4 to 79 mm hr^{-1} covering ranges typically observed across stream networks (Ensign & Doyle, 2006; Wollheim et al., 2006, 2008). We also present a test case using Wax Lake Delta (Supplemental Material) where we compare simulated concentrations from our modeling approach with field observations.

2.2. Synthetic Delta Creation

The simulated deltas were built in Delft3D using lognormal distributions of incoming sediment size with median grain diameters (D_{50}) varying from 0.01 to 1 mm, resulting in deltas with unique morphologies (Figure 1). Details of the delta modeling are discussed in Caldwell and Edmonds (2014). In brief, delta evolution was simulated in

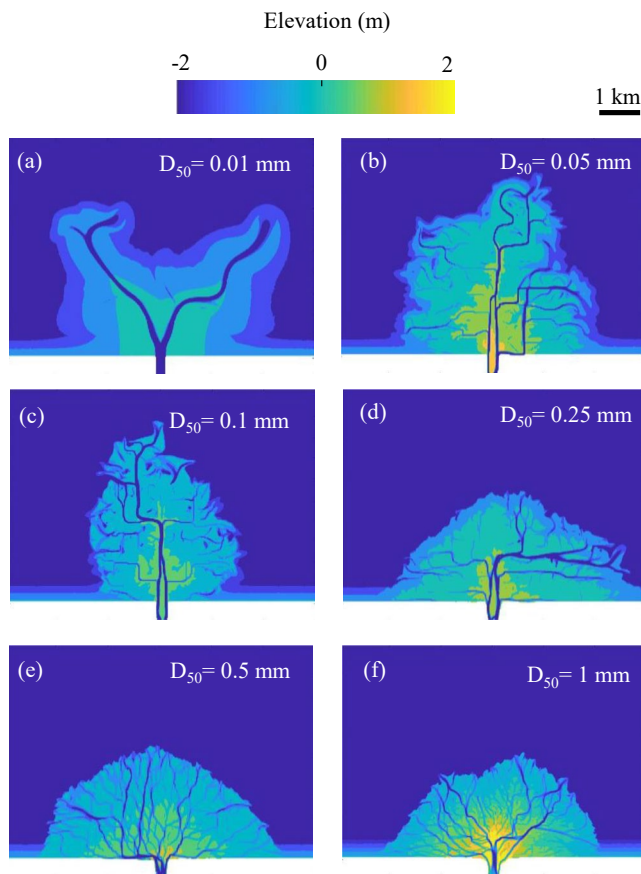


Figure 1. Six synthetic deltas with unique geomorphologies were created following Caldwell and Edmonds (2014) by varying the median grain size (D_{50}) in incoming sediment. Synthetic deltas here are statistically similar to real ones in terms of their shapes and channel network organization (Edmonds et al., 2011). Elevations are referenced to mean sea level. Because of transient water levels, positive elevations do not necessarily indicate subaerial exposure.

holland et al., 1985; Ye et al., 2017). It is inversely related to the Damkohler number, or the balance between downstream transport and biogeochemical demand, through water depth, h . Uptake length was calculated across the delta grid as:

$$S_w = \frac{|v| \times h}{V_f}. \quad (3)$$

Delta topset slope (Caldwell & Edmonds, 2014) was analyzed as an indicator of the elevation range available for wetland establishment and its influence on nitrate removal (Cahoon et al., 2011; Carle et al., 2014, 2015; Ma et al., 2018). Topset area was calculated to test the effect of increased benthic exposure on removal. Topset area was defined as the delta area enclosed upstream by the 0 m elevation contour. The delta network configuration as measured by nER and calculated by Tejedor et al. (2015) was analyzed to understand how the diversity of flux of bifurcating channels affects removal. N_{ap} and LI (calculated by Tejedor et al., 2017) were used to measure how dynamic and topologic channel complexity influences removal, respectively. For every channel mouth, N_{ap} describes the total number of possible pathways a parcel of water can take to arrive at the outlet, LI quantifies the amount of flux lost from a subnetwork of interest to the rest of the delta through shared bifurcations.

Lastly, to understand how the temporal evolution of a growing delta affects nitrate removal, we simulated reactive transport at 9 different time-steps on the synthetic delta simulation created using median grain size of 1.0 mm

response to river and sediment discharge entering a static body of water that was 7.5 km by 5.625 km (model grid cell resolution was 25 m by 25 m). The basin initially consisted of a floor with a slope of 0.000375 to the north. The initial conduit at the inlet, located on the southern face of the model, was 250 m wide, 2.5 m deep, and 500 m long with specified incoming water discharge of $1,000 \text{ m}^3 \text{ s}^{-1}$. The west, north, and east boundaries of the domain were designated as open with constant water elevation.

Elevation from model output, steady flow fields, and water depths were used as inputs to solve the unsteady reactive transport equation (Equation 1) for nitrate concentration using the process library configuration in the water quality module of Delft3D (Delft3D-WAQ). Concentrations were solved until they reached steady state (i.e., no change in concentration with time), which typically required 40 model days. Only the final steady concentrations were reported. Mass transfer velocities were specified according to elevation (Equation 2). Thus, as the delta topography changed with delta growth mass transfer velocities changed accordingly. We assume that similar biogeochemical and ecological processes underpin the kinetics of nitrate removal in our synthetic deltas (for example, the synthetic deltas are intended to represent scenarios with similar temperatures and climates, pH, elevation-dependent plant and microbial communities, and other factors that influence nitrate removal kinetics). The river inlet boundary was assigned a specified concentration of 1 mg N L^{-1} (taken as the median concentration value observed at USGS gauge 07381590 on the Lower Atchafalaya at Morgan City rounded to the nearest whole number). The distal boundaries were treated as advective flux boundaries, as the flow was always directed outward.

2.3. Metrics Controlling Nitrate Removal

Nitrate retention in each delta was calculated as the percent difference between the mass flux of nitrate entering the model domain through the river inlet and the mass flux of nitrate leaving through the distal boundaries. Uptake length (S_w [L]) represents the distance a nitrate molecule would travel in the water column before being processed (Newbold et al., 1981). It is a measure of the efficiency of nitrate removal from the water column that has been widely used across aquatic environments (Ensign & Doyle, 2006; Mul-

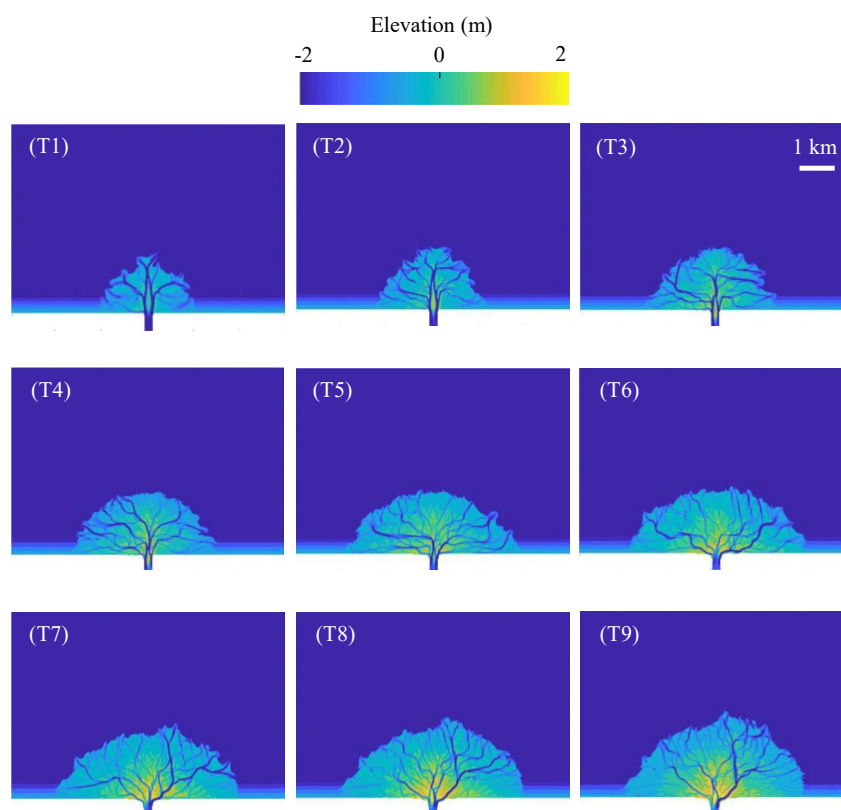


Figure 2. One synthetic delta was created using 1 mm D_{50} . Snapshots of the model at nine timesteps of growth (T1–T9) were used to simulate nitrate removal with delta evolution.

(D_{50} = 1.0 mm, Figure 2). We compared nitrate removal rates with topset area and channel network complexity metrics (N_{ap} and LI) over the 9 time-steps. Topset slope and nER were not available for the growing delta simulations.

3. Results

3.1. Static Delta Models

Patterns of nitrate removal were similar across the six synthetic deltas, but the magnitudes of retention varied. For example, nitrate levels remained relatively high across all six synthetic deltas but were efficiently processed (concentrations approaching 0 mg N L⁻¹) in discrete areas within some of the shallowest water depths (near the subaerial portions of deltas, which appear white in Figure 3). On the synthetic delta that was fully submerged (Figure 3a), nitrate concentrations nowhere fell below 80% of incoming levels. Nitrate uptake lengths were generally several times longer than delta lengths in all simulations, especially within channels, indicating little potential for removal during transport through the delta. Median uptake lengths were greater than 60 km (Figure 4). Uptake lengths were greatest within the channels and least within subaerial regions. Damköhler numbers ranged from 2.74×10^{-17} ($D_{50} = 0.01$) to 964 ($D_{50} = 0.1$) across the deltas. Average Damköhler numbers across all deltas were less than one indicating the systems were transport dominant (Table 1).

Nitrate removal across the six synthetic deltas ranged from 1.1 to 11 metric tons per day, representing only 1.3%–13% of incoming nitrate (Table 1), similar to the amount of removal simulated for Wax Lake Delta (Supplemental Material). This also compares well with whole stream removal rates of 10% measured in the Elbe River, a low-lying eighth order river (Ritz et al., 2018). In general, nitrate removal was positively correlated with delta topset slope, nER , and N_{ap} (R^2 of 0.87, 0.91, and 0.84, respectively) (Figure 5). Nitrate removal was negatively correlated with LI (R^2 of 0.30).

Contrary to our hypothesis, nitrate removal was not positively correlated with topset area of the six different deltas, but instead, decreased with increasing area (Figure 5b, R^2 of 0.61). This relationship stems from the fact that

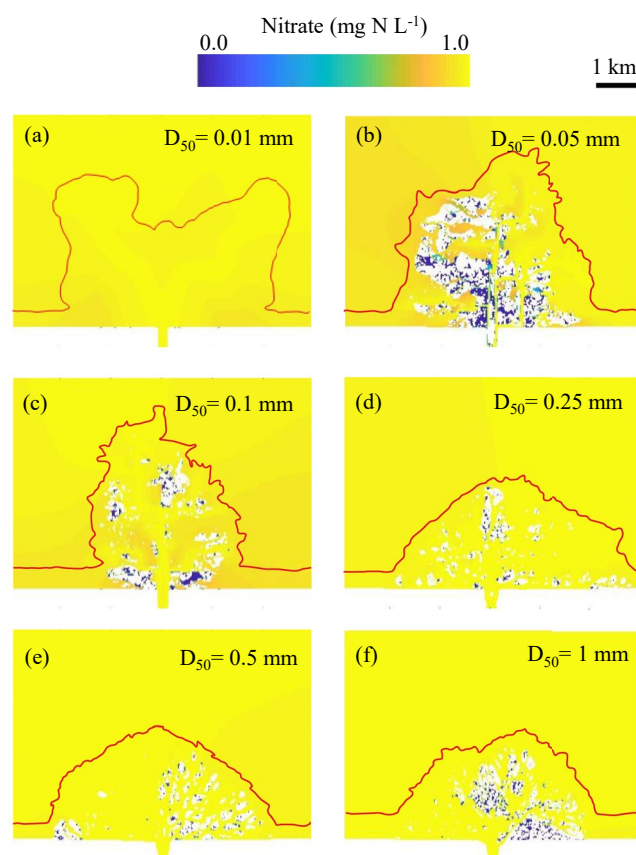


Figure 3. Nitrate concentration remained relatively high (close to inlet concentration of 1.0 mg N L^{-1}) in all six models. Concentrations were lowest near subaerial (white) areas. White areas represent dry cells where surface water concentrations were not computed because water depth was 0 m. Red lines represent geomorphic delta fronts.

the largest deltas tend to have more submerged, low-lying areas that are inadequate to support highly reactive wetlands. In other words, the smaller deltas in this study tend to have greater slopes and a greater portion of their topset areas characterized by high nitrate removal potential, leading to greater overall removal (Figure 5). It is worth noting that the median elevation alone is not a good predictor of nitrate removal (Figure 5g) because the median holds little information about the distributions of the highest elevations, which are the hotspots of greater nitrate demand (Equation 2). Similarly, max elevation may represent an extreme of the DEM which is not reflective of the overall highs in the wetland. Topset slope is more indicative of these hotspots and overall removal (Figure 5a).

3.2. Growing Delta Simulation

The growing delta simulation allowed for comparison of nitrate removal under the same grain size and morphologic characteristics but along a trajectory through time. When considering one delta ($D_{50} = 1 \text{ mm}$) where morphology evolves in a self-similar way, nitrate removal does increase with delta size as expected. The total delta area increased from 2.4 to 6.6 km^2 as the synthetic delta developed over timesteps 1–9 (Figure 6a). As the delta increased in size, nitrate retention increased from 0.47% to 3.2% (Figure 6b). The delta also increased in nitrate removal efficiency on an areal basis as the percent of nitrate removed per unit area increased from 0.20 (percent removed per m^2) at timestep 1 to 0.48 at timestep 9, peaking at 0.61 at timestep 8 (Figure 6c). In other words, as the delta grew in age and size, it built a larger delta top wetland that grew in demand for nitrate. Removal increased linearly with N_{ap} , or the number of alternate flow pathways, but there was no correlation between nitrate removal and LI .

4. Discussion

4.1. Controls of Nitrate Removal in Simulated Deltas

Simulated deltas with more of their topset areas at or above sea level generally have greater areas available for dense vegetation establishment and thus more biogeochemically active sites for nutrient removal (Figure 7). This is also clearly illustrated by examining the least submerged delta, which only removed 2.5% of the incoming nitrate load (1.2% – 4.9% , depending on Equation 2) (Figure 3a). Densely vegetated, shallow deltaic areas have many biophysical conditions that promote various pathways for removal, including direct uptake by plants (Saeed & Sun, 2012) but also indirect effects of plant-water-sediment-microbial interactions. For example, more denitrification may occur in microsites on plant material in the water column and at the sediment-water interface (Holmes et al., 1996). Plant detritus also provides more organic matter that acts as a terminal electron donor for denitrification (Vymazal et al., 1999; Weisner et al., 1994), and contributes to the development of soils with high potential respiration rates (Henry & Twilley, 2014).

Our model results are relatively robust against uncertainties in nitrate removal kinetics (Equation 2). To check the sensitivity of our results to Equation 2, we ran additional simulations where we doubled the relationship between the kinetic uptake rate constant and elevation, resulting in average kinetic uptake rates for each delta of 17 – 79 mm hr^{-1} , which approach some of the larger observed values in streams and wetlands (Ensign & Doyle, 2006; Mulholland et al., 2009) and are within the range of values observed in subtropical estuaries (Bernot et al., 2003; Pina-Ochoa & Álvarez-Cobelas, 2006). This doubling approximately doubled delta-wide removal rates (Table 1) yet still only resulted in a modest reduction in nitrate export from the delta (1.3% – 13%). We acknowledge that the parameterization of Equation 2 warrants further testing, and measurements of reaction kinetics in intertidal locations and deeper areas like channels and other deltas are important areas for future research. However, we expect that the general form of the relationship should hold. After all, several studies have shown that elevation acts as a

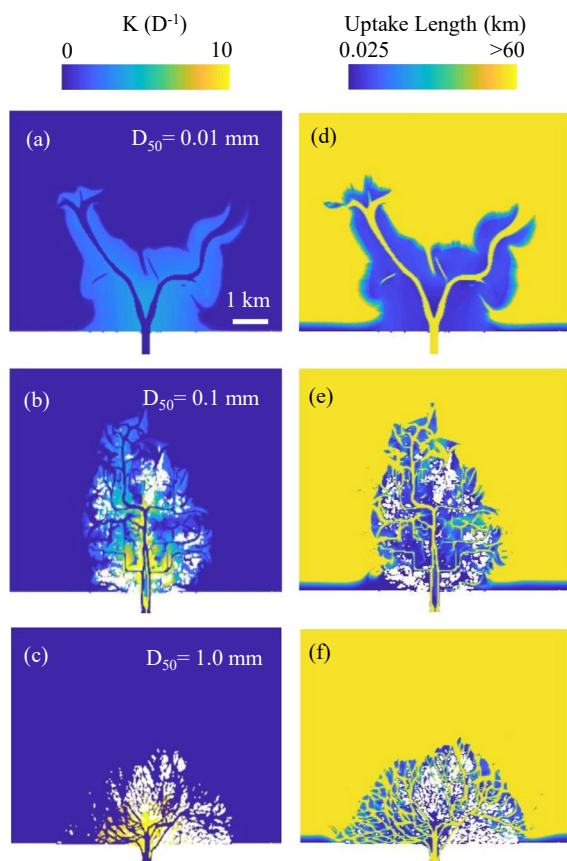


Figure 4. (a)–(c) Mass transfer velocity (V_p) (d)–(f) and corresponding uptake length for deltas formed under fine, intermediate, and coarse grain sizes. Despite the increase in the area of highly reactive wetlands (high k) with grain size, uptake length (S_w) remains large across all deltas, showing a lack of efficient removal over the delta length scale. White areas represent dry (depth = 0 m) locations where k and S_w were not calculated.

Table 1
Geometrics of Synthetic Deltas and Nitrate Removal

D_{50} (mm)	Slope	Area (km ²)	nER	N_{ap}	LI	^a Total removal (%)	Mean uptake length (km)
0.01	0.00081	7.6	0.83	24	0.20	2.5 (1.3–4.9)	1.8×10^{12}
0.05	0.00053	9.5	0.69	7	0.16	3.8 (2.0–7.1)	3.7×10^{11}
0.1	0.00048	1.0	0.76	4	0.22	3.5 (1.8–6.7)	2.1×10^{12}
0.25	0.00049	8.6	0.67	2	0.22	4.7 (2.4–9.0)	2.5×10^{11}
0.5	0.00039	10.6	0.69	1	0.23	4.2 (2.1–8.1)	9.0×10^8
1	0.00016	11.2	0.67	1	0.29	6.7 (3.4–13)	4.7×10^5

Note. All simulations were run at steady state an incoming discharge of 1,000 m³ Day^{−1} and nitrate flux of 86.4 tons Day^{−1}.

^aRanges based on 0.5 and 2 times coefficient a (Equation 2) are in parentheses.

master variable influencing biogeochemical activity (Carle et al., 2015; Johnson et al., 1985; Knights et al., 2020; Ma et al., 2018) because it positively correlates with vegetation density (Carle et al., 2015; Ma et al., 2018), and nitrate assimilation into plant biomass can account for a substantial portion of removal in wetlands (Vymazal, 2011). Also, greater denitrification rates, associated with older, more reduced sediment, are found at higher elevations on prograding deltas (Henry & Twilley, 2014; Li et al., 2020). Soils near and marginally above mean sea level provide ideal locations for highly active wetlands to develop (Ma et al., 2018). In contrast, active live vegetation is sparser in deep environments of embayments and channels. An important percentage of the nitrate removed is likely attributed to plant conversion to biomass as spatially distributed nitrate removal rates positively trend with vegetation greenness which is greatest along island heads and levees in Wax Lake Delta (Knights et al., 2020).

While delta topset slope explains 87% of the variability between retention rates (Figure 5a), two deltas with similar slopes had noticeably different removal rates of 3.45% and 4.67% (Figures 1c and 1d). These differences may be due to the distribution of flow within the two deltas. The delta with greater nER (median grain size of 0.25 mm, Figure 1d) was more efficient at removing nitrate than the delta with a lower nER (median grain size of 0.1 mm, Figures 1c and Table 1). Deltas with high nER have channel junctions with more asymmetric flux partitioning (Tejedor et al., 2016), meaning that the distribution of nutrient fluxes to the basin should be more diverse and heterogeneous. As nER increases, the diversity of nitrate flux to the basin increases, which increases the chance that nitrate will be delivered to nitrate sinks of high reactivity zones (Powers et al., 2012). This finding also implies that similar sloping deltas could have markedly different retention rates if a disproportionate amount of flux is routed through high reactivity zones (shallowly submerged island areas, in this case).

Contrary to our hypothesis, removal decreases with delta top area across the six morphologically different deltas (Figure 5b). To examine the relationship between delta area and nitrate removal independently from the confounding effects of elevation-dependent removal kinetics, we repeated our simulations using uniform removal kinetics (6.39 mm hr^{−1}, representative of the average V_f measured in Wax Lake Delta by Knights et al. (2020)) across each delta top, which was defined by the 0 m elevation contour (Olliver et al., 2020) and prescribed no reactions outside the delta top. For these simulations, nitrate removal does increase with delta area as expected—the larger the delta top wetland, the more nitrate it can remove (Figure 5h). Thus, both steeper deltas and larger deltas should remove more nutrients, all other factors held constant. This relationship is observed in the growing-delta analysis where nitrate removal increases with delta age and size as the delta morphotype remained relatively consistent with growth (Figure 6). However, there is a tradeoff between delta top area and delta top elevation in our synthetic deltas and natural deltas with the same sediment volume. This finding has important implications for management practices aimed at creating new delta land (Paola et al., 2011). Because nitrate removal potential is greatest in high-standing wetlands with older soils (Henry & Twilley, 2014; Knights et al., 2020), it could be more efficient from a nutrient management standpoint to construct smaller deltas with greater proportions of high-standing area than larger low-lying deltas.

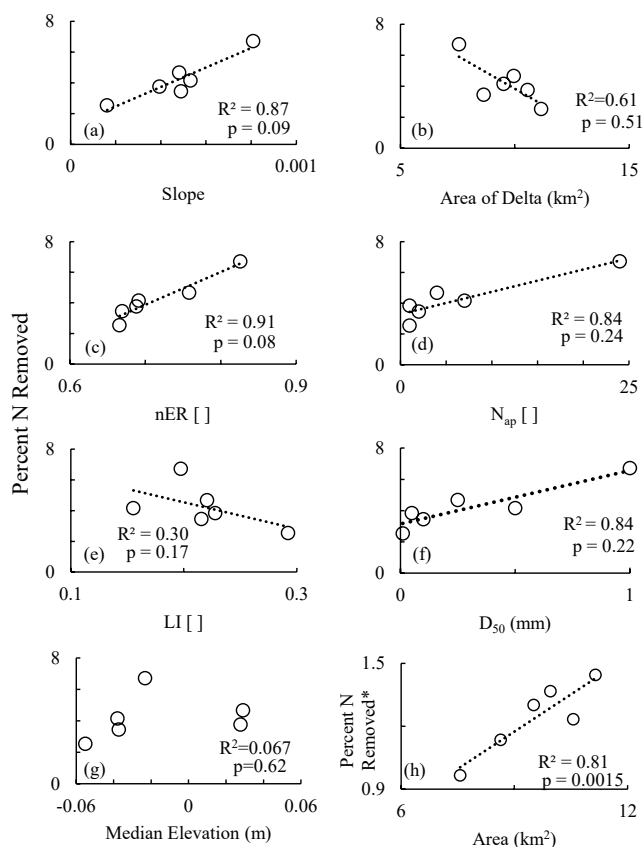


Figure 5. (a), (c), and (d) The percent removal is positively correlated to slope, nER , N_{ap} , and LI . (b) Percent removed is negatively correlated to delta area. (e) There is a weak and negative relationship between LI and removal rate. (f) The percent of nitrate removed from each delta increases as median grain size increases. (g) There is no correlation with median elevation and percent removal (h) * Percent N removed under uniform removal kinetics. Removal does increase with area when reaction kinetics are uniform.

4.2. Coastal Water Quality Implications

For all deltas in this study, nitrate retention rates amounted to a small portion of the incoming load under the long-term steady discharge conditions that created these deltas. Even when kinetic rate constants were doubled, removal roughly doubled and still represented a small fraction of the incoming nitrate load (4.9%–13%, Table 1). Similarly, in our test case for Wax Lake Delta (Supplemental Material), only 2.1%–6.6% of the incoming nitrate load was removed. Removing nitrate requires water to be moved to shallow, biogeochemically reactive areas (DeLaune et al., 2005; Powers et al., 2012; Hiatt et al., 2018). Thus, changing the kinetic uptake rate does not influence removal if water is not being transported to those reactive areas (Bernhardt et al., 2017). The implication is that river-dominated deltas in temperate regions may have only a modest capacity to buffer nitrate fluxes to the coast (Table 1). Low retention rates can be attributed to these deltas being mostly transport-dominated (i.e., discharge is generally too great to allow time for removal). The few areas such as island levees with very short uptake lengths (on the order of tens of meters) cannot contribute much to removal as they are not well connected hydrologically to channels where most discharge is focused (Powers et al., 2012). As a proof of concept, we simulated one scenario ($D_{50} = 1$, Figure 1f) where we drastically increased nitrate removal kinetics within the delta top wetland ($V_f = 150 \text{ mm hr}^{-1}$ or 25 times the mean observed value in Wax Lake Delta) and maintained no reactions in the channels and portions of the delta below mean sea level. This simulation was not designed to replicate realistic removal rates but to give insight into transport limitations on removal. Under the extreme conditions of this scenario, all nitrate that escaped the channels and entered the wetlands was fully removed under the extreme biogeochemical demand that was prescribed outside the channels, and no nitrate that remained in the channels was removed. Nitrate removal increased from 6.7% to 22.3%, suggesting that at most, only a quarter of flow from channels makes it onto delta islands. This finding aligns well with estimates for Wax Lake Delta that 23%–54% of channel flux enters delta islands (Hiatt & Passalacqua, 2015; Olliver et al., 2020). Even if removal kinetics in island retention zones were much greater than in Equation 2, less than 25% of nitrate would interact with these hotspots of reactivity unless hydrologic connectivity were greater. In river-dominated deltas, the natural

distribution of flow through channel and island networks may impose fundamental limits on hydrologic connectivity and thus nitrate retention.

4.3. The Role of Sediment Grain Size

Given the importance of the delta elevation profile for nutrient removal services in deltas, sediment grain size emerges as a key parameter that may influence nutrient removal services in deltas. In our simulations, the steepest deltas had the coarsest sediments, as expected. Greater bed shear stresses (and thus greater topset slopes) are needed to transport coarser material, leading to more aggradation (Parker et al., 1998; Whipple et al., 1998). In contrast, fine sediments form gently sloping deltas, even though finer-grained sediment is more cohesive because the low settling rate of fine-grained sediment results in greater sediment bypass and more distal deposition (Caldwell & Edmonds, 2014). This relationship has important implications for coastal land reclamation projects. To achieve better water quality outcomes, river diversions can be designed to divert coarser sediment loads, leading to the construction of steeper deltas (Paola et al., 2011).

Another implication is that deltas draining active coastal margins with high relief catchments and presumably coarser sediments may have greater nitrate removal potential, all other factors held constant (e.g., temperature, plant communities, river discharge, and incoming nitrate concentrations). The Sacramento-San Joaquin River delta located at an active margin may fall into this category. Fine-grained, gently sloping deltas in passive margins

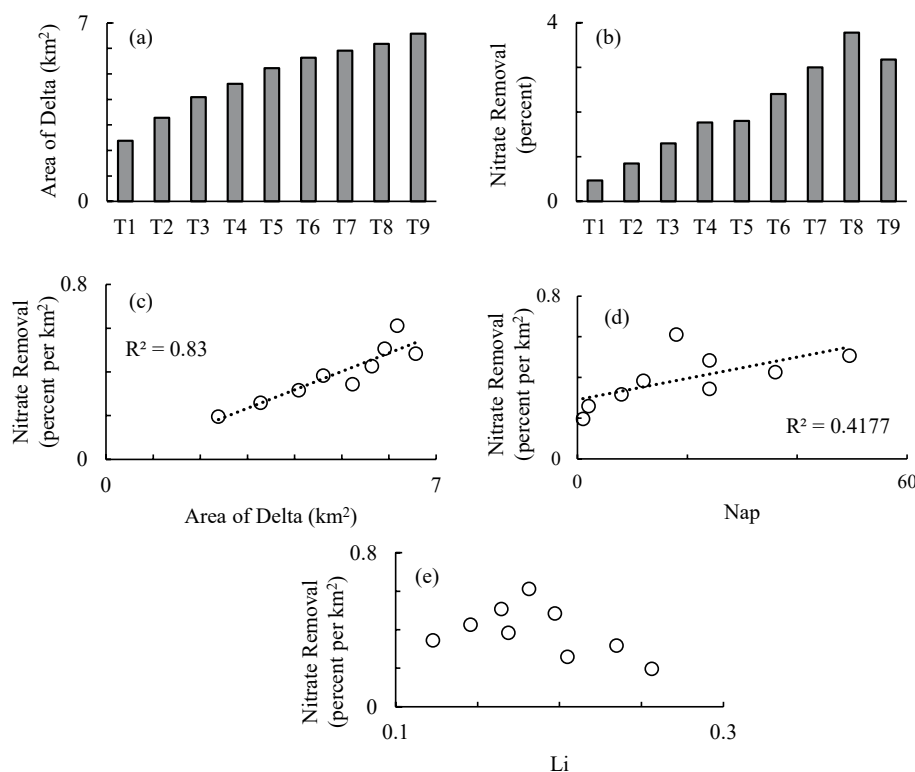


Figure 6. One delta ($D_{50} = 1$ mm) was arrested at nine timesteps of growth (T1–T9). (a) Delta area and (b) the percent of nitrate removal increases with time. (c) The percent of nitrate removal relative to delta area also increases with time (d) Removal rate is positively correlated with number of alternative pathways. (e) There is no correlation between leakage index and removal as the delta grows.

such as the Orinoco River Delta may have comparatively reduced nutrient removal capacity, relative to their size. There is a need for more studies to assess nitrate removal in natural deltas around the world to understand how their removal efficiencies relate to delta topset slope and grain size, among other morphologic factors.

4.4. Areas for Further Study

The synthetic delta simulations here are simplistic versions of natural deltas and examine only a limited range of potential controls on nutrient removal. For example, we examined an incoming nitrate concentration representative of mixed upstream land use. The efficiency of nitrate removal in streams and wetlands is inversely proportional to background concentration (Hall et al., 2009; Mulholland et al., 2009; Tank et al., 2008). Therefore, removal efficiency in synthetic and natural deltas would likely improve if incoming nitrate concentration were reduced, although connectivity may still ultimately limit substantial removal.

Advective nitrate transport along groundwater flow paths is not explicitly modeled in this study, although the shallowest, shortest timescales of surface water-groundwater mixing and nitrate removal within the benthic zone are implicitly represented in V_f . For example, the chambers used for deriving V_f (Equation 2) were open on the bottom to allow some degree of small-scale fluid exchange and nitrate removal immediately beneath the sediment-water interface. The benthic and hyporheic zones where surface water and groundwater mix are known regions of nitrogen processing, especially in the upper several centimeters of sediment where mixing is most efficient (Harvey et al., 2013). Longer subsurface flow paths also contribute to nitrate removal (i.e., Duff & Triska, 1990; Gu et al., 2007; Zarnetske et al., 2011; Kolbjørn Jensen et al., 2017), but a groundwater modeling study for synthetic deltas with similar grain size suggests that subsurface residence times and fluxes are too long and slow along these flow paths to substantially influence the nitrate budget for surface water (Sawyer et al., 2015). Due to the likely limited influence of groundwater flow—outside of small-scale advective exchange that is captured by the benthic chambers and inherent in Equation 2—we exclude groundwater flow at this stage.

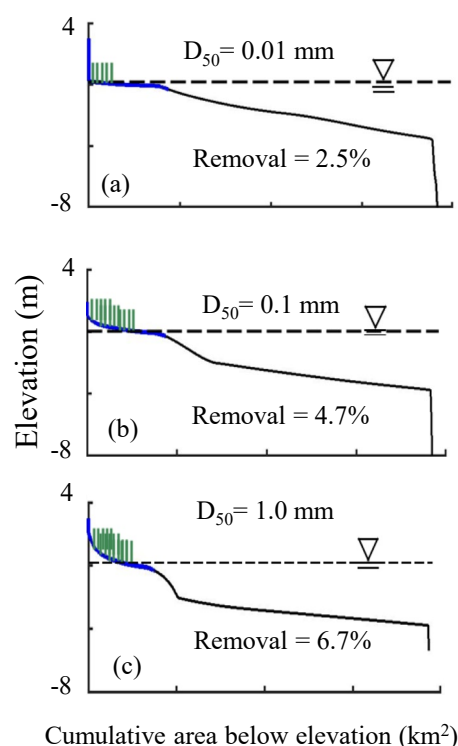


Figure 7. (a)–(c) Cumulative area curves of the delta-top (blue) and basin (black). The horizontal line represents mean sea level. The percent of nitrate removed increases with delta top slope and median elevation above mean sea level. The green vegetation is added only as a representation of wetlands in this figure. There is no explicit representation of vegetation in the Delft3D model.

In the model, we manipulate median grain size while keeping all other variables constant. However, inherent spatial autocorrelations between grain size and other environmental variables known to influence removal rates like organic matter content, permeability, and dissolved oxygen levels likely exists (Mulholland et al., 2008; Seitzinger et al., 2006). As grain size decreases, organic matter content, pH and DO tend to increase, decrease, and decrease, respectively. These can have opposing and additive influences on nitrate removal (Milliman, 1994; Strayer et al., 1997), the modeling of such is beyond the scope of this study.

The models are most representative of sub-tropical river-dominated deltas during summer flow conditions, and removal efficiencies are likely to vary over seasons, in tidal- or wave-dominated deltas, or in high-latitude deltas. Over seasons, it is unlikely that removal efficiency would increase in winter because colder temperatures lead to slower denitrification kinetics (Bachand & Horne, 1999; Bremner & Shaw, 1958), and winter senescence results in limited assimilation into biomass. At higher latitudes, removal efficiency could be lower due to both colder temperatures and permafrost, which can restrict liquid water interactions with organic-rich soil. Further analyses of other types of deltas under different hydrodynamics are necessary to understand nutrient fate and inform managerial practices.

5. Conclusion

Nitrate retention in river-dominated deltas is likely limited to a small fraction of the incoming load (in our simulations, ~1%–13%). Nitrate uptake lengths are several times larger than the delta length scale, indicating that most nitrate is transported to sea without removal. High-standing portions of deltas with emergent vegetation and well-developed soils play an outsized role in nutrient removal, and therefore delta topset slope has a greater influence on nitrate removal than any other geometric parameter examined here. These results suggest that diverting sediments with a coarser grain size distribution

should be considered when land reclamation projects intended to improve water quality are a priority. This would thereby achieve steeper slopes and greater nitrate removal potential. Future research considering a wider range of temporal effects (seasons, floods, and tides) on both reactivity and hydraulic connectivity can improve estimations of nitrate fate in deltas.

Data Availability Statement

Data archiving in HydroShare is underway and will be made public upon acceptance. A temporary copy of our data is in the Supporting Information. Additional data for this study is available at Knights (2021).

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