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Assessing legacy nitrogen in groundwater using numerical models of the Long Island aquifer system, New York

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1 Abstract

Nitrogen transported along groundwater flow paths in coastal aquifers can contribute substantially to nitrogen loading into surface water receptors, particularly in hydrologic systems dominated by groundwater discharge. Nitrogen entrained in the aquifer is a function of land use and associated nitrogen sources at the time of groundwater recharge, which may differ considerably from present-day sources. Legacy nitrogen can result in substantial discrepancies between observed present-day nitrogen loading to surface water receptors and loading estimated from present-day sources. Additionally, legacy nitrogen can continue to discharge into surface waters after nitrogen mitigation actions have been undertaken. Here, we use a numerical modeling framework to compare three methods of estimating time-varying historical nitrogen loads to four water bodies (receptors) on eastern Long Island, New York. The methods span a range of data requirements and process complexity, from instantaneous receptor loads calculated from steady-state groundwater contributing areas, to transient loads estimated by explicitly simulating legacy groundwater nitrogen transport over a century with large changes in nitrogen sources and hydrologic conditions. The effects of legacy nitrogen on estimated receptor loads varied temporally and spatially within the study area. Depending on antecedent nitrogen inputs and hydrologic conditions, historical annual nitrogen loads estimated from transient simulations accounting for legacy nitrogen can be quite similar (<10% difference) or substantially different (±100%) from those estimated from simpler instantaneous methods. Continued input of present-day nitrogen sources using methods that account for legacy nitrogen results in asymptotic increases in receptor nitrogen loads over time, indicating that simulated present-day receptor nitrogen loads are not in equilibrium with present-day inputs. For these receptors in disequilibrium, models simulating transient groundwater nitrogen transport could be used to account for legacy nitrogen lag times to help resource managers evaluate the potential effectiveness of proposed nitrogen mitigation actions.

2 Introduction

Nearshore coastal waters adjacent to developed areas are at risk for eutrophication arising from excess nutrients from onshore sources that can result in the degradation of water quality and clarity, growth of harmful algal blooms (Anderson et al., 2021), and loss of marine habitats. Nearshore coastal waters like estuaries and bays can have limited exchange with open coastal waters and receive freshwater from groundwater discharge and surface-water inflows that can introduce excess nutrients from anthropogenic sources. The nutrient of generally greatest concern in these marine ecosystems is nitrogen (Ryther and Dunstan, 1971), which can be introduced from several onshore sources, including residential and agricultural fertilizers, wastewater, and atmospheric deposition (Carpenter et al., 1998). The economic and recreational importance of inshore coastal waters, which provide habitat for commercial and recreational fisheries, makes eutrophication and habitat loss an increasing concern to local communities and stakeholders. Coastal eutrophication is a major ecological and societal issue in the northeastern United States, from Chesapeake Bay to New England, arising from the region's large population, historical agricultural practices, rapid urbanization, and presence of inshore marine waters with limited circulation and exchange with the ocean (Kennish, 2009; Paerl et al., 2006). Nitrogen in large population areas is generally derived from nonpoint anthropogenic sources (Van Drecht et al., 2001), and nitrogen mitigation actions often are costly and complex, requiring large-scale engineering solutions, land use changes, and innovative technologies. Due to legacy nitrogen in soils and groundwater, nitrogen mitigation actions have not always resulted in water quality improvements on expected timelines (Ascott et al. 2021). Therefore, estimating the degree and rate of nitrogen load reductions from potential mitigation actions using approaches that account for the effects of legacy nitrogen could help inform development of more effective mitigation strategies (Basu et al., 2022).

There are two components of legacy nitrogen: biogeochemical accumulation of nitrogen in soils and substrates, and nitrogen dissolved in groundwater (Van Meter et al., 2016). The latter component is the result of the time needed for nitrogen to be transported by advection through the aquifer to eventually discharge into surface waters (Puckett et al., 2011). The importance of legacy nitrogen within the aquifer relative to more recent nitrogen inputs is most significant in hydrologic systems where groundwater is the dominant contributor of water to surface water receptors, travel times through the aquifer are long, and there have been substantial changes in the location and amount of nitrogen entering the aquifer over the span of those travel times. The relative contribution of groundwater legacy nitrogen to total loads to a receptor is a function of transient changes in both the groundwater flow paths and nitrogen inputs to groundwater and therefore varies through time. Research on terrestrial nitrogen loads to surface waters has increasingly shown that legacy nitrogen should be accounted for in nitrogen management policy choices (Ascott et al., 2021; Van Meter et al., 2016), but quantification of legacy nitrogen in aquifer systems is rarely included in assessments of nitrogen loading to surface waters (Basu et al., 2022).

The nitrogen loading model (NLM) framework was developed as a method of approximating nitrogen loads to surface water receptors in groundwater-dominated hydrologic systems (Valiela et al., 1997). NLM has been used in numerous locations to estimate receptor nitrogen loads from non-point sources (Barclay and Mullaney, 2021; Bowen and Valiela, 2004; Suffolk County Department of Health Services and CDM Smith, 2020; Kelly et al., 2021; Kinney and Valiela, 2011; Latimer and Charpentier, 2010; Valiela et al., 2016; Lloret et al., 2022). NLM approaches take a receptor contributing area and sums current potential terrestrial nitrogen loads, reduced by attenuation factors, within that contributing area to calculate an "instantaneous" load. Receptor contributing areas are delineated through contouring of land topography (Kelly et al., 2021), contouring of groundwater potentiometric surfaces (Kinney and Valiela, 2011) or through simulations of steady-state groundwater flow and particle tracking (Barclay and Mullaney, 2021; Suffolk County Department of Health Services and CDM Smith, 2020; Valiela et al., 2000). Thus, the NLM approach typically represents loading conditions at a theoretical fixed point in time represented by the steady-state flow conditions or potentiometric surface. This approach can be used to calculate loads in areas with limited data (Kelly et al., 2021), or in more data-abundant areas with fine resolution contributing areas available via detailed groundwater models and fine resolution nitrogen inputs estimated from detailed land use records (Suffolk County Department of Health Services and CDM Smith, 2020).

However, NLM does not explicitly account for the nitrogen residence times within groundwater systems, compounding nitrogen contributions to surface waters that can occur as nitrogen moves along groundwater flow paths of different lengths that eventually discharge to the same surface water receptor, or spatiotemporal changes in nitrogen sources. Thus, determining when and where instantaneous loads calculated from NLM can provide reasonable approximations of time-varying surface water nitrogen loading could help resource managers balance science-informed decision making with the fiscal and time constraints imposed on stakeholders. Process-based simulations of steady-state groundwater flow and nitrogen transport have been used to estimate receptor nitrogen loads accounting for groundwater lag times in comparison to NLM instantaneous loads (Walter, 2013). Nitrogen loads simulated with a solute-transport model to an estuarine system on Cape Cod, MA that assumes constant nitrogen sources, steady-state flow conditions, and no initial mass in the aquifer eventually approached the instantaneous loads calculated for the same receptors using NLM from watersheds produced by the steady-state flow model (Walter, 2013). The simulated times required to reach the instantaneous loads depended on the distribution of steady-state groundwater travel times within the contributing areas. Walter (2013) reported simulated nitrogen loads to surface water bodies that approached loads approximated using NLM and were about 90 percent of the instantaneous loads after about 70 years of transport time. Nitrogen loads to public-supply wells had shorter travel times and were within that same fraction of the instantaneous load after about 20 years of transport. The results suggest that in a steady-state flow field, model-generated watersheds and NLM will approximate nitrogen loads to receptors from solute-transport models over sufficiently long (decadal) transport times. However, NLM instantaneous loads will not account for changes in hydrologic stresses and changes in nitrogen inputs during that transport time.

A comprehensive assessment of legacy nitrogen contributions to surface water receptors requires estimates of both historical nitrogen inputs and historical hydrologic conditions, particularly in hydrologic systems with long groundwater travel times or substantial historical changes in hydrologic stresses and land use. Simulations of transient flow and time-varying nitrogen inputs have not been used in assessments of legacy nitrogen due to the difficulty in obtaining these historical estimates. The Long Island, New York, aquifer system is characterized by a wide distribution of groundwater travel times and large changes in land use over the last century. Recently, the U.S. Geological Survey has compiled historical datasets for land use, nitrogen inputs, and hydrologic conditions from 1900 to 2019 (Walter et al., 2024; Monti et al., 2024; Finkelstein et al., 2022). These new datasets permit a detailed comparison of receptor nitrogen loads estimated using three approaches within a single modeling framework: numerical simulations of groundwater nitrogen transport under (1) steady-state and (2) transient groundwater flow, and (3) NLM using contributing areas from those flow models. The single modeling framework is an important element of this study, as different modeling frameworks may each be reasonable representations a hydrologic system yet produce different, yet reasonable, receptor nitrogen load estimates due to differences in model resolution, present-day nitrogen source terms, and nitrogen attenuation. Therefore, an assessment of legacy nitrogen contributions should be done by comparing approaches built on a single modeling framework. Comparing the approaches allowed us to assess the relative contributions of legacy and annual nitrogen inputs to select surface water discharge boundaries over the last century and into the future. Assessment of the differences in approaches also provides information about the utility of developing more complex models to estimate receptor nitrogen loads as compared to instantaneous load methods like NLM.

2.1 Study site

The Long Island aquifer system is at the northern edge of the Northern Atlantic Coastal Plain aquifer system along the eastern coast of the United States (Figure 1A). The Long Island aquifer system consists of a wedge of unconsolidated Pleistocene and Cretaceous sediments overlying relatively impermeable bedrock, with a maximum freshwater thickness of approximately 1,800 feet (550 m). Two extensive clay formations, the Gardiners Clay and Raritan Formation confining units, separate the aquifer system into three major units: (1) the Lloyd aquifer, (2) the Jameco and Magothy aquifers, and (3) upper glacial aquifer, which together serve as the sole drinking water source for more than 3 million people. The hydrogeology of the Long Island aquifer system is described in detail by Stumm et al. (2024), as is the groundwater flow system by Walter et al. (2020, 2024). Nitrogen contamination is primarily a concern in the unconfined Jameco, Magothy, and upper glacial aquifers, which receive water solely from precipitation and are susceptible to contaminants from land surface.

Long Island is a rapidly urbanizing region that has experienced a large population increase over the last century—from about 1.5 million residents in 1900 to more than 8 million in 2020 (Monti et al., 2024)—which has resulted in large changes in hydrologic stresses and land use on Long Island. Pumping from the Long Island aquifer system has increased from about 20 Mgal/d in the early 20th century to more than 400 Mgal/d in the early 21st century. As a result, the water table has been drawn down historically by as much as 80 feet, and saltwater has intruded the aquifer in some areas (Walter et al., 2024). The large amount of pumping has perturbed hydraulic gradients, groundwater age distributions, and the advective transport of anthropogenic contaminants, including nitrogen.

Urbanization also has resulted in large changes in land use and the potential for non-point source contamination to the aquifer. Generally, there has been an east-west trend in urbanization, from parts of New York City in Kings and Queens Counties in western Long Island to rural areas in Suffolk County on eastern Long Island (Figure 1A). About 28 percent of land on Long Island was considered developed (urban) in 1938 with high-density residential development limited to parts of New York City (Figure 1B). Developed land had increased to about 70 percent of total land area by 2019, including much of eastern Long Island (Monti et al., 2024) (Figure 1C). Agriculture has been important historically on Long Island, and in 1938, about 22 percent of the total land area of Long Island was farmed, generally in central and eastern Long Island (Figure 1B). The conversion of agricultural to residential land use, starting generally in the mid-20th century, decreased agricultural land use to less than 5 percent of total land area in 2019 (Figure 1C). Open, forested land decreased from about 50 percent to about 26 percent of total land area between 1900 and 2019 (Monti et al., 2024).

Nitrogen inputs at land surface vary by land use, and changes in land use have resulted in large changes in the amount and location of nitrogen inputs into the aquifer. Residential sources include septic wastewater and lawn fertilizer; agricultural sources include livestock and crop fertilizer. An additional source of nitrogen is atmospheric deposition, which is independent of land use. Monti et al. (2024) used spatially and temporally variable land use and nitrogen source loading to estimate annual nitrogen sources across Long Island.

The coastal waters on Long Island are important recreational and economic resources that have been adversely affected by the discharge of nitrogen-contaminated groundwater (Monti and Scorca 2003) resulting in loss of critical ecosystem habitat and associated fisheries (Hoagland et al 2002). An area of particular concern is the fresh and marine waters of the Peconic Estuary Watershed (Figure 1), which are currently being evaluated for potential nitrogen management actions (Peconic Estuary Partnership 2020). This study focuses on four water bodies that are broadly representative of the hydrologic settings in and around the Peconic Estuary Watershed (Figure 1): the Peconic River, Great Peconic Bay, eastern Long Island Sound, and the Shinnecock, Moriches, and Narrow Bays within the South Shore Estuary Reserve (hereafter referred to as “eastern South Shore Estuary”).

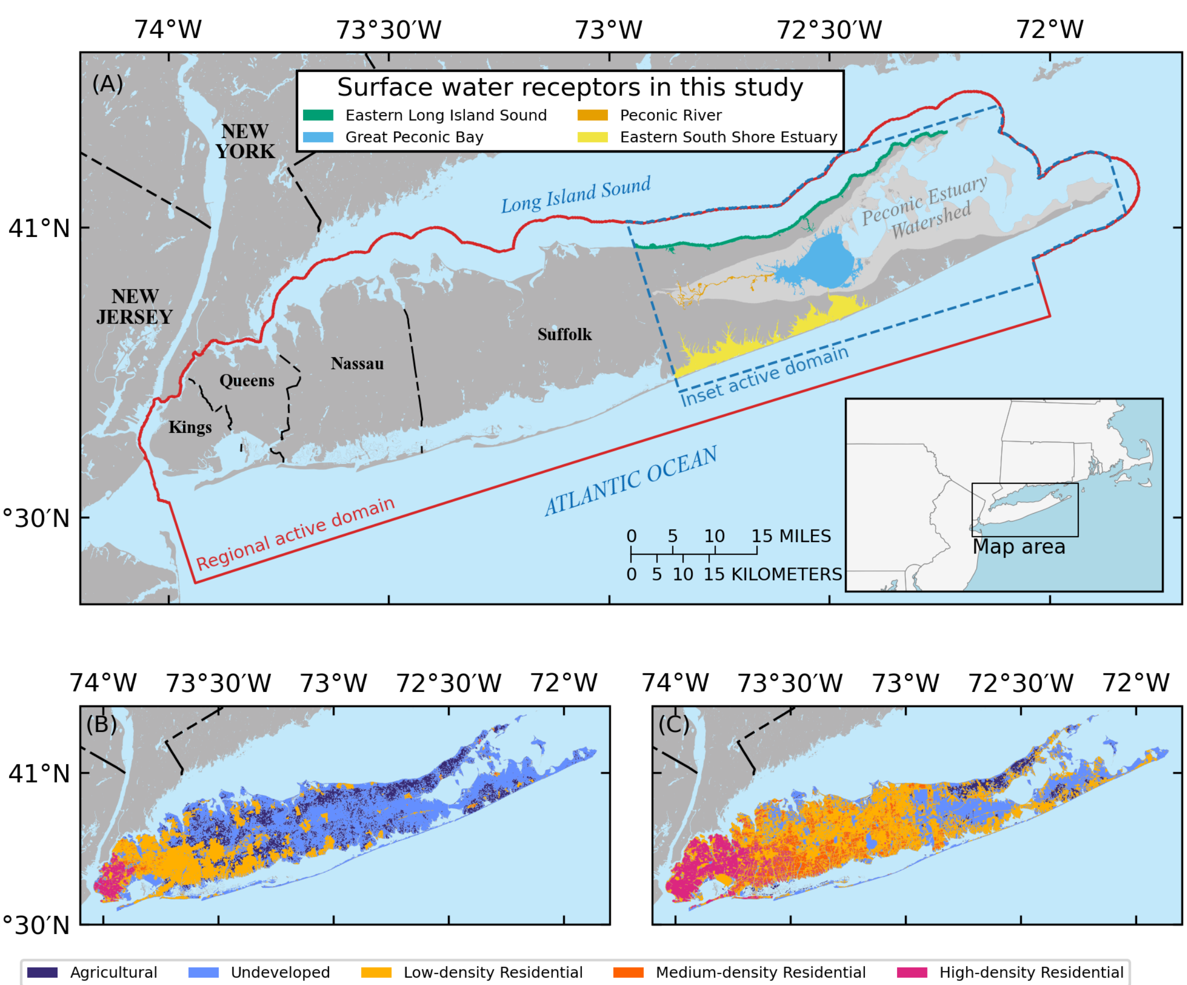

Figure 1. Study area showing (A) regional model active domain, inset model active domain, the Peconic

Estuary Watershed in light gray (Peconic Estuary Partnership, 2020), and delineations of the surface water receptors Peconic River (orange), Great Peconic Bay (blue), eastern Long Island Sound (green), and eastern South Shore Estuary (yellow). Land use on Long Island in (B) 1938 and (C) 2019 (Monti et al., 2024).

3 Methods

3.1 Historical nitrogen inputs and attenuation

Historical nitrogen inputs were estimated by Monti et al. (2024) for the entirety of Long Island on a 500-foot by 500-foot (154.4 m) cell grid using land use records, agricultural censuses, and population censuses. Nitrogen input arrays for six non-point sources were estimated: septic systems, atmospheric deposition, residential and golf course fertilizer, agricultural fertilizer, livestock waste, and household pet waste (Figure 2A).

For this study, nitrogen inputs were reduced by lumped attenuation fractions representing removal of nitrogen due to plant uptake, volatilization, and denitrification (Table 1 and Figure 2B). The lumped attenuation fractions, which represent the proportion of nitrogen removed, align with those chosen by nitrogen management decision makers on Long Island based on literature review and expert knowledge (Table 1; Suffolk County Department of Health Services and CDM Smith, 2020). Although this study does not explore the inherent uncertainty in the attenuation fractions, the exact values selected do not affect our analysis of legacy groundwater nitrogen effects because the fractions are applied uniformly across the tested methods of estimating receptor nitrogen loads.

In addition to the attenuation fractions, impervious surfaces were assumed to eliminate nitrogen transport from the ground surface to the water table due to overland flow in urban areas being routed to stormwater sewer systems. Therefore, atmospheric deposition and pet waste nitrogen inputs in each cell were further attenuated by that cell's fraction of impervious land cover. An impervious land cover fraction array was estimated for each year in the 1900-2019 period by extrapolating conterminous U.S. data available for five years: 1974, 1982, 1992, 2002, and 2012 (Falcone, 2017). The data were mapped to the Long Island grid, at a uniform resolution of 500 feet (Monti et al., 2024). Arrays of impervious surface for years between the existing five arrays were generated using linear interpolation. For years prior to 1974, the 1974 impervious land cover fraction array was scaled by annual changes in human population arrays estimated for each pre-1974 year (Monti et al., 2024).

To serve as inputs to the models, 120 total annual nitrogen input arrays (also covering the entire model domain at a uniform resolution of 500 feet) were created for 1900-2019 by per-cell summation of the six attenuated sources for each year.

Table 1. Attenuation fractions used to calculate attenuated nitrogen inputs from potential inputs estimated for 1900-2019 (Monti et al., 2024). Selected values are based on fractions determined by a technical advisory committee (Suffolk County Department of Health Services and CDM Smith, 2020). For nitrogen inputs on morainal deposits (Walter et al., 2024), 0.15 was added to the attenuation fraction, also in accordance with the advisory committee (Suffolk County Department of Health Services and CDM Smith, 2020).

Non-point source	**Nitrogen attenuation fraction**
Septic	0.16
Atmospheric deposition	0.60
Residential and golf course fertilizer	0.70
Agricultural fertilizer	0.60
Livestock waste	0.50
Household pet waste	0.50

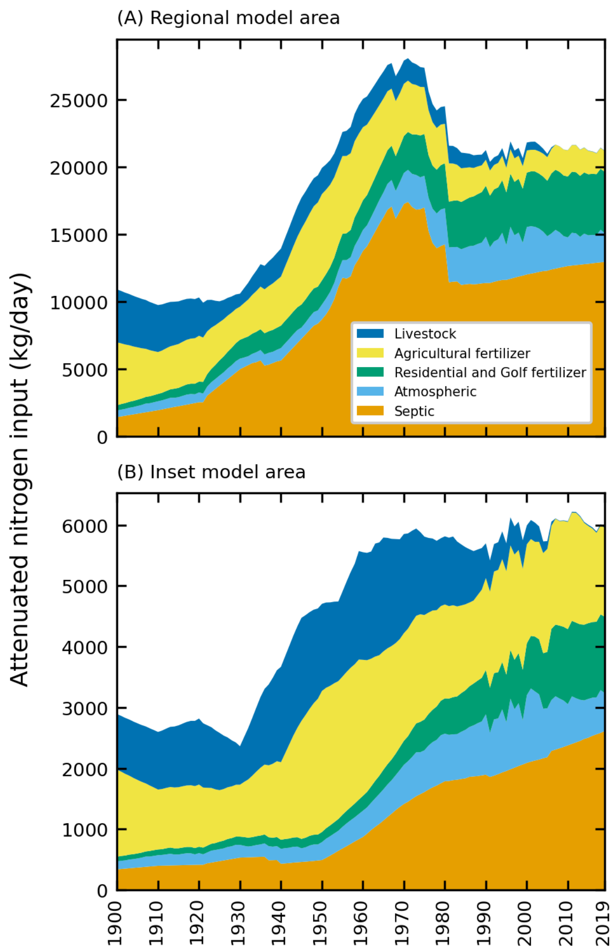

Figure 2. 1900-2019 changes in attenuated nitrogen inputs in kilograms per day (kg/day) estimated for (A) the regional model area and (B) the inset model area using nitrogen attenuation fractions from Table 1. Note that the two y-axis scales are not the same to allow for easier inspection of year-to-year changes. Pet waste was not included because the loads were too small to be visible.

3.2 Numerical simulations of groundwater flow and nitrogen transport accounting for transport lags

Numerical simulations of groundwater flow and nitrogen transport were performed using a set of models developed from a previously published transient regional model that extends across the entirety of Long Island (Walter et al., 2024). The regional model was used in its original transient form to simulate nitrogen transport from 1900-2019, but two steady-state versions were also developed. The steady-state regional models were used both to delineate surface water receptor contributing areas then used to estimate instantaneous loads via NLM, and to numerically simulate 1900-2019 nitrogen transport under steady-state flow conditions. The transient regional groundwater flow and transport model was also coupled to an inset model developed to contain the eastern Long Island surface water receptors that were the focus of this study (Figure 1). The inset model was used to simulate potential future nitrogen transport under possible nitrogen management scenarios to inform community decision making. The details of the various models are described below, and their connections and dependencies are outlined in a flow diagram in the Supporting Information (Figure S1).

3.2.1 Groundwater flow

A previously developed MODFLOW 6 (Langevin et al., 2017) model of transient, variable density groundwater flow in the Long Island aquifer system from 1900-2019 (Walter et al., 2024) was used to simulate historical nitrogen transport from 1900-2019. The regional model covers the entirety of Long Island (Figure 1), discretizing the 3-D aquifer system into 20 layers, 348 rows, and 1309 columns, with a uniform horizontal cell discretization of 500 feet. The 1900-2019 period is discretized into 120 annual stress periods, each divided into three time steps. This variable density groundwater flow model was calibrated against historical records of groundwater elevations, stream baseflows, and groundwater chloride concentrations (Walter et al., 2024). The Long Island aquifer system is bounded laterally by a dynamic saltwater interface, and the simulation of groundwater flow requires the simulation of the time-varying position of that interface. The model solves for the dynamic interface position using the MODFLOW 6 Buoyancy package (Langevin et al., 2017, 2022) coupled with the MODFLOW 6 Groundwater Transport Model, as described in detail in Walter et al. (2024).

An additional nine layers were added to the 20-layer regional model by halving the nine layers representing the Magothy aquifer (layers 7 through 15) into eighteen layers using the Python package FloPy version 3.8.0 (Bakker et al., 2016, 2024). The additional layers were added to reduce the effects of numerical dispersion on simulated vertical nitrogen transport in that formation. Each new layer pair retained the aquifer properties of the corresponding original layer, and wells within the original nine layers were relocated to one of the new layers based on the elevations of well screen centers. This modified regional model is documented in Jahn and Walter (2025) and simulates transient variable density groundwater flow under time-varying annual hydrologic stresses from 1900 to 2019 in an identical fashion to the original regional model, resulting in functionally identical groundwater system budgets (Figure S2).

An inset model domain of the 29-layer regional model domain (Figure 1) was created using a 250-by-250-foot (76.2 m) grid discretization to simulate potential future nitrogen transport under theoretical nitrogen management scenarios. The inset model was developed to assist local communities on eastern Long Island in making science-based decisions on nitrogen management with the Peconic Estuary Watershed (Figure 1). The finer discretization better resolves the extents of the surface water receptors and their resulting simulated nitrogen loads, and represents dispersion and advection at a more accurate scale. Model boundary cell representations of surface water bodies in the inset model were manually discretized to the new grid using satellite imagery, following the method described by Walter et al. (2024). Each finer resolution inset cell inherited aquifer properties directly from the corresponding "parent" cell in the regional model. The inset and regional model grids were tightly coupled using the local grid refinement (LGR) utility available in FloPy, which calculates all of the necessary flow and transport exchanges at the interfaces of the regional and inset models (Hughes et al., 2023).

Steady state versions of the regional and inset models were also developed. Two steady-state versions of the regional model were used to delineate surface water body contributing areas and explore the effects of simulating historical nitrogen transport using a simpler flow modeling approach. Both steady-state version of the regional model used 1900-2019 mean annual recharge rates, but one had 2010-2019 mean annual well pumping conditions and one had no pumping. A steady-state version of the inset model was generated using the 2010-2019 mean annual recharge and well pumping conditions to simulate potential future nitrogen transport.

Groundwater contributing areas and travel times for the Peconic River, Great Peconic Bay, Long Island Sound, and South Shore Estuary were defined from the steady-state models using the particle-tracking software MODPATH 7 (Pollock, 2016) and documented in Jahn and Walter (2025). The delineations of each receptor were constrained to the domain of the inset model (Figure 1A) to facilitate estimation of loads using both the regional and inset models. Thus, the western parts of Long Island Sound and South Shore Estuary are not considered in this study. One particle was assigned to each water table cell, defined as the uppermost active layer in each row and column combination. The particles were tracked through the steady-state flow fields until termination in either one of the surface water body boundary cells or after 100,000 years of tracking, whichever occurred first. The groundwater contributing areas were used to calculate instantaneous nitrogen loads, as described in Section 3.3.

3.2.2 Nitrogen transport

Nitrogen transport in the Long Island aquifer system was simulated with the MODFLOW 6 Groundwater Transport Model (Langevin et al., 2022), which solves for cell-by-cell nitrogen concentrations using a finite-difference approach. Sediment porosity varied by geologic unit and ranged from 0.29 to 0.4 (Walter et al., 2024). The Long Island aquifer system generally is oxic, and nitrate is assumed to be the predominant species of nitrogen in the aquifer (Walter, 1997). Nitrate is chemically conservative, and transport is predominately by advection, which refers to the transport of a solute with average groundwater velocity. An additional component of transport is dispersion, which refers to the spreading of mass resulting from sediment heterogeneity. The finite-difference approximation introduces numerical dispersion in simulated concentrations; the estimated impact of that numerical dispersion on the simulated longitudinal dispersivity, as determined from simulated velocities and the discretization of time and space, is about 250 feet (Walter et al., 2024). This approximation is assumed to reasonably represent actual dispersivity at the scale of the analysis; additional dispersion was not specified.

Flow fields from the steady-state and transient regional models described above were used to simulate 1900-2019 nitrogen transport using 120 annual stress periods. Nitrogen concentrations at the water table were calculated by multiplying each annual recharge array, which account for the physical flow processes prior to the water table, by the corresponding annual attenuated nitrogen array, which account for potential attenuation along the transport pathways. Recharge nitrogen concentration arrays were calculated for each year in 1900-2019 by dividing each annual total attenuated nitrogen input array by the corresponding annual volumetric recharge array (Walter et al., 2024). To provide initial aquifer nitrogen concentrations for the 1900-2019 nitrogen transport model, a "warm start" model was run under steady-state 1900 hydrologic conditions with transient total nitrogen input arrays estimated for 1790 to 1899 by linearly reducing the 1900 total nitrogen input array back to the earliest reliable population estimate for Long Island: 36,949 in 1790 (Bureau of the Census, 1901). The average annual fractional change in Long Island population between 1900 and 1790 (0.00886) was used to scale the 1900 nitrogen concentrations for each year between 1790 and 1900. The final aquifer nitrogen concentrations in the warm start model were used as initial concentrations for the 1900-2019 nitrogen transport simulations.

To provide a smoother transition from historical nitrogen transport simulation at the regional model discretization to future transport simulation at the inset model discretization, a coupled transient transport simulation was completed for the 2010-2019 period using 10 annual stress periods and initial 2010 aquifer conditions (groundwater elevations, densities, and nitrogen concentrations) drawn from the transient regional model. Potential future nitrogen transport was simulated using the steady-state (mean 2010-2019 conditions) inset flow fields described above and potential future nitrogen inputs.

Nitrogen transport through the steady-state inset flow field was simulated using annual stress periods for a hypothetical future 2020-2119 period, with initial 2020 aquifer nitrogen concentrations drawn from the final nitrogen concentrations simulated by the 2010-2019 coupled models. Two future nitrogen source scenarios were simulated using the steady state inset flow field: 1) continued 2019 sources and 2) removal of terrestrial anthropogenic nitrogen sources (i.e., the only input after 2019 was atmospheric deposition). Scenario results are documented in Jahn and Walter (2025).

3.3 Estimation of receptor nitrogen loads

Nitrogen loads to receptors were estimated for the Peconic River, Great Peconic Bay, Long Island Sound, and South Shore Estuary (Figure 1A, Section 2.1). The delineations of each receptor were constrained to the domain of the inset model (Figure 1) to facilitate estimation of future loads using the inset model. Thus, the western parts of the Long Island Sound and South Shore Estuary are not considered in this study.

Two approaches were used to estimate nitrogen loads to receptors. The simpler approach, an instantaneous load using NLM, involved summing the attenuated nitrogen inputs within each receptor contributing area estimated from steady-state particle tracking described in Section 3.2.1. Thus, an instantaneous total nitrogen load was calculated for each year in the 1900-2019 period. The approach used for the numerical simulations required more steps. First, nitrogen loads were calculated for each cell within a surface water body receptor, which were defined in the groundwater flow model as either a drain boundary, general head boundary, or constant head boundary. MODFLOW 6 can record both concentrations in and groundwater fluxes across boundary cells, so the nitrogen concentration in each receptor cell was multiplied by the cell groundwater flux to calculate the nitrogen load reaching that receptor cell. The cell-specific loads within the receptor delineations (Figure 1) were summed to calculate a total annual groundwater nitrogen load to each receptor.

4 Results

4.1 Receptor contributing area travel times and nitrogen inputs

Contributing areas to surface water receptors are a function of the groundwater flow field, which is in turn defined by hydrogeology, recharge rates, and anthropogenic withdrawals by pumping wells. A set of contributing areas to the four receptors are defined under the steady-state flow field that develops under mean annual recharge rates from 1900-2019 in the absence of pumping wells (Figure *3*A). The Peconic River has the smallest contributing area, followed by Great Peconic Bay, then Long

Island Sound and South Shore Estuary with similarly sized areas (Table S1). The addition of pumping wells (mean annual rates from 2010-2019) that intercept natural groundwater flow paths (Figure *3*B) reduces the size of contributing areas: the contributing areas when pumping wells are included are slightly smaller, with areal reductions of 18.4% for Peconic River, 4.5% for Great Peconic Bay, 10.7% for Long Island Sound, and 13.9% for South Shore Estuary (Table S1).

The spatial distribution of steady-state travel times generally increases with distance from surface water bodies, with travel times <1 year near the coastline and >100 years near groundwater flow divides (Figure *4*). The addition of pumping removes flow paths from the receptor contributing areas (Figure *4*B) but has a limited effect on the travel times that remain within the contributing areas, resulting in similar travel time distributions under both pumping and non-pumping conditions (Table S1). With pumping, the median travel times for all the receptors are between 15 and 26 years, but the interquartile ranges vary substantially: from 44 years for Great Peconic Bay to 187 years for South Shore Estuary. The Peconic River and South Shore Estuary travel times have a more bimodal distribution than Great Peconic Bay and Long Island Sound, and the most pronounced differences between all four distributions occur around the 75th percentile (Table S1, Figure S3).

Nitrogen inputs within the steady state contributing areas change during the 1900-2019 period. The septic, residential fertilizer, and atmospheric nitrogen input trends in the four contributing areas are largely similar (Figure 5), mirroring the trend seen in the inset model domain as a whole of steady increases in those three inputs after 1950 (*Figure 2*A). The agricultural fertilizer and livestock input histories differ more between the contributing areas. Great Peconic Bay and South Shore Estuary contributing areas show similar trends to the whole inset domain, with increasing and then consistent combined agricultural and livestock inputs until the 1980-2000 period, when agriculture and livestock inputs begin to decrease (Figure 5B and D). The Peconic River and Long Island Sound agricultural and livestock input histories differ from the broader inset domain trends: there was a significant spike in livestock nitrogen input from 1940-1970 in the Peconic River contributing area (Figure 5A) due to intensive duck farming (Monti et al., 2024), and agricultural nitrogen fertilizer inputs have continued to increase through the 2010s in the Long Island Sound contributing area due to continued agricultural activities on the North Fork of Long Island (Figure 1B and C).

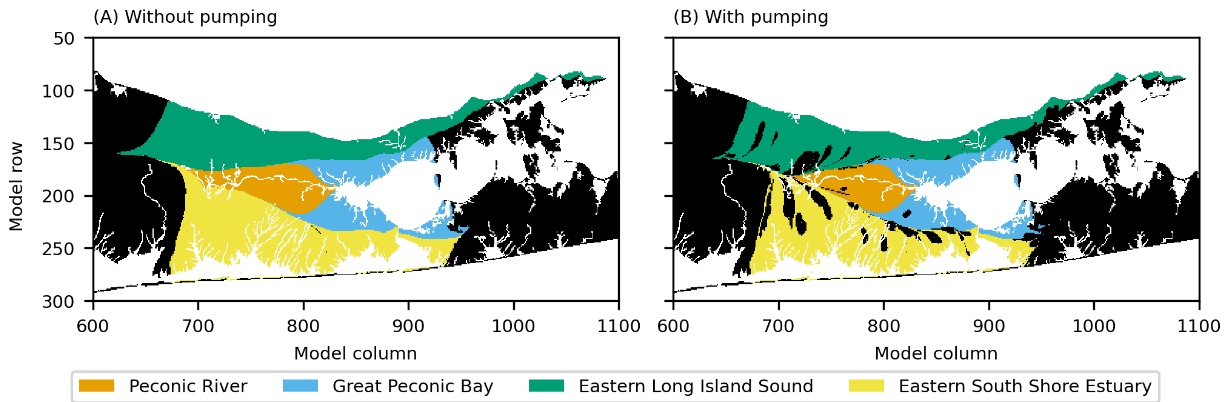

Figure 3. Simulated steady-state contributing areas under mean 1900-2019 recharge conditions (A) without pumping wells and (B) with 2010-2019 mean pumping for the Long Island aquifer system. Black areas are those outside of the contributing areas of the four considered water surface water receptors. Additional black areas in panel B are due to changes in the flow field due to pumping wells.

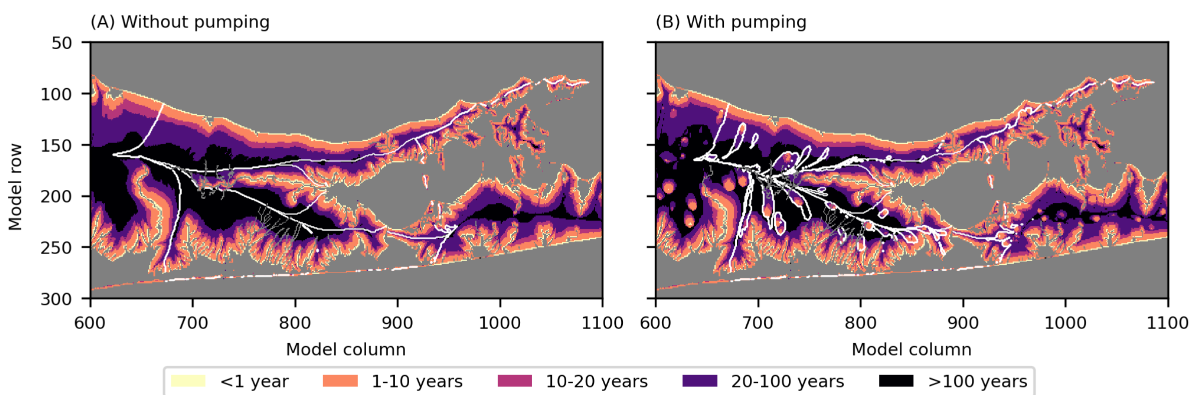

Figure 4. Steady-state groundwater travel times under mean 1900-2019 recharge conditions (A) without pumping wells and (B) with 2010-2019 mean pumping for the Long Island aquifer system. The white lines correspond to the boundaries of the contributing areas in Figure 3.

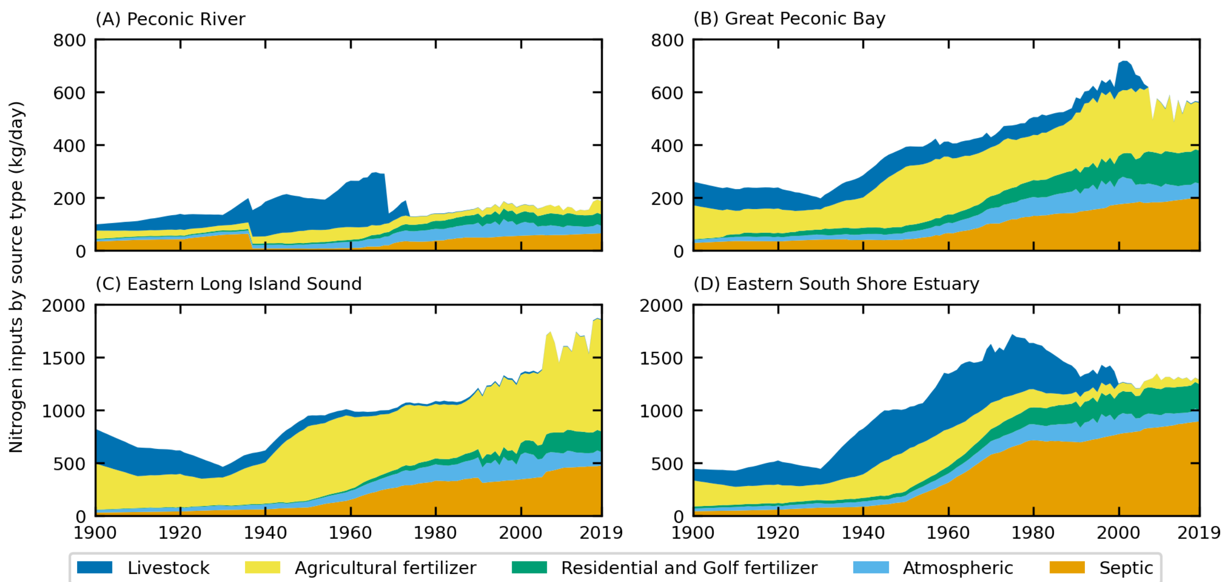

Figure 5. Nitrogen inputs within steady state contributing areas (equivalent to instantaneous loads) under mean 1900-2019 recharge conditions with mean 2010-2019 pumping (Figure 3B) for (A) Peconic River, (B) Great Peconic Bay, (C) Long Island Sound, and (D) South Shore Estuary for the Long Island aquifer system. Inputs are derived from Monti et al. (2024).

4.2 Simulated nitrogen within the Long Island aquifer system

The total simulated nitrogen mass in the Long Island aquifer system is the difference between the amount of nitrogen input at land surfaces and the nitrogen discharged to surface water receptors, withdrawn from pumping wells, or discharged to seawater along the freshwater-saltwater interface. From 1900 to 2019, nitrogen mass within the Long Island aquifer system nearly doubled (Figure *6*). The increases in aquifer nitrogen are a function of both increases in nitrogen inputs and nitrogen traveling along longer flow paths reaching deeper parts of the aquifer. There are two broad inflection points under both flow conditions: the rate of mass accumulation in the aquifer increases between 1930-1940 and decreases after 1970-1980 to a rate slightly higher than the pre-1930 accumulation rate. Both changes are associated with the significant changes in nitrogen inputs from septic systems, which increased approximately six-fold between 1920 and 1970 due to population growth on Long Island (Figure *2*). Following sewer system installations in the 1970s, the septic system nitrogen inputs were reduced (Monti et al., 2024), and the aquifer nitrogen accumulation rate decreased. However, despite total nitrogen inputs remaining relatively steady after 1980, nitrogen mass continues to accumulate in the Long Island aquifer system during that time (Figure *6*) due to groundwater travel times longer than the 120-year simulated period (Figure S3). As this much older, essentially nitrogen-free, predevelopment groundwater is replaced by younger groundwater with higher nitrogen concentrations, the total nitrogen mass within the aquifer increases and will continue to increase until the older nitrogen-free water is replaced by younger nitrogen-containing water. Because of this long-term groundwater system “memory,” gains in aquifer nitrogen mass during periods of high nitrogen input are not immediately lost when nitrogen inputs are reduced. Storage of nitrogen in groundwater has previously been described for four major river basins, with simulated nitrogen accumulation in the Long Island aquifer system following a similar pattern as accumulation simulated for groundwater in the Mississippi River basin (Liu et al 2024)

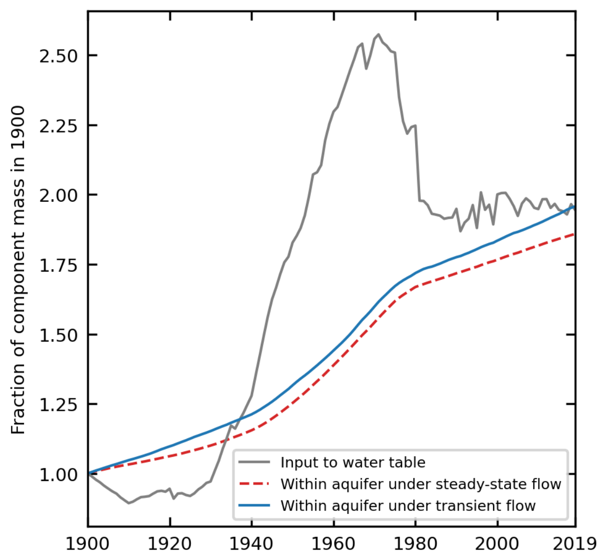

Figure 6. Temporal changes in annual attenuated nitrogen inputs to (gray line, sum of the inputs from the separate sources in Figure 2) and simulated nitrogen mass within the entire Long Island aquifer system under steady-state (dashed red line) and transient (blue line) flow conditions.

4.3 Estimated historical nitrogen loads to receptors

Maximum calculated nitrogen loads to surface water receptors varied substantially depending on the estimation approach (Figure 7). The largest estimated annual instantaneous load estimated from NLM (which does not explicitly account for legacy nitrogen) over the period 1900-2019 in the Peconic River were 313 kg/day (in 1966), 725 kg/day in Great Peconic Bay (in 2001), 2,004 kg/day in Long Island Sound (in 2018), and 1,858 kg/day in South Shore Estuary (in 1975). The largest nitrogen loads estimated using the transport model under steady-state flow conditions typically occurred within one year of the largest estimated instantaneous loads, but at different magnitudes. The largest load under steady-state flow transport was substantially higher in Peconic River (683 kg/day, 1966), slightly lower in Great Peconic Bay (641 kg/day, 2002), substantially lower in Long Island Sound (978 kg/day, 2019), and similar in South Shore Estuary (1756 kg/day, 1976), than the instantaneous loads. The largest loads under transient flow transport also occurred within one year of the other methods, except for South Shore Estuary, which experienced peak load in 1983, 7-8 years after the peak loads from instantaneous and steady-state flow transport loads. Nitrogen loads under transient flow conditions were generally higher than those under steady-state conditions, except for loads to the Peconic River for 1910-1970, when significantly higher nitrogen loads were simulated under steady-state flow (Figure 7).

The instantaneous loads calculated from contributing areas without pumping and with pumping (top and bottom, respectively, of gray filled areas in Figure 7) represent the range of nitrogen loads that would be calculated from NLM in any given year during 1900-2019 period. The simulation with no pumping generally represents the earlier part of the 20th century when there were relatively few groundwater withdrawals in eastern Long Island. Simulations with pumping represent increased withdrawals associated with population growth in the later part of the 1900-2019 period. Interannual variability in simulated nitrogen loads was typically greatest under transient flow conditions, as expected given the potential for compounding variability in both flow and nitrogen inputs. However, after 2000, the instantaneous load variability for Long Island Sound was greater than the transient load variability (Figure 7C), indicating that an increase in nitrogen input variability during that time may be muted by transient changes in hydrologic conditions. Interannual variability was generally smallest under steady state flow conditions, indicating that consistent flow patterns smooth out variability in nitrogen inputs.

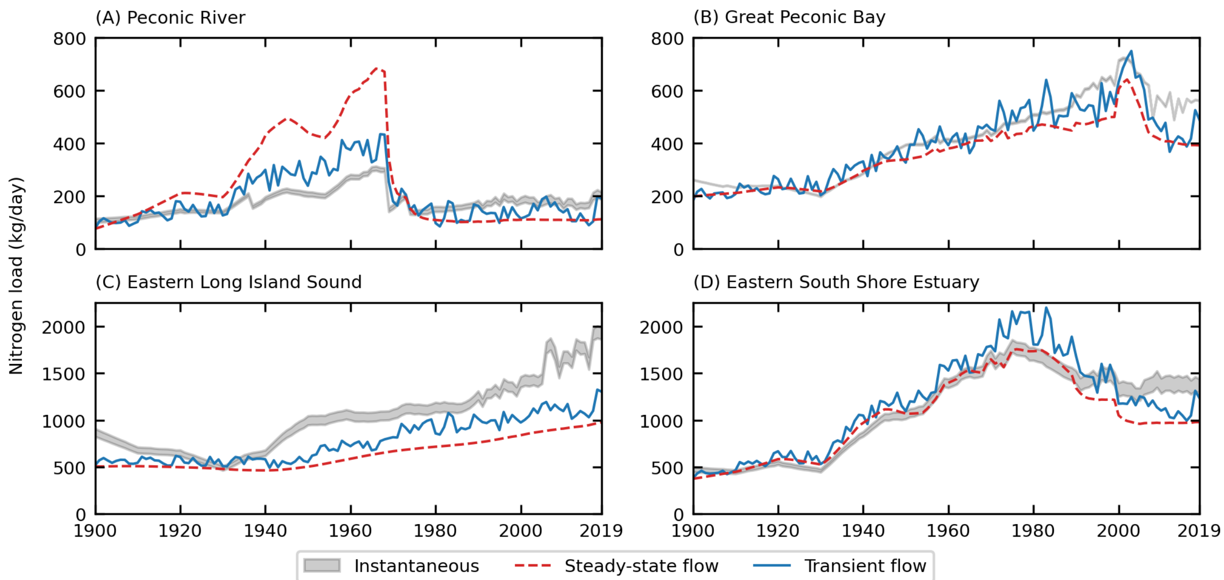

Figure 7. Simulated annual receptor nitrogen loads from 1900-2019 for the (A) Peconic River, (B) Great Peconic Bay, and (C) South Shore Estuary, and (D) Long Island Sound. The gray filled curves represent the instantaneous loads calculated for contributing areas from steady-state flow fields without pumping wells (top of filled area) and with pumping wells (bottom of filled area).

4.4 Estimated nitrogen load responses to management actions

Two potential management actions were simulated with the steady-state 2010-2019 mean annual conditions inset model: 1) continued 2019 nitrogen sources, and 2) removal of all land use-based nitrogen sources (atmospheric deposition retained). Although unrealistic, the removal action is an end-member scenario that allows us to better understand the least amount of time required to reach desired reductions in current receptor nitrogen loads (Suffolk County Department of Health Services and CDM Smith, 2020). In all four receptors, continued 2019 sources result in continued increases in nitrogen loads over time, which shows that the MODFLOW 6 simulated 2019 loads are not yet in equilibrium with the 2019 nitrogen inputs within the watersheds (Figure *8*). The nitrogen loads simulated with MODFLOW 6 asymptotically approach but never reach the instantaneous 2019 nitrogen loads because of groundwater travel times greater than the 100 simulated years of transport (Figure S3). Great Peconic Bay comes closest with a nitrogen load of 386 kg/day in 2119, about 94% of the instantaneous nitrogen load of 412 kg/day (Figure *8*B). In contrast, the Peconic River reaches 113 kg/day by 2119, only about 61% of the instantaneous nitrogen load of 187 kg/day (Figure *8*A). The Peconic River and Long Island

Sound showed similar asymptotic behavior, reaching approximately 87% and 86% of their respective instantaneous nitrogen loads by 2119.

The amount of time needed for the simulated nitrogen loads to reach 30%, 50%, and 70% reductions from current nitrogen loads varies between receptors by as much as 1-3 decades (Figure *8*). Nitrogen loads in the South Shore Estuary were reduced by 70% after 20 years of transport, nitrogen loads in the Peconic River reached a 70% reduction after 33 years, and Great Peconic Bay and Long Island Sound both reached a 70% reduction after approximately 45 years. Although the 70% reduction time is similar for Great Peconic Bay and Long Island Sound, the earlier reduction rates differ. Nitrogen loads in Great Peconic Bay reached a 30% reduction within 4 years and 50% reduction within 13 years, similar to the Peconic River and South Shore Estuary. Meanwhile, Long Island Sound had an initial response to nitrogen management that was about a decade slower as a result of a larger proportion of nitrogen inputs to groundwater pathways with longer travel times (Figure *9*, discussed further in Section 5.1), reaching 30% and 50% reductions after 13 and 24 years, respectively.

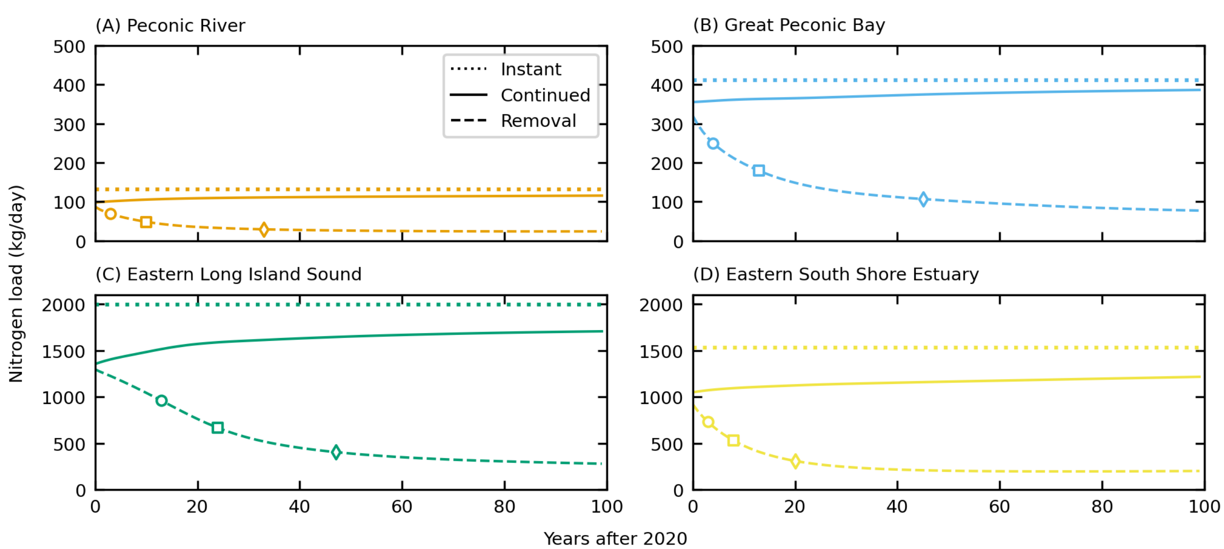

Figure 8. Simulated future receptor nitrogen loads under continued 2019 inputs (solid lines) and removal of land-use nitrogen inputs (dashed lines), with corresponding 2019 instantaneous loads for reference (dotted lines). Markers on the dashed lines represent percent reductions from 2020 loads: 30% (circles),

50% (squares), and 70% (diamonds). Note the differences between the y-axis scale of panels A and B, and the scale of panels C and D.

5 Discussion

5.1 Legacy nitrogen in groundwater

Nitrogen loads to receptors vary as a function of the unique temporal and spatial distributions of nitrogen inputs within receptor contributing areas. How much those antecedent inputs change over time and how much of the nitrogen is input to longer and deeper groundwater flow paths will determine the importance of legacy nitrogen to the present-day nitrogen loads to receptors. Generally, when antecedent nitrogen inputs are higher than current nitrogen inputs, estimated nitrogen loads that account for legacy nitrogen conditions will be higher than those that do not account for legacy nitrogen. The opposite can generally be expected when antecedent nitrogen inputs are lower than current nitrogen inputs, due to receptors receiving both younger groundwater with high nitrogen concentrations and older groundwater with lower nitrogen concentrations, resulting in a dilution of total receptor nitrogen load. Differences in nitrogen loads estimated by NLM, steady-state groundwater flow simulations, and transient groundwater flow simulations highlight periods when and locations where quantifying legacy nitrogen could improve accuracies of simulated load estimates.

The time-varying total nitrogen inputs are mapped to the steady-state groundwater travel time bands (Figure *5*) to assess the temporal changes in the spatial distribution of the inputs and their relative proportioning to the travel time bands (Figure *9*). The temporal patterns in these proportions (Figure *9*) align clearly with the patterns in the differences between the instantaneous nitrogen loads from NLM and those from the numerical simulations (Figure *10*). Differences between the instantaneous loads and steady-state simulated loads represent nitrogen transport through the aquifer functioning as temporal sinks and sources of nitrogen to surface water receptors. If a steady-state flow load is lower than the concurrent instantaneous loads, there is a net temporal sink for that year that cannot be accounted for in NLM (i.e., some nitrogen inputs from that year went to longer groundwater flow paths and/or older groundwater discharging that year had lower nitrogen concentrations relative to that year's inputs). If an annual steady-state flow load is higher than the concurrent instantaneous load, there is a net temporal source for that year (i.e., older groundwater discharging that year had higher nitrogen concentrations relative to that year's inputs). This is also true for differences between the transient flow loads and instantaneous loads, with the additional effect due to the annual changes in hydrologic conditions.

The NLM and steady-state flow simulation approaches estimated similar nitrogen loading (typically <10% difference) for Great Peconic Bay and South Shore Estuary during 1920-1980 (Figure *10*B and D), when the contributing areas for those two receptors had consistent year-to-year spatial distributions of nitrogen inputs. So although total nitrogen inputs increased significantly within the two contributing areas from 1920-1980 (Figure 5B and D), how those total nitrogen inputs were proportioned based on steady-state groundwater travel time did not change substantially (Figure 9B and D). These consistent spatial distributions, along with few groundwater ages greater than 20 years, results in similar estimates from NLM and the steady-state flow simulations for those two receptors. Although the instantaneous loads from NLM do not explicitly account for legacy N, minimal changes in nitrogen proportioning by steady-state travel times allows the instantaneous loads to approximate a steady-state simulation result for contributing areas with nitrogen inputs consistently proportioned by groundwater travel times.

Compared to 1920-1980, larger differences in the instantaneous loads and the loads simulated under steady-state flow occur in both Great Peconic Bay and South Shore Estuary during the periods 1900-1919 and 1981-2019, when steady-state nitrogen loads were as much as 25% lower than instantaneous loads (*Figure 10*). Lower simulated steady state loads during 1900-1919 are likely due to the initial (1900) aquifer concentrations simulated from the 1790-1899 warm start simulation being lower than the 1900 nitrogen inputs. The instantaneous load in 1900 was estimated from land use for that year, whereas the initial concentrations in 1900 were derived from the same land use but sequentially decreased using population decreases from 1900 to 1790 (refer to Section 3.2). Lower simulated steady-state loads after 1980 may be due to the increase in the proportion of nitrogen inputs to older travel time bands after 1980 (Figure *9*B and D), which would result in lower loads in subsequent years until that older water reached receptors.

Differences between loads simulated for steady-state flow and instantaneous loads from NLM are more pronounced in nitrogen load estimates for the Peconic River (Figure *10*A). The difference between Peconic River steady state flow nitrogen loads and instantaneous nitrogen loads exceeds 100% from 1930 to 1970 and is associated with livestock nitrogen inputs (Figure 5A) concentrated within the <1 year travel time bands during 1930 to 1970 (Figure *9*). The large nitrogen loads in the <1 year groundwater bands are compounded by legacy nitrogen transported to the Peconic River along longer groundwater flow paths. This additional component of nitrogen is not accounted for in the instantaneous nitrogen load calculations, which only sum up inputs from within each year, thus resulting in significantly lower nitrogen loads. A similar trend exists in the transient flow nitrogen loads but is dampened by the transient hydrologic conditions that adjust the nitrogen input travel time band location each year, reducing the amount of nitrogen input to groundwater travel times <1 year observed in the steady-state flow simulation.

Nitrogen loads to Long Island Sound simulated under steady-state flow were consistently less than instantaneous loads, by as much as 50%, during the 1900-2019 period (Figure 10C). The Long Island Sound contributing area has the largest fraction of nitrogen associated with long travel times (Figure 9C), resulting in a transport lag as nitrogen moves along longer groundwater paths and a consistent lack of agreement between steady-state and instantaneous loads. Nitrogen loads estimated using NLM should also be larger than those simulated under steady-state transport (representing an overestimate using NLM) if nitrogen inputs in the watershed increased consistently over time, which is consistent with inputs in the Long Island Sound steady-state contributing area (Figure 5C). The watershed is one of the few remaining intensive agricultural areas on Long Island and agricultural nitrogen inputs have continued to increase, primarily within interior areas with large travel times (*Figure 9*C).

Nitrogen loads simulated under steady-state and transient flow conditions generally follow similar trends, but the addition of time-varying pumping and recharge results in interannual variability superimposed on the underlying trends arising from changing nitrogen sources (Figure 7). Deviations between steady-state and transient flow conditions arise due to differences between the mean hydraulic stresses simulated under steady-state and the actual annual stresses for the 1900-2019 period, one result of which is more total nitrogen within the aquifer under transient flow conditions than steady-state (Figure *6*). The largest deviations in steady-state and transient loads are to the Peconic River prior to the early 1970s. This large deviation may be due to particularly large differences between the transient groundwater travel times for that period and the steady-state travel time distributions under mean conditions. The 1900-2019 mean annual recharge used in the steady-state flow simulation is higher than the transient recharge during the 1930-1970 period (Figure S4) and combined transient pumping rates likely result in contributing areas in the transient simulation that vary substantially from the steady-state contributing areas. The lower transient recharge during that period would result in lower groundwater gradients and slower groundwater velocities, and thus smaller < 1 year travel time bands than those for the steady-state simulation (*Figure 4*). This in turn would result in a relatively larger fraction of nitrogen inputs to older travel time bands, enhancing overall nitrogen lag times.

Simulated nitrogen loads seem to generally align with historical observations of estuary health degradation on Long Island. For example, the first coastal eutrophication observed on Long Island were chlorophyte blooms, known as green tides, in parts of the South Shore Estuary in the early 1950s (Ryther, 1989). Green tides were associated with duck farming adjacent to streams and estuaries, which resulted in large increases in nitrogen inputs from livestock starting in the late 1940s (Figure 5, Monti et al., 2024) that contributed to increases in nitrogen loads to the South Shore Estuary (Figure 7). Historically large shellfish harvests in eastern Long Island waters that occurred from the mid-1950s to the mid-1980s indicated good ecological health (MacKenzie, 1997). During that same period, human populations and associated nitrogen sources increased substantially (Monti et al., 2024), indicating that the lag due to nitrogen transport through the aquifer buffered ecological health for several decades, even as terrestrial nitrogen loading increased. Brown tides in Great Peconic Bay were first observed in the mid-1980s (Cosper et al., 1989; Nuzzi and Waters, 1989) following a long period of steady increases in simulated nitrogen loads to surface waters since the 1940s (Figure 7). Transient nitrogen loads to receptors, including Great Peconic Bay, show a series of peaks between 1970 and 1985, resulting from periods of high recharge (Figure 7). The largest transient nitrogen load occurred in 1983, which corresponds to the wettest year for the period 1900-2019, further demonstrating the importance of interannual variations in groundwater flow rates on nitrogen loading and resulting algal blooms (Laroche et al., 1997). Simulated nitrogen loads remained high through the early 2000s, generally corresponding to the first occurrences of red tides and harmful algal blooms in eastern Long Island waters between 2002 and 2006 (Gobler et al., 2008).

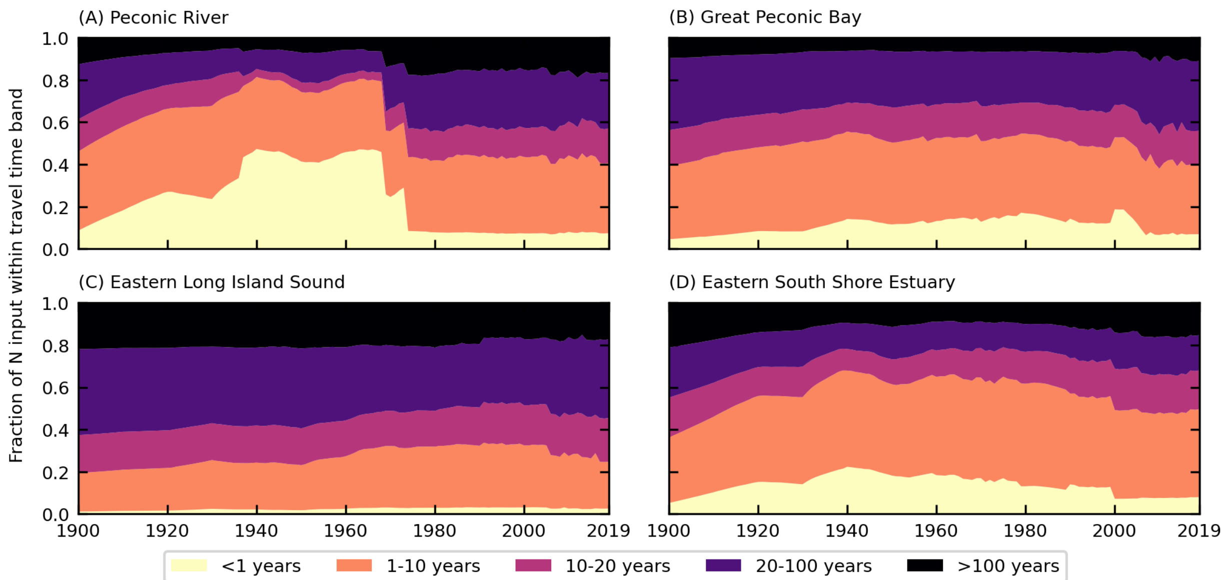

Figure 9. Annual nitrogen inputs grouped by the steady-state groundwater travel time bands in Figure 4, normalized to the total annual nitrogen input for each year. For reference, total annual nitrogen inputs within each contributing area are shown as gray lines *Figure 7*.

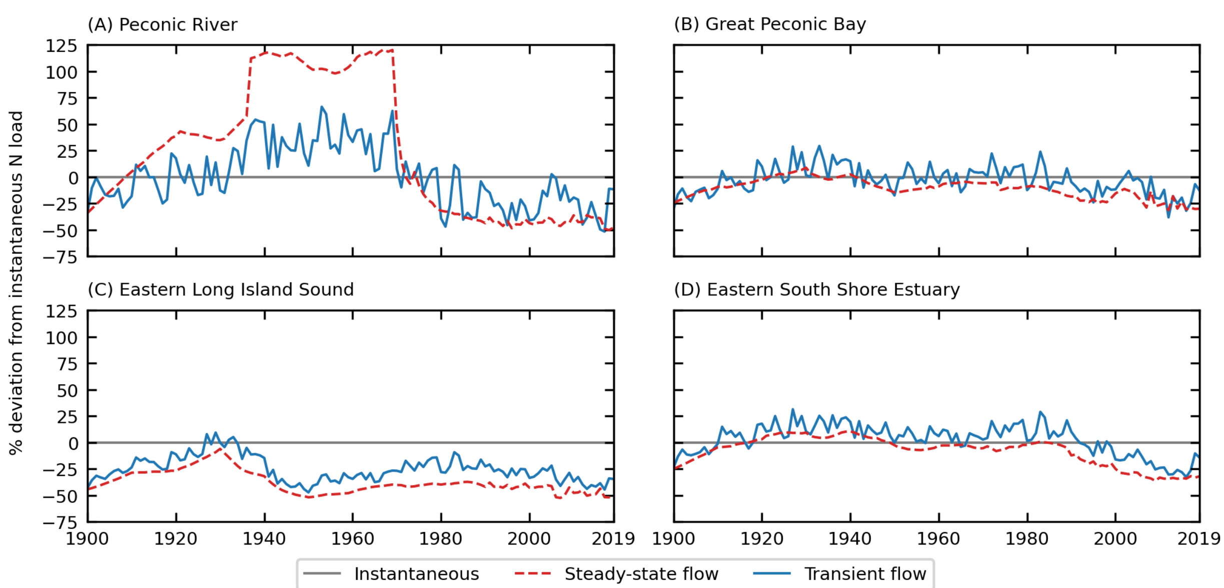

Figure 10. Annual loads shown as a percent deviation from the mean of the instantaneous loads calculated from contributing areas with and without pumping. Differences between the instantaneous

loads and steady-state simulated loads are due to nitrogen transport through the aquifer functioning as temporal sinks and sources of nitrogen to surface water receptors. If a steady-state flow load is lower than the concurrent instantaneous loads, there is a net temporal sink for that year that cannot be accounted for in NLM (i.e., some nitrogen inputs from that year went to longer groundwater flow paths and/or older groundwater discharging that year had lower nitrogen concentrations relative to that year's inputs). If an annual steady-state flow load is higher than the concurrent instantaneous load, there is a net temporal source for that year (i.e., older groundwater discharging that year had higher nitrogen concentrations relative to that year's inputs). This is also true for differences between the transient flow loads and instantaneous loads, with the additional effect due to the annual changes in hydrologic conditions.

5.2 Considerations for nitrogen management

Simulations that account for time-varying nitrogen inputs and groundwater travel times are rigorous tools available for assessing environmental nitrogen loading. However, developing transient simulations of groundwater flow and nitrogen transport for aquifer systems with long travel times is an intensive effort, requiring decades-spanning estimates of both hydrologic stress and nitrogen inputs. Thus, study context and management objectives could help determine whether simpler approaches, such as instantaneous loads estimated from NLM, provide an adequate understanding of a system or whether more data-intensive approaches are needed.

The suitability of a given method to adequately estimate receptor nitrogen loading will likely depend on 1) the physical characteristics of the aquifer system, 2) the degree to which the aquifer system is affected by anthropogenic stresses like groundwater withdrawals, and 3) the time-varying characteristics of nitrogen inputs to the aquifer. Physical characteristics of the aquifer include complexities in the geologic framework and water transmitting properties and the distribution of hydraulic gradients, velocities, and travel times. The contributing areas used to estimate instantaneous loads are static features, but watersheds in natural systems are dynamic and change in response to temporal changes in recharge and withdrawals (Figure 3; Reilly and Pollock, 1995; Walter, 2013). The associated changes in hydraulic gradients and groundwater flow paths affect the advective transport of nitrogen through the aquifer to receptors. Additionally, changes in nitrogen inputs over a period that spans a substantial part of the typical groundwater travel times within an aquifer would likely result in greater disagreement between instantaneous loads and those simulated by transient groundwater models.

Instantaneous loads estimated using NLM methods may be reasonable approximations of loads from process-based simulations in some coastal hydrologic systems, such as those that are: 1) dominated by surface-water drainages resulting in relatively static watersheds following topographic surface drainages, or 2) dominated by groundwater discharge through thin permeable aquifers characterized by high recharge rates and short travel times. Barclay and Mullaney (2021) found that watershed delineations and travel time bands in a coastal aquifer system in southern Connecticut greatly informed understanding of nitrogen transport. The median travel time through the aquifer was on the order of a year and potential lag times for transport were on a seasonal scale. Instantaneous loads have been used to estimate Total Maximum Daily Loads (TMDLs) for estuaries on Cape Cod as part of the Massachusetts Estuaries Project (Massachusetts Department of Environmental Protection 2024) using NLM methods and simulated contributing areas to wells and surface waters (Walter et al., 2004). The Cape Cod hydrologic system is groundwater-dominated, with an aquifer that is thin, permeable, and generally characterized by small travel times. Walter (2013) evaluated instantaneous loads for an estuarine system on Cape Cod and found that loads simulated under steady-state conditions were within 20% of instantaneous loads after about 25 years of transport time. The reasonableness of instantaneous loads in systems like coastal Connecticut and Cape Cod is a function of the small changes in nitrogen inputs over the span of short groundwater travel times.

Some watersheds on Long Island may require methods that account for legacy nitrogen to more fully evaluate nitrogen management actions. The underlying aquifer system is thick and geologically complex and is bounded laterally by a dynamic freshwater-saltwater interface. Travel times through the aquifer can exceed hundreds of years and solutes entering the aquifer at the water table can move through the aquifer for decades before discharging into streams and coastal receptors (Walter et al. 2024). The aquifer has historically been highly perturbed by large groundwater withdrawals and the redistribution of anthropogenic recharge (Walter et al., 2024). Likewise, land use and associated nitrogen sources have changed significantly, particularly since 1900, with conversion of forested land first to agricultural, then to residential land uses (Monti et al., 2024). Many of these changing nitrogen sources have occurred over the span of travel times in the aquifer, and as a result, there is a large mass of legacy nitrogen in the aquifer (Figure 6). This legacy nitrogen will continue to discharge after nitrogen mitigation is undertaken, affecting the response of the system to those actions (Ascott et al., 2021).

As discussed in Section 5.1, estimates of annual receptor nitrogen loads that do not account for contributions from legacy nitrogen (i.e., instantaneous loads), could accurately estimate actual nitrogen loads when the legacy nitrogen portion and relative magnitude of annual receptor nitrogen loads has remained unchanged for decades. However, large annual changes in nitrogen inputs (Figure 5), low fractions of nitrogen inputs to groundwater with travel times <1 year (i.e., shorter than the annual time frame on which instantaneous loads are calculated) (Figure 9), and occasionally large differences between annual instantaneous and simulated loads (Figure 10) demonstrate the dynamic effects of legacy nitrogen in the Long Island aquifer system and the utility of advanced approaches for evaluating the potential efficacy of nitrogen management actions.

The contribution of legacy nitrogen to total receptor nitrogen load in a given year is a function of specific hydrologic conditions and historical nitrogen inputs within a receptor's contributing area, which can vary greatly within an aquifer system, as demonstrated in this study. Our results of simulated nitrogen transport in the Long Island aquifer system indicated that high annual variability in both nitrogen input magnitudes and nitrogen input partitioning by groundwater travel times in a contributing area increases the effect of legacy nitrogen on estimates of receptor nitrogen loads. Similar legacy nitrogen dynamics in groundwater have been noted in four major river basins across the globe (Liu et al., 2024). Therefore, accounting for legacy sources of nitrogen could improve accuracies of models used to predict ecosystem responses to potential nitrogen management actions. In this study, we developed and used a groundwater flow and transport model to simulate a historical period (1900-2019) encompassing most groundwater travel times in the Long Island aquifer system to predict current nitrogen loads while accounting for the effect of legacy sources of nitrogen. Stakeholders can use the developed model to predict time-dependent responses of nitrogen loads and evaluate the effectiveness of proposed nitrogen management actions. Complete realization of improvements to ecological health may lag management actions by several years or decades in watersheds with long travel times (Figure 8). Understanding these lags can help decision makers manage public expectations related to planned resource management activities.

Prior to analyzing nitrogen management proposals, groundwater flow and transport models can be used to assess receptor response dynamics by simulating the response to full removal of terrestrial nitrogen sources (Figure 8). Although such a scenario is likely not a realistic action, it serves as a preliminary analysis that can be performed on multiple water bodies to identify those that may respond more readily to proposed management actions and those that may require more time to reach a desired outcome. Receptor nitrogen load response times to management actions are a function of groundwater travel times within the contributing area and the fraction of nitrogen inputs to areas with small travel times (Figure 8 and Figure 9). These simulated response times can help stakeholders make more informed decisions about the allocation of limited resources to maximize long-term benefits. Additionally, groundwater flow and transport modeling analyses that fully account for legacy nitrogen can provide estimates of the spatial distribution of nitrogen mass fluxes across the beds of surface water bodies (Figure 11). Simulated nitrogen mass fluxes for both current and potential future conditions could be used to identify local areas with potentially high flux rates, which, in turn, could help determine candidate locations for more spatially focused nitrogen mitigation like permeable reactive barriers.

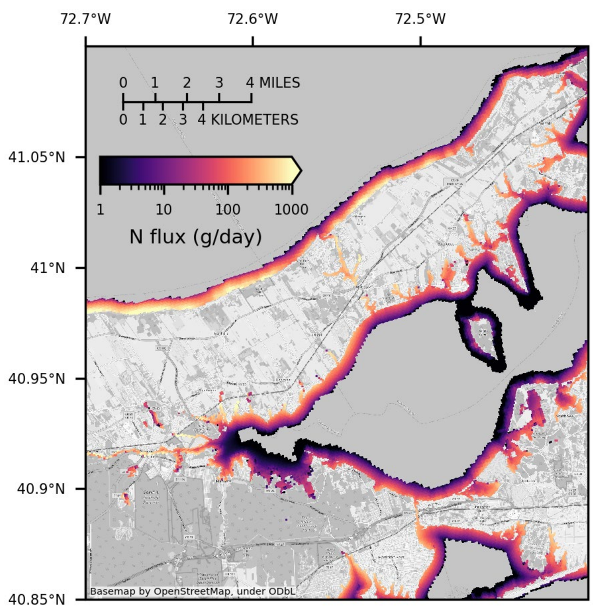

Figure 11. Example of per model cell spatial distributions of groundwater nitrogen fluxes to surface water bodies that can be estimated using transient simulations of nitrogen transport. Base map by OpenStreetMap, under ODbL (openstreetmap.org/copyright).

5.3 Limitations

This study focused on larger water bodies with relatively broad groundwater travel time distributions than those typically observed in smaller water bodies. Narrower travel time distributions with predominantly shorter travel times will likely lead to more agreement between instantaneous loads from NLM and transient loads from numerical models. The numerical models used in this study are subject to the limitations associated with simulating groundwater flow in a complex system like the Long Island aquifer system, including (1) the simplifying assumptions inherent in the conceptual model underlying the numerical model, (2) the discretization and representation of hydrologic boundaries, (3) representation of hydraulic stresses and intrinsic aquifer properties, and (4) the accuracy of parameters estimated from model calibration (Walter et al., 2024). In addition to flow field uncertainties, nitrogen transport within the flow field was assumed to have spatially and temporally uniform attenuation rates, regardless of where the attenuation is assumed to occur along the transport pathway. This assumption may result in both underestimates and overestimates of actual receptor nitrogen loads, depending on actual subsurface hydrogeochemical conditions. The use of groundwater flow and transport models does not account for overland nitrogen transport, but in a groundwater-dominated hydrologic system such as the one on Long Island, we assumed that overland flow had a negligible effect on attenuation. The magnitudes of the estimated historical nitrogen input, though based on the best available data, are subject to a significant amount of uncertainty, especially for inputs earlier in the 20th century (Monti et al., 2024). The actual locations of those inputs have also been discretized to the model grid, resulting in inputs distributed uniformly across model cells by land use, which may smear areas of high nitrogen inputs that are smaller than the 500-foot cell size. However, assumptions about input locations are likely to be less important than the uncertainty associated with input magnitudes and attenuation rates (Barclay and Mullaney, 2021).

6 Conclusions

Nitrogen loads to receptors vary as a function of the unique temporal and spatial distributions of nitrogen inputs within receptor contributing areas. In some watersheds, legacy nitrogen in groundwater can result in differences between nitrogen loads estimated by methods accounting for nitrogen transport in groundwater and those estimated from summations of present-day sources (i.e., instantaneous loads from NLM). In other watersheds, steady antecedent nitrogen inputs and hydrologic conditions can lead to close agreement between instantaneous load estimates and the loads estimated from transport simulations under steady-state and transient flow conditions. The transient flow simulations in this study provided insight into historical interannual variability in receptor nitrogen loads that could help improve understanding future variability and environmental impacts of these loads. Legacy nitrogen in groundwater continues to discharge into surface waters after nitrogen mitigation actions have been undertaken, and adding transient simulations that account for time-varying nitrogen inputs and legacy nitrogen in an aquifer system can help resource managers evaluate the efficacies of potential nitrogen mitigation actions.

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9 Supporting Information

Table S1. Steady-State contributing areas and travel times.

	Without wells							**With wells**						
		travel time (years)							**travel time (years)**					
Surface water body	**Area (sq km)**	**median**	**IQR**	**5%**	**25%**	**75%**	**95%**	**Area (sq km)**	**median**	**IQR**	**5%**	**25%**	**75%**	**95%**
Peconic River	76	27	122	0.9	7.0	129	307	62	26	151	0.9	7.2	158	327
Great Peconic Bay	111	15	44	0.8	4.4	49	256	106	15	44	0.9	4.4	49	260
Long Island Sound	234	28	86	1.2	7.7	94	257	209	26	87	1.1	7.3	95	260
South Shore Estuary	237	22	183	0.9	5.9	189	370	204	18	187	0.9	5.3	193	396

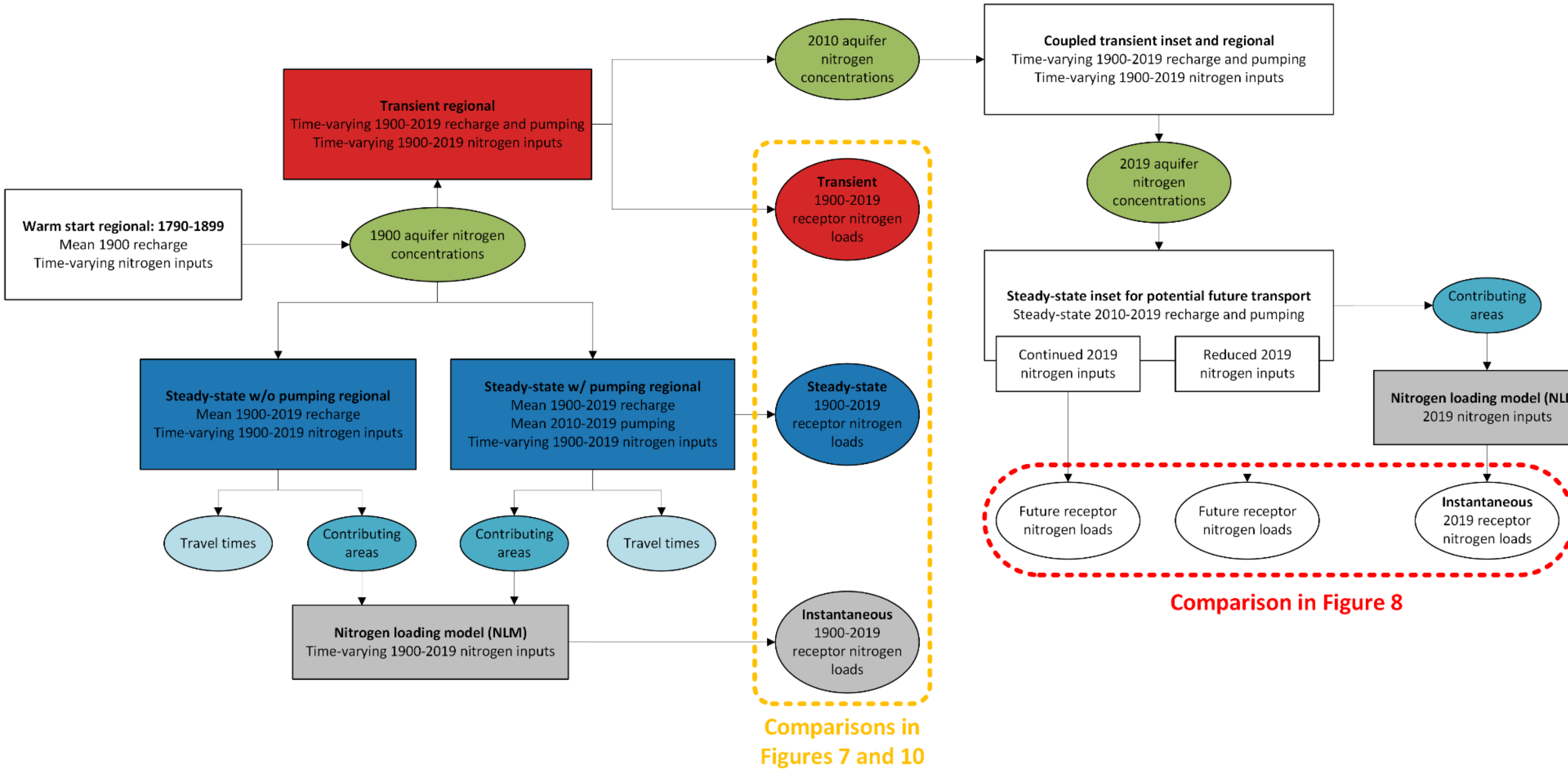

Figure S1. Flow diagram of the connections and dependencies between the models in this study.

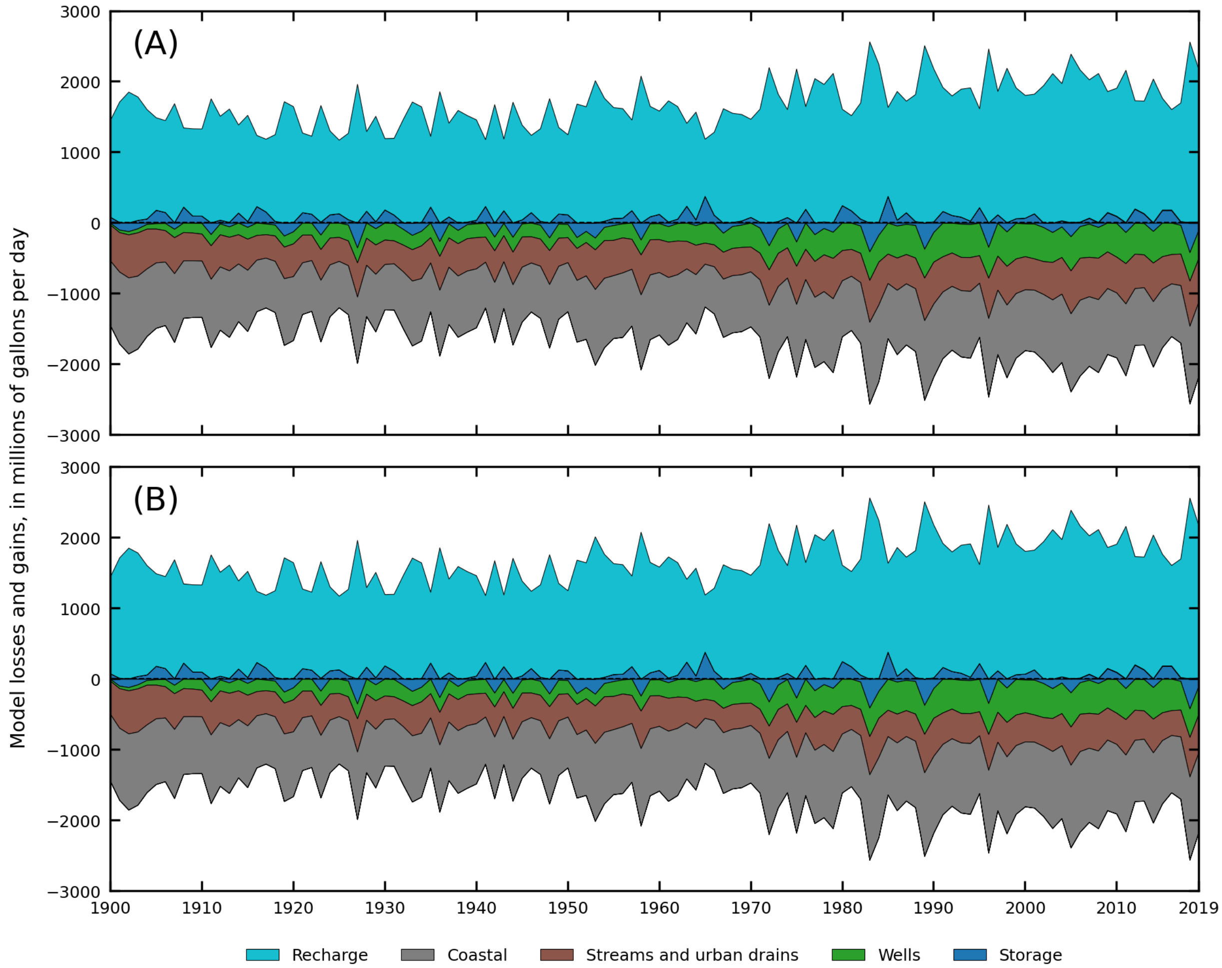

Figure S2. MODFLOW 6 volumetric water budgets for the (A) original 20-layer regional model (Walter et al. 2024) and (B) modified 29-layer regional model (this study).

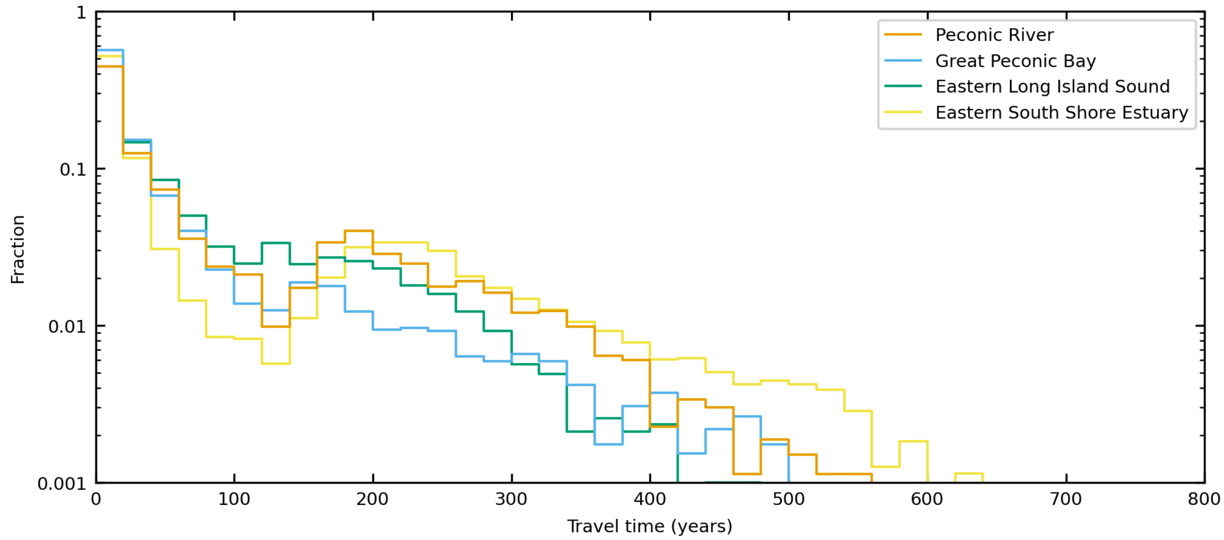

Figure S3. Histograms of groundwater travel times within the four surface water receptors under steady-state conditions with 1900-2019 mean recharge rates and 2010-2019 mean pumping rates. Note the log scale on the y-axis.

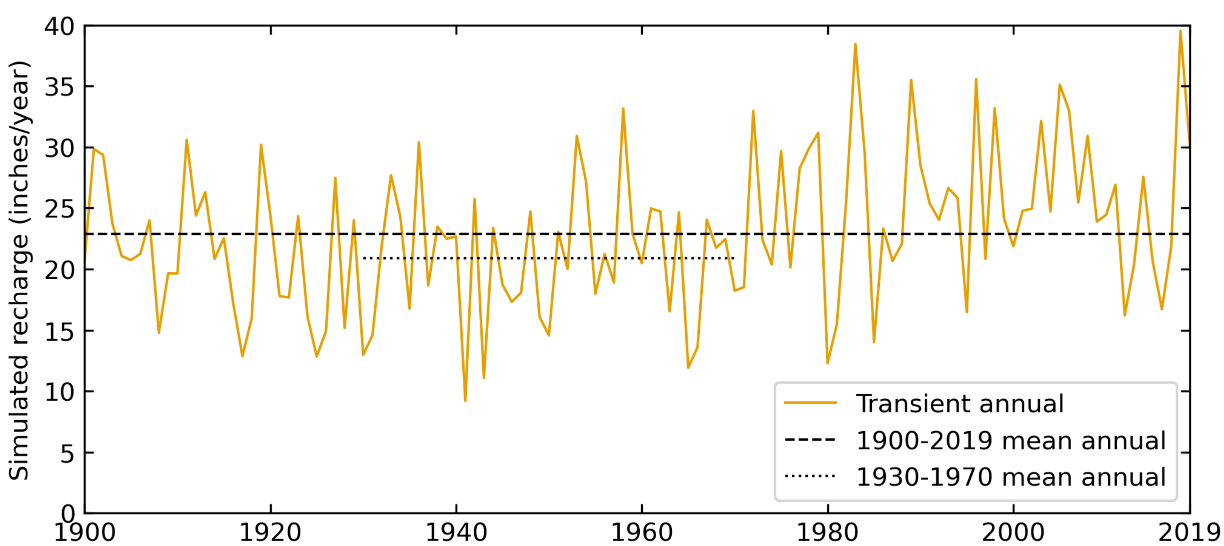

Figure S4. Annual recharge rates for the Peconic River steady-state contributing area.