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A Novel Modeling Framework to Track Contaminant Sources and Transport in a Groundwater-Dominated Watershed

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Abstract. Backwards particle-tracking is widely used in groundwater modeling to gain insights into transport processes in terms of source areas, travel paths, and travel times. In this study we develop a novel modeling framework to apply this approach to SWAT-MODFLOW, a coupled surface water-groundwater model, using the particle-tracking model MODPATH. We tested the framework by applying MODPATH to a SWAT-MODFLOW model of a groundwater-dominated watershed and tracking particles backwards from a river reach with significant spring discharges to the locations where they entered the MODFLOW domain. By combining the source area information of the particles with the

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nitrate loading from the SWAT model, we were able to simulate nitrate concentrations in the discharged groundwater as well as to simulate changes in nitrate concentrations resulting from different land use and management scenarios. Critically, our approach developed here permits simulation of travel time distributions and source areas of water volumes and nitrate masses. We show how these source and age components can be used to guide the prioritization of water quality mitigation strategies that consider both intrinsic (i.e., hydrogeological) vulnerability as well as specific management actions that could be taken to improve surface water and groundwater quality.

1 Introduction

Groundwater contamination is a widespread global problem. Nitrate pollution in particular can lead to eutrophication of water bodies and health problems in humans [Hansen et al., 2017; Zhou et al., 2015]. Elevated nitrate concentrations in groundwater are typically linked to the use of fertilizers in agriculture, animal waste, and effluent from human wastewater disposal systems [Almasri, 2007; Craswell, 2021]. Alternative mitigation strategies to decrease the nitrate loading to groundwater have different costs, benefits, and implementation-response timelines. Assessing the downstream response to a change in nitrate loading is complex, as the fate and transport of nitrate in the subsurface is determined by coupled biological, chemical, and hydrogeological processes that vary widely in time and space [Correa-González et al., 2023; Lake et al., 2003; Wriedt and Rode, 2006].

The recently developed SWAT-MODFLOW-RT3D (SMR) code [Wei et al., 2019], is a numerical simulation tool that accounts for most processes relevant to groundwater nitrate transport. In essence, SWAT simulates flow and solute transport near the surface (including recharge to groundwater),

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MODFLOW simulates groundwater flow, and RT3D simulates solute transport in the groundwater domain by means of solving the advection-dispersion-reaction equation. Compared to using only MODFLOW and RT3D, the key advantage of SMR is that the nitrate loading to the groundwater domain does not need to be specified as a boundary condition. Instead, this loading is implicitly simulated as a function of weather, soil and hydrologic conditions, land use and management systems. Thus, SMR permits assessment of nitrate fate and transport under scenarios of changes in land use (including changes in agricultural practices) and climate [Wei et al., 2019]. However, SMR only simulates spatiotemporal variations in nitrate concentrations and does not provide direct information about source areas or travel times of groundwater and nitrate. This means that while it can be used to simulate the outcome of alternative strategies, it provides little information to guide optimal water quality improvement strategies and does not provide immediate insights into why the outcomes of alternative mitigation strategies may differ. Furthermore, as a fully coupled transport model, SMR is computationally expensive, especially for large regions or highly resolved model domains.

In contrast, particle-tracking is widely regarded as a useful and relatively computationally efficient tool for solute transport that can simulate the source and age components of groundwater as well [Engdahl et al., 2016; V Kaandorp et al., 2018; Maxwell et al., 2019; Wilusz et al., 2020]. Backward particle-tracking provides insight into the intrinsic vulnerability of specific regions to groundwater pollution [Medici et al., 2021; Molson and Frind, 2012]. Namely, using the final location of the back-tracked particles, it is possible to map pollutant source areas and associated travel times. Source areas with higher contributions to the discharge volumes and shorter travel times are typically classified as more vulnerable. However,

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applying particle-tracking to simulate nitrate transport typically requires setting an explicit boundary condition for nitrate mass loading [V P Kaandorp et al., 2021; Kauffman, 2001; Mullaney, 2007].

Given the complexity of nitrate transport in groundwater and the limitations of current modeling approaches, there is a need for more efficient and flexible methods that not only simulate nitrate concentrations but also provide insights into groundwater source areas, travel times, and vulnerability to pollution. In this study, we propose a novel modeling framework based on SWAT-MODFLOW [Bailey et al., 2016] and MODPATH [Pollock, 2016], a particle-tracking code specifically designed for MODFLOW models. Using this approach, we simulate the age and source components of discharged groundwater. SWAT-MODFLOW is the same model code as SWAT-MODFLOW-RT3D except that the RT3D computations are omitted. Thus, SWAT-MODFLOW simulates the nitrate loading to the aquifer but does not simulate subsequent transport in the MODFLOW domain. By combining particle-tracking results with the simulated nitrate loading we can simulate nitrate concentrations in discharged groundwater. The age and source components of the discharged groundwater provide information about the intrinsic vulnerability of the groundwater domain, which together with nitrate loadings, can be used to consider alternative mitigation strategies and estimate how long they will take to affect nitrate concentrations in discharge areas. Our novel approach thus overcomes some of the limitations of SMR while keeping the useful feature of simulating the nitrate loading as a function of land use and weather conditions, rather than specifying it as a boundary condition.

We apply this modeling framework to the Santa Fe River Basin in north-central Florida, which is fed in large part by discharge from springs, many of which have been identified as impaired because of high nitrate levels [FDEP, 2018]. According to the Florida Springs and Aquifer Protection Act, the Florida

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Department of Environmental Protection (FDEP) is required to delineate a priority focus area (PFA) for each impaired spring system by considering groundwater travel times, hydrogeology, and nutrient loads [FDEP, 2018]. A basin management action plan (BMAP) is then designated for the PFA to reduce pollutant loading, which can be accomplished by adoption of best management practices, improvement in wastewater treatment and disposal, conservation programs, and other water quality improvement projects. Given this context, this study aims to: (1) develop a novel modeling framework that combines SWAT-MODFLOW and MODPATH to simulate groundwater flow and nitrate transport in terms of age and source components; (2) apply and test this framework in the Santa Fe River Basin to quantify the contributions of different land uses to nitrate pollution and assess groundwater travel times; and (3) provide actionable insights into the prioritization of nitrate mitigation strategies based on the travel time and source area contributions to groundwater nitrate pollution.

2 Modeling Framework

2.1 Overview of Modeling Framework

Our overall model integration framework and workflow is summarized in Figure 1, and each step is described in detail below. Briefly, the first step is to build, calibrate, and validate one or more SWAT-MODFLOW models of the target domain. Next, a stand-alone MODFLOW model, adapted from the SWAT-MODLFOW model(s) in step 1, is developed on a coarser temporal timescale and used along with MODPATH to simulate particle-tracking in the final step. While the modeling framework uses three

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existing simulation codes (SWAT-MODFLOW, MODFLOW, and MODPATH), some additional codes for pre- and post-processing are also required (Figure 1).

2.2 SWAT-MODFLOW Model(s)

The Soil Water Assessment Tool (SWAT) (Arnold et al., 1998) is a widely used watershed model. In SWAT, the watershed is divided into hydrologic response units (HRUs), which are areas within a subbasin that share the same slope, soil, and land use characteristics. In SWAT-MODFLOW (Bailey et al., 2016), SWAT is coupled with the well-established groundwater modeling code MODFLOW-NWT (Niswonger et al., 2011; Harbaugh, 2005) from the United States Geological Survey (USGS). The process of creating the linkage files needed to build a SWAT-MODFLOW model is described in existing literature (Bailey et al., 2016; Park and Bailey, 2017; Park et al., 2019). These linkage files define how to partition fluxes from HRU's to MODFLOW grid cells. As explained in more detail below, it can be useful to build more than one SWAT-MODFLOW model, each based on a static land use map, to analyze changes in nitrate loading due to changes in land use.

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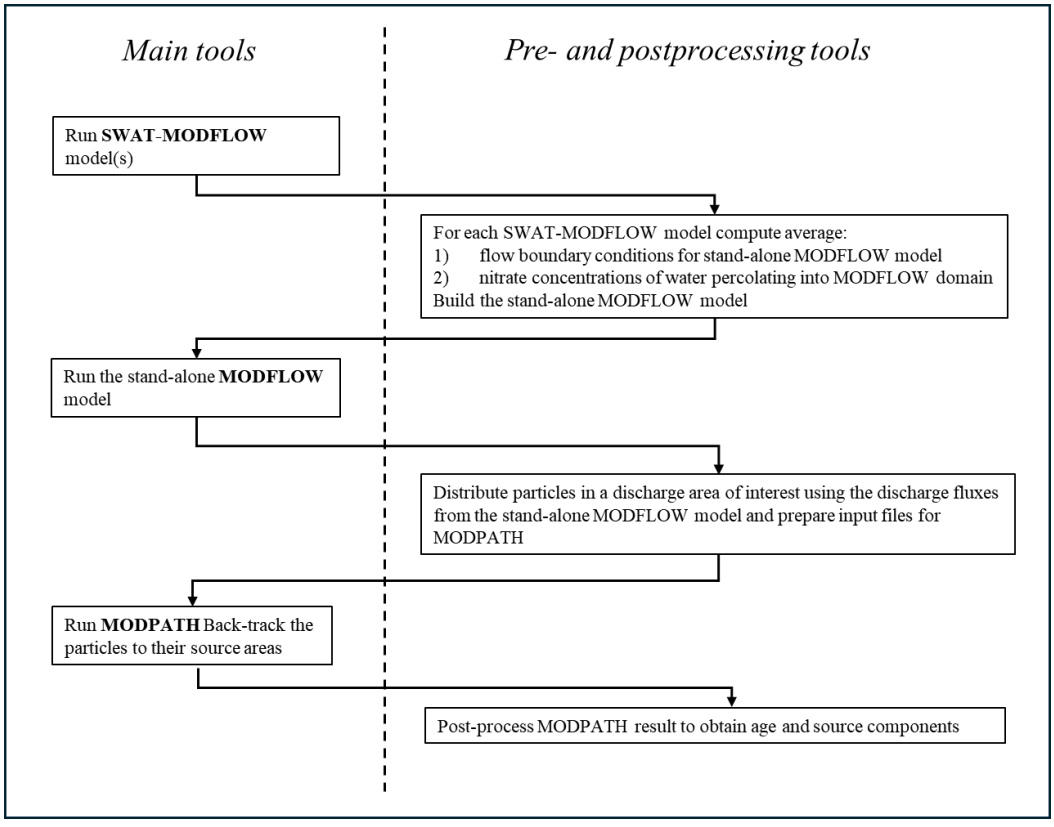

Figure 1. Schematic illustrating the modeling framework.

2.3 Stand-Alone MODFLOW Model

MODPATH is a particle-tracking code for MODFLOW that is based on computationally efficient semi-analytical solutions that track each particle through a single cell in a single step as long as the flow field remains constant during this tracking step (Pollock, 2016). However, for flow fields that vary on a daily basis, as is the case in SWAT-MODFLOW, this process can be computationally inefficient, particularly since the time period covered by the flow fields must be long enough to simulate complete particle trajectories. Our modeling framework circumvents these issues by using a separate, stand-alone

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MODFLOW model. From the SWAT-MODFLOW output, we compute the average flow boundary conditions for effective groundwater recharge and well pumping over longer time periods, reducing the number of changes in the flow field and enhancing computational efficiency by limiting the number of semi-analytical tracking steps. As explained further in Appendix A, due to an error in the original SWAT-MODFLOW code, we modified the code to transfer water between MODFLOW groundwater cells and SWAT river reaches using differences between groundwater elevation and the river bottom. As a result, transient river stages in SWAT do not affect the MODFLOW simulations, obviating the need to average their stages for the stand-alone MODFLOW model. We note that our approach assumes boundary conditions can be averaged over longer periods without significantly affecting particle-tracking results; this assumption is reasonable if boundary conditions fluctuate around a mean on timescales much smaller than groundwater travel times.

The stand-alone MODFLOW model allows us to easily simulate transitions between alternative land use and land management scenarios. Namely, we can define multiple stress periods to account for different boundary conditions as computed from specific SWAT-MODFLOW models with different land use and land management configurations. Depending on groundwater travel time distributions, it may be necessary to account for historic land uses when simulating groundwater nitrate concentrations emerging in river reaches at a particular point in time. That is, some particles may need to be tracked though a time period when the land use was different from what it is during the time when the particles emerge in the river. Therefore, in this study we define an initial steady-state stress period using the averaged boundary conditions computed from a SWAT-MODFLOW model that represents pre-development land use conditions. Changes from pre-development conditions are then defined in subsequent stress periods, each

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requiring one or more separate SWAT-MODFLOW models to compute average boundary conditions. As discussed later in section 3.3, it is possible to combine multiple SWAT-MODFLOW models to compute the average boundary conditions for a stress period.

In this study, we use steady state stress periods in the stand-alone MODFLOW model. Switching between steady-state flow models to reflect changes in land use assumes that the change in the flow boundaries has a relatively immediate effect on the flow field compared to groundwater travel times. However, if needed, the methodology can incorporate transient stress periods to account for a more gradual change in the flow field. It is also noted that if changes in land use occur gradually over a relatively long time, an interpolation procedure could be used to define a gradual change in boundary conditions.

2.4 Particle-Tracking

In this study, we exclusively use backward tracking of particles. This is the most efficient method for computing the travel time distributions and nitrate concentration within the discharge area of interest at a particular point in time since all the tracked particles will contribute to these computations. Using the head file created by the stand-alone MODFLOW model, we reconstruct the groundwater discharges to stream reaches in our area of interest, which occur at either river or drain cells. We distribute particles across these discharge cells in proportion to their flow rate such that each particle corresponds to the same volumetric discharge, ensuring that each particle contributes equally to the travel time distribution. Within each discharge cell we distribute the particles across the discharge area using a uniform random number generator.

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Travel time distributions of the discharged water volumes are then directly computed from the standard output from MODPATH by counting the number of particles with a travel time less than τ, and dividing by the total number of particles:

$$g_v(\tau,t) = \frac{1}{n}\sum_{i=1}^{n} \mathbf{1}_{\tau_i(t)\le\tau} \quad (1)$$

where $g_v(\tau,t)$ is the cumulative distribution function for particles at time t, n is the number of particles, τ_i is the travel time of particle i and **1** the indicator function (which has a value of 1 when the condition $\tau_i(t) \le \tau$ is met and 0 otherwise).

2.5 Linking Particles to Nitrate Loading

The concentration, c, of the discharged water is a function of the travel time distribution and is given by the following response function (Benettin et al., 2022; Rodriguez et al., 2021):

$$c(t) = \int_0^\infty \iint_A c_{in}(x,y,t-\tau) e^{-\kappa\tau} p_v(\tau,t)\, dx dy d\tau \quad (2)$$

where c_{in} is the nitrate concentration of groundwater discharged in the region of interest at time t, τ is the travel time, k is the denitrification rate, A the source area and $p_v(\tau,t)$ is the fraction of water parcels with

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travel times equal to τ, as derived from the travel time probably density function. It is noted that equation 2 implies that denitrification is a first-order rate process.

Using the particle-tracking results, the concentration of each discharged particle at time t is computed from the following numerically equivalent equation:

$$c_{\text{out},i}(t) = c_{\text{in},i}(t - \tau_i)e^{-k\tau_i} \quad (3)$$

where $c_{\text{in},i}(t - \tau_i)$ is the nitrate concentration associated with the endpoint of particle i at time t- τ_i. The concentration of discharged groundwater at time t is computed from:

$$c(t) = \frac{1}{n}\sum_{i=1}^{n} c_{\text{out},i}(t) \quad (4)$$

To compute $c_{\text{in},i}$ it is important to consider the relationship between SWAT HRUs and MODFLOW raster cells. As shown in Figure 2, HRUs overlap with MODFLOW cells and can be subdivided into different "overlap areas". These overlap areas are defined in linkage files and dictate how the fluxes from SWAT HRUs are distributed among MODFLOW cells. The nitrate concentrations $c_{\text{in},i}$ at the surface of the MODFLOW domain are computed on these overlap areas using values for the nitrate mass loading and the effective groundwater recharge flux (defined as percolation flux minus groundwater evaporation flux) into the MODFLOW domain. The effective groundwater recharge flux is first computed per MODFLOW raster cell using HRU values for the percolation values and MODFLOW values for the groundwater evaporation fluxes. Subsequently, for each overlap area the HRU percolation value is scaled such that the total percolation for each MODFLOW cell equals the effective groundwater recharge flux. The nitrate

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mass flux from each HRU is then distributed across the associated overlap areas according to the scaled percolation water fluxes. Finally, the nitrate concentrations in the overlap areas are computed from the nitrate mass loadings and effective groundwater recharge fluxes. Using this procedure, the nitrate concentrations at the surface of the MODFLOW domain are constructed for each stress period.

Using the endpoint of each particle i as provided by MODPATH, it is straightforward to locate the MODFLOW cell from which the particle exited the domain. To assign the particle to a particular overlap area, and thus a particular HRU and land use, a transfer probability function is computed for each overlap area such that the probability equals the ratio of the effective groundwater recharge flux for that overlap area to the effective groundwater recharge flux for the entire MODFLOW cell. Thus, this probability function accounts for a non-uniform distribution of overlap area fluxes. A random number generator is used to allocate the particle to a particular overlap area with the MODFLOW cell from which the particle originated and its associated concentration $c_{\mathrm{in},i}$. Transfer probability functions for particles for distributing particles based on volumetric fluxes have previously been used in surface-subsurface flow models and matric-fracture flow models (De Rooij et al., 2012; Visser et al., 2009; Pan and Bodvarsson, 2002).

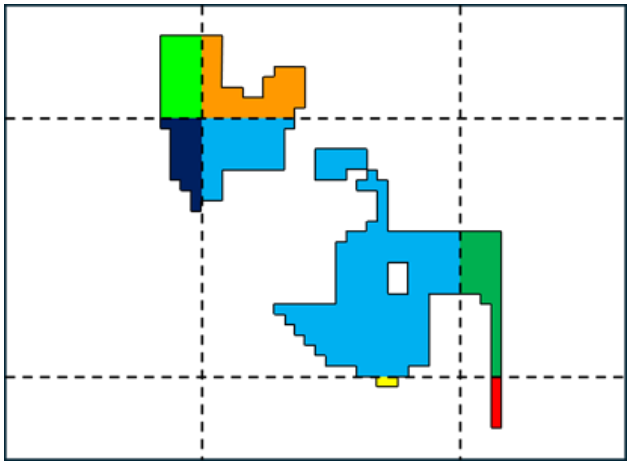

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Figure 2. A single HRU subdivided into overlap areas (each color represents a separate overlap area) by its intersection with the MODFLOW raster (black dashed lines).

Since we can link each particle with a nitrate concentration in the source area, and each particle is assumed to represent the same volumetric flux, it is then relatively straightforward to compute the travel time distributions of discharged nitrate masses:

$$g_{\mathrm{m}}(\tau,t)=\frac{1}{\sum_{j=1}^{n} c_{\mathrm{out},j}(t)}\sum_{i=1}^{n} c_{\mathrm{out},i}(t)\mathbf{1}_{\tau_i\le\tau} \tag{5}$$

where $g_m(\tau,t)$ is the cumulative travel time distribution of nitrate mass for particles. Note that once particles are associated with HRUs, we can also associate a land use with each particle and thus construct travel time distributions for each land use component.

2.6 Intrinsic Vulnerability Mapping

Backward particle-tracking results are useful to gain insight into the intrinsic vulnerability of watersheds (Molson and Frind, 2012; Medici et al., 2021). Using the final location of the back-tracked particles, it is possible to map source areas as well as the travel times. Source areas with higher contributions to the groundwater discharge volumes and shorter travel times may be classified as more vulnerable. In this study, the source area contributions and the travel times were mapped to a raster with the same resolution as used for the HRU mapping in SWAT. Source area contributions were mapped as a fraction of the total number of particles released and thus sum to unity. Using the simulated nitrate loadings for each HRU, we also create raster maps that indicate how much each raster cell contributes to the nitrate mass in the

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discharge area of interest. Nitrate mass contributions are mapped as a fraction of the total nitrate mass discharged and thus also sum to unity. Together with the travel times the raster map of nitrate mass contributions can be used to prioritize mitigation strategies for areas with higher contributions and shorter travel times.

3 Application of Modeling Framework

3.1 Santa Fe River Basin

We applied the SWAT-MODFLOW and MODPATH modeling framework to the Santa Fe River Basin (SFRB) in north-central Florida. Figure 3a illustrates the location and extent of our SWAT-MODFLOW model of the SFRB. The degree of confinement of the Upper Floridan Aquifer (UFA) is mapped into three distinct regions: a confined region, a semi-confined region, and an unconfined region (Figure 3b). In the confined region, the UFA is overlain by the Intermediate Confining Unit (ICU) and the Surficial Aquifer System (SAS). The confined region is dominated by surface water runoff, with abundant surface drainage features including lakes, streams, and wetlands. In the unconfined region the ICU is absent, and the limestone rocks of the UFA are overlain by a thin layer of sand (Arthur et al., 2005). The unconfined region is dominated by groundwater flow and characterized by karst features such as sink holes and springs and the absence of smaller streams feeding the main Santa Fe River (Hunn and Slack, 1983). The UFA is underlain by the Middle Confining Unit (MCU) and the Lower Floridan Aquifer (LFA) (Williams and Kuniansky, 2015).

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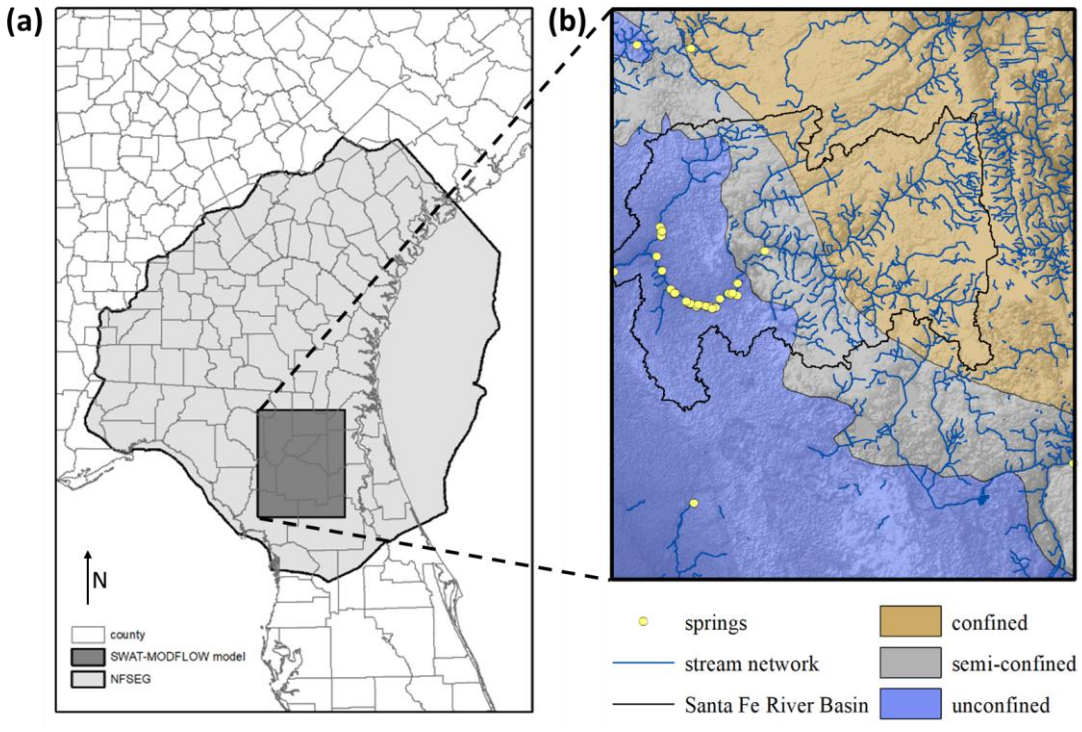

Figure 3. a) NFSEG and SWAT-MODFLOW Model domain locations in North Florida. b) Map illustrating the extent of the confined, semi-confined and unconfined zone, stream network, the Santa Fe River Basin and major springs in the SWAT-MODFLOW domain.

3.2 Model Setup

The MODFLOW model domain was clipped from the larger North Florida Southeast Georgia (NFSEG) steady-state MODFLOW model (year 2001), developed by the St John's River and Suwannee River Water Management Districts (Durden et al., 2019) . The extent of the NFSEG model is shown in Figure 3a. Natural flow boundaries to the Floridan Aquifer are not present in the close vicinity of the SFRB, so

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to limit the size of the clipped model, we selected a rectangular model domain large enough to encompass the springsheds of several large springs along the Santa Fe River (Figure 4). Along the perimeter of the model, we applied fixed-head boundary conditions based on steady-state values of the NFSEG model. Pumping from municipal and domestic self-supply wells were taken from the NFSEG model.

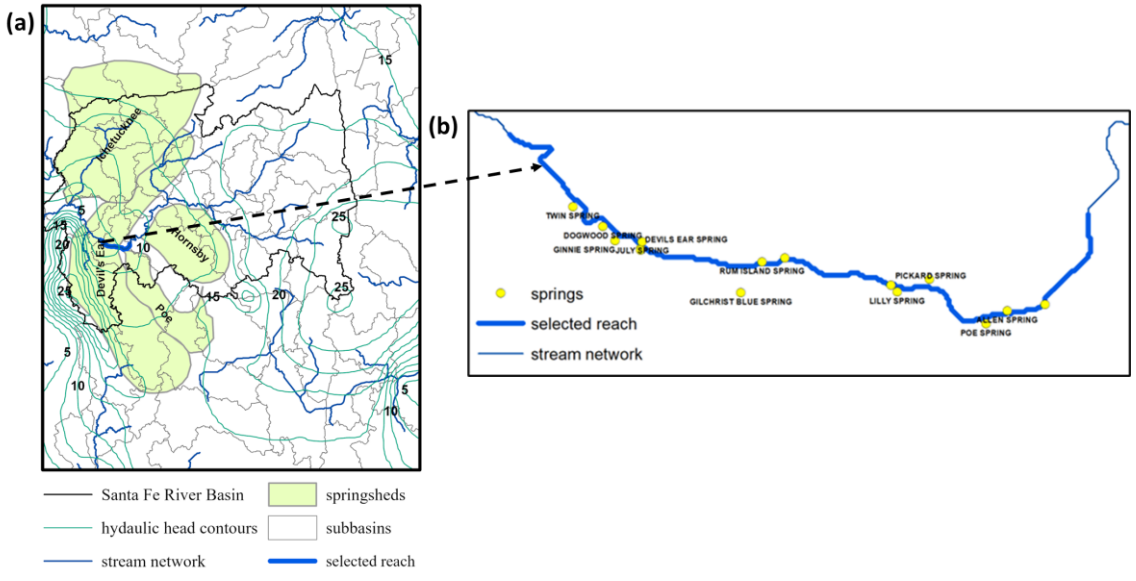

Figure 4. a) Map of Santa Fe River subbasins, springsheds,stream network and springs within the SWAT-MODFLOW model domain. b) Springs along the selected river reach.

The NFSEG was developed as a steady-state model, however, SWAT-MODFLOW requires a transient MODFLOW model with specific storage coefficients. We assumed a value of 1E-4 m^{-1} for all layers (Srivastava et al., 2014). Since the first layer is confined/unconfined, specific yields must also be defined, which we set to 0.25. Finally, we assumed porosity values of 0.25 throughout the model domain (Budd and Vacher, 2004). As discussed in Appendix B, reasonable travel times were only obtained after increasing the NFSEG Upper Floridan Aquifer horizontal hydraulic conductivity and decreasing the

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NFSEG horizontal hydraulic conductivities in the Middle Confining Unit and the Lower Floridan Aquifer to maintain the same effective horizontal conductivity across all layers.

While denitrification in the Floridan Aquifer is thought to be highly variably (Heffernan et al., 2012; Henson et al., 2019; Yang et al., 2023), Heffernan et al. (2012) found an average 32% reduction in nitrate in groundwater emerging from Upper Floridan Aquifer spring vents. Based on these studies we assumed a first-order denitrification process with a half-life of 50 years which yields a 32% loss in groundwater emerging from springs with a travel time of about 20 years, which is within the 16-27 year spring water age estimated by Katz et al. (1999) and Katz (2004) using chlorofluorocarbon measurements. Previously calibrated parameters governing denitrification in the SWAT domain were taken from (Rath, 2021).

3.3 Intrinsic Vulnerability Land-Use Scenarios

We used a sequence of three land-uses to explore intrinsic vulnerability and nitrate mitigation priorities in the SFRB. The first land use ("pre-development") mimics pre-development conditions. Until the end of the 19th century, much of Florida remained undeveloped and low-density longleaf pines covered much of the region (Volk et al., 2017). Around 1920, most of the old growth forests were logged and converted to higher density production forestry or agriculture. Thus, for the pre-development land use we replaced all current agricultural and forestry lands with low-density longleaf pine and replaced all current rural area and urban areas with grasslands. Moreover, in this scenario there was no groundwater pumping for municipal use, agriculture or domestic self-supply and no wastewater effluent discharge from septic tanks or wastewater treatment plants. The second land use ("current conditions", Figure 5) represents the current land use of the study region derived from the Statewide Land Use / Land Cover map of the Florida

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Department of Environmental protection (FDEP) for the year 2017 with water, nutrient and other residential, agricultural and silvicultural management practices defined through a stakeholder engagement process (Bartels and Furman, 2023). Pumping from municipal and domestic self-supply wells was taken from the NFSEG model and nitrate loading from septic tanks and wastewater effluent disposal, as well as fertilizer usage on agricultural, silvicultural and residential lands, was simulated by SWAT. The third land use ("restoration forestry") represents a scenario in which all current agricultural and forestry lands are replaced with low-density longleaf pines, but rural residential and urban land uses remain. The SWAT-MODFLOW model with current land use conditions was calibrated to stream flows at USGS gauge stations in the Santa Fe River, hydraulic head contour maps (Fdep, 2023) and remote sensing estimates of evapotranspiration (Senay and Kagone, 2019) using NLDAS weather data from 1980 to 2018. During the calibration period (2010-2018), the Nash–Sutcliffe efficiency (NSE) was 0.68 for daily streamflow at the Ft. White gauging station (station ID 02322500), and overall groundwater elevation and ET NSE values were of 0.70 and 0.83, respectively. For the validation period (1980-2009), NSE was 0.45 for daily streamflow and overall NSE for ET was 0.82. Details regarding SWAT-MODLFOW model development and calibration for the SFRB are documented and available as a CUAHSI HydroShare (Reaver et al., 2025).

Using the three land use periods, we constructed a stand-alone MODFLOW model with three steady-state stress periods (pre-development, followed by current conditions, followed by restoration forestry) using the averaged boundary conditions from each model that was run from 1980 to 2018. Since the flow field changed between stress periods, the particle-tracking results depend on the time of release. To simulate the long-term effects of a future land use based on restoration forestry, the third period was set to 100

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years. The length of the second period was 80 years such that if we release particles at the end of this period, we will simulate results for the present. We thus assume that current land use and management practices go back 80 years. The first period extended as long as necessary for MODPATH to back-track particles to reach their aquifer entry point. Each particle-tracking simulation was carried out with a million particles.

After construction of the stand-alone MODFLOW model, we back-tracked particles from a Sante Fe River reach containing a large number of first- and second-order magnitude springs including Ginnie Spring, July Spring, Devil's Ear Spring, Gilchrist Blue Spring, Rum Island Spring and Poe Spring (Figure 4b). All of these springs are classified as impaired, with Nitrate-Nitrogen (NO3-N) concentrations ranging from 1.5 to 6 times above the Numeric Nutrient Criteria (NNC) of 0.35 mg/l NO3-N (Fdep, 2018). As a result, state regulations mandate the establishment of a Basin Management Action Plan (BMAP) designed to reduce nitrate loadings and achieve the NNC within 20 years.

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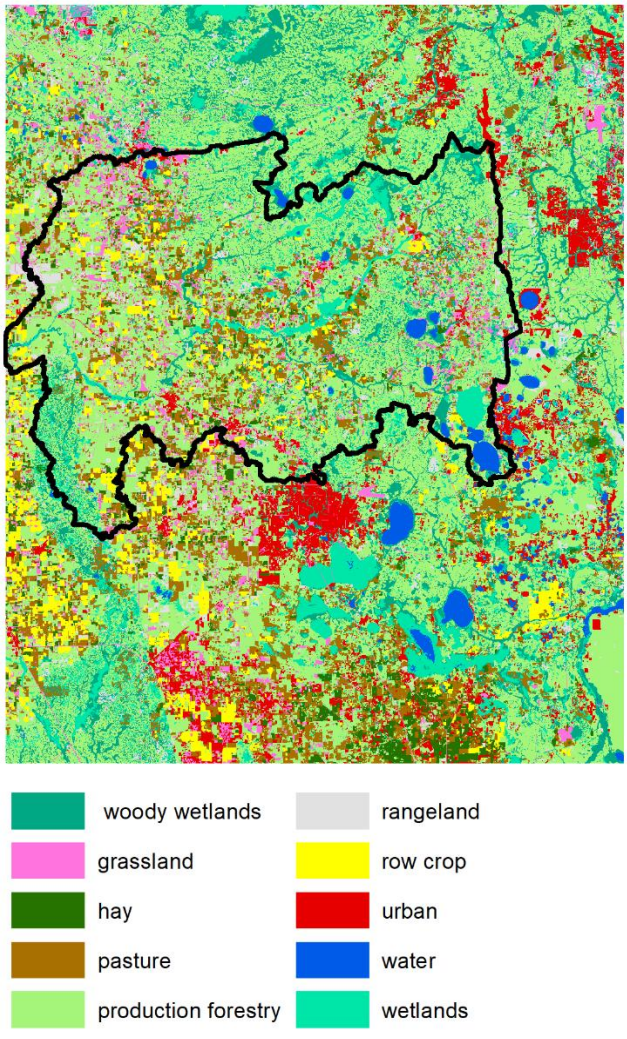

Figure 5. Land use map for current conditions within the SWAT-MODFLOW domain.

After observing the travel times and nitrate load contributions to this river reach for the current land use conditions and the response to conversion to restoration forestry across the entire domain, we simulated a targeted nitrate load mitigation strategy. This strategy included the removal of all septic tanks and fertilized lawns and the conversion of all agricultural lands within a 30-year travel time contour to

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restoration forestry. For this simulation we did not simulate an additional scenario with a separate SWAT-MODFLOW model. Instead, we derive the last stress period from the existing three scenarios. Namely, we replaced the agricultural HRU values for percolation and nitrate loading within the 30-year travel time contour with corresponding HRU values from the restoration forestry scenario and reduced the amount of pumping needed for agriculture accordingly. We also replaced the HRU values for percolation and nitrate loading from septic tanks and rural areas with the corresponding the grassland values from the pre-development scenario. Finally, using the relationship between SWAT HRUs and MODFLOW raster cells as discussed in section 2.5, we computed the average boundary conditions for the third period in our stand-alone MODFLOW model. Exchanging HRU values between different scenarios is valid because in SWAT-MODFLOW the computations on the HRUs are independent of the MODFLOW computations. Since this methodology avoids the need for running a separate SWAT-MODFLOW model, it is more efficient.

4 Results

4.1 Groundwater and Nitrate Travel Time Distributions

Figure 6a illustrates the travel time distributions (TTDs) for current conditions (t=80 years). After parameter modifications detailed in Appendix B, the median water travel time for the current condition scenario is about 32 years, which corresponds reasonably well with age values of 16-27 years derived from chlorofluorocarbons measurements (Katz et al., 1999). Figure 6b illustrates the TTDs of the corresponding nitrate mass based on the assumed half-life of 50 years. The median nitrate mass age of 19

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years is lower than the median water age. It is noted the difference in TTD's between water volumes and nitrate masses is not only caused by denitrification but also by differences in the source areas between the water volumes and the nitrate masses.

Since each back-tracked particle comes from a specific land use, we can partition the areas below the TTDs according to cumulative land use fractions as illustrated in Figure 6a and 6b. In the current condition, production forestry is the land use that contributes the largest volume of discharged water (Fig. 6a), whereas row crops, pasture, and septic tanks contribute the largest nitrate mass (Fig. 6b). Rural and urban lands also contribute to the nitrate mass due to fertilized lawns. We can also construct TTDs for each source component separately (Fig. 7a), illustrating that certain source components have a maximum age of 80 years; these components are associated with land use types that did not exist during pre-development conditions.

Figure 7b illustrates how the concentration of nitrate in the discharged groundwater changes with time over the three scenario periods, partitioned according to source components. As expected, the concentration of nitrate increases after switching from pre-development to current conditions and decreases after the switch to restoration forestry. The lag between changes in nitrate loading and nitrate concentration in discharged water is a consequence of groundwater travel times. The NNC is achieved approximately 60 years after the switch to restoration forestry.

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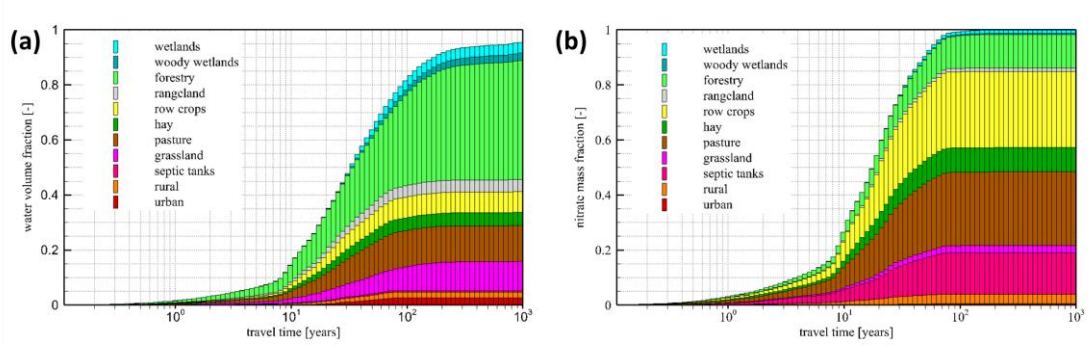

Figure 6. Travel time CDF of water volumes (a) and nitrate masses (b) within the selected river reach with source components for the current conditions.

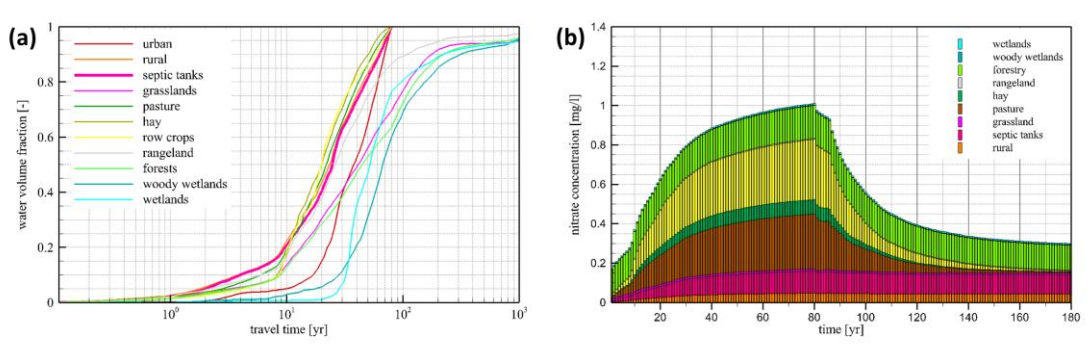

Figure 7. a) Travel time CDF's of water volumes per source component and b) Nitrate concentration in discharged groundwater as a function of time assuming transition from predevelopment conditions to current conditions at year 0 and transition from current conditions to restoration forestry at year 80.

4.2 Intrinsic Vulnerability and Mitigation Prioritization

Figure 8a and 8b show maps of the fraction of particles back-tracked to the source areas and the average travel time, respectively. These maps are for particles released at the end of the current condition period,

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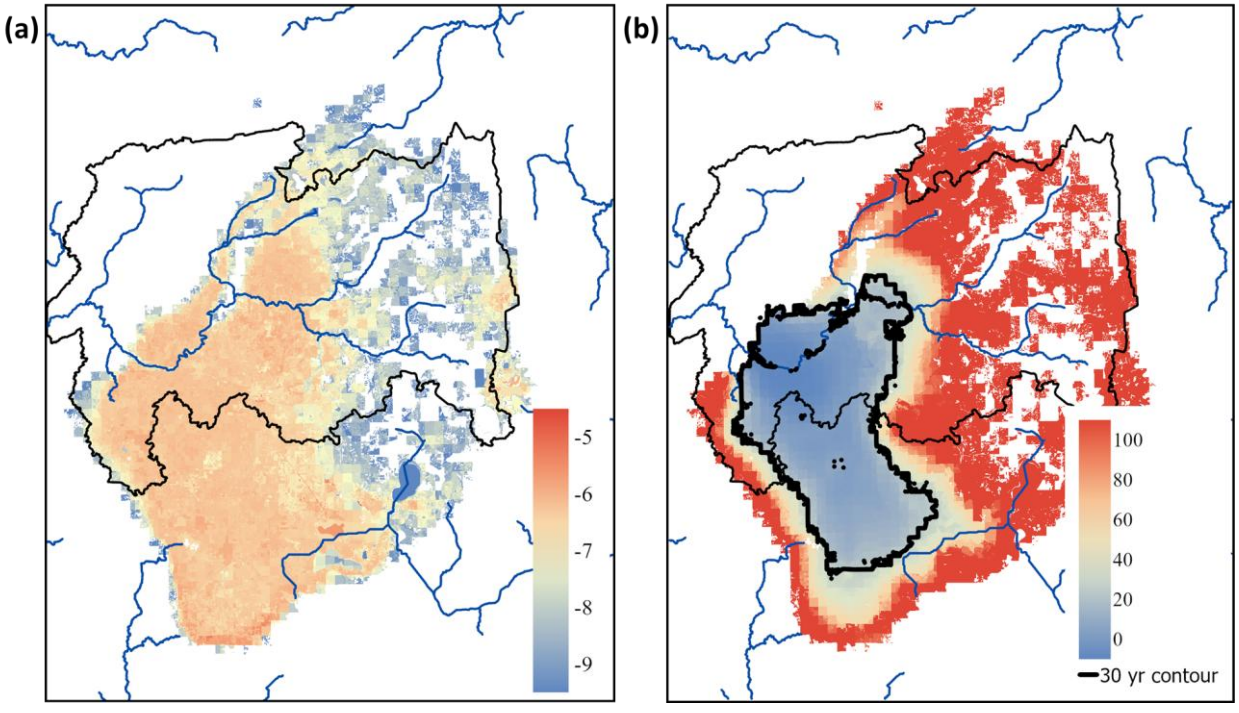
388 just before the switch to restoration forestry. Since each particle represents an equal volume of water, a
389 higher fraction of particles represents a higher contribution to the discharge volume. These maps are
390 important qualitative indicators for intrinsic vulnerability. Namely areas with high particle fractions and
391 low travel times are most vulnerable to contaminants as these areas would contribute relatively quickly
392 and significantly to the contaminant concentration in the discharge area.



393

394 **Figure 8**. a) Map of water volume fractions back-tracked to the source areas (log scale) and b) Travel
395 time map (in years) for current condition.

396

397 Since we have the nitrate loadings from the SWAT model, we can compute how much each particle
398 contributes to the total concentration in the discharged groundwater, which provides insights into which
399 areas contribute the most to the nitrate concentrations in discharged groundwater. This is illustrated in

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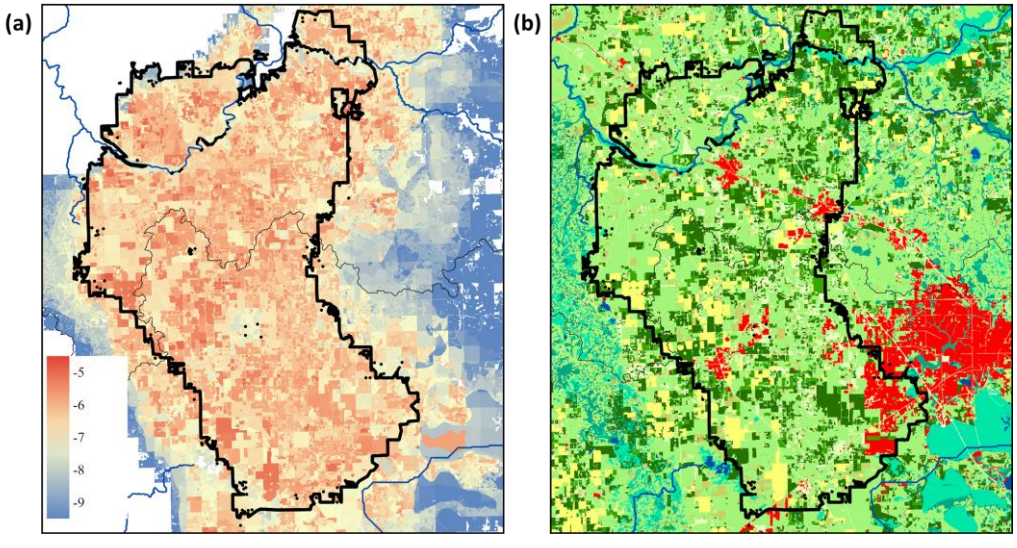
Figure 9a for particles released at the end of the current condition period. Similar to the results shown in Figure 7b, Figure 9a and 9b indicate that agriculture is the largest contributor to nitrate concentrations, since the most significant contribution areas correspond to agricultural land uses. Together with the travel time map in Figure 8b, the map of mass contributions may be used to design potential mitigation strategies.

A potential mitigation strategy that results in meeting the mandated reduction to the NNC more quickly and with less land use change than the full restoration forestry scenario is to remove all septic tanks and fertilized lawns within rural lands and to convert all agriculture land uses within the travel time contour of 30 years into restoration forestry. The decrease in nitrate concentration with time due to this mitigation strategy is shown in Figure 10. In this case the NNC is achieved after 50 years, and 163507 hectares of the current condition 196315 hectares of agriculture remain in production contributing to the regional economy.



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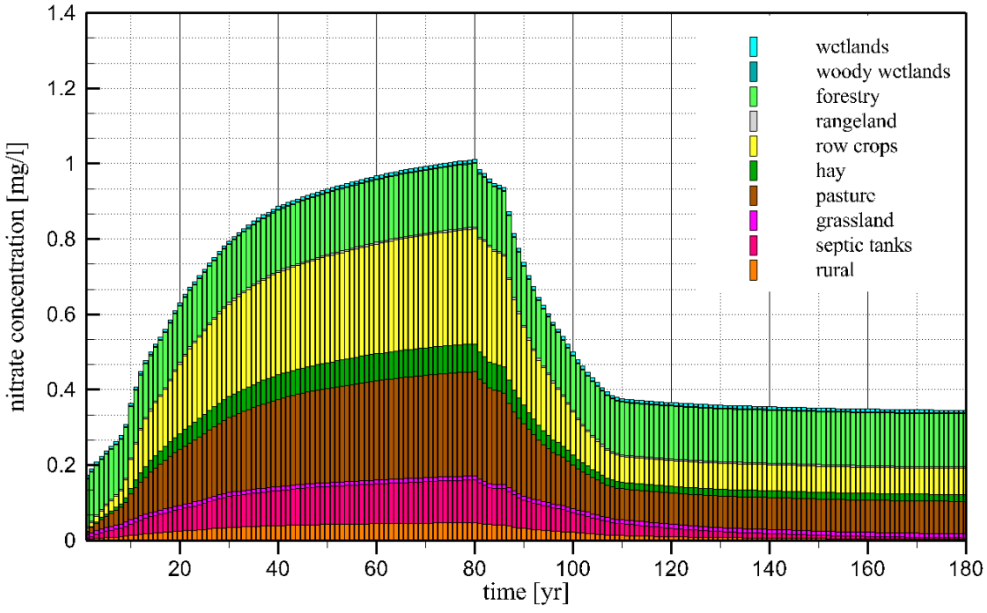
414 **Figure 9**. a) Map of nitrate mass fraction back-tracked to the source areas (log scale) and b) Land use
415 map (see Figure 5 for legend) for current condition.



416

417 **Figure 10**. Nitrate concentration in discharged groundwater as a function of time for targeted restoration
418 which 1) replaces agricultural and production forestry within the 30 years travel time contour with
419 restoration forestry and 2) removes all septic tanks and replaces fertilized rural lawns with unfertilized
420 grassland.

421 **5 Discussion**

422 Numerical models have been widely recognized as useful tools to assess the fate and transport of nitrate
423 in groundwater (Rawat et al., 2022). The recently developed SWAT-MODFLOW-RT3D code (Wei et
424 al., 2019), is a particularly useful numerical simulation tool since nitrate loading to the groundwater

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domain can be simulated for different land use and climate scenarios (Wei et al., 2019). In this study, we propose an alternative novel modeling framework. Instead of simulating transport in the groundwater domain by means of an advection-dispersion-reaction equation as in SWAT-MODFLOW-RT3D, we simulate transport by backward particle-tracking using SWAT-MODFLOW and MODPATH. This idea is motivated by a range of other studies that have shown that particle-tracking is a useful tool to gain insights into the source areas and travel times of groundwater (Engdahl et al., 2016; Kaandorp et al., 2018; Maxwell et al., 2019; Wilusz et al., 2020) and into the intrinsic vulnerability of groundwater systems (Medici et al., 2021; Molson and Frind, 2012). Whereas particle-tracking to simulate nitrate transport typically requires an explicit boundary condition for nitrate mass loading (Kaandorp et al., 2021; Mullaney, 2007; Kauffman, 2001), SWAT-MODFLOW provides this loading as a function as weather, soil and hydrologic conditions, land use and management within our coupled modeling framework.

5.1 Framework Application and Advantages

We applied our modeling framework to a river reach in the Santa Fe River Basin in north-central Florida which is fed by a large number of first and second magnitude springs that are impaired by elevated nitrate concentrations. Initial application of our modeling approach to simulate the age and source components for water volumes in this reach provided useful insights into the underlying flow model which resulted in significant changes to the calibrated model parameters (see Appendix B for details). After these modifications we simulated a median groundwater travel time of about 30 years (Figure 6) which corresponds well with values of 16-27 years as derived from chlorofluorocarbons measurements (Katz et al., 1999). TTDs can be constructed for groundwater volumes as well as nitrate masses. Using the source

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components of the particles the simulated TTDs can be portioned according to land use type (Figure 7) or separate TTDs can be constructed for each land use separately (Figure 8a).

Our modeling framework can account for changes in land use over time. By back-tracking particles at different times and using different nitrate loading maps, we can simulate the change in groundwater nitrate concentrations in a discharge zone of interest as a function of time. In our model application, we show that there is a significant lag time between a change in nitrate loading and the subsequent reduction in nitrate concentrations at the springs (Figure 8b). This lag time is a direct result of the travel time distribution of discharged water. An understanding of this lag time is imperative for sustaining consensus within communities with respect to mitigation efforts. Specifically, if the lag is not well understood, delays in water quality improvement may erroneously be attributed to an improper mitigation plan. The final locations of the back-tracked particles can be used to establish water and nitrate source density maps by counting the number of these locations on a user-defined raster. The particle density on such maps (Figure 8a) indicate how much different areas contribute to groundwater discharge. Together with the travel time maps (Figure 8b) these density maps are indicative of intrinsic vulnerability. However, since we have the simulated nitrate loadings, we can go further and establish maps indicating the nitrate mass contributions for different areas (Figure 9a). These contributions together with the travel time map are of direct use for prioritizing mitigation strategies. In this study, we show an example of a mitigation strategy based on targeting those areas which contribute most significantly to the nitrate discharge. Our modeling framework permits us to simulate the effect of this mitigation strategy in terms of a change of nitrate concentrations in discharged groundwater as a function of time (Figure 10). Results indicate that the mandated reduction to the NNC of 0.35 mg/l NO3-N within 20 years may not be realistic. If all

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agricultural lands (196315 hectares) and forestry (561904 hectares) are replaced with low-density longleaf pines, our simulation results indicate that reaching the NNC takes about 60 years. If all agricultural lands within the 30-year travel time contour are replaced with low-density longleaf pines (32808 hectares), all septic tanks are removed, and all lawn fertilization in rural areas is eliminated it takes about 50 years to reach the NNC.

With respect to SWAT-MODFLOW-RT3D, our modeling framework retains the feature that nitrate loadings to the groundwater domain can be simulated as a function of land use and weather conditions. Contrary to SWAT-MODFLOW-RT3D which can simulate the nitrate concentrations in the entire SWAT-MODFLOW domain, our modeling approach only simulates nitrate concentrations in particular groundwater discharge zones of interest. However, in terms of designing mitigation strategies, our modeling framework offers a clear advantage as it provides the source and age components needed to guide optimal water quality improvement strategies.

5.2 Limitations and Uncertainties

It is important to highlight assumptions made in this study that may limit our findings. The particle-tracking results, and thus the simulated TTDs and nitrate concentrations, depend on the flow fields as simulated by the SWAT-MODFLOW models. In this study we use the equivalent porous medium concept model to a groundwater system characterized by numerous karstic features. Our modeling approach makes simplifying assumptions regarding historical and current land use and assumes that the effective recharge and nitrate loading to MODFLOW are reasonably well simulated by SWAT-MODFLOW. Travel times are only determined for the saturated portion of the MODFLOW domain and do not account

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for travel times through dry MODFLOW cells or the soil layers in SWAT. The application of MODPATH also requires the assumption that reasonable porosity values can be specified and that nitrate transport is purely advective without dispersion. Finally, travel times for nitrate masses as well as simulated nitrate concentrations depend on the assumed denitrification processes and parameters. Possibilities for improving on these assumptions should be investigated in future work. With regard to our Santa Fe River Basin models, future explorations into model equifinality and uncertainty would be useful. For example, in this study we only use a single set of model parameters for the groundwater domain. The uncertainty about these parameters is high, especially within the Upper Floridan Aquifer, due to the presence of karstic voids related to the dissolution of carbonate rocks. In fact, it is reasonable to question the validity of using a single equivalent porous medium layer for this aquifer. However, when using SWAT-MODFLOW alternative representations for the Upper Floridan Aquifer are limited. SWAT-MODFLOW cannot handle turbulent flow in karstic conduits. Moreover, little information exists about the location and geometry of karstic conduits. Nonetheless, alternative representations within the limitations of SWAT-MODFLOW may be possible. For example, the aquifer could be divided into a number of separate layers of porous medium types. By doing so, the duality of flow regimes within the aquifer could be represented; slower flow in the porous limestone matrix and faster albeit laminar flow in karstified horizons. Regardless of representation, however, the uncertainty about the model parameters can only be accounted for by running multiple model realizations. Likewise, we acknowledge that we did not explore the sensitivity of the model parameters that govern the denitrification processes.

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6 Conclusions

This study presents a modeling framework that integrates MODPATH with SWAT-MODFLOW to simulate groundwater nitrate transport in terms of source areas and travel time distributions. By combining backward particle tracking with nitrate loading simulated by SWAT, the approach provides a computationally efficient alternative to fully coupled reactive transport models. Application of the framework to the Santa Fe River Basin demonstrates its effectiveness. Results show that groundwater travel time distributions strongly control nitrate delivery to spring-fed river reaches. Simulated travel times have a median on the order of three decades, indicating substantial lag times between reductions in nitrate loading and measurable improvements in water quality.

In addition to simulating nitrate concentrations at locations of interest, the framework spatially identifies the source areas and travel times that govern nitrate delivery. Linking particle source locations with simulated nitrate loading enables mapping of nitrate contributions from different land uses and quantifying their relative influence on concentrations in groundwater. Results show that mitigation strategies focused on source areas with shorter travel times reduce nitrate concentrations more efficiently than uniform basin-wide land-use changes, although improvements may still occur over multi-decadal time scales due to subsurface transport lags. By explicitly resolving both the age and source components of nitrate transport, the proposed framework provides a practical and transferable tool for prioritizing water quality mitigation strategies in groundwater-dominated watersheds.

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Appendix A

For this study, we made a significant change to the current SWAT-MODFLOW Version 3 code (https://swat.tamu.edu/software/swat-modflow/) regarding the water stages in the river. The original SWAT-MODFLOW code is designed with the intention that the reach water stages as computed by SWAT are used as river stages by MODFLOW. However, in the current SWAT-MODFLOW code, the reach stages are not transferred correctly to corresponding river cells. A river cell may be connected to one or more reaches. At a junction of reaches, for example, a cell can be connected to three reaches. To set the river stage in such a cell, the intention is to find a weighted average of the water depths in the reaches based on their length within the river cell. In the part of the code where this is supposed to be computed, however, the computation is incomplete and thus incorrect. For each cell the code finds the number m, where m is the number of reaches connected to the river cell. This cell is then connected to reaches 1 to m. However, these numbers are not the actual reach numbers to which the cell needs to be connected. After we corrected this coupling issue using the correct reach numbers, however, the river stages became highly oscillatory as a result of the sequential non-iterative coupling between SWAT and MODFLOW. Basically, the water stages are transferred without any further communication between the models during the time step. It is worthwhile to mention that GSFLOW (Markstrom et al., 2008), another model for coupling surface and subsurface flow, uses a sequential iterative approach which is more robust. Here, we circumvent the instability problem, by setting the river stages in MODFLOW equal to the river bottom. This implies that the river exchange flux between SWAT and MODFLOW becomes independent on the reach stages as computed by SWAT. It can be argued that this is reasonable except during periods with relatively extreme variations in the reach stages.

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Appendix B

As mentioned in section 3.2, reasonable travel times are only obtained when we increase the horizontal hydraulic conductivity in the Upper Floridan Aquifer and decrease the horizontal hydraulic conductivity in the Middle Confining Unit and the Lower Floridan Aquifer with respect to the original values in the NFSEG model. To illustrate this in more detail, we run a stand-alone model with the original NFSEG hydraulic conductivity values for the current conditions. Figure B1 shows the TTD for water volumes and indicates the median travel time using the original hydraulic conductivity values is about 150 years. This result does not correspond well with values obtained from CFC measurements (Katz et al., 1999). Upon closer examination of the particle-tracking results, (Figure A1b) it is observed that many particles move through the Lower Floridan aquifer. Contrary to this simulated result the water discharged at the springs is believed to travel mainly through the Upper Floridan Aquifer since it has been subjected more to karstification than the Lower Floridan Aquifer, especially in the unconfined zones (Williams and Kuniansky, 2015). To force more flow through the Upper Floridan Aquifer and obtain reasonable travel times we decrease the horizontal hydraulic conductivity of the Lower Floridan Aquifer and Middle Confining Unit by a factor of 100 and increase the horizontal hydraulic conductivity of the Upper Floridan such that the equivalent horizontal hydraulic conductivity of all three layers remains the same. As a result, our new model and the original model yield similar hydraulic head fields. This is important since the NFSEG is calibrated to hydraulic heads.

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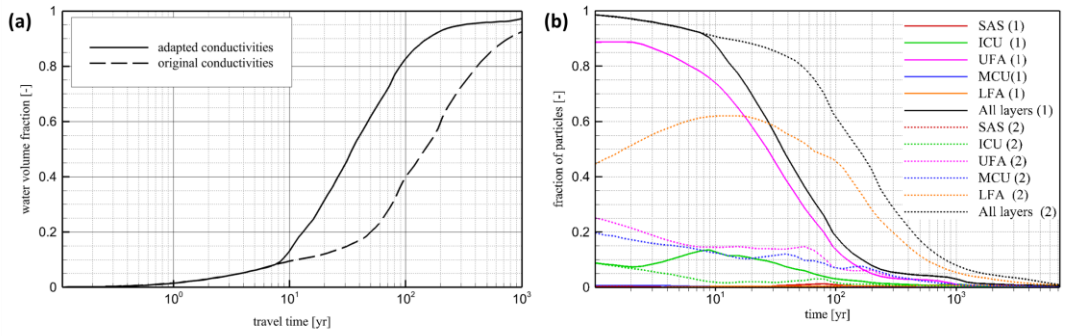

Figure B1. a) Travel time CDF's using original and adapted horizontal conductivities and b) Fraction of particles (with respect to total number of released particles) in each layer as a function of time for original conductivities (solid lines, 1) and adapted conductivities (dashed lines 2).

Code and data availability

The SWAT-MODFLOW model used in this study is available as a CUAHSI HydroShare (Reaver et al., 2025).

Author contributions

RdR: conceptualization, methodology, software, data curation, formal analysis, validation, visualization, writing (original draft); NR: methodology, data curation, formal analysis, validation, writing (review and editing); DL: methodology, software, formal analysis, validation, DK: conceptualization, methodology, formal analysis, validation, funding acquisition, supervision, writing (review and editing), and WG

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conceptualization, methodology, formal analysis, validation, funding acquisition, project administration, supervision, writing (review and editing).

Competing interests

The authors declare there are no conflicts of interest for this manuscript.

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