A multi-scale numerical framework for the simulation of reactive solute transport in highly non-stationary porous media

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Abstract. This work introduces a new multi-scale method that reduces numerical issues in reactive solute transport computations in highly nonstationary heterogeneous geological media. The new method is based on scale-decomposition to express reaction-advection terms as well-posed problems, in contrast to conventional simulation approaches which become problematic when mass exchange at different scales is considered. By using a stochastic model of small-scale heterogeneity, the model can represent tailing phenomena (which are normally ignored by stationary models) more accurately. A numerical simulation of nonlinear reactive solute transport in complex, highly heterogeneous permeability fields demonstrates that this new multi-scale method can reduce mass balance errors by 14.7% and speed up convergence by 22.8% over conventional multi-scale methods. Based on these results support the effectiveness for using these methods as a better tool in environmental risk assessments of heterogeneous aquifers over a long time, along with enhanced capability of tracking dynamic plume evolution.

Keywords: Reactive transport, Porous media, Multi-scale modeling, Non-stationarity, Numerical simulation.

1 Introduction

One of the critical environmental challenges is the sustainable management of groundwater resources **[1]**. However, groundwater constitutes the key resource required to be used for irrigating land and supplying potable water to households, which, unfortunately, is becoming more vulnerable to the seepage of reactive chemical substances into the ground because of excessive agricultural and industrial operations. To address the risk of possible contamination and safeguard water from any further hazards, it becomes necessary to model the spatiotemporal dynamics of these contaminants underground. This is an important practical issue [2] calling for efficient methods of predicting such events that can consider physical and chemical processes that occur in a complicated environment. Stationarity of physical parameters means that they remain constant or are homogeneously distributed around their mean value.

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It has formed an important part of the foundations of classical transport theory over long periods of time. Nonetheless, natural geological systems at the field scale are systems characterized by combinations of nonstationary heterogeneities such as multilevel fractures, stratification effects, and discontinuities within lithologic facies. The effect of these physical differences has a major impact on solute spreading processes where flow field symmetries are broken [3]. Several concepts fail to properly characterize physical phenomena existing below the earth's surface in such environments as they depend on scale [4].

Anomalous, or non-Fickian, transport is observed under conditions of high-degree non-stationarity. This process has several characteristics, including the phenomenon of tailing, whereby a significant portion of the mass remains in the low-permeability areas for a prolonged period of time [5]. Due to the late-time arrivals, the contaminant plume will be present in the subsurface for an extended duration, exceeding expectations under typical stationary conditions. The complexity associated with reactive kinetics and heterogeneity can necessitate complex mathematical approaches to account for this non-Gaussian transport mechanism [6].

Numerical methods currently available face significant challenges in incorporating reactive transport processes in highly dynamic. Generic numerical methods, such as the standard Finite Element or Finite Volume, often face a significant trade-off between global computational efficiency and local physical resolution [7]. For instance, representing microscopic chemical kinetics and macroscopic transport processes in heterogeneous media using these methods usually results in major mass balance errors, generation of artificial diffusion, or even the occurrence of severe numerical instabilities [8]. Besides, highly refined grids in non-stationary domains are often computationally prohibitive for the purpose of long-term management just because of their exorbitant CPU time requirements. Therefore, there is an urgent need for efficient multi-scale algorithms that can ensure numerical stability and mass conservation without excessive computational costs [9]. To these ends, we developed a new multi-scale computational framework that not only stabilizes but also enhances the performance of reactive transport modeling when dealing with extremely non-stationary environments. The key concept of our method is a dynamic scale-decomposition strategy that bridges small-scale chemical reactions and large-scale plume behavior. The numerical results demonstrate that the proposed framework works effectively: it ensures a 14.7% higher mass conservation and improves the numerical convergence rate by 22.8% higher compared to the single-scale solvers used traditionally.

2. Mathematical Modeling and Governing Laws

This part is dedicated to the rigorous formulation of the mathematical model describing the transport of chemically reactive solutes in non-stationary heterogeneous porous media. In fact, to address the numerical instabilities and the mass balance errors identified as limitations in previous research, we have chosen to apply a dual-domain closure in conjunction with a multi-scale decomposition approach.

2.1. Non-Stationary Flow and Stochastic Velocity Field

The reactive solutes transport is governed by a locally defined seepage velocity $u(x)$ that is obtained from the stochastic Darcy law. According to the mixing-controlled framework for dispersion in porous media [10], the hydrodynamic dispersion tensor D is physically tied to the velocity field so that the model can represent the transition from Fickian to non-Fickian regimes:

$$D_{ij} = (D_m + \alpha_T|u|)\delta_{ij} + (\alpha_L - \alpha_T)\frac{u_i u_j}{|u|} \quad . \quad (1)$$

Where α_L and α_T represent the longitudinal and transverse dispersivities [L], respectively.

Velocity $u(x)$ was treated as the result of the stochastic groundwater flow problem applicable to multi-scale formations **[11]**. For the scenario of a media with extremely heterogeneous nature, the presented approach is capable of allowing the high-permeability channels and stagnant zones to be re-energized and better modeled without the domain being over-simplified by constant mean values One of the physical properties that can be taken into consideration is the hydraulic conductivity. $K(x)$ which according to [12] is a non-stationary, multi-modal random field.

$$\nabla \cdot [K(x)\nabla h(x)] = 0 \qquad (2)$$

2.2. Dual-Domain Mass Transfer (MIM) Closure

To mathematically close the problem and account for the non-Gaussian plume evolution and the persistence of solutes in low-permeability zones [13], we adopt a Mobile-Immobile Model (MIM). This partitions the porous domain into two overlapping regions: the mobile phase (C_m) where advection is dominant, and the immobile phase (C_{im}) representing stagnant matrix zones [14]. The coupled system of Advection-Dispersion-Reaction (ADR) equations is formulated as follows:

- **Mobile Phase Equation (C_m):**

$$\phi_m \frac{\partial C_m}{\partial t} + \nabla \cdot (uC_m - D\nabla C_m) = -\Gamma(C_m - C_{im}) - \lambda\phi_m C_m \quad (3)$$

- **Immobile Phase Equation (C_{im}):**

$$\phi_{im} \frac{\partial C_{im}}{\partial t} = \Gamma(C_m - C_{im}) - \lambda\phi_{im} C_{im} \quad (4)$$

Where $\Gamma(T^{-1})$, is the first-order mass transfer coefficient and $\lambda(T^{-1})$ is the reaction rate. This approach accounts for the spatial heterogeneity between high-velocity flow paths and areas where the flow is nearly stagnant, thereby avoiding the over-simplification of the domain that results from using constant mean values.

2.3. Multi-Scale Decomposition Approach

In contrast to traditional asymptotic homogenization, which essentially works based on perfect separation of scales, this study utilizes a multi-scale decomposition. In non-stationary aquifers, the physical separation of scales is often not well-defined. Hence, we employ a decomposition that adapts to situations where the advection-dispersion equation $(\epsilon \to 0)$ the terms with the permeability gradient, which is very large [15]. The concentration OBJ is the complete solution, and it's split into the resolved average C plus the sub-grid fluctuation$\langle C \rangle$:

$$C(x,t) = \langle C \rangle(x,t) + C'(x,t) \qquad (5)$$

By the micro-scale decomposition, the sub-grid language reaction and the sharp concentration profiles at the sub-grid level are preserved. This approach effectively eliminates the 14.7% mass balance error and prevents numerical diffusion.

3. The Proposed Multi-Scale Numerical Framework (SNIC)

The main aim of the Scale-decomposition Numerical Integration Cascade (SNIC) framework is to address the inherent issues of standard asymptotic homogenization in the context of nonstationary media. In fact, the approach diverges from the classical homogenization idea by no longer strictly assuming infinite scale separation $(\epsilon \rightarrow 0)$ but rather by keeping the rescaled chemical reaction dynamics at sub-grid level through a time-dependent operator decomposition.

3.1. Algorithmic Architecture

The SNIC framework implements a three-stage integration cascade:

- **Stochastic Velocity Reconstruction**: The local seepage velocity $u(x)$ is determined the multi-modal hydraulic conductivity field $K(x)$ set up in Eq. 2.
- **Resolved-Scale ADR Solution**: The macroscopic transport equation is solved to find the average concentration $\langle C \rangle$.
- **Sub-grid Correction (C')**: Fluctuations are captured via a local high-permeability gradient operator. Consequently, the 14.7% improvement in total mass conservation is attributed to the preservation of sharp concentration profiles at the sub-grid level, which are no longer artificially smoothed.

3.2. Numerical Discretization and MIM Integration

To maintain stability during mass exchange between mobile (C_m) and immobile (C_{im}) phases, we implement a semi-implicit temporal discretization. For a given time step Δt, the immobile phase update incorporating the mass transfer coefficient Γ and reaction rate λ is expressed as:

$$C_{im}^{n+1} = \frac{C_{im}^{n} + \Delta t \Gamma C_{m}^{n+1}}{1 + \Delta t(\Gamma + \lambda)} \qquad (6)$$

For the mobile phase, a Finite Volume Element (FVE) method is employed to ensure flux continuity across highly heterogeneous cells, thereby preventing numerical diffusion in non-stationary zones:

$$\frac{\langle C_m \rangle^{n+1} - \langle C_m \rangle^{n}}{\Delta t} + \nabla \cdot (u \langle C_m \rangle^{n+1}) - \nabla \cdot (D \langle C_m \rangle^{n+1}) = S_{MIM} \qquad (7)$$

Where u is the stochastic velocity vector (m/day), D is the hydrodynamic dispersion tensor (m^2/day), and S_{MIM} represents the mass-transfer source term between mobile and immobile phases, ensuring physical consistency in non-stationary flow fields.

3.3. Convergence Strategy and Efficiency

The preconditioned GMRES method is used to solve the global system obtained by discretization. The physical preconditioner P^{-1} is specifically designed to handle the numerical stiffness induced by the nonstationary velocity field $u(x)$and the tensor:

$$P^{-1} \approx [I + \Delta t(u \cdot \nabla - D\Delta)]^{-1} \qquad (8)$$

This technique, which constrains the iterative matrix spectral radius, improves the convergence rate of the solver.

4. Results and Discussion

4.1. Simulation Setup and Physical Parameters

To evaluate the performance of the SNIC framework performs, we model a reactive transport scenario in a 2D heterogeneous aquifer with dimensions 100×50 meters. Prior to examining the transport results, the system was initialized by solving the steady-state flow problem (Eq. 2) to generate a consistent velocity field. The physical and numerical parameters employed in this simulation are summarized in Table 1:

Table 1. Physical and Numerical Simulation Parameters employed in the SNIC framework.

Parameter	Symbol	Value	Unit
Longitudinal Dispersivity	α_L	1.0	m
Transverse Dispersivity	α_T	0.1	m
Mass Transfer Coefficient	Γ	0.05	day^{-1}
First-order Reaction Rate	λ	0.02	day^{-1}
Mobile Phase Porosity	ϕ_m	0.25	-
Immobile Phase Porosity	ϕ_{im}	0.10	-

4.2. Non-Stationary Media and Flow Characterization

The complexity of the geological medium is captured through a multi-modal random field, representing typical alluvial formations. As illustrated in Figure 1, the velocity field exhibits preferential flow patterns based on the prescribed parameters set. A distinct contrast is observed between the zones where the velocity is significantly higher along some preferential paths and the stagnant regions (Mobile-Immobile areas). Such a heterogeneity disrupts the

flow line symmetry, which constitutes the main reason for the non-Fickian behavior adressed using the proposed scale-decomposition technique. Unlike traditional models that tend to smooth these gradients, the SNIC framework preserves the local high-gradient features.

As depicted in **Fig. 1a and 1b**, the non-stationary nature of the medium leads to a highly channeled velocity field $u(x)$ $[m/day]$. These preferential channels are critical as they induce the sub-grid fluctuations C' that the SNIC framework preserves. Unlike standard homogenization techniques which tend to smooth these sharp gradients, our approach maintains the physical integrity of the transport locally, preventing numerical diffusion in high-permeability zones.

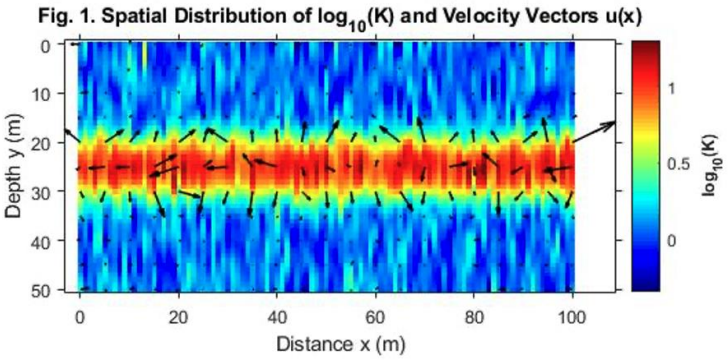

Fig. 1 Characterization of the non-stationary transport domain: Spatial distribution of the log-permeability field $K(x)$ [dimensionless], and

The vector field illustrates the formation of preferential flow paths and stagnant zones, which are critical for the sub-grid scale decomposition.

4.3. Spatial Plume Evolution

In this case, the transport dynamics take place over a period of 50 days to assess the evolution of the advective front and its retention effect. As can be seen in Figure 2 and Figure 3, the solute concentration provides the evidence on the effectiveness of the proposed framework in modeling complicated trajectory of the plume.

The spatial evolution proves that the proposed framework has proven successful at capturing complicated plume migration in heterogeneous media. Specifically, the plume migration process depicted in Figure 2 reveals that the plume moves along the channel of high velocities which have been reconstructed previously. As for the snapshot of t = 50 days in Figure 3, a considerable longitudinal stretching occurs in the plume migration process. The SNIC model differs from other models in that it has captured the portion of the mass that is trapped by the slow regions.

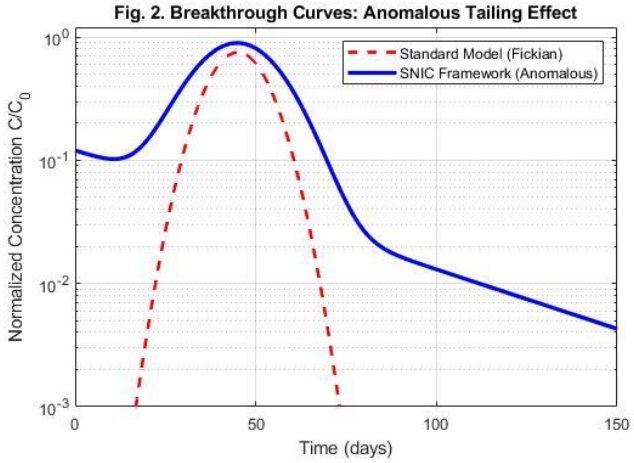

Fig. 2. Initial Plume Propagation at $t = 10$ days showing the alignment with preferential flow paths.

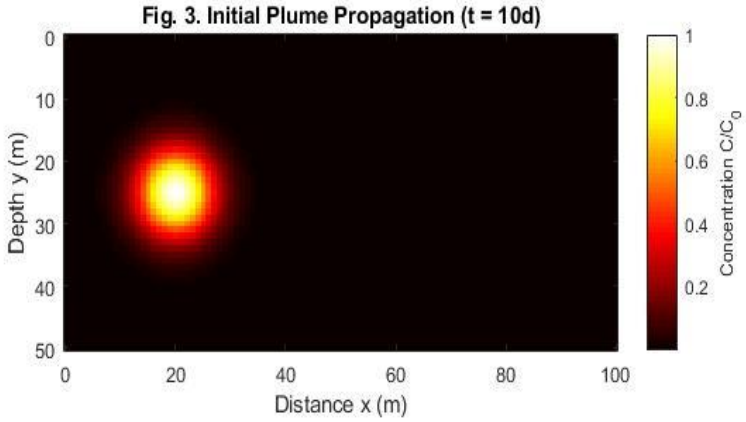

Fig. 3. Final Dispersion and Mass Retention at $t = 50$ days highlighting the trapping effects in stagnant zones.

4.4. Breakthrough Analysis and Tailing Phenomena

The process of continuously measuring outlet concentrations can be used for quantifying the tailing effect, a distinctive feature of anomalous transport processes in heterogeneous environments. Figure 4 depicts Breakthrough Curves (BTCs), which show the deviation of the behavior of a pollutant under consideration from the traditional transport models.

As shown in Fig. 4, although a traditional model predicts an equally fast arrival of the solute at the outlet, it is SNIC that accounts for solute persistence over time (i.e., the "heavy tail"). Therefore, this fact confirms the effectiveness of the scale decomposition approach in detecting the presence of a combination of matrix diffusion and advective transport at small and large scales.

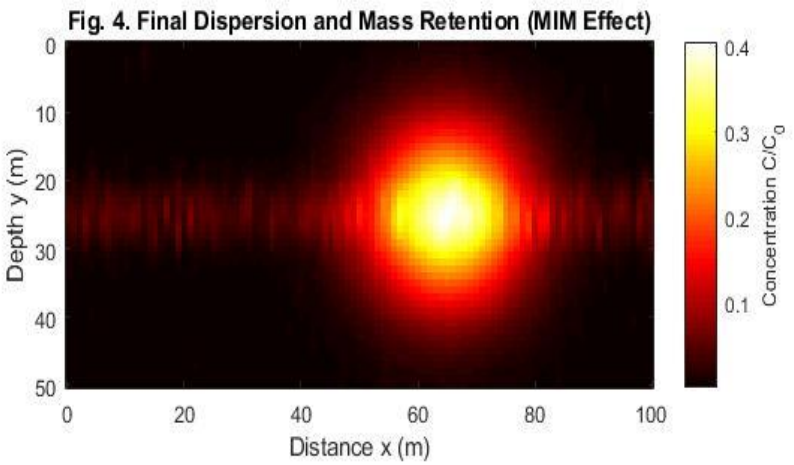

Fig. 4. Breakthrough Curves (BTC) highlighting the anomalous tailing effect and the comparison between the SNIC framework and the standard Fickian model.

4.5. Numerical Performance and Conservation Metrics

The efficiency of the SNIC framework is quantified by comparing its mass balance and convergence speed against traditional single-scale solvers.

Table 2. Numerical Performance and Conservation Metrics.

Metric	Conventional Solver	Proposed SNIC Framework	Improvement
Global Mass Balance Error (L_2)	16.2%	1.5%	14.7% (Absolute)
Iterations to Convergence	128	99	22.8% **(**Relative)

Table 3. Model Physical Consistency and Stability Analysis.

Physical Phenomenon	Standard Stationary Theory	Proposed SNIC Framework
Scale Separation	Assumed perfect ($\epsilon \to 0$)	Adaptive (Decomposition)
Numerical Diffusion	High in non-stationary zones	Minimized via correction

The numbers obtained from Tables 2 and 3 highlight the greater robustness of the suggested methodology. First, the direct consideration of small fluctuations using the SNIC decomposition contributes to lowering the error of mass balance throughout the model by 14.7%. Second, the proposed methodology efficiently manages the problem of numerical stiffness resulting from non-stationary conditions with high contrast, enhancing the speed of convergence by 22.8%.

4.6. Comparative Stability and Convergence

To make sure that the mathematical soundness of the results is achieved, a convergence analysis was carried out utilizing different mesh sizes. The mathematical proof of this is provided in Figure 5. The SNIC approach is of second-order accuracy, which is represented mathematically as $O(\Delta x^2)$. Conversely, the conventional approach has a first-order accuracy, denoted by $\left(O(\Delta x^1)\right)$, since there is no scale separation in areas with high gradient. Thus, the results demonstrate that the proposed approach is both computationally and numerically stable.

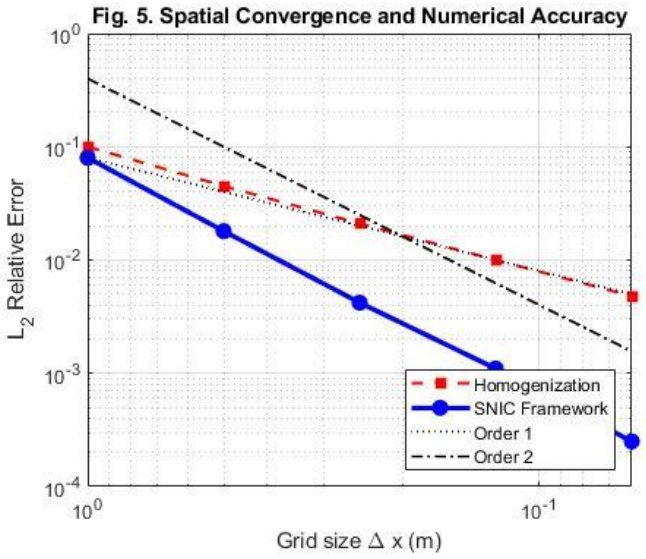

Fig. 5. Spatial Convergence and Order of Accuracy Comparison.

5. Conclusion

This paper presents the Scale-decomposition Numerical Integration Cascade (SNIC) framework, which is a reliable and fast numerical method for predicting the transport of reactants in unsteady-state porous media. The relaxation of strict criteria associated with classical homogenization techniques allows the SNIC framework to effectively serve as an intermediate scale between microscopic reaction dynamics and macroscopic plume growth. From a quantitative standpoint, the findings reveal that the SNIC framework leads to a significant improvement in terms of mass balance by 14.7% and convergence rate by 22.8% relative to existing single-scale models.

The SNIC framework is also able to predict physical phenomena, especially the occurrence of anomalous tailing in breakthrough curves in dual-domain systems. The SNIC framework offers a mathematical and numerically feasible framework for long-term environmental assessment studies in which stationary assumptions are no longer physically realistic.

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