



## RESEARCH LETTER

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## Key Points:

- We identify two fundamental time scales associated with diffusive transport problems
- We provide a theoretical basis for explaining the differences between steady state and steady shape concepts
- We present a novel theory for quantifying the time scales needed for a diffusive system to approach steady shape and steady state conditions

## Supporting Information:

- Supporting Information S1

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## Understanding time scales of diffusive fluxes and the implication for steady state and steady shape conditions

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**Abstract** The diffusion equation is one of the most commonly used models for describing environmental problems involving heat, solute, and water transport. A diffusive system can be either *transient* or *steady state*. When a system is transient, the dependent variable (e.g., temperature, concentration, or hydraulic head) varies with time; whereas at steady state, the temporal variations are negligible. Here we consider an intermediate state, called *steady shape*, corresponding to the situation where temporal variations in diffusive fluxes are negligible but the dependent variable may remain transient. We present a general theoretical framework for identifying *steady shape* conditions and propose a novel method for evaluating the time scale needed for a diffusive system to approach both steady shape and steady state conditions.

### 1. Introduction

Environmental transport problems involving heat, solute, or groundwater flow are typically classified as being either *transient* or *steady state*. Transient problems involve temporal variations in the dependent variable (temperature, concentration, or hydraulic head), whereas steady state problems do not involve any temporal variations. In some geoscience fields (e.g., groundwater), previous researchers have postulated the existence of another state, often called *steady shape*, which has been used to describe the situation where the shape of the solution is steady, but the dependent variable may still vary with time; hence, the system remains transient [Bohling *et al.*, 2002; Heath, 2009]. Heath [1983] refers to this as a *steady shape* condition, while Kruseman and Ridder [1990] refer to this as a *transient steady state* condition.

An important feature of the steady shape condition is that it is thought to be an intermediate state that arises well before the system reaches steady state. In the literature, it is also broadly assumed that the transport process during the steady shape period can be analyzed by using simpler mathematical models that are normally used for describing steady problems [Bohling *et al.*, 2002; Butler, 1988]. The ability to use simpler models can have profound practical consequences since it can considerably reduce computational time and effort. However, despite its use, steady shape approximations are invoked heuristically, without any fundamental theoretical understanding of when steady shape conditions are relevant. The objective of this work is to address this fundamental limitation. To achieve this, we develop a formal theoretical basis for defining steady shape conditions and use this definition to examine the relevance of steady shape conditions when modeling a range of diffusive transport problems.

The cornerstone of the diffusion equation is the concept of flux, which, as defined by Fourier's law, relates the transport of an extrinsic property (e.g., heat) with the spatial gradient of an intrinsic property (e.g., temperature) [Carslaw and Jaeger, 1959; Crank, 1979]. The macroscopic quantity flux, which is proportional to the spatial gradient of the dependent variable, quantifies the amount of an extrinsic property transported across a unit area per unit time. The flux concept has been used for modeling several types of environmental transport systems including solute transport (where solute flow is linked with concentration gradient) and groundwater flow (where the groundwater flow is linked with hydraulic head gradient). In the groundwater literature, researchers have identified that pumping problems appear to first reach an intermediate steady shape condition where hydraulic fluxes are effectively steady, while the hydraulic head continues to vary with time [Bohling *et al.*, 2002; Butler, 1988]. Understanding how fluxes might approach steady state conditions at a different rate when compared to how the hydraulic head approaches steady state conditions is a fundamental issue that could shed some light on how steady shape conditions develop.

To quantify the time scales required for a diffusive process to approach steady state, we employ a concept called mean action time (MAT), which was originally proposed for analyzing heat transport [McNabb and Wake, 1991; McNabb et al., 1991]. Later researchers adapted this approach for modeling more general transport problems [Ellery et al., 2012a; Jazaei et al., 2014, 2016; Landman and McGuinness, 2000; McGuinness et al., 2000; McNabb and Keady, 1994; Simpson et al., 2013]. However, all of these studies focus on quantifying the time scale associated with the dependent variable (e.g., hydraulic head, concentration, or temperature). Here we adapt the diffusion equation to model transient changes in diffusive fluxes. We hypothesize that the time scale associated with the dependent variable could be different from the time scale associated with the flux variable. These differences in time scales will be quantified by using MAT theory to develop a formal mathematical basis for understanding the differences between steady shape and steady state conditions.

## 2. Development of a General Mathematical Framework of Modeling Steady State Fluxes

We first review the mathematical framework needed for computing the time scale required for a diffusive variable to approach steady state. We then extend the framework to compute the time scale required for the flux to approach steady state. In this study we use nondimensional variables to generalize the analysis. All dimensional variables are primed ( $'$ ), and dimensionless variables are unprimed.

The aim of our analysis is to quantify the time scales required for a diffusive variable,  $\phi'$ , and the associated flux,  $J' = -K' \partial \phi' / \partial x'$  (where the constant  $K' > 0$  is a parameter), to effectively asymptote to their respective steady states. We begin with the dimensional form of the diffusion equation,

$$\frac{\partial \phi'(x', t')}{\partial t'} = D' \frac{\partial^2 \phi'(x', t')}{\partial x'^2} + W', \quad 0 < x' < L', \quad (1)$$

where  $\phi'(x', t')$  is the dependent variable at location  $x'$  and time  $t'$ ,  $D' > 0$  is the diffusivity,  $W'$  is the spatially uniform source/sink term, and  $L'$  is the length of the domain. We define three dimensionless variables,

$$x = \frac{x'}{L'}, \quad \phi(x, t) = \frac{\phi'(x', t')}{\Phi'}, \quad t = \frac{t' D'}{L'^2}. \quad (2)$$

Using these variables, the nondimensional governing equation is

$$\frac{\partial \phi(x, t)}{\partial t} = \frac{\partial^2 \phi(x, t)}{\partial x^2} + w, \quad 0 < x < 1, \quad (3)$$

where  $w = W' L'^2 / D' \Phi'$  and  $\Phi'$  is a characteristic value of the dependent variable.

To quantify how the flux varies with time and position, we differentiate both sides of equation (3) with respect to  $x$ . Writing  $g(x, t) = \partial \phi(x, t) / \partial x$ , we obtain

$$\frac{\partial g(x, t)}{\partial t} = \frac{\partial^2 g(x, t)}{\partial x^2}, \quad 0 < x < 1, \quad (4)$$

where the dimensionless flux is  $J = -g(x, t)$ . Note that the equations governing the evolution of the dependent variable and the flux variable are similar for this Cartesian problem, except that there is no source/sink term in equation (4). However, the solutions of equations (3) and (4) will differ because they involve a different set of initial and boundary conditions.

## 3. Application of the MAT Theory to Evaluate the Time Scales of Diffusive Fluxes

To derive an expression for the MAT of  $\phi(x, t)$ , we begin by considering two fundamental quantities [Jazaei et al., 2014, 2016; McNabb and Wake, 1991; Simpson et al., 2013]

$$F_\phi(t|x) = 1 - \left[ \frac{\phi(x, t) - \phi_\infty(x)}{\phi_0(x) - \phi_\infty(x)} \right], \quad t \geq 0, \quad (5)$$

$$f_\phi(t|x) = \frac{dF_\phi(t|x)}{dt} = - \frac{\partial}{\partial t} \left[ \frac{\phi(x, t) - \phi_\infty(x)}{\phi_0(x) - \phi_\infty(x)} \right], \quad t \geq 0, \quad (6)$$

where  $\phi_0(x)$  is the initial value of  $\phi(x, t)$  (initial condition) and  $\phi_\infty(x)$  is the long-time steady value of  $\phi(x, t)$  (final steady state condition). MAT theory depends on interpreting  $F_\phi(t|x)$  as a cumulative distribution function (CDF), and therefore, this approach is valid for transitions where  $F_\phi(t|x)$  increases monotonically from  $F_\phi(0|x) = 0$  and approaches  $F_\phi(t|x) = 1$ , as  $t \rightarrow +\infty$ . Under these conditions,  $f_\phi(t|x)$  can be interpreted as a probability density function. The mean, or first moment of this distribution, gives a time scale that is called the MAT [Ellery et al., 2012a, 2012b; Jazaei et al., 2014, 2016; Simpson et al., 2013]. This time scale provides an estimate of the amount of time required for  $\phi(x, t)$  to asymptote from  $\phi_0(x)$  to  $\phi_\infty(x)$ . The dimensionless MAT is

$$M_\phi(x) = \int_0^\infty t f_\phi(t|x) dt. \quad (7)$$

One of the advantages of working with the MAT framework is that it is possible to solve for  $M_\phi(x)$  without solving equation (3) for  $\phi(x, t)$  [Landman and McGuinness, 2000; Simpson et al., 2013]. To solve for  $M_\phi(x)$  we require explicit expressions for  $\phi_0(x)$  and  $\phi_\infty(x)$ .

We now extend the MAT theory to evaluate the time scale associated with the transition in terms of the diffusive flux by defining

$$F_g(t|x) = 1 - \left[ \frac{g(x, t) - g_\infty(x)}{g_0(x) - g_\infty(x)} \right], \quad t \geq 0, \quad (8)$$

$$f_g(t|x) = \frac{dF_g(t|x)}{dt} = -\frac{\partial}{\partial t} \left[ \frac{g(x, t) - g_\infty(x)}{g_0(x) - g_\infty(x)} \right], \quad t \geq 0, \quad (9)$$

where  $g_0(x)$  is the initial value of  $g(x, t)$  and  $g_\infty(x)$  is the steady value of  $g(x, t)$ . Once again, we interpret  $F_g(t|x)$  as a CDF provided that  $F_g(t|x)$  increases monotonically from  $F_g(0|x) = 0$  and asymptotes to  $F_g(t|x) = 1$  as  $t \rightarrow +\infty$ . The MAT of the flux variable is

$$M_g(x) = \int_0^\infty t f_g(t|x) dt. \quad (10)$$

To compute  $M_g(x)$ , we first evaluate  $M_\phi(x)$  and then develop a relationship between  $M_\phi(x)$  and  $M_g(x)$ . To accomplish this, we combine equations (9) and (10) and interchange the order of differentiation. Since  $g(x, t) = \partial\phi(x, t)/\partial x$ , we obtain

$$M_g(x) = \frac{-1}{g_0(x) - g_\infty(x)} \frac{\partial}{\partial x} \int_0^\infty t \frac{\partial[\phi(x, t) - \phi_\infty(x)]}{\partial t} dt. \quad (11)$$

Combining equations (6) and (11) gives

$$M_g(x) = \frac{1}{g_0(x) - g_\infty(x)} \frac{d}{dx} \left[ (\phi_0(x) - \phi_\infty(x)) \int_0^\infty t f_\phi(t|x) dt \right]. \quad (12)$$

The integral in equation (12) is  $M_\phi(x)$ , as in equation (7). Therefore, equation (12) simplifies to

$$M_g(x) = M_\phi(x) + \frac{C_\phi(x)}{dC_\phi(x)/dx} \left( \frac{dM_\phi(x)}{dx} \right), \quad (13)$$

where  $C_\phi(x) = \phi_\infty(x) - \phi_0(x)$  and  $dC_\phi(x)/dx = g_\infty(x) - g_0(x)$ . Note that equation (13) is an important contribution of this work because it describes an explicit relationship between the time scales associated with the dependent variable,  $M_\phi(x)$ , and the time scale associated with the flux variable,  $M_g(x)$ . In the supporting information document, we extend this analysis to solve a diffusion problem in radial coordinates and a two-dimensional diffusion problem in Cartesian coordinates (see Texts S2 and S3 in the supporting information).

#### 4. Case Studies

We apply the MAT expressions developed here to quantify the time scales associated with the dependent variable and the flux variable in two test cases. Case I involves a diffusive problem with no source/sink term and a Dirichlet boundary condition at  $x = 1$ . The initial condition,  $\phi(x, 0)$  or  $\phi_0(x)$ , is spatially uniform. Transient conditions are induced by applying a constant flux at  $x = 0$ . This test case could represent the hydraulic head

in a confined aquifer with uniform initial condition, a fixed head at the right boundary, and a constant flux at the left boundary. Furthermore, this case could also represent the temperature distribution in thermal conductor with constant initial temperature, a fixed temperature at the right boundary, and the left boundary losing heat at a constant rate.

Case II involves a spatially uniform initial condition, a zero flux condition at  $x=0$ , and fixed head boundary conditions at  $x=1$ . A constant source/sink term is included to induce a transient transition. This case could represent hydraulic head mounding in an aquifer with uniform initial condition and receiving areal recharge from above, or it could represent a thermal conductor heated from above. The initial and boundary conditions in these two test cases are

$$\text{Case I : IC } \phi_0(x) = 1; \text{ BCs } \frac{\partial \phi(0, t)}{\partial x} = \alpha \text{ and } \phi(1, t) = 1; w = 0, \quad (14)$$

$$\text{Case II : IC } \phi_0(x) = 1; \text{ BCs } \frac{\partial \phi(0, t)}{\partial x} = 0 \text{ and } \phi(1, t) = 1; w \neq 0, \quad (15)$$

where  $\alpha$  and  $w$  are constants.

To solve for the MAT, we apply integration by parts to equation (7) and note that  $\phi(x, t)$  approaches  $\phi_\infty(x)$  exponentially fast as  $t \rightarrow +\infty$  [Crank, 1979; Simpson et al., 2013]; then differentiating the result twice with respect to  $x$ , and combining the resulting expression with equation (3), leads to a second-order boundary value problem for  $M_\phi(x)$ ,

$$\frac{d^2 [M_\phi(x) C_\phi(x)]}{dx^2} = -C_\phi(x). \quad (16)$$

To solve equation (16) we need to evaluate  $C_\phi(x) = \phi_\infty(x) - \phi_0(x)$ , where  $\phi_\infty(x)$  is found by evaluating equation (3) at steady state. The  $C_\phi(x)$  functions for the two cases are

$$\text{Case I : } C_\phi = \alpha(x - 1), \quad (17)$$

$$\text{Case II : } C_\phi = \frac{w(-x^2 + 1)}{2}. \quad (18)$$

The boundary conditions required to solve equation (16) are [Jazaei et al., 2016; Simpson et al., 2013]

$$\text{Case I : } \frac{d M_\phi(0)}{dx} - M_\phi(0) = 0; \quad \frac{d M_\phi(1)}{dx} = 0, \quad (19)$$

$$\text{Case II : } \frac{d M_\phi(0)}{dx} = 0; \quad 2 \frac{d M_\phi(1)}{dx} + M_\phi(1) = 0. \quad (20)$$

Further details of these boundary conditions are given in Text S1. The MAT expressions for the dependent variable, obtained by using equations (16)–(20), are

$$\text{Case I : } M_\phi(x) = \frac{1}{6} (2 + 2x - x^2), \quad (21)$$

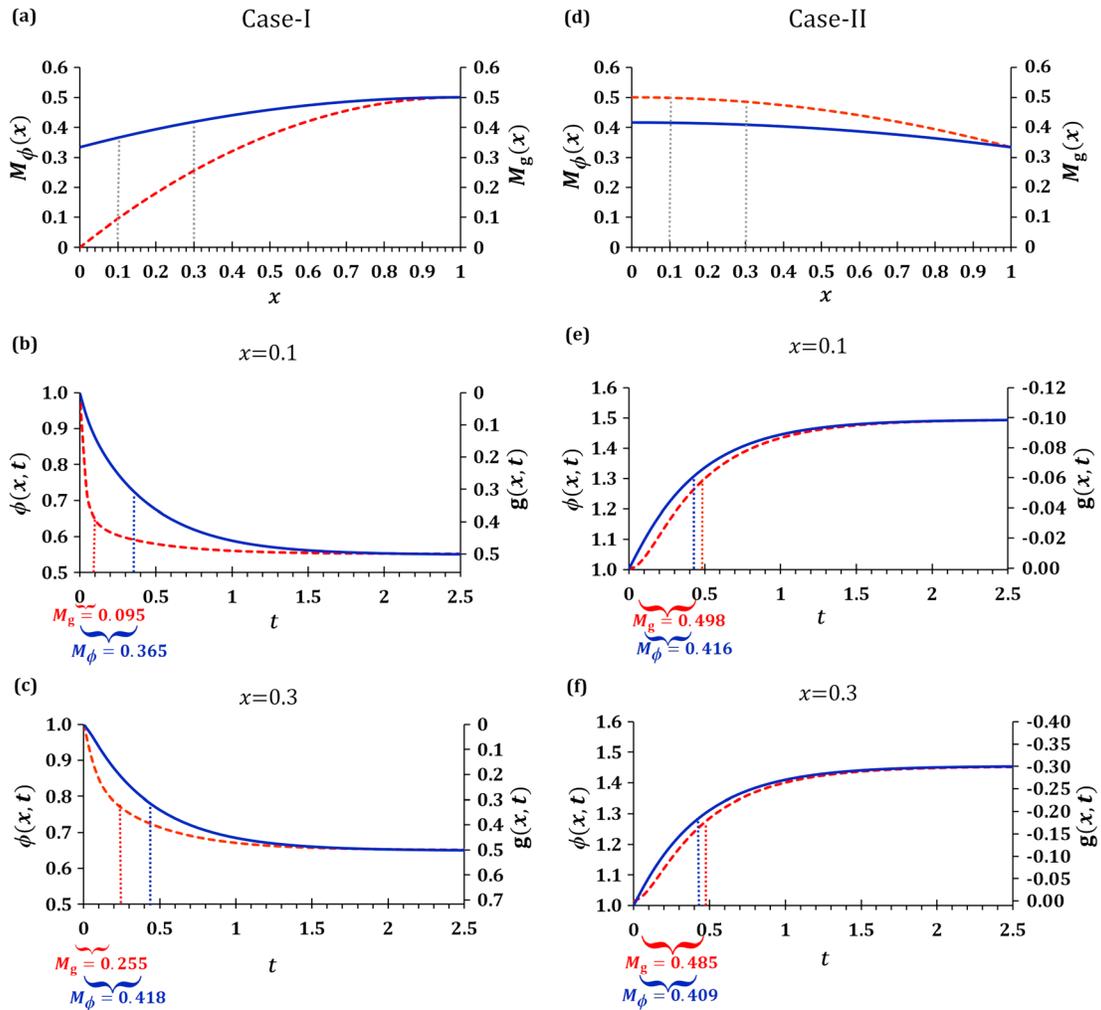
$$\text{Case II : } M_\phi(x) = \frac{1}{12} (5 - x^2). \quad (22)$$

In contrast, MAT expressions for the flux, obtained by using equations (13), (21), and (22), are

$$\text{Case I : } M_g(x) = \frac{1}{2} (2x - x^2), \quad (23)$$

$$\text{Case II : } M_g(x) = \frac{1}{6} (3 - x^2). \quad (24)$$

Note that these MAT expressions give nondimensional time scales. If required, the dimensional time is given by  $\tau'(x') = M(x'/L')(L'^2/D')$ .



**Figure 1.** (left) Case I results. (right) Case II results. (a and d) The spatial variation in MAT for the dependent variable,  $M_\phi(x)$  (solid blue), and the spatial variation in MAT for the flux,  $M_g(x)$  (dashed red) for Case I and Case II, respectively. (b and c) Results show the numerical solutions of  $\phi(x, t)$  (solid blue) and  $g(x, t)$  (dashed red) for Case I at  $x=0.1$  and  $x=0.3$ , respectively. (e and f) Results show the numerical solutions of  $\phi(x, t)$  (solid blue) and  $g(x, t)$  (dashed red) for Case II at  $x=0.1$  and  $x=0.3$ , respectively. Parameters are  $\Delta x = 0.0005$ ,  $\Delta t = 0.05$ ,  $w = 1$ , and  $\alpha = 0.5$ .

## 5. Results

### 5.1. Case I

Figure 1a shows the time scales required for the dependent variable and the diffusive flux variable, computed using equations (21) and (23), as functions of position. The data show considerable differences between the two time scales, with  $M_g(x) < M_\phi(x)$ . This is consistent with the conventional assumption that steady shape conditions arise before steady state conditions. To validate these findings we also solve the governing partial differential equations, with appropriate initial and boundary conditions, using an implicit finite difference method to estimate  $\phi(x, t)$  and  $g(x, t)$  for the two test problems. These two functions, evaluated at two fixed spatial locations, are given in Figures 1b and 1c. The figures also show the time scales,  $M_\phi(x)$  and  $M_g(x)$ , computed by using the appropriate exact MAT expressions. These solutions confirm that the MAT elegantly characterize the time scales required for  $\phi(x, t)$  and  $g(x, t)$  to approach to their respective steady states. Furthermore, the numerical results confirm that in this case, the time scales associated with the dependent variable and its flux can be different and that  $g(x, t)$  approaches steady state faster than  $\phi(x, t)$ .

### 5.2. Case II

Figure 1d shows the time scales required for the dependent variable and the flux variable to approach their respective steady states at all locations for the second test problem. Unlike Case 1, the difference in the two

time scales is relatively small, meaning that the steady shape condition is not so relevant in this case. Numerical data for  $\phi(x, t)$  and  $g(x, t)$  are given in Figures 1e and 1f, confirming that  $g(x, t)$  approaches steady state almost similar to  $\phi(x, t)$ .

### 5.3. Concluding Remarks

In this work we quantify differences in the time scales associated with the dependent variable and the flux variable to reach steady conditions in a diffusive transport system. These two time scales are associated with the development of steady state and steady shape conditions, respectively. We employ the theory of MAT to quantify these differences. These results are general and applicable to any problem that can be modeled by using the linear diffusion equation for which the transient solution asymptotes to a steady state solution. This includes problems such as groundwater flow in confined aquifers and heat flow in thermal conductors. In the groundwater literature, researchers have pointed out the possibility of groundwater flow systems reaching an intermediate condition, often called steady shape [Bohling *et al.*, 2002; Butler, 1988; Heath, 2009]. However, there has been no theoretical basis for objectively identifying steady shape conditions nor has there been any theoretical framework developed to quantify the differences in time scales associated with steady shape and steady state conditions. Interestingly, of the two test problems we consider, only the first problem exhibits a clear and prolonged steady shape transition period. Therefore, the concept of steady shape conditions may not always be relevant. Understanding the relevance of steady shape conditions is difficult without the kinds of analytical tools presented in this work. Without a formal mathematical definition of steady shape conditions, and exact expressions for  $M_\phi(x)$  and  $M_g(x)$ , we are limited to studying numerical solutions of transient partial differential equations on a case-by-case basis. In contrast, our formal mathematical framework provides very general insight and can be adapted to apply to a range of other problems with different boundary conditions, initial conditions, and multidimensional problems (which are discussed in Texts S2 and S3).

Previous analyses of transient diffusive problems have primarily employed scaling arguments to argue that the time scale required to reach steady state or steady shape conditions is proportional to  $L^2/D'$ , where  $L'$  is a characteristic length scale [Cussler, 1997; Gelhar and Wilson, 1974; Heath, 2009; Manga, 1999; Turcotte and Schubert, 1982]. However, it is unclear how to choose the proportionality constant. For example, Heath [2009] presents an empirical formula to estimate the critical time (in minutes) required for steady shape conditions to develop near a pumping well:  $\tau' = 7500r^2/D_a'$ , where  $r'$  is distance (m) from the pumping well and  $D_a'$  is the aquifer diffusivity ( $\text{m}^2/\text{d}$ ). Cussler [1997] states that diffusive systems are expected to approach steady state conditions when the Fourier number,  $L^2/D't'$ , is much less than unity. Our analysis shows that these empirical arguments neglect the fact that the proportionality term in the time scale relationship is not a constant, but it depends on position. Furthermore, the functional relationship itself is different for steady state or steady shape conditions. Our proposed mathematical framework provides an objective method for deriving analytical expressions for these relationships. Our analysis also quantifies the role of boundary conditions and demonstrates their importance in determining steady state and steady shape time scales.

A key aspect of our work is that we make use of the fact that if  $\phi(x, t)$  evolves according to a linear diffusion equation, then the diffusive flux also evolves according to a linear diffusion equation, however with a different set of boundary conditions and a modified source term. These subtle differences in boundary conditions lead to the differences in the time scales. For example, if a diffusion equation describing the evolution of  $\phi(x, t)$  involves a constant flux boundary condition, then the corresponding equation describing the evolution of  $\partial\phi/\partial x$  will involve a Dirichlet boundary condition. However, there are many cases for which transforming the boundary conditions for the equation governing the evolution of the dependent variable into the boundary conditions associated with the evolution of the flux is challenging. For example, consider a one-dimensional diffusion problem with two Dirichlet boundary conditions. Without solving the transient equation for  $\phi(x, t)$ , only the value of the dependent variable at the boundaries is known, whereas the value of the flux at the boundaries is unknown. This situation leads to complications while solving equation (10). An important contribution in this work is the development of equation (13), which circumvents this issue. In summary, the use of equation (13) avoids the need for solving equation (4). For example, Simpson [2017] recently analyzed the critical time scales for a problem involving morphogen gradients, which requires explicit analysis of the partial differential equation governing the evolution of the gradient variable. This approach cannot be used to

solve the problems considered in the present study involving Dirichlet boundary conditions for the dependent variable. In contrast, our approach circumvents these issues.

Although all analyses presented here focus on one-dimensional Cartesian problems, the method can be extended to radial problems and multidimensional Cartesian problems. Two specific examples are presented in Texts S2 and S3. Furthermore, the applications described here involve homogeneous transport processes, where  $D'$  and  $W'$  are constants. However, our framework also applies to more general heterogeneous problems provided that  $D'(x')$  and  $W'(x')$  are sufficiently differentiable. Since the linear diffusion equation is routinely used to model several practical science and engineering problems including consolidation (geotechnical literature) and corrosion (material science literature) processes, the proposed theoretical framework has practical relevance to a variety of practical problems across many applied fields.

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