A Nonlocal Extension of Dispersion Analysis for Closures in Reactive Flows

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Abstract

Nonlocal algebraic closure models associated with both unresolved advective transport and nonlinear reaction terms in a Reynolds-averaged Navier-Stokes context are presented in this work. In particular, a system of species subject to binary reactions and transport by advection and diffusion are examined by expanding upon analysis originally developed for binary reactions in the context of Taylor dispersion of scalars. This work extends model forms from weakly-nonlinear extensions of that dispersion theory and the role of nonlocality in the presence of reactions is studied and captured by analytic algebraic expressions. These expressions can be incorporated into an eddy diffusivity matrix that explicitly capture the influence of chemical kinetics on the closure operators. Furthermore, we demonstrate that the model form derived in a laminar context directly translates to an analogous setup in homogeneous isotropic turbulence. We show that this framework improves prediction of mean quantities compared to previous results.

Keywords: Nonlocal Models; RANS Closures; Scalar Transport
1. Introduction

In considering the temporal evolution of a binary reaction system with two independent scalars with corresponding concentrations labeled as $C_1$ and $C_2$ in the presence of an imposed divergence-free velocity field, $U$, the governing equations for the scalars can be formulated as

$$\frac{\partial C_i}{\partial t} + \nabla \cdot (UC_i) = D_m \nabla^2 C_i - AC_i C_j \phi_i, \quad (1)$$

where $A$ is a reaction coefficient and $D_m$ is a scalar diffusivity that is identical for both indexed scalars. In examining the evolution of the concentrations of passive scalars in such an incompressible flow, a one-way coupling between the fluid momentum and the scalars can be assumed in writing the transport equations given by Equation (1). This assumption is physically realizable in the dilute species limit, where the effects of heat release from the reaction are so small that neither the fluid density nor the reaction kinetics are significantly affected.

While this form of the equation incorporates all effects present in the system, in practice, the Reynolds-averaged Navier-Stokes (RANS) space, is far less expensive to solve. This equation exists in the Reynolds-averaged Navier-Stokes (RANS) space, where the effects of heat release from the reaction are so small that neither the fluid density nor the reaction kinetics are significantly affected. In this study, we build off of the work of [1] to develop a model in the RANS context that can provide algebraic closures to the scalar evolution equations for a binary reactant setup. This is done in the spirit of dispersion analysis, as first demonstrated by Taylor in [2, 3] and since extended by many others, cf. [4–7]. By considering the case of a parallel flow as an analytical prototype, [1] extends Taylor’s analysis to write local model forms for the case of dispersion of scalars undergoing binary reactions, which, in particular, lead to scalar flux closures that can be written as

$$u_i^\prime C_j^\prime = -[D_{eff}] \left[ \frac{\partial C_j}{\partial x_i} \right], \quad (3)$$

where $D_{eff}$ is some generalized effective eddy diffusivity matrix and gradients of both reactants play a role. In particular, the effective eddy diffusivity involves the superposition of two scalar gradients multiplied by a prefactor dependent on both scalar fields. The equations for the scalar flux are written as

$$\begin{bmatrix} u_i^\prime C_1^\prime \\ u_i^\prime C_2^\prime \end{bmatrix} = - \begin{bmatrix} D_{11} & D_{12} \\ D_{21} & D_{22} \end{bmatrix} \frac{\partial}{\partial x} \begin{bmatrix} C_1^\prime \\ C_2^\prime \end{bmatrix}, \quad (4)$$

where $D_{kl}$ represents the diffusivity coefficient associated with the flux of the $k$-th species due to a gradient in the species. These coefficients are written explicitly as

$$\begin{align*}
D_{11} &= \frac{D^0 (1 + AC_1 \tau_{mix})}{1 + A \tau_{mix} C_1 + A \tau_{mix} C_2} \\
D_{12} &= - \frac{D^0 A \tau_{mix} C_1}{1 + A \tau_{mix} C_1 + A \tau_{mix} C_2} \\
D_{21} &= - \frac{D^0 A \tau_{mix} C_2}{1 + A \tau_{mix} C_1 + A \tau_{mix} C_2} \\
D_{22} &= \frac{D^0 (1 + AC_2 \tau_{mix})}{1 + A \tau_{mix} C_1 + A \tau_{mix} C_2} 
\end{align*} \quad (5)$$

Here, $\tau_{mix}$ denotes the characteristic mixing time associated with a particular turbulent flow. These two quantities, the mixing time and eddy diffusivity, are measured from a non-reactive flow using methods such as the macroscopic forcing method (MFM) of [6, 8].

The most novel implication of the derivation approach used to obtain this model, however, is that it...
invokes an implied model for the fluctuating fields themselves. As a result, it is possible to determine the actual local form of the fluctuations and to close not only the scalar transport term, but also the unclosed reaction terms in the standard RANS equations. This closure expression can be written as

$$\frac{A}{u_{rms}} \left( D_{11} \frac{\partial C_1}{\partial x} + D_{12} \frac{\partial C_2}{\partial x} \right) \left( D_{21} \frac{\partial C_1}{\partial x} + D_{22} \frac{\partial C_2}{\partial x} \right),$$

(6)

where $u_{rms}$ is the measured root-mean-squared velocity field of the underlying flow. As such, we can fully close the scalar transport equation for both scalars using a single framework.

The basic weakly nonlinear model offers an alternative approach to perturbation expansions based on directly linearizing the equations performed by works like [9]. It recovers the scaling relationship between diffusivity and Damköhler number derived in other numerical works and experiments, as in [7, 10–12], and the model based on [13] explicated in [14].

This provides a description of the local model that solves the reduced-degree-of-freedom system resulting from the RANS equations. The three algebraic components describe an entire model form, and require only information about the underlying flow, specifically $D^0$ and $u_{rms}$.

3. The model problem

In order to develop insights into a model form that captures nonlocal closure terms in a binary mixture, we propose the following illustrative setup that can be reasonably confident in the utility of these simplifications.

3.1. Nonlocal formulation

To derive nonlocal insight, let us examine this problem in the context of a linear reaction, as studied in works like [13]. Here, we are following the basic steps of the MFM procedure that is fully explained in [6] and complete details can be found in that reference.

For this particular setup, the concentration of one of the scalars, $C_2$, is held constant in the entire domain at unity and does not evolve, while the maximum value of $C_1$, at the left side of the domain, is held to be at least an order of magnitude less than that of $C_2$. As $C_2$ is a constant, we will denote the reaction rate as $A_L = AC_2$, denoting the product of a standard binary species reaction coefficient times the concentration of the constant species. The fluctuations for $C_1$ are now governed by a transport equation as

$$\frac{\partial C'_1}{\partial t} + \frac{\partial(u' C'_1)}{\partial x_1} + u'(y) \frac{\partial C'_1}{\partial x_2} = D_m \frac{\partial^2 C'_1}{\partial x_1^2} - A_L C'_1,$$

(8)

where $C'_1 = \overline{C_1} + C'_1$ and so on. This equation is derived by finding the full transport equation for $C_1$ and subtracting from it the evolution equation for $C_1$. Note that this equation is linear in the scalar concentration, and so therefore can be described as evolving via a linear operator acting on the scalar field. If we further regard the term on the left-hand side that deals with fluctuations of fluctuations as negligible and add fully turbulent flow with reacting scalars, this model problem captures the essential competing physics of mixing, transport, and reactions that govern mean scalar concentrations in the more realistic case. However, we can now formulate nonlocal closures with interpretable forms in this sandbox context.

In particular, past work in [1] and work on a similar problem in [6] has shown that this setup allows for the formulation of solutions that translate directly from a laminar flow topology to a turbulent context, so we can be reasonably confident in the utility of these simplifications.
an arbitrary forcing in time and the axial $x-$ direction to the equation, we can write the transport equation as

$$\frac{\partial C_1'}{\partial t} + \cos(x_2)\frac{\partial C_1'}{\partial x_1} = \frac{\partial^2 C_1'}{\partial x_2^2} + \frac{1}{Pe} \frac{\partial^2 C_1'}{\partial x_1^2} - ALC_1' + s(x_1, t),$$

(9)

where $s = \exp(\omega t + ikx_1)$ is a unit forcing term added to the right-hand side and $Pe = uL/D_m$ is a Péclet number. We can then also expand the concentration field as $C_1' = \hat{c}(\omega, k; x_2)\exp(\omega t + ikx_1)$ as suggested by the imposed velocity field to get a final governing equation of

$$\left[\omega + \frac{k^2}{Pe} + AL - \frac{\partial^2}{\partial x_2^2} + ik\cos(x_2)\right] \hat{c} = 1,$$

(10)

where $\omega$ is a temporal frequency and the bracketed terms represent a linear operator that acts on the scalar field. Note that we can now write this equation simply as $L\hat{c}(\omega, k; x_2) = 1$, where $L$ denotes the linear operator.

Following the process of MFM as described in [6], gives us the exact macroscopic operator. We note that we can also rewrite this equation more generally as

$$\left[Z - \frac{\partial^2}{\partial x_2^2} + ik\cos(x_2)\right] \hat{c} = 1,$$

(11)

where $Z$, which we call a “multi-physics coefficient,” is a complex number that incorporates the effects of...
axial diffusion, chemical reaction, and unsteadiness. If we were to apply the over-barred averaging operator to this equation, only $\overline{Z}$ and the advective term will remain, allowing us to find the operator that acts on $\overline{Z}$.

We can solve this ordinary differential equation directly for each $\omega$ and $k$ and get $\hat{c}$, the scalar magnitude, using appropriate boundary conditions. Now, after applying the averaging operator to the measured solution field, we have found the operator that acts on the full Fourier field. Thus, the macroscopic closure operator that captures purely unresolved effects in Fourier space can be computed by subtracting the mean field effects, so that we can write

$$\overline{\mathcal{T}} = 1/\overline{c} - \overline{Z}$$

(12)

where the overbar represents averaging. This operator is now a function of the real wavenumber, $k$, and $Z$, which can take complex values.

3.2. The closure model form

To showcase the real and imaginary parts of the operator, we will consider $\overline{Z}$ to take the form of a scalar multiplied by $1 + i$, and we can plot the real and imaginary components of the macroscopic operator as in Figure (2). If we consider purely the steady limit ($\omega = 0$), we can get Figure (3), which expresses the true steady macroscopic closure operator over a range of $Z$ values. In this limit, we see that the effect of increasing the reaction coefficient in Equation (9) is to suppress the projection of perturbations to the macroscopic space.

While we have calculated the values for the macroscopic operator directly, the practical utility of this process is that one can now fit analytical curves to the actual calculated MFM operator. We do this by examining the asymptotic limits of said operator at zero and high $k$ and matching those limits for intermediate wavenumbers. One analytic expression that matches the limit values of $\overline{\mathcal{T}}$ is

$$\overline{\mathcal{T}} = \left((1 + Z)^2 + k^2\right)^{1/2} - 1 - Z,$$

(13)

which is a reasonable approximation to the true operator across decades of wavenumbers. We want to consider the steady limit, and a fit that approximates the MFM data can be expressed as

$$\overline{\mathcal{T}} = \frac{k^2}{\sqrt{4(1 + Z)^2 + k^2}}$$

(14)

which allows us to consider the steady RANS problem. However, we want to modify this expression slightly in order to match previously explored model forms in the local linear limit as well as in the nonlocal but nonreacting limits.

For a linear reaction problem, [13] explicated a model form that we would like to match with our new generalized nonlocal operator. In the limit of nonreactive flow, we would also like to recover the nonreactive scalar transport model derived in [6]. To do so, we can rewrite the operator derived here for the linear reaction setup as

$$\overline{\mathcal{T}} = \frac{\hat{L}_{nr}}{1 + Z\tau_{mix}} = \frac{0.5k^2/(1 + 0.25k^2)}{1 + Z/(1 + 0.25k^2)}.$$  

(15)

where the subscript $nr$ denotes the non-reactive macroscopic operator. In the absence of reaction, the only term that survives is $\overline{\mathcal{T}}_{nr}$, which matches the nonreactive but nonlocal scalar transport model derived in [6].

This model for the eddy diffusivity is plotted in Figures (2-3) against the true, measured MFM operator. We see that the role of the multi-physics coefficient is to suppress the magnitude of the macroscopic operator, which is captured in Equation (15). In the specific steady case where $Z = A_L$ and there is no axial diffusion, we recover the model form suggested by the local work of [13] in the case of $k = 0$.

Furthermore, Equation (15) isolates a nonlocal form for the mixing time scale,

$$\tau = (1 + 0.25k^2)^{-1/2},$$

(16)

and this assumed model form is plotted in Figure 4. The colored lines represent the real measured mixing time as suggested by the middle expression in Equation (15) from a range of $Z = A_L$ values. We see the mixing time is highly scale-dependent, and is largely agnostic to the presence of a reaction. While not covered here, the mixing time is sensitive to the steadiness of the underlying flow.

From Figures (2-3), we can see that as the magnitude of reaction coefficient or the axial diffusivity
increases, the magnitude of the nonlocal behavior of the closures rolls off.

4. Turbulent Connection

We have thus far written a Fourier space operator, but we must translate it into physical space to draw conclusions about its utility. We start by noting that each \( i \) and \( \tau \) represent a spatial derivative, while each \( i \omega \) denotes a time derivative. Now we can use the notation of \( D^0 \) and \( l \), an eddy mixing length, from [8] to transform our operator derived from a laminar context into one suited for a fully turbulent flow. Such an operator for the linear reaction problem is given by

\[
D_{\text{ef}} = \frac{D}{1 + A_L \tau} = \frac{D^0}{\sqrt{1 - l^2 \frac{\partial^2}{\partial x_1^2}}} \quad (17)
\]

where \( I \) represents an identity operator and which assumes that \( Z = A_L \) only, as described in the previous section. We are concerned with the steady-state limit only here, and at the high \( Pr \) limit, where we can neglect all terms in \( Z \) except this scalar reaction coefficient.

It is important to note that while this model appears similar to a fractional step operator, it is derived analytically by considering the asymptotic limits of a governing equation solution. In addition, while this operator is structurally similar to the work of [13], it is far more general and allows for definitions for mixing time and eddy diffusivity that incorporate nonlocality.

Based on the translation of the laminar model to a turbulent context, we can write that

\[
D = \frac{D^0}{\sqrt{1 - l^2 \frac{\partial^2}{\partial x_1^2}}} \quad (18)
\]

and

\[
\tau = \frac{\tau_{\text{mix}}}{\sqrt{1 - l^2 \frac{\partial^2}{\partial x_1^2}}} \quad (19)
\]

using values for the eddy diffusivity, mixing time, and mixing length as defined and tabulated in [8].

In previous work, [1] derived model forms for closures of the binary reaction problem involving \( \tau_{\text{mix}} \) and \( D^0 \). With the observation that we can write independent expressions for eddy diffusivity and mixing time, an extension of those parameters to the binary reaction case seems straightforward by adapting the model forms of Equation (5) for the binary reaction problem. The newly stated nonlocal eddy diffusivity, \( D \), and nonlocal mixing time, \( \tau \), can be used in place of the previously used local eddy diffusivity, \( D^0 \) and the local mixing time, \( \tau_{\text{mix}} \). This means, for example, we can write the first element of the diffusivity matrix as

\[
D_{11} = \frac{D(1 + A_L \tau)}{1 + A_L \tau_0} \quad (20)
\]

Having conceived this model form, we seek to test its applicability for the binary reactant problem. In particular, we replace the steady, parallel flow examined in the previous section with three dimensional (3D) homogeneous isotropic turbulence (HIT) in an elongated domain. To generalize the model problem, the code of [13] in an incompressible mode was adapted for this work to simulate HIT in a 3D domain of size \((2\pi)^3\). The resulting flow field is periodically extended in the axial \( x \)-direction to generate a \( 20\pi \times 2\pi \times 2\pi \) computational domain for scalar transport. The scalar transport equations are solved with \( C_1 = C_{\text{ref}} \) at \( x = -L_1/2 \) and \( C_2 = C_{\text{ref}} \) at \( x = L_1/2 \). This provides a realistic reaction zone in the middle of the domain, far from the boundaries.

For our flow simulations, we adopted parameters from [8] for a case of \( Re_\lambda = 26 \), which was obtained by setting the HIT box size of \( L = 2\pi \), the turbulent forcing parameter following the prescription of [13] equal to 0.2792, and the kinematic viscosity to \( \nu = 0.0263 \).

This setup allows adoption of the values for non-reactive eddy diffusivity. For this specific case, we use values matching those in [1], which fall within error bounds reported by [8]. These values, chosen to match constant scalar flux values outside the reaction zone, are \( D^0 = 0.86u_{\text{rms}}/\epsilon = 0.96 \), \( l = 1.23u_{\text{rms}}/\epsilon = 1.41 \), and \( \tau_{\text{mix}} = 0.86u_{\text{rms}}/\epsilon = 1.02 \), where \( \epsilon = 0.79 \) is turbulent kinetic energy dissipation rate and \( u_{\text{rms}} = 0.97 \) is the single-component root-mean-squared velocity.

For scalar transport, we consider molecular diffusivity \( D_m = 0.0263 \), matching \( \nu \), \( C_{\text{ref}} = 1 \) and a reaction coefficient of \( \alpha = 100 \). This leads to \( Pr \equiv D^0/D_m = 37 \) and \( Da \equiv \alpha \tau_{\text{mix}} C_{\text{ref}} = 102 \).

4.1. A priori analysis

First, let us use a priori analysis to examine the ensemble-averaged binary reaction problem in the defined turbulent context. In Figure (5), we can see the plotted closures for the case, as well as the mean scalar concentrations. The nonlocal ROM recovers from some of the errors in capturing the true transport closures that are incurred by the local ROM model of [1]. In particular, we see that the nonlocal model performs far better than the local model for the reaction closure term.

For these specific closure terms, the standard Gradient Diffusion model appears to match the DNS data more closely than the local ROM and some regimes of the nonlocal ROM. However, an overall assessment that considers both reaction and transport closures reveals the advantage of the nonlocal ROM, as the Gradient Diffusion transport closure model offers no answers to the equally vital reaction closure question, and the local model does not accurately match the
peak of the DNS reaction closure term. Quantifying this advantage can be accomplished by performing a posteriori analysis and comparing one-dimensional model predictions against DNS data.

4.2. A posteriori analysis

In this section, having developed confidence in our nonlocal ROM, we can now solve Equation (2) directly by invoking the closure models heretofore presented. The problem setup and parameters used in this section are identical to those for the a priori analysis.

The only exception is that for the RANS equation, instead of Dirichlet boundary conditions for the scalars, Neumann boundary conditions with slopes matching the DNS profiles are used. This is done because the DNS develops axial boundary layers near the Dirichlet conditions due to local outflow advection. For the DNS, we have ensured these artificial effects do not pollute the reaction zone results by ensuring the boundaries are far from the reaction zone. These boundary layers are absent in the RANS case as the mean velocity is zero. Appropriate matching of RANS solutions to the DNS ones should consider concentration profiles outside of these artificial boundary layers. We have done so by matching the slopes of the RANS concentration profiles to those of the DNS outside of the boundary layers, but far from the reaction zones.

In Figure (6), we can see the a posteriori results. In particular, we compare the predicted mean profile of $C_1$ to the results derived from the DNS described in the previous section. As the problem exhibits mirror symmetry in the ensemble-averaged sense about the midplane, results for $C_2$ are omitted as they provide
the same insight.

It is clear that when all closures are considered, the local ROM outperforms the standard Gradient Diffusion model, while the nonlocal model outperforms the local model, especially for $X = x/L_1 \approx 0$, where the mean reaction zone, the “flame,” exists.

5. Conclusions

In this work, we have introduced a framework to extend analysis created to study the dispersion of a single passive tracer to analysis of system of species undergoing binary reactions with nonlocal closures. The nonlocality for the binary reaction problem is derived from solving the corresponding linear reaction problem exactly to extract scale-dependent measures of eddy diffusivity and mixing time. The resulting model parameters and their incorporation into the model form of \[ X \] introduces closures to both advective flux and reaction terms with nonlinear interactions between the mean state of all species, even though the primary set of equations being solved undergo a linearization procedure.

A particularly salient highlight from the \textit{a posteriori} analysis is that there is reduced scalar leakage of $C_1$ to the right of the flame-zone, where we expect it to exponentially decay to zero concentration. As this leakage often misrepresents the mean reaction zone width, it is particularly important to capture.

It is also salient to reiterate that while the capturing of nonlocality produces model parameters that appear notionally similar to a fraction power operator, they are analytically explainable as fitting the high and low wavenumber limits of diffusion.

A future avenue of further inquiry is applying the proposed ROM to a Large-Eddy Simulation context as a subgrid-scale model, where mean space variables are replaced with filtered variables and quantities like $\urms$, are evaluated at the grid scale. In addition, while this work examines the steady-state limit of the nonlocal operator, the use of a generalized multi-physics coefficient in the model derivation allows the definition of an operator capable of modeling transient behaviour.

Acknowledgements

Support for this work was provided by the National Science Foundation Graduate Research Fellowship Program under Grant No. 1656518, the Stanford Graduate Fellowships in Science and Engineering, and National Science Foundation Extreme Science and Engineering Discovery Environment resources under Grant No. CTS190057.

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