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Lagrangian analysis for turbulent transport in variable-density turbulence

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Lagrangian analysis of materially conserved scalars is applied to the problem of turbulent transport in variable density flows. The consequences of an additional material conserved quantity, the density, is generally not acknowledged and leads to significant and meaningfully different expressions for turbulent transport in the moment equations. The formal Lagrangian analysis produces gradient transport expressions substantially different from those obtained by the physically intuitive “argument by analogy” method used in computational models. Various intuitive arguments, in Favre and Reynolds averaged settings, are contrasted to the formal Lagrangian results. Using expressions from the formal analysis, we derive consistent gradient transport closures for the turbulent transport terms that appear in the first and second order Favre moment equations. Results for coupled multi-species turbulent transport are given. The analysis is limited to variable density turbulence in which the dilatation of the fluctuating velocity is small. The results are applicable to turbulent combustion and to stellar convection problems in which the density fluctuations are on the order of the mean density.

I. INTRODUCTION

Moment closures for turbulent transport invoke eddy-viscosity or linear gradient transport models. Gradient transport models are generally argued for on the basis of mixing-length ideas [16] and Lagrangian analyses by Taylor[28] and Corrsin[8] of the transport of a materially conserved passive scalar in constant density flows. In addition to the requirement of material conservation, gradient transport is valid when the characteristic scale of the turbulent transport mechanism is smaller than that associated with the inhomogeneity of the mean scalar [9]. Engineering models also apply the gradient transport model to turbulent fluctuations of velocities, scalars as well as higher moments of these quantities [10, 22].

Closures involving higher order moments are commonly encountered in variable density turbulence and turbulent combustion [26, 27]. Unclosed moments in these regimes are often expressed in Favre-averaged variables. In an argument by analogy, gradient transport models developed for constant density turbulence are used for transport models of Favre-fluctuating quantities. This strategy is applied not just for k-epsilon models but also for second moment closures in which several additional equations are carried. All such gradient transport models are based on an *argument by analogy* with the constant density modeling and are inconsistent with a formal Lagrangian analysis. There are several problematic issues with developing turbulent transport models in Favre variables based on argument by analogy to the Reynolds variables in constant density flows.

There are several problematic issues with developing turbulent transport models in Favre variables based on argument by analogy to the Reynolds variables. The two most salient issues are

1. The fluctuating velocity in Reynolds averaged coordinates, u'_i has a zero mean. The analogous fluctuating velocity in Favre averaged variables, u''_i does not have a zero mean. It is not a centered variable and argument by analogy for quantities not based on centered variables are specious.
2. In variable density turbulence¹, the density is an additional material conserved variable and the consequences of this additional constraint on turbulent transport is substantial and is not accounted for in any transport models in Favre treatments.

To address these shortcomings not accounted for in the Favre argument by analogy, we work only with centered variables in a formal Lagrangian analysis and account for the additional constraint required by a materially conserved density.

To close higher moments such as $\overline{\rho'c'u'_k}$, we generalize the Lagrangian arguments to use simultaneous material conservation of the density ρ and the conserved scalar ρc . A formal Lagrangian analysis using this method produces some very interesting and unexpected formal results. For example, the Lagrangian gradient transport model for the

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¹ We limit our work to the case of negligible fluctuating dilatation.

third moment in the turbulent flux is bilinear in the gradients of mean density and mean mass fraction. Our general principle result is that the turbulent fluxes of second moments have the form

$$\langle \rho' c' v_k \rangle = \langle \xi_i \xi_j v_k \rangle \bar{\rho}_{,i} \bar{C}_{,j} \quad (1)$$

where ξ_i, v_i are Lagrangian objects. In comparison, the conventional model for the second moment flux [11] results in an expression that is proportional to the down gradient of the second moment $\overline{\rho' c'}$. Simultaneous dependence of the turbulent flux on density and scalar gradients is consistent with the density-fluctuation-correlation in variable-density turbulent jets and wakes [5].

The formal Lagrangian results not only apply to variable-density turbulent transport but to all third and higher moments. An example is the turbulent flux of the Reynolds stress $\overline{u'_i u'_j u'_k}$ (or its trace $\overline{u'_i u'_i u'_k}$) in constant-density turbulence. While several improved closures for third and higher moments have been proposed in the literature [3, 15, 29, 32], the present work is novel and mathematically formal application of Lagrangian analysis to extend gradient transport to turbulent flux of second and higher order moments. The formal results are applicable to the class of physical problems where the assumptions of fine-grained turbulence and fluctuations in the presence of mean gradients hold, as is typical of gradient-transport and mixing-length modeling hypotheses.

Within the context of conserved scalars, we find that formal gradient transport can result in counter-gradient fluxes even with isotropic eddy viscosity models for the Lagrangian transport coefficient. This is not the case for the gradient transport by analogy for Favre fluxes. Counter-gradient fluxes are observed in variable-density mixing and combustion [13, 14, 23]. In such flow regimes, the scalar is, in the general case, linked to the density field and therefore becomes an active scalar. In the simplest case this issue is made clear by the relation between mass fraction and density used in the variable-density turbulence analyses of isothermal and isobaric binary mixing [6, 19].

$$\frac{1}{\rho} = v = \frac{c}{\rho_1} + \frac{1-c}{\rho_2}, \quad \rho = \frac{\rho_2}{1+rc}, \quad r = \frac{\rho_2}{\rho_1} - 1 > 0 \quad (2)$$

$$\overline{\rho c' u'_k} = -\frac{1+r\bar{C}}{r} \overline{\rho' u'_k} \quad (3)$$

More generally, for multispecies/multimaterial mixing under such conditions, the species mass fluctuations are coupled to the mass flux via mass conservation and equation-of-state constraints,

$$\sum_{\alpha} (\rho c_{\alpha})' = \rho', \quad \sum_{\alpha} \frac{(\rho c_{\alpha})'}{\rho_{\alpha}} = 0 \quad (4)$$

where c_{α} denotes the mass fraction of the specie α and ρ_{α} is the corresponding specie microdensity [20].

In the next few sections, the constant density Lagrangian analysis (see also [21]) is reviewed as a foundation that we extend to multiple materially conserved objects. The gradient transport expressions are derived using formal Lagrangian analysis and compared to the heuristic arguments used in models. We then move on to the variable density case. Our results are presented in the context of a first order Favre moment k-epsilon type closure as well second order Favre moment closures. Our general style of presentation is to contrast the formal Lagrangian results with expressions based on *arguments of analogy* approach.

II. A REVIEW OF LAGRANGIAN GRADIENT TRANSPORT IN CONSTANT-DENSITY TURBULENCE

The Lagrangian gradient transport is a widely-invoked hypothesis in turbulent transport. It is also commonly referred to as the mixing-length and sweeping decorrelation hypothesis [7, 30]. We review constant-density Lagrangian gradient transport in this section to introduce nomenclature and basic relations.

Gradient transport hypothesis is based on material conservation in a Lagrangian trajectory. Let c be a materially conserved scalar. Then

$$\frac{D}{Dt}(c) = 0 \implies c(t_0; a_i) = c(t_0 + t; a_i) \quad (5)$$

where a_i is the initial position of a Lagrangian particle, and the particle is displaced by ξ_i from a_i in the time period t .

For simplicity, let us choose our co-ordinate system so that $t_0 = 0$. We can decompose the variable c in terms of its Eulerian mean \bar{C} and fluctuation c' as

$$c'(t; a_i) + \bar{C}(t; a_i) = c'(0; a_i) + \bar{C}(0; a_i) \quad (6)$$

For clarity, we drop the information in the parentheses that specifies the Lagrangian time co-ordinate and use the subscript 0 to imply the quantities at the beginning of the trajectories $t = 0$. Material conservation of the scalar during the Lagrangian trajectory is then expressed as

$$c' + \bar{C} = c'_0 + \bar{C}_0 \quad (7)$$

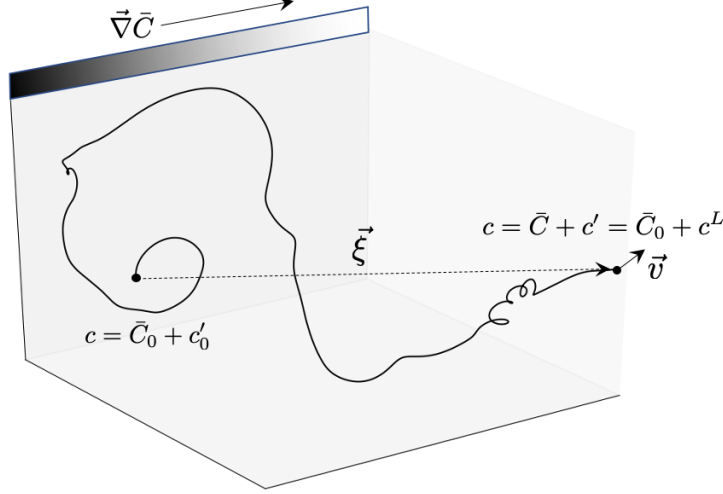


FIG. 1. Illustration of a Lagrangian trajectory in stationary homogeneous isotropic turbulence: the Lagrangian displacement vector ξ_i , Lagrangian velocity v_i and the Lagrangian fluctuation c^L (introduced in the next section) are labelled.

Eulerian mean scalar fields \bar{C} and \bar{C}_0 are independent of time and only a function of the spatial co-ordinate. In the presence of mean scalar gradient $\bar{C}_{,j}$, the mean scalar field can be expressed as a linear function of the Lagrangian particle displacement vector

$$\bar{C} - \bar{C}_0 = \bar{C}(a_i + \xi_j) - \bar{C}(a_i) = \xi_j \bar{C}_{,j} \quad (8)$$

With Eq. 8, we can express $\bar{C} - \bar{C}_0$ in Eq. 7 in terms of the Lagrangian particle displacement vector ξ_i such that

$$\begin{aligned} c' + \bar{C}_0 + \xi_j \bar{C}_{,j} &= c'_0 + \bar{C}_0 \\ c' - c'_0 &= -\xi_j \bar{C}_{,j} \end{aligned} \quad (9)$$

The displacement vector ξ_j in homogeneous turbulence is a stochastic variable. To obtain a single point closure for the statistical average of turbulent scalar flux in homogeneous turbulence, we take the moment of Eq. 9 with the Lagrangian velocity at the end of the Lagrangian trajectory $v_k(t; a_i) = u_k(\xi_i, t)$. We then have

$$\langle c' v_k \rangle - \langle c'_0 v_k \rangle = -\langle \xi_j v_k \rangle \bar{C}_{,j} \quad (10)$$

The angled bracket operator $\langle \cdot \rangle$ represents averaging over an ensemble of Lagrangian trajectories in the turbulent flow-field such that $c(x, t) = c(t; a_i)$. The term $\langle v_k c_0 \rangle = \langle v_k(t) c(0) \rangle$ represents the correlation of the Eulerian fluctuation in the scalar value at the beginning of the Lagrangian trajectory with the velocity fluctuation at the end of the Lagrangian trajectory. For a passive scalar, the objects at initial and final points of the trajectory decorrelate over the time-scale of particle transport and we have

$$\langle c_0 v_k \rangle = 0 \quad (11)$$

Eq. 11 results in the well-known gradient transport equation for the passive scalar

$$\langle c' u'_k \rangle = \overline{c' u'_k} = \langle c' v_k \rangle = -\langle \xi_j v_k \rangle \bar{C}_{,j} \quad (12)$$

The object $\langle \xi_j v_k \rangle$ is the Lagrangian particle displacement flux and is the unclosed object. The single-point turbulent scalar flux has now been re-expressed in terms of a temporal Lagrangian cross-correlation, which can be obtained from

theoretical and/or data-driven models of Lagrangian dynamics. The Lagrangian particle displacement flux can be expressed in terms of the cross-correlation using the definition of the displacement vector in homogeneous turbulence.

$$\frac{d\xi_i}{dt} = v_i, \quad \xi_i(t) = \int_0^t v_i(t') dt' \quad (13)$$

$$\langle v_k \xi_j \rangle = \langle v_k(t) \int_0^t v_j(t') dt' \rangle = \int_0^t \langle v_k(t) v_j(t') \rangle dt' \quad (14)$$

Here the trajectory ensemble average commutes with the integral over the trajectory. The cross-correlation can be normalized in the form

$$\langle \xi_j v_k + \xi_k v_j \rangle = \int_0^t \langle v_j(t') v_k(t) + v_j(t) v_k(t') \rangle dt' \quad (15)$$

$$= \sigma_v^2 \int_0^t \mathcal{R}_{v_j, v_k}(\tau) d\tau = \sigma_v^2 \mathcal{T}_{jk} \quad (16)$$

Here $\sigma_v = 2k = \langle v_k v_k \rangle = \langle u'_k u'_k \rangle$ is the velocity variance or twice the turbulent specific kinetic energy, $\mathcal{R}_{v_j, v_k}(\tau)$ is the normalized two-time Lagrangian correlation function and \mathcal{T}_{jk} is the resulting Lagrangian time-scale tensor. Therefore, gradient transport for a passive scalar can be expressed as

$$\overline{c' u'_k} = -k \mathcal{T}_{jk} \overline{C}_{,j} \quad (17)$$

70 For homogeneous shear flow, additional moments are involved [18]. We now look into how Eq 17 is modeled for moment closures and subgrid-scale closures in numerical computations.

A. Constant density moment closure: the long time limit

75 Moment closures capture mean-gradient turbulent transport over length-scales much larger than the transport process i.e. over regions with $\langle v_i(t; a_i) \rangle = 0$ and the Lagrangian time-scales are large so that the $t \rightarrow \infty$ limit for the cross-correlation be employed. The Lagrangian time-scale tensor is therefore, a constant and depends on the state of turbulence.

In engineering models, the Lagrangian time-scale tensor is modeled as an isotropic tensor

$$\mathcal{T}_{jk} \approx T^L \delta_{jk} \quad (18)$$

The Lagrangian time-scale, in Eq 18 is often modeled using an Eulerian time-scale $C_\mu k / \epsilon$ resulting in the well-known gradient transport expression

$$\overline{c' u'_k} = -C_\mu \frac{k^2}{\epsilon} \overline{C}_{,k} \quad (19)$$

The approximation of the Lagrangian time-scale using an Eulerian time-scale is valid in turbulent flows with a single time-scale. In turbulent flows with competing time-scales, due to different physical mechanisms, the Eulerian time-scale may be a poor approximation.

An anisotropic engineering approximation for the time-scale tensor may also be employed. One such approximation uses the Reynolds stress anisotropy as a proxy for the time-scale tensor anisotropy.

$$\mathcal{T}_{jk} \approx T^L \left(\frac{\overline{u'_j u'_k}}{2k} \right) \quad (20)$$

B. Constant density subgrid-closures: the small-time limit

In the context of large-eddy simulations, the time-scale of the turbulent processes being modeled is imposed by observer's resolution and is typically constrained by the numerical grid resolution Δ . When gradient transport is modeled over small transport length-scales due to high resolution, the small-time or the ballistic dispersion limit of the integral in Eq. 15 can also become relevant. In such a case, the upper limit of the integral over the Lagrangian

trajectory, t is small and as $t = t_\Delta \rightarrow 0$, the two-time correlation tensor $\langle v_j(t') v_k(t) \rangle$ can be approximated with $\langle v_j(t=0) v_k(t=0) \rangle \approx \overline{u'_j u'_k}$. Eq. 15 then results in

$$\langle v_k \xi_j + v_j \xi_k \rangle = \int_0^{t_\Delta} \langle v_k(t_\Delta) v_j(t') + v_k(t') v_j(t_\Delta) \rangle dt' \quad (21)$$

$$\approx \langle v_j v_k \rangle \int_0^{t_\Delta} dt' = \overline{u'_j u'_k} t_\Delta \quad (22)$$

An important point to be noted from this limit is that gradient transport at length-scales much larger than the turbulent transport length-scale $S_T = T^L / \sigma_v$ can assume a constant Lagrangian particle displacement flux tensor. At small length-scales however, when $\Delta \ll S_T$, the displacement flux tensor scales with $\Delta = t_\Delta / \sigma_v$.

The small-time limit of gradient transport has also been previously received attention in the context of modeling near-field transport of scalar atmospheric constituents near and within vegetation canopies which serve as spatially-distributed scalar sources [17].

III. LAGRANGIAN GRADIENT TRANSPORT FOR SECOND MOMENT TURBULENT FLUX IN CONSTANT DENSITY TURBULENCE

Gradient transport for variable-density turbulence requires closures of Favre fluxes. Due to the density weighted nature of the fluxes, third moments arise as unclosed terms. These third moments are turbulent fluxes of second moments. For example, the third moment $\overline{\rho' c' u'_k}$ appears in the transport of conserved scalar $\overline{\rho \tilde{C}} = \overline{\rho} \overline{C} + \overline{\rho' c'}$ and is the turbulent flux of $\overline{\rho' c'}$.

The primary objective of this section concerns with derivation of the formal Lagrangian gradient transport hypothesis for the turbulent flux of the second moment. In this section, we consider second moments in constant-density turbulence and use the results in this section as a pedagogical tool before its application to closures in variable-density turbulence.

We consider two scalar species, c_α and c_β . All three variables, c_α , c_β and $c_\alpha c_\beta$ are materially conserved in the non-diffusive limit.

$$\frac{Dc_\alpha}{Dt} = 0, \quad \frac{Dc_\beta}{Dt} = 0, \quad \frac{D}{Dt}(c_\alpha c_\beta) = 0 \quad (23)$$

The object $\overline{c'_\alpha c'_\beta}$ represents the covariance between two species c_α and c_β in multispecies/multimaterial transport. The transport equation for the second moment $\overline{c'_\alpha c'_\beta}$ is

$$\overline{c'_\alpha c'_\beta, t} + (\overline{c'_\alpha c'_\beta} \overline{U}_k + \overline{c'_\alpha c'_\beta u'_k},_k + \overline{c'_\alpha u'_k} \overline{C}_{\beta, k} + \overline{c'_\beta u'_k} \overline{C}_{\alpha, k} = \varepsilon_{\alpha\beta} \quad (24)$$

The unclosed term of focus is the second moment turbulent flux $\overline{c'_\alpha c'_\beta u'_k}$. The key idea is to derive the formal gradient transport expression for $\overline{c'_\alpha c'_\beta u'_k}$ and apply it to second moment fluxes involving density fluctuations. By substituting c_α with ρ and c_β with a scalar c , we can use the result derived for $\overline{c'_\alpha c'_\beta u'_k}$ to obtain the corresponding formal gradient transport for objects of the form $\overline{\rho' c' u'_k}$. The result derived in this section will also serve as the template for closure of several second and third moment turbulent fluxes in the variable-density turbulence section.

A. Gradient transport by analogy for second moment turbulent flux in constant-density turbulence

We first review the gradient transport by analogy for $\overline{c'_\alpha c'_\beta u'_k}$ and discuss why this commonly employed gradient transport expression is not formally correct. The gradient transport expression for second moment turbulent flux $\overline{c'_\alpha c'_\beta u'_k}$ using analogy to the passive scalar flux in Eq. 12 is [11, 12]

$$\overline{c'_\alpha c'_\beta u'_k} = - \langle \xi_j v_k \rangle \overline{(c'_\alpha c'_\beta),_j} \approx - C_\mu \frac{k}{\epsilon} \overline{u'_j u'_k} \overline{(c'_\alpha c'_\beta),_j} \quad (25)$$

From a Lagrangian perspective, this model approximates an empirical gradient transport of the form

$$\overline{c'_\alpha c'_\beta u'_k} = - \langle \xi_j v_k \rangle \overline{(c'_\alpha c'_\beta),_j} \quad (26)$$

Eq. 26 is empirical because an argument is employed that $(c'_\alpha c'_\beta)' = c'_\alpha c'_\beta - \overline{c'_\alpha c'_\beta}$ is a materially conserved scalar. Gradient transport for the passive scalar (Eq. 12) is then conveniently employed. However, $(c'_\alpha c'_\beta)'$ is not materially conserved as its Lagrangian derivative is not zero in the presence of mean species gradients.

$$\frac{D}{Dt}(c'_\alpha c'_\beta)' = -(c'_\alpha u'_k)' \overline{C}_{\beta,k} - (c'_\beta u'_k)' \overline{C}_{\alpha,k} \quad (27)$$

In the Appendix, we discuss more about the limitations of the assumptions that lead to Eq. 26 from both perspectives, Eulerian (Sec VI G) and Lagrangian (Sec VI D). We will now derive formal gradient transport expression for $c'_\alpha c'_\beta u'_k$ using Lagrangian principles discussed in the previous section.

B. Lagrangian gradient transport for binary passive scalar fluctuation in constant-density turbulence

In this subsection, we derive formal gradient transport expression for the turbulent flux of the second moment $c'_\alpha c'_\beta u'_k$ using Lagrangian analysis. As we mention before, the conserved scalars in this setup are c_α , c_β , $c_\alpha c_\beta$.

$$\frac{Dc_\alpha}{Dt} = 0, \quad \frac{Dc_\beta}{Dt} = 0, \quad \frac{D}{Dt}(c_\alpha c_\beta) = 0 \quad (28)$$

We introduce the notation of the Lagrangian fluctuations to avoid cumbersome expressions. The Lagrangian fluctuation is defined as the difference between a quantity at a time t in the Lagrangian trajectory and its Eulerian mean at the initial point in the trajectory. Therefore, Lagrangian fluctuations can be defined for both, total quantities and as well as their Eulerian means. For example, for a scalar c , the Lagrangian fluctuation of the total quantity is denoted by c^L and the Lagrangian fluctuation of the Eulerian mean is denoted by \overline{C}^L

$$c^L(t; a_i) = c(t; a_i) - \overline{C}(0; a_i), \quad (29)$$

$$\overline{C}^L(t; a_i) = \overline{C}(t; a_i) - \overline{C}(0; a_i) \quad (30)$$

The passive scalar gradient transport equation Eq. 12 can be rewritten using Lagrangian fluctuations as

$$\langle c^L v_k \rangle = \langle c'_0 v_k \rangle = 0 \quad (31)$$

In other words, the flux of the Lagrangian fluctuation of a passive scalar is zero. The zero flux results from material conservation of the scalar and a decorrelation of the Lagrangian velocity from the initial point Eulerian fluctuation of the scalar. Expanding $c^L = c' + \overline{C}^L$ and using $\overline{C}^L = \xi_j \overline{C}_{,j}$ for mean gradient homogeneous turbulence,

$$\langle c' v_k \rangle = -\langle \overline{C}^L v_k \rangle = -\langle \xi_j v_k \rangle \overline{C}_{,j} \quad (32)$$

For binary fluctuations, using $c_\alpha^L = c'_{\alpha 0}$ and $c_\beta^L = c'_{\beta 0}$, we can conveniently write

$$c_\alpha^L c_\beta^L = c'_{\alpha 0} c'_{\beta 0} \quad (33)$$

A formal proof of Eq. 33 starting from material conservation of the object $c_\alpha c_\beta$ is presented in the Appendix. Additionally, we also show there that $(c_\alpha c_\beta)^L = (c_\alpha c_\beta)'_0$ is equivalent to Eq. 33. Next, expanding the Lagrangian fluctuations in Eq. 33 as $c^L = c' + \overline{C}^L$, we have

$$c'_\alpha c'_\beta + c'_\alpha \overline{C}_\beta^L + c'_\beta \overline{C}_\alpha^L + \overline{C}_\alpha^L \overline{C}_\beta^L = c'_{\alpha 0} c'_{\beta 0} \quad (34)$$

Eq. 34 can be re-expressed using $c'_\alpha = c'_{\alpha 0} - \overline{C}_\alpha^L$ and $c'_\beta = c'_{\beta 0} - \overline{C}_\beta^L$ to obtain

$$c'_\alpha c'_\beta - \overline{C}_\alpha^L \overline{C}_\beta^L = c'_{\alpha 0} c'_{\beta 0} - c'_{\alpha 0} \overline{C}_\beta^L - \overline{C}_\alpha^L c'_{\beta 0} \quad (35)$$

In the case of constant mean species gradients, a linear spatial variation of \overline{C}_α and \overline{C}_β exists and we express the Lagrangian fluctuations of Eulerian means using Lagrangian particle displacement vector as $\overline{C}_\alpha^L = \xi_i \overline{C}_{\alpha,i}$ and $\overline{C}_\beta^L = \xi_j \overline{C}_{\beta,j}$ to write

$$c'_\alpha c'_\beta = \xi_i \xi_j \overline{C}_{\alpha,i} \overline{C}_{\beta,j} + c'_{\alpha 0} c'_{\beta 0} - c'_{\alpha 0} \xi_i \overline{C}_{\beta,i} - c'_{\beta 0} \xi_j \overline{C}_{\alpha,j} \quad (36)$$

$$\langle c'_\alpha c'_\beta v_k \rangle = \langle \xi_i \xi_j v_k \rangle \overline{C}_{\alpha,i} \overline{C}_{\beta,j} \quad (37)$$

The moments of final-point Lagrangian objects with initial point scalar Eulerian fluctuations vanish. As a consequence, $\langle c'_{\alpha 0} c'_{\beta 0} v_k \rangle = 0$, $\langle c'_{\alpha 0} \xi_i v_k \rangle = 0$ and $\langle c'_{\beta 0} \xi_j v_k \rangle = 0$. Eq. 37 is the primary result of the paper, and has been formally derived using material conservation of c_α, c_β along Lagrangian trajectories. Eq. 37 will later be used to propose formal gradient transport expressions for all second moment turbulent fluxes encountered in variable-density turbulence.

The result in Eq. 37 is interesting in that gradient transport for second moment turbulent flux is proportional to the product of mean gradients of the two objects in the second moment. Compare this to the gradient transport by analogy to the passive scalar, which is, as intuition suggests, proportional to the down gradient of the second moment. Inclusion of mean gradient terms in second moment turbulent fluxes have also been proposed earlier in the literature to improve the conventional gradient transport expressions [31]. However formal gradient transport naturally brings mean gradients into the equation, not as an additional ‘proposed tensor basis’. Numerical experiments suggest that gradient of first order moments can improve modeling accuracy (Appendix VII). The bilinear form of the mean gradient and the higher order Lagrangian moments, are both new objects that have not been studied previously. Future investigations into their properties are critical to close turbulent fluxes of high order Eulerian moments with gradient transport.

C. Comparison of formal and gradient transport by analogy for second moment turbulent flux

Formal Lagrangian analysis results in expression for second moment turbulent flux that is bilinear in mean gradients. This expression is different from the expression by analogy (Harlow-Hirt-Daly [11, 12]) proposed by applying an analogy between the passive scalar and the second moment. Eqs. 38-39 provides a comparison between these two expressions.

$$\text{Analogy (Eq. 26): } \langle c'_\alpha c'_\beta u'_k \rangle = -\langle \xi_j v_k \rangle (\overline{c'_\alpha c'_\beta})_{,j} \quad (38)$$

$$\text{Formal Analysis (Eq. 37): } \langle c'_\alpha c'_\beta u'_k \rangle = \langle \xi_i \xi_j v_k \rangle \overline{C}_{\alpha,i} \overline{C}_{\beta,j} \quad (39)$$

In the second-moment modeling, second moment turbulent fluxes of type $\overline{u'_i u'_j u'_k}$, $\overline{\phi' u'_j u'_k}$ and $\overline{\phi' \phi' u'_k}$ require closure. Here ϕ is a scalar. Using the sweeping hypothesis for u_i , we can compare the two gradient transport expressions for these three classes of moments.

$$\text{Analogy: } \overline{u'_i u'_j u'_k} = -\langle \xi_p v_k \rangle (\overline{u'_i u'_j})_{,p} - \langle \xi_p v_i \rangle (\overline{u'_j u'_k})_{,p} - \langle \xi_p v_j \rangle (\overline{u'_k u'_i})_{,p} \quad (40)$$

$$\text{Formal Analysis: } \overline{u'_i u'_j u'_k} = \langle \xi_p \xi_q v_k \rangle \overline{U}_{i,p} \overline{U}_{j,q} + \langle \xi_p \xi_q v_i \rangle \overline{U}_{j,p} \overline{U}_{k,q} + \langle \xi_p \xi_q v_j \rangle \overline{U}_{k,p} \overline{U}_{i,q} \quad (41)$$

$$\text{Analogy: } \overline{\phi' u'_j u'_k} = -\langle \xi_p v_k \rangle (\overline{\phi' u'_j})_{,p} - \langle \xi_p v_j \rangle (\overline{\phi' u'_k})_{,p} \quad (42)$$

$$\text{Formal Analysis: } \overline{\phi' u'_j u'_k} = \langle \xi_p \xi_q v_k \rangle \overline{\Phi}_{,p} \overline{U}_{j,q} + \langle \xi_p \xi_q v_j \rangle \overline{\Phi}_{,p} \overline{U}_{k,q} \quad (43)$$

$$\text{Analogy: } \overline{\phi' \phi' u'_k} = -\langle \xi_p v_k \rangle (\overline{\phi' \phi'})_{,p} \quad (44)$$

$$\text{Formal Analysis: } \overline{\phi' \phi' u'_k} = \langle \xi_p \xi_q v_k \rangle (\overline{\Phi}_{,p} \overline{\Phi}_{,q}) \quad (45)$$

The third moment $\overline{\phi' \phi' u'_k}$ provides an useful perspective on the difference in the two expressions. The expression by analogy is proportional to gradient in the scalar variance, while the formal expression is proportional to flux of the scalar variance itself, indirectly, via the product of the mean gradients.

We can also assess the formal gradient transport from the point of view of second moment transport. Substituting Eq. 38 into the third moment in Eq. 24, GT by analogy results in

$$\overline{c'_\alpha c'_\beta},_t + (\overline{c'_\alpha c'_\beta} \overline{U}_k)_{,k} = -\overline{c'_\alpha u'_k} \overline{C}_{\beta,k} - \overline{c'_\beta u'_k} \overline{C}_{\alpha,k} + \varepsilon_{\alpha\beta} + (\langle \xi_p v_k \rangle \overline{c'_\alpha c'_\beta},_p)_{,k} \quad (46)$$

while substitution with the formal result in Eq. 37 leads to

$$\overline{c'_\alpha c'_\beta},_t + (\overline{c'_\alpha c'_\beta} \overline{U}_k)_{,k} = -\overline{c'_\alpha u'_k} \overline{C}_{\beta,k} - \overline{c'_\beta u'_k} \overline{C}_{\alpha,k} + \varepsilon_{\alpha\beta} - (\langle \xi_p \xi_q v_k \rangle \overline{C}_{\alpha,p} \overline{C}_{\beta,q})_{,k} \quad (47)$$

With the expression by analogy, the third moment accounts for turbulent diffusion in the second moment equation. Whereas, the non-isotropic approximation for $\langle \xi_i \xi_j v_k \rangle$ in the formal expression yields terms proportional to mean gradients. The role of the third moment then becomes reminiscent of a production/destruction term. In particular, the ‘energy’ associated with $\overline{c'_\alpha c'_\beta}$ with formal gradient transport is affected by the term

$$\text{Analogy: } (\overline{c'_\alpha c'_\beta})^2_{,t} + \dots = -2 \langle \xi_i v_k \rangle (\overline{c'_\alpha c'_\beta})_{,i} (\overline{c'_\alpha c'_\beta})_{,k} \quad (48)$$

$$\text{Formal: } (\overline{c'_\alpha c'_\beta})^2_{,t} + \dots = 2 \langle \xi_i \xi_j v_k \rangle \overline{C}_{\alpha,i} \overline{C}_{\beta,j} \overline{c'_\alpha c'_\beta},_k \quad (49)$$

The energetics associated with third moment is no longer straightforward and depends on product of four tensors. The modeling choice for the object $\langle \xi_i \xi_j v_k \rangle$ will affect the computational stability of the second moment equation. Therefore, future work will investigate the Lagrangian third moment $\langle \xi_i \xi_j v_k \rangle$ to develop stable and accurate models for second moment turbulent fluxes.

Assumptions for the gradient transport expression in Eq. 37: We reiterate and list the basic assumptions on the flow state using which the formally exact expression in Eq. 37 is derived.

- **High Reynolds number turbulence:** The correlation length-scale of the Lagrangian fluid particle is small so that molecular diffusion for the transported quantity can be neglected.
- **Mean gradient as fluctuation production mechanism:** A linearly varying mean field is assumed along the particle path and so the materially conserved scalar analysis assumes the existence of a constant mean gradient. This, on the level of principle precludes Eulerian fluctuations in flows with no mean gradients.
- **Fine grained turbulence:** For Lagrangian statistical analysis to hold, the length-scale of the transport mechanism must be small compare to the mean-gradient length-scale. This is also required for the fluctuations to become decorrelated between the initial and final points in the trajectory.

For third moments of the type in Eqs. 40 and 42, the flows must contain mean velocity gradients for turbulence production. This is a consequence of the sweeping hypothesis applied to fluid velocity, which is inconsistent with Lagrangian gradient transport assumptions in the absence of a mean velocity gradient. This limits the use of formal Lagrangian gradient transport to flows where velocity fluctuations are associated with mean velocity gradient. When used in moment-closure modeling of buoyancy-driven or decaying turbulent layers, these principles are not valid. However, in large-eddy simulations and scale-resolving simulations, formal gradient-transport expressions remain valid and useful since turbulence producing coherent structures provide non-zero mean velocity gradients.

D. Third order Lagrangian correlation

The Lagrangian third moment in Eq. 37 contains a quadratic term in Lagrangian particle displacement vector. This term is further expressed in terms of a bicorrelation tensor as

$$\langle v_i \xi_j \xi_k \rangle + \langle v_j \xi_k \xi_i \rangle + \langle v_k \xi_i \xi_j \rangle = \int_0^t \int_0^t \langle v_i(t) v_j(t'_1) v_k(t'_2) \rangle + \langle v_j(t) v_k(t'_1) v_i(t'_2) \rangle + \langle v_k(t) v_i(t'_1) v_j(t'_2) \rangle dt'_1 dt'_2 \quad (50)$$

$$= \mathcal{S}^v (\sigma^v)^3 \int_0^t \int_0^t \mathcal{B}_{v_i v_j v_k}(\tau_1, \tau_2) d\tau_1 d\tau_2 \quad (51)$$

The bicorrelation tensor \mathcal{B}_{ijk} is defined as

$$\mathcal{B}_{ijk}(\tau_1, \tau_2) = \frac{\langle v_i(t) v_j(t + \tau_1) v_k(t + \tau_2) \rangle + \langle v_j(t) v_k(t + \tau_1) v_i(t + \tau_2) \rangle + \langle v_k(t) v_i(t + \tau_1) v_j(t + \tau_2) \rangle}{\mathcal{S}^v (\sigma^v)^3} \quad (52)$$

and \mathcal{S}^v is the skewness of the Lagrangian velocity field, defined as

$$\mathcal{S}^v = \frac{\langle v_i^3 \rangle}{(\sigma^v)^{3/2}} \quad (53)$$

Approximations to the Lagrangian correlation object may be devised using first moment gradients, as well as second moments such as Reynolds stress and turbulent mass flux. A data-driven approach can then be used to identify the optimal bases. In the present work, we do not focus on the closure of the Lagrangian moments.

IV. LAGRANGIAN GRADIENT TRANSPORT FOR VARIABLE-DENSITY TURBULENCE

Variable-density turbulence is a turbulent transport regime in which the inertial effects due to density differences in the fluid affect the state of turbulence. In the incompressible limit, the density differences can relate to differences in material composition, temperature and/or phase [24]. Simulations of variable-density turbulence employ conserved variable sets which results in Favre solution variables. The convective terms of these Favre solution variables involve unclosed third moments, as we discussed in the previous section.

The conserved scalar equation and its Favre averaged form is

$$(\rho c)_{,t} + (\rho c u_k)_{,k} = 0 \quad (54)$$

$$(\bar{\rho} \tilde{C})_{,t} + \left(\bar{\rho} \tilde{C} \tilde{U}_k + \bar{\rho} \widetilde{c'' u_k''} \right)_{,k} = 0 \quad (55)$$

The molecular diffusion terms are not included as we focus on the turbulent transport arising from the convective term. The third moment $\overline{\rho' c' u_k'}$ appears in the Favre turbulent flux as a consequence of transport of $\overline{\rho' c'}$, which is a part of the Favre solution variable $\bar{\rho} \tilde{C} = \bar{\rho} \bar{C} + \overline{\rho' c'}$. The Favre turbulent flux $\bar{\rho} \widetilde{c'' u_k''}$ is

$$\bar{\rho} \widetilde{c'' u_k''} = \bar{\rho} \overline{c' u_k'} + \overline{\rho' c' u_k'} - \bar{\rho} \overline{c''} \overline{u_k''} \quad (56)$$

Modeling within the Favre framework is common because Favre variables re-express the turbulent fluxes as a second Favre moment. An implicit assumption in modeling the second Favre moment is that the physics can be represented solely on the basis of mass-weighted variables. The closures are then borrowed from constant-density turbulence and applied using arguments by analogy.

A. Gradient transport by analogy for Favre turbulent fluxes

Engineering models almost exclusively employ gradient transport by analogy for Favre fluxes. However, the analogy for Favre fluxes is not consistent with formal Lagrangian analysis. In the Appendix (Sec VI A), we discuss why gradient transport cannot provide closed expressions for moments involving Favre variables.

The analogy to passive-scalar gradient transport (Eq. 12) in Favre variables is

$$\text{Passive scalar : } \overline{c' u_k'} \equiv \langle c' v_k \rangle \bar{C}_{,j} \quad (57)$$

$$\text{Favre analogy : } \bar{\rho} \widetilde{c'' u_k''} \equiv \langle \rho c'' v_k'' \rangle \tilde{C}_{,j} \quad (58)$$

where Favre-fluctuating Lagrangian velocity is defined as

$$v_k'' = v_k + \frac{\overline{\rho v_k}}{\bar{\rho}} \quad (59)$$

The analogy is obtained by using a Favre decomposition for the scalar c instead of the Reynolds decomposition. Then, similar to constant-density passive scalar Lagrangian gradient transport (Eq. 12), the moment with c'' is replaced with a moment with $-\xi_j \tilde{C}_{,j}$.

Eqs. 60-64 provide gradient transport expressions obtained by applying the analogy for second and third order Favre moments. The unclosed second Favre moments on the LHS appear in the governing equations for conserved scalar $\bar{\rho} \tilde{C}$, momentum $\bar{\rho} \tilde{U}_i$, total energy $\bar{E} = \bar{\rho} (\tilde{\mathcal{E}} + \widetilde{u_k u_k}/2)$. The third Favre moments appear in the total energy, Favre stress and Favre scalar variance $\bar{\rho} \widetilde{c'' c''}$ equations. The governing equations for the relevant Favre first and second order moments are provided in the Appendix (Sec VI F).

$$\bar{\rho} \widetilde{c'' u_k''} = -\langle \rho \xi_p v_k'' \rangle \tilde{C}_{,p} \quad (60)$$

$$\bar{\rho} \widetilde{u_i'' u_k''} = -\langle \rho \xi_p v_k'' \rangle \tilde{U}_{i,p} - \langle \rho \xi_i v_k'' \rangle \tilde{U}_{j,p} \quad (61)$$

$$\bar{\rho} \widetilde{e'' u_k''} = -\langle \rho \xi_p v_k'' \rangle \tilde{\mathcal{E}}_{,p} \quad (62)$$

$$\bar{\rho} \widetilde{u_i'' u_j'' u_k''} = -\langle \rho \xi_p v_k'' \rangle \tilde{R}_{ij,p} - \langle \rho \xi_p v_i'' \rangle \tilde{R}_{jk,p} - \langle \rho \xi_p v_j'' \rangle \tilde{R}_{ki,p} \quad (63)$$

$$\bar{\rho} \widetilde{c'' c'' u_k''} = -\langle \rho \xi_p v_k'' \rangle \widetilde{c'' c''}_{,p} \quad (64)$$

Here, $e = \bar{\mathcal{E}} + e'$ denotes the specific internal energy. For the third order Favre moment of the form $\widetilde{c''_\alpha c''_\beta u_k''}$, gradient transport by analogy is

$$\bar{\rho} \widetilde{c''_\alpha c''_\beta u_k''} = \langle \rho c''_\alpha c''_\beta v_k'' \rangle = -\langle \rho \xi_j v_k'' \rangle (\widetilde{c''_\alpha c''_\beta})_{,j} \quad (65)$$

The object $\langle \rho \xi_j v_k'' \rangle$ in Eq. 58 is often approximated as an isotropic object using Favre kinetic energy $2\bar{\rho} \tilde{k} = \overline{\rho u_k'' u_k''}$ as $\langle \rho \xi_p v_k'' \rangle \approx \bar{\rho} \tilde{k} \frac{\tilde{k}}{\epsilon}$

$$\bar{\rho} \widetilde{c'' u_k''} = -\langle \rho \xi_p v_k'' \rangle \tilde{C}_{,p} \approx -\bar{\rho} C_\mu \frac{\tilde{k}^2}{\epsilon} \tilde{C}_{,k} \quad (66)$$

In anisotropic closures, the object $\langle \rho \xi_j v_k'' \rangle$ is approximated using the Favre stress $\bar{\rho} \tilde{R}_{ij} = \overline{\rho u_i'' u_j''}$ and the Eulerian time-scale \tilde{k}/ϵ .

$$\bar{\rho} \widetilde{c'' u_k''} = -\langle \rho \xi_p v_k'' \rangle \tilde{C}_{,p} \approx -\bar{\rho} C_\mu \frac{\tilde{k}}{\epsilon} \tilde{R}_{kp} \tilde{C}_{,p} \quad (67)$$

B. Gradient transport analogy for Reynolds turbulent fluxes

The gradient transport closures may also be applied to the Favre turbulent fluxes in their expanded Reynolds form, in which moments with density are explicitly accounted for. Different second and third Favre moments in their Reynolds form are expanded exactly as

$$\bar{\rho} \widetilde{c'' u_k''} = \bar{\rho} \overline{c' u_k'} + \overline{\rho' c' u_k'} + \bar{\rho} a_i c'' \quad (68)$$

$$\bar{\rho} \widetilde{u_i'' u_k''} = \bar{\rho} \overline{u_i' u_k'} + \overline{\rho' u_i' u_k'} - \bar{\rho} a_i a_k \quad (69)$$

$$\bar{\rho} \widetilde{u_i'' u_j'' u_k''} = \overline{\rho' u_i' u_j' u_k'} + \bar{\rho} (\overline{u_i' u_j' u_k'} - (\tilde{R}_{ij} a_k + \tilde{R}_{jk} a_i + \tilde{R}_{ki} a_j + a_i a_j a_k)) \quad (70)$$

$$\bar{\rho} \widetilde{c'' c'' u_k''} = \overline{\rho' c' c' u_k'} + \bar{\rho} (\overline{c' c' u_k'} - \overline{c'' c''} a_k + 2 \overline{c'' c'' u_k''} - \overline{c'' c''} a_k) \quad (71)$$

Consider the second Favre moment $\bar{\rho} \widetilde{c'' u_k''}$ in the conserved scalar equation (Eq. 55). It comprises of three second moments and one third order moment (Eq. 56). The scalar flux $\overline{c' u_k'}$ is closed using gradient transport (Eq. 12) in the general case. The density-species covariance $\overline{\rho' c'}$, can be transported or estimated using a transported variance. The third moment is closed using Eq. 26, which we have earlier referred to as *gradient transport by analogy for second moment turbulent flux*. Note that the analogy is made in this case by applying the sweeping hypothesis to the zero-mean binary fluctuation $\rho' c' - \overline{\rho' c'}$.

$$\overline{\rho' c' u_k'} = -\langle \xi_p v_k \rangle \overline{(\rho' c')},_p \quad (72)$$

Second moment turbulent fluxes appear in transport equations for first Favre moments, conserved scalar $\bar{\rho} \tilde{C}$ and momentum $\bar{\rho} \tilde{U}_i$ as well as second moments such as the turbulent mass-flux $\overline{\rho' u_i'}$, the density-specific-volume covariance $\overline{\rho' v'}$ and the scalar variance $\overline{c' c'}$. When the sweeping hypothesis is applied to the zero-mean binary fluctuations $\rho' u_i' - \overline{\rho' u_i'}$, $\rho' v' - \overline{\rho' v'}$ and $c' c' - \overline{c' c'}$, Eq. 26 results in

$$\overline{\rho' u_i' u_k'} = -\langle \xi_p v_k \rangle \overline{(\rho' u_i')},_p - \langle \xi_p v_i \rangle \overline{(\rho' u_k')},_p \quad (73)$$

$$\overline{\rho' v' u_k'} = -\langle \xi_p v_k \rangle \overline{(\rho' v')},_p \quad (74)$$

$$\overline{c' c' u_k'} = -\langle \xi_p v_k \rangle \overline{(c' c')},_p \quad (75)$$

Similarly, third moment turbulent fluxes (which are fourth order moments) can also be closed using gradient transport by analogy. These fourth order moments appear in transport equations of second Favre moments. For example, $\overline{\rho' u_i' u_j' u_k'}$ appears in Favre stress $\tilde{R}_{ij} = \overline{u_i'' u_j''}$ equation and the fourth moment $\overline{\rho' c' c' u_k'}$ appears in Favre scalar variance $\widetilde{c'' c''}$ equation. The corresponding transport equations are provided in the Appendix (Sec VIF). The gradient transport expressions for these two fourth moments are obtained by applying the analogy to the zero-mean ternary fluctuations $\rho' u_i' u_j' - \overline{\rho' u_i' u_j'}$ and $\rho' c' c' - \overline{\rho' c' c'}$ respectively.

$$\overline{\rho' u_i' u_j' u_k'} = -\langle \xi_p v_k \rangle \overline{(\rho' u_i' u_j')},_p - \langle \xi_p v_i \rangle \overline{(\rho' u_j' u_k')},_p - \langle \xi_p v_j \rangle \overline{(\rho' u_i' u_k')},_p \quad (76)$$

$$\overline{\rho' c' c' u_k'} = -\langle \xi_p v_k \rangle \overline{(\rho' c' c')},_p \quad (77)$$

¹⁹⁰ We reiterate that the sweeping hypothesis for zero-mean binary (and ternary) fluctuations is not valid formally.

C. Formal closures from Lagrangian gradient transport

In contrast to gradient transport expressions using analogy, we present closures for third and fourth moments based on the formal Lagrangian gradient transport analysis that leads to Eq. 37. Formal Lagrangian gradient transport for

the turbulent flux of the third moment is derived in the Appendix, and follows a procedure similar to that used for the second moment turbulent flux in Eq. 37.

Using Eqs. 37 and 121, we have the following closures for third and fourth Reynolds moments. The third moments $\overline{\rho' c' u'_k}$ and $\overline{\rho' u'_i u'_k}$ appear in the transport equations for first Favre moments, conserved scalar $\bar{\rho} \tilde{C}$ and momentum $\bar{\rho} \tilde{U}_i$ respectively.

$$\overline{\rho' c' u'_k} = \langle \xi_p \xi_q v_k \rangle \bar{\rho}_{,p} \bar{C}_{,q} \quad (78)$$

$$\overline{\rho' u'_i u'_k} = \langle \xi_p \xi_q v_k \rangle \bar{\rho}_{,p} \bar{U}_{i,q} + \langle \xi_p \xi_q v_i \rangle \bar{\rho}_{,p} \bar{U}_{k,q} \quad (79)$$

The third moments $\overline{c' u'_k}$ and $\overline{\rho' v' u'_k}$ appear in the transport equations for density-specific-volume covariance and Favre scalar variance respectively.

$$\overline{c' u'_k} = \langle \xi_p \xi_q v_k \rangle \bar{C}_{,p} \bar{C}_{,q} \quad (80)$$

$$\overline{\rho' v' u'_k} = \langle \xi_p \xi_q v_k \rangle \bar{\rho}_{,p} \bar{v}_{,q} \quad (81)$$

The fourth moments $\overline{\rho' u'_i u'_j u'_k}$ and $\overline{\rho' c' c' u'_k}$ appear in the Favre stress $\bar{\rho} \tilde{R}_{ij} = \bar{\rho} \widetilde{u''_i u''_j}$ and Favre scalar variance $\bar{\rho} \widetilde{c'' c''}$ equations respectively.

$$\overline{\rho' u'_i u'_j u'_k} = \langle \xi_p \xi_q \xi_r v_k \rangle \bar{\rho}_{,p} \bar{U}_{i,q} \bar{U}_{j,r} + \langle \xi_p \xi_q \xi_r v_i \rangle \bar{\rho}_{,p} \bar{U}_{j,q} \bar{U}_{k,r} + \langle \xi_p \xi_q \xi_r v_j \rangle \bar{\rho}_{,p} \bar{U}_{k,q} \bar{U}_{i,r} \quad (82)$$

$$\overline{\rho' c' c' u'_k} = \langle \xi_p \xi_q \xi_r v_k \rangle \bar{\rho}_{,p} \bar{C}_{,q} \bar{C}_{,r} \quad (83)$$

In Eqs. 78-83, the sweeping hypothesis has been applied to the scalars ρ , v , c and the turbulent velocity vector u_i . These results are valid for flows with negligible dilatation at the time-scales relevant to turbulent transport.

The turbulent fluxes in variable-density flows, however, unlike constant-density passive scalar fluxes experience dynamic effects in the presence of pressure-gradient driven acceleration [25]. As a consequence, Lagrangian gradient transport for moments involving density and associated ‘active’ scalars involve unclosed terms when dynamic effects are important. We discuss this in Appendix Sec. VI H.

D. Comparison of different gradient transport expressions for Favre fluxes

The gradient transport expressions in Eqs. 78-83 for the second/third moment turbulent fluxes can be used to derive formal Lagrangian transport expression for Favre turbulent fluxes.

Eqs. 84-86 show different gradient transport expressions for the Favre conserved scalar flux.

Favre conserved scalar flux $\bar{\rho} \widetilde{c'' u''_k}$ in $\bar{\rho} \tilde{C}$ transport

$$\text{Analogy(F)} : \bar{\rho} \widetilde{c'' u''_k} = -\langle \rho \xi_p v''_k \rangle \tilde{C}_{,p} \quad (84)$$

$$\text{Analogy(R)} : \bar{\rho} \widetilde{c'' u''_k} = -\langle \xi_p v_k \rangle \bar{\rho} \tilde{C}_{,p} + \bar{c''} (\langle \xi_p v_k \rangle \bar{\rho}_{,p} + \bar{\rho} a_k) \quad (85)$$

$$\text{Formal} : \bar{\rho} \widetilde{c'' u''_k} = -\langle \xi_p v_k \rangle \bar{\rho} \tilde{C}_{,p} + \bar{c''} (\langle \xi_p v_k \rangle \bar{\rho}_{,p} + \bar{\rho} a_k) + (\langle \xi_p v_k \rangle \overline{\rho' c'})_{,p} + \langle \xi_p \xi_q v_k \rangle \bar{\rho}_{,p} \bar{C}_{,q} \quad (86)$$

In ‘Analogy(F)’ equation, we have used gradient transport by analogy for Favre fluxes, Eq. 60. In ‘Analogy(R)’ and ‘Formal’ equations, we have applied gradient transport to Reynolds fluxes. The difference between ‘Analogy(R)’ and ‘Formal’ is the choice of closure for the second moment turbulent flux $\overline{\rho' c' u'_k}$. In ‘Analogy(R)’, the flux is closed using analogy (Eq. 26), while in ‘Formal’, we use formal Lagrangian analysis derived in Eq. 37.

There is an important difference in applying gradient transport to Favre vs Reynolds fluxes. The Lagrangian particle displacement flux tensor in the Favre analogy $\langle \rho \xi_j v''_k \rangle$, involves Lagrangian fluctuating velocity and is a dynamical variable due to density. We have shown in Appendix that the analogy using Favre fluctuating velocity is not valid (Sec VI A). Formal gradient transport, instead, involves transport coefficients only in Reynolds (centered) fluctuations such as $\langle \xi_j v_k \rangle$ and $\langle \xi_i \xi_j v_k \rangle$. Physically, this is a consequence of the fact that Lagrangian analysis is a kinematic description of transport.

D1. Beyond down-gradient conserved scalar flux

We now discuss the additional physics captured in gradient transport expressions in Eq. 85 and Eq. 86, in comparison to the popular Favre analogy (Eq. 84) to constant-density gradient transport. In Eq. 86, there are two new terms in addition to the Favre mean scalar gradient. The first term which results from applying gradient transport to centered fluctuations, or Reynolds moments, is $\bar{\rho}a_k + \langle \xi_p v_k \rangle \bar{\rho}_{,p}$. Note that gradient transport for mass flux without assumptions on initial-final point Lagrangian moments implies $\bar{\rho}a_k + \langle \xi_p v_k \rangle \bar{\rho}_{,p} = \langle \rho'_0 v_k \rangle$. Therefore, the term $\bar{\rho}a_k + \langle \xi_p v_k \rangle \bar{\rho}_{,p}$ incorporates the non gradient transport physics associated with the mass flux. Gradient transport by analogy for Favre moments does not contain this term, and therefore implicitly asserts gradient transport closure for the turbulent mass flux. This assertion is inaccurate during the mass-flux production/destruction phase in buoyancy-driven or decaying variable-density turbulence. Numerical simulations of buoyancy-driven turbulent mixing layers indicate that this term can be significant for large ($\mathcal{O}(10)$) density contrasts (see Appendix VII).

The second term, that appears as a consequence of invoking formal Lagrangian gradient transport for the third moment, is $(\langle \xi_p v_k \rangle (\bar{\rho}'c'),_p + \langle \xi_p \xi_q v_k \rangle \bar{\rho}_{,p} \bar{C}_{,q})$. This term is the difference between the formal and the empirical gradient transport expressions for the third moment $\overline{\rho'c'u'_k}$. The term may also be recast in terms of Lagrangian fluctuations as $\langle (\bar{\rho}^L \bar{C}^L + \bar{\rho}'c'^L)v_k \rangle \approx \langle (\rho'c' + \bar{\rho}'c'^L)v_k \rangle$. If the binary fluctuation $\rho'c'$ was a materially conserved quantity, this term would reduce to zero. However, this is not the case in the presence of mean scalar and density gradients as can be inferred from Eq. 27.

D2. Gradient transport expressions for Favre fluxes of momentum, stress and scalar variance

Next, we write the three gradient transport expressions (two analogies and the formal expression) for Favre stress and third Favre moments, in a manner similar to how Eqs 84-86 are presented for the Favre conserved scalar flux. For the Favre analogy, we also show the modeling approximation employed in second moment closures for the Lagrangian particle displacement flux tensor. For the ternary fluctuation flux, formal gradient transport derived in Eq. 121 is employed. The Favre stress appears in transport of Favre velocity. The stress flux appears in transport of Favre kinetic energy and Favre stress, while the scalar variance flux appears in the transport of Favre scalar variance. The transport equations are provided in the Appendix (Sec VIF).

Favre stress $\bar{\rho} \widetilde{u''_i u''_k}$ in $\bar{\rho} \widetilde{U}_i$ transport equation

$$\begin{aligned} \text{Analogy(FW): } \bar{\rho} \widetilde{u''_i u''_k} &= -\langle \rho \xi_p v''_i \rangle \widetilde{U}_{k,p} - \langle \rho \xi_p v''_k \rangle \widetilde{U}_{i,p} \\ &\approx -C_\mu \bar{\rho} \frac{\tilde{k}}{\epsilon} (\widetilde{R}_{ip} \widetilde{U}_{k,p} + \widetilde{R}_{kp} \widetilde{U}_{i,p}) \end{aligned} \quad (87)$$

$$\begin{aligned} \text{Analogy(RU): } \bar{\rho} \widetilde{u''_i u''_k} &= -\langle \xi_p v_k \rangle \bar{\rho} \widetilde{U}_{i,p} - a_i (\langle \xi_p v_k \rangle \bar{\rho}_{,p} + \bar{\rho} a_k / 2) \\ &\quad - \langle \xi_p v_i \rangle \bar{\rho} \widetilde{U}_{k,p} - a_k (\langle \xi_p v_i \rangle \bar{\rho}_{,p} + \bar{\rho} a_i / 2) \end{aligned} \quad (88)$$

$$\begin{aligned} \text{Formal: } \bar{\rho} \widetilde{u''_i u''_k} &= -\langle \xi_p v_k \rangle \bar{\rho} \widetilde{U}_{i,p} - \bar{\rho} (a_i a_k / 2 - \langle \xi_p v_k \rangle a_{i,p}) + \langle \xi_p \xi_q v_k \rangle \bar{\rho}_{,p} \bar{U}_{i,q} \\ &\quad - \langle \xi_p v_i \rangle \bar{\rho} \widetilde{U}_{k,p} - \bar{\rho} (a_i a_k / 2 - \langle \xi_p v_i \rangle a_{k,p}) + \langle \xi_p \xi_q v_i \rangle \bar{\rho}_{,p} \bar{U}_{k,q} \end{aligned} \quad (89)$$

Favre stress flux $\bar{\rho} \widetilde{u_i'' u_j'' u_k''}$ in $\bar{\rho} \widetilde{R_{ij}}$ transport equation

$$\begin{aligned} \text{Analogy(F): } \bar{\rho} \widetilde{u_i'' u_j'' u_k''} &= -\langle \rho \xi_p v_k'' \rangle \widetilde{R_{ij,p}} - \langle \rho \xi_p v_i'' \rangle \widetilde{R_{jk,p}} - \langle \rho \xi_p v_j'' \rangle \widetilde{R_{ki,p}} \\ &\approx -C_\mu \bar{\rho} \frac{\tilde{k}}{\epsilon} (\widetilde{R_{kp}} \widetilde{R_{ij,p}} + \widetilde{R_{ip}} \widetilde{R_{jk,p}} + \widetilde{R_{jp}} \widetilde{R_{ki,p}}) \end{aligned} \quad (90)$$

$$\begin{aligned} \text{Analogy(R): } \bar{\rho} \widetilde{u_i'' u_j'' u_k''} &= -(\langle \xi_p v_k \rangle \bar{\rho} \widetilde{R_{ij,p}} + \langle \xi_p v_i \rangle \bar{\rho} \widetilde{R_{jk,p}} + \langle \xi_p v_j \rangle \bar{\rho} \widetilde{R_{ki,p}}) \\ &\quad + \widetilde{R_{ij}} \langle \xi_p v_k \rangle \bar{\rho}_{,p} + \widetilde{R_{jk}} \langle \xi_p v_i \rangle \bar{\rho}_{,p} + \widetilde{R_{ki}} \langle \xi_p v_j \rangle \bar{\rho}_{,p} \\ &\quad - \widetilde{R_{ij}} (\bar{\rho} a_k + \langle \xi_p v_k \rangle \bar{\rho}_{,p}) - \widetilde{R_{jk}} (\bar{\rho} a_i + \langle \xi_p v_k \rangle \bar{\rho}_{,p}) - \widetilde{R_{ki}} (\bar{\rho} a_j + \langle \xi_p v_k \rangle \bar{\rho}_{,p}) \\ &\quad - \langle \xi_p v_k \rangle (\bar{\rho} a_i a_j)_{,p} - \langle \xi_p v_i \rangle (\bar{\rho} a_j a_k)_{,p} - \langle \xi_p v_j \rangle (\bar{\rho} a_k a_i)_{,p} - \bar{\rho} a_i a_j a_k \end{aligned} \quad (91)$$

$$\begin{aligned} \text{Formal: } \bar{\rho} \widetilde{u_i'' u_j'' u_k''} &= \bar{\rho} \langle \xi_p \xi_q v_k \rangle \overline{U_{i,p} U_{j,q}} + \bar{\rho} \langle \xi_p \xi_q v_i \rangle \overline{U_{j,p} U_{k,q}} + \bar{\rho} \langle \xi_p \xi_q v_j \rangle \overline{U_{k,p} U_{i,q}} \\ &\quad - \langle \xi_p \xi_q \xi_r v_k \rangle \bar{\rho}_{,p} \overline{U_{i,q} U_{j,r}} - \langle \xi_p \xi_q \xi_r v_i \rangle \bar{\rho}_{,p} \overline{U_{j,q} U_{k,r}} - \langle \xi_p \xi_q \xi_r v_j \rangle \bar{\rho}_{,p} \overline{U_{k,q} U_{i,r}} \\ &\quad - \bar{\rho} \widetilde{R_{ij}} a_k - \bar{\rho} \widetilde{R_{jk}} a_i - \bar{\rho} \widetilde{R_{ki}} a_j - \bar{\rho} a_i a_j a_k \end{aligned} \quad (92)$$

Favre scalar variance flux $\bar{\rho} \widetilde{c'' c'' u_k''}$ in $\bar{\rho} \widetilde{c'' c''}$ transport equation

$$\begin{aligned} \text{Analogy(F): } \bar{\rho} \widetilde{c'' c'' u_k''} &= -\langle \rho \xi_p v_k'' \rangle \widetilde{c'' c''}_{,p} \\ &= -C_\mu \bar{\rho} \frac{\tilde{k}}{\epsilon} \widetilde{R_{kp}} (\widetilde{c'' c''})_{,p} \end{aligned} \quad (93)$$

$$\begin{aligned} \text{Analogy(R): } \bar{\rho} \widetilde{c'' c'' u_k''} &= -\bar{\rho} \langle \xi_p v_k \rangle \widetilde{c'' c''}_{,p} + \overline{c' c'} \langle \xi_p v_k \rangle \bar{\rho}_{,p} \\ &\quad - \widetilde{c'' c''} (\bar{\rho} a_k + \langle \xi_p v_k \rangle \bar{\rho}_{,p}) + 2\bar{\rho} \overline{c' c'} \widetilde{c'' u_k''} - \bar{\rho} \overline{c' c'} \overline{c'' a_k} \end{aligned} \quad (94)$$

$$\begin{aligned} \text{Formal: } \bar{\rho} \widetilde{c'' c'' u_k''} &= \bar{\rho} \langle \xi_p \xi_q v_k \rangle \overline{C_{,p} C_{,q}} - \langle \xi_p \xi_q \xi_r v_k \rangle \bar{\rho}_{,p} \overline{C_{,q} C_{,r}} \\ &\quad - \bar{\rho} \widetilde{c'' c''} a_k + 2 \bar{\rho} \overline{c' c'} \widetilde{c'' u_k''} - \bar{\rho} \overline{c' c'} \overline{c'' a_k} \end{aligned} \quad (95)$$

The results in Eqs. 84-95 provide a useful perspective on the complexity of objects that formal gradient transport produces. Compared to Favre analogy, the formal expressions include the explicit terms with turbulent mass flux and higher-order Lagrangian objects contracted with the mean density gradients. The formal expression therefore provides insight into how the mean density gradients can produce counter-gradient diffusion in the turbulent flux, as observed in the turbulent flux of kinetic energy in recent variable-density jet experiments [4].

V. CONCLUSIONS AND SUMMARY

Historically the formal Lagrangian analysis of a materially conserved scalar has been used to model the scalar turbulent transport in constant density turbulence. That result is then applied to the turbulent transport of other quantities despite the fact that these other quantities are not materially conserved. In variable density turbulence treated in Favre variables, most gradient transport models for the Favre turbulent fluxes use an *argument by analogy* to produce expressions that are mathematical analogues to the constant density passive scalar. One can expect that it would be unlikely that constant-density Lagrangian analysis of a passive scalar would carry over to a conserved scalar in the variable-density case. Argument by analogy to the constant density case is not formally valid because it does not account for the additionally conserved scalar, density, and does not work with centered variables. This raises the unexplored question of “What does a formal Lagrangian analysis for turbulent transport in variable density turbulence produce?” This paper gives the mathematical details and conclusions of such a formal analysis. Computational models using these formal results requires a deeper investigation of various moments of the Lagrangian particle displacement vector and is the subject of a succeeding paper.

We can expect that the variable density case will lead to substantial differences based on several ideas all of which we have explored:

- In an analysis for materially conserved species in variable density turbulence with negligible dilatation, there is an additional materially conserved quantity, the density. The appearance of density conservation constraint

plays a substantial role in the expression of turbulent fluxes. The most important consequence of an additional conservation principle in a formal Lagrangian analysis is that the turbulent flux is 1) no longer a down gradient diffusion and 2) involves a bilinear term in means species gradient and mean density gradient and 3) the turbulent transport depends on the relative orientation of the mean density and mean species gradients.

$$\overline{\rho'c'u'_k} = \langle \xi_i \xi_j v_k \rangle \bar{\rho}_{,i} \bar{C}_{,j} \quad (96)$$

- In the gradient transport by analogy for Favre turbulent fluxes, u''_i is erroneously assumed to play the same role as u'_i , a fluctuation object with a zero mean. However, the turbulent mass flux $\bar{\rho} a_k = -\bar{\rho} \overline{u''_k} \neq 0$ is an independent dynamical variable. Gradient transport in the *argument by analogy* for the Favre flux asserts gradient transport for the turbulent mass flux which ignores the active scalar effects experienced by the variable-density fluid in the presence of external and pressure-gradient driven acceleration. Numerical simulations of buoyancy-driven mixing layers show that these effects are sizeable for large density differences and must be modeled. Excluding active scalar effects is also inconsistent with second moment models where the mass flux is explicitly transported.
- The formal Lagrangian gradient transport model of Eq. 96 has important implications for modeling of turbulent fluxes in the second moment transport equations. In gradient transport by analogy, these third moments are modeled as diffusion of the second moments. The formal Lagrangian gradient transport expression, however, is not strictly a gradient diffusion of second order moments. Consequently, it can represent transfer of ‘energy into and out of the second moments.
- In contradistinction to the models arrived at in the *argument by analogy* method, the formal Lagrangian analysis for the turbulent fluxes in Favre variables has three distinct physics components. The first relates to dynamics of the turbulent mass flux and results from explicit accounting of moments with density fluctuations. The second is the dynamics associated the binary (and ternary) fluctuations, embodied in the third (and higher) moments involving the Lagrangian particle displacement vector (eg. $\langle \xi_i \xi_j v_k \rangle$). The third important physical distinction is that the Lagrangian particle displacement objects are kinematic in the formal expression and do not involve the density. This is unlike the expression by analogy, which involves density velocity product in the particle displacement moment and is thus related to particle momentum, a dynamical quantity that does not appear in a formal Lagrangian analysis.

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VI. APPENDIX

A. Favre decomposition in Lagrangian gradient transport

We analyze Lagrangian gradient transport in Favre averages and fluctuations. We demonstrate why moments with Favre fluctuations cannot result in closed form gradient transport expressions. We consider the Favre decomposition form of Eq. 9

$$c'' - c'_0 = -\tilde{C}^L \quad (97)$$

which is equivalent to $c' - c'_0 = -\overline{C}^L$. Moments of Eq. 97 with v_k, v''_k and $\rho v''_k$ are evaluated below. Here, $v''_k = v_k + \overline{u''_k}$. Moment with $\rho v''_k$ results in the Favre turbulent scalar flux $\langle \rho c'' v'' \rangle = \overline{\rho c'' u''_k}$.

$$\text{Moment with } v_k : \quad \langle c'' v_k \rangle = -\langle \tilde{C}^L v_k \rangle + \langle v_k c''_0 \rangle = -\langle \tilde{C}^L v_k \rangle + \langle v_k \overline{c''_0} \rangle \quad (98)$$

Using two-point mean-fluctuation decorrelation, we can assume $\langle v_k \overline{c''_0} \rangle = 0$. If a linear variation of the Favre mean scalar exists, we have $\langle c'' v_k \rangle = -\langle \xi_j v_k \rangle \tilde{C}_{,j}$.

Moment with v_k'' :
$$\langle c'' v_k'' \rangle = -\langle \tilde{C}^L v_k'' \rangle + \langle c_0'' v_k'' \rangle \quad (99)$$

We write $v_k'' = v_k''^L + \overline{u_{k0}''}$, where $v_k''^L = v_k + \overline{u_k''^L}$ and $c_0'' = c_0' + \overline{c_0''}$. Moments of final-point Lagrangian objects (denoted with the superscript L) with initial-point Eulerian objects vanish. So, we have

$$\langle c'' v_k'' \rangle = \langle (c_0' + \overline{c_0''})(v_k''^L + \overline{u_{k0}''}) \rangle = \overline{c_0''} \overline{u_{k0}''} \quad (100)$$

Similarly, we can expand $\langle \tilde{C}^L v_k'' \rangle$ as $\langle \tilde{C}^L v_k \rangle + \langle \tilde{C}^L \overline{u_k''^L} \rangle$ to obtain

$$\langle c'' v_k'' \rangle = -\langle \tilde{C}^L v_k \rangle - \langle \tilde{C}^L \overline{u_k''^L} \rangle + \overline{c_0''} \overline{u_{k0}''} \quad (101)$$

Therefore, due to the non-zero initial-point means, Lagrangian analysis cannot close the gradient transport of $\langle c'' v_k'' \rangle$.

Moment with $\rho v_k''$:
$$\langle \rho c'' v_k'' \rangle = -\langle \rho \tilde{C}^L v_k'' \rangle + \langle \rho c_0'' v_k'' \rangle \quad (102)$$

Similarly, moments with $\rho v_k''$ are unclosed. Writing $\rho = \rho^L + \overline{\rho_0}$, we have

$$\langle \rho c_0'' v_k'' \rangle = \langle (\rho^L + \overline{\rho_0})(c_0' + \overline{c_0''})(v_k''^L + \overline{u_{k0}''}) \rangle \quad (103)$$

We can write $\rho^L = \rho_0'$ as it is a materially conserved scalar. Since moments of Lagrangian fluctuations with initial-point Eulerian objects vanish, the non-zero terms in the RHS of Eq 103 are $\overline{\rho_0} \overline{c_0''} \overline{u_{k0}''}$ and $\overline{\rho_0'} \overline{c_0'} \overline{u_{k0}''}$. Next, the term $\langle \rho \tilde{C}^L v_k'' \rangle$ is expanded as

$$\langle \rho \tilde{C}^L v_k'' \rangle = \langle (\rho^L + \overline{\rho_0}) \tilde{C}^L (v_k''^L + \overline{u_{k0}''}) \rangle = \langle (\rho_0' + \overline{\rho_0}) \tilde{C}^L (v_k''^L + \overline{u_{k0}''}) \rangle = \overline{\rho_0} (\langle \tilde{C}^L v_k \rangle + \langle \tilde{C}^L \overline{u_k''^L} \rangle) \quad (104)$$

resulting in

$$\langle \rho c'' v_k'' \rangle = -\overline{\rho_0} (\langle \tilde{C}^L v_k \rangle + \langle \tilde{C}^L \overline{u_k''^L} \rangle) + \overline{\rho_0} \overline{c_0''} \overline{u_{k0}''} + \overline{\rho_0'} \overline{c_0'} \overline{u_{k0}''} \quad (105)$$

Therefore, analogy of the passive-scalar gradient transport in Favre variables is not valid. For reference, the analogy invoked in Favre variables can be expressed in the form $\langle \rho c'' v_k'' \rangle = -\langle \rho \rangle \langle \tilde{C}^L v_k \rangle$

B. The Lagrangian fluctuation of the conserved binary scalar

In Eq. 33, we write $c_\alpha^L c_\beta^L = c_{\alpha 0}' c_{\beta 0}'$ using Eq. 31. Here, we prove the identity $c_\alpha^L c_\beta^L = c_{\alpha 0}' c_{\beta 0}'$ (Eq. 33) independently starting from material conservation of $c_\alpha c_\beta$ along its Lagrangian trajectory.

$$c_\alpha c_\beta = (c_\alpha c_\beta)_0 \quad (106)$$

We carry out a Lagrangian decomposition of the LHS and an Eulerian decomposition of the RHS in Eq. 106. A key step is to express the mean-fluctuation decomposition for the product in a form where the expansion only consists of means, total quantities and the binary fluctuation. For the Eulerian decomposition of the conserved scalar at the initial point in the trajectory, we have

$$c_{\alpha 0} c_{\beta 0} = c_{\alpha 0}' c_{\beta 0}' + \overline{C_{\alpha 0}} c_{\beta 0} + c_{\alpha 0} \overline{C_{\beta 0}} - \overline{C_{\alpha 0}} \overline{C_{\beta 0}} \quad (107)$$

Similarly, the Lagrangian decomposition at the end of the trajectory $c = c^L + \overline{C_0}$ results in

$$c_\alpha c_\beta = c_\alpha^L c_\beta^L + \overline{C_{\alpha 0}} c_\beta + c_\alpha \overline{C_{\beta 0}} - \overline{C_{\alpha 0}} \overline{C_{\beta 0}} \quad (108)$$

Equating the right hand sides in Eqs. 107 and 108 as $c = c_0$, we obtain the second moment turbulent flux counterpart to the scalar flux in Eq. 31. Alternatively, we can also show that

$$(c_\alpha c_\beta)^L = (c_\alpha c_\beta)_0' \equiv c_\alpha^L c_\beta^L = c_{\alpha 0}' c_{\beta 0}' \quad (109)$$

using $(\overline{C_\alpha} \overline{C_\beta})^L = \overline{C_\alpha^L} \overline{C_{\beta 0}} + \overline{C_{\alpha 0}} \overline{C_\beta^L} + \overline{C_\alpha^L} \overline{C_\beta^L}$.

C. Mean field dependence on Lagrangian particle displacement vector in variable-density flows

An important step in gradient transport closures is the dependence of mean fields on the Lagrangian particle displacement vector. The Lagrangian moments appearing in gradient transport stem from this functional dependence. *Linear \bar{C} and $\bar{\rho}$ fields:* With the passive scalar assumptions, formal Lagrangian analysis yields

$$\langle \rho' c' v_k \rangle = \langle \bar{\rho}^L \bar{C}^L v_k \rangle \quad (110)$$

With linear variation of the form $\bar{\rho}^L = \bar{\rho}_{,j} \xi_j$ and $\bar{C}^L = \bar{C}_{,j} \xi_j$, we have a quadratic dependence of $\bar{\rho}^L \bar{C}^L$ on the displacement vector.

Linear $\bar{\rho}\bar{C}$ and $\bar{\rho}$ fields: With a linear variation of the form $\bar{\rho}\bar{C}^L = \bar{\rho}\bar{C}_{,j} \xi_j$ and $\bar{\rho}^L = \bar{\rho}_{,j} \xi_j$, we do not have a linear variation of \bar{C}^L . Instead, \bar{C}^L is expanded on dependent variables as

$$\bar{C}^L = \frac{\bar{\rho}\bar{C}^L - \bar{\rho}'c'^L - \bar{\rho}^L\bar{C}_0}{\bar{\rho}_0 + \bar{\rho}^L} \quad (111)$$

We must make an additional assumption about the variation of $\bar{\rho}'c'$. If a linear variation for $\bar{\rho}'c'$ is assumed, we have

$$\bar{C}^L = \xi_p \frac{\bar{\rho}\bar{C}_{,p} - \bar{\rho}'c'_{,p} - \bar{\rho}_{,p}\bar{C}_0}{\bar{\rho}_0 + \xi_q \bar{\rho}_{,q}} \quad (112)$$

295 which shows that that \bar{C}^L and consequently $\bar{\rho}^L \bar{C}^L$ cannot result in closed form gradient transport expressions.

Linear \tilde{C} and $\bar{\rho}$ fields: In the case of linear Favre mean field \tilde{C} , \bar{C}^L can simply be expressed as $\bar{C}^L = \tilde{C}^L + \bar{c}''^L$. The spatial variation of \bar{C}^L will depend on the spatial variation of the object \bar{c}''^L . When \bar{c}''^L varies linearly, we have $\bar{C}^L = \xi_q(\tilde{C}_{,q} + \bar{c}''_{,q})$ such that a quadratic dependence of $\bar{\rho}^L \bar{C}^L$ is retained, as is the case with a linear \bar{C} field.

D. Mean gradient inconsistency in Eq. 26

We consider the validity of Eq. 26 using gradient transport applied to the conserved scalar $c_\alpha c_\beta$. A straightforward expression for $\overline{c'_\alpha c'_\beta u'_k}$ is suggested using the following arguments

$$\overline{(c_\alpha c_\beta)' u'_k} = -\langle \xi_j v_k \rangle (\overline{c_\alpha c_\beta})_{,j} \quad (113)$$

$$\implies \bar{C}_\alpha \overline{c'_\beta u'_k} + \bar{C}_\beta \overline{c'_\alpha u'_k} + \overline{c'_\alpha c'_\beta u'_k} = -\langle \xi_j v_k \rangle (\overline{c_\alpha c_\beta})_{,j} \quad (114)$$

$$\implies \overline{c'_\alpha c'_\beta u'_k} = -\langle \xi_j v_k \rangle (\overline{c'_\alpha c'_\beta})_{,j} \quad (115)$$

where we have expanded the Eulerian fluctuation $(c_\alpha c_\beta)'$ and used the gradient transport hypothesis for c_α, c_β . There is an inconsistency in the logic which makes this argument, and Eq. 115 invalid. The following three relations for the mean fields cannot hold true simultaneously.

$$\bar{C}_\alpha^L = \bar{C}_{\alpha,j} \xi_j, \quad \bar{C}_\beta^L = \bar{C}_{\beta,j} \xi_j, \quad \overline{c_\alpha c_\beta}^L = \overline{c_\alpha c_\beta}_{,j} \xi_j \quad (116)$$

300 If the mean fields $\bar{C}_\alpha, \bar{C}_\beta$ are linear in the displacement vector, $\overline{c_\alpha c_\beta}$ will generally have a quadratic dependence on the Lagrangian particle displacement vector thereby making Eq. 113 invalid.

E. Turbulent flux of ternary fluctuations

We consider three materially conserved scalars c_α, c_β and c_γ . The turbulent flux of ternary fluctuations is $\overline{c'_\alpha c'_\beta c'_\gamma u'_k}$. We derive the Lagrangian gradient transport closure for this turbulent flux. The following quantities are materially conserved.

$$\frac{Dc_\alpha}{Dt} = \frac{Dc_\beta}{Dt} = \frac{Dc_\gamma}{Dt} = \frac{D}{Dt}(c_\alpha c_\beta) = \frac{D}{Dt}(c_\beta c_\gamma) = \frac{D}{Dt}(c_\alpha c_\gamma) = \frac{D}{Dt}(c_\alpha c_\beta c_\gamma) = 0 \quad (117)$$

The ternary counterpart to the mean-fluctuation decomposition in Eq. 107 is the identity

$$c_\alpha c_\beta c_\gamma = c'_\alpha c'_\beta c'_\gamma + \overline{C}_\alpha \overline{C}_\beta \overline{C}_\gamma + \overline{C}_\alpha c_\beta c_\gamma + c_\alpha \overline{C}_\beta c_\gamma + c_\alpha c_\beta \overline{C}_\gamma - \overline{C}_\alpha \overline{C}_\beta c_\gamma - c_\alpha \overline{C}_\beta \overline{C}_\gamma - \overline{C}_\alpha c_\beta \overline{C}_\gamma \quad (118)$$

which results in $c_\alpha^L c_\beta^L c_\gamma^L = c'_{\alpha 0} c'_{\beta 0} c'_{\gamma 0}$, the ternary counterpart to Eq. 33. This relation can also be derived using Eq. 33 using induction. Using a decomposition of the form $c'_0 = c' + \overline{C}^L$ in Eq. 118, the expression $c_\alpha^L c_\beta^L c_\gamma^L$ can be expanded as

$$c'_\alpha c'_\beta c'_\gamma + \overline{C}_\alpha^L \overline{C}_\beta^L \overline{C}_\gamma^L + \overline{C}_\alpha^L c'_{\beta 0} c'_{\gamma 0} + c'_{\alpha 0} \overline{C}_\beta^L c'_{\gamma 0} + c'_{\alpha 0} c'_{\beta 0} \overline{C}_\gamma^L - c'_{\alpha 0} \overline{C}_\beta^L \overline{C}_\gamma^L - \overline{C}_\alpha^L c'_{\beta 0} \overline{C}_\gamma^L - \overline{C}_\alpha^L \overline{C}_\beta^L c'_{\gamma 0} = c'_{\alpha 0} c'_{\beta 0} c'_{\gamma 0} \quad (119)$$

We then take moments with the velocity at the final position. For passive scalars, the moments with initial point Eulerian and final point Lagrangian fluctuations are zero. Here, we do not distinguish between unary, binary or ternary fluctuations. For example, the moment of ternary final point fluctuation with a unary initial point fluctuation is taken to be zero. Similarly, the moment of a binary final point fluctuation with a binary initial point fluctuation is taken to be zero. Therefore, for passive scalars, we have

$$\langle c'_\alpha c'_\beta c'_\gamma v_k \rangle + \langle \overline{C}_\alpha^L \overline{C}_\beta^L \overline{C}_\gamma^L v_k \rangle = 0 \quad (120)$$

Using linear dependence of the Lagrangian fluctuations of the means on the dispersion vector, the gradient transport closure for the ternary fluctuation flux is

$$\langle c'_\alpha c'_\beta c'_\gamma v_k \rangle = -\langle \xi_p \xi_q \xi_r v_k \rangle \overline{C}_{\alpha,p} \overline{C}_{\beta,q} \overline{C}_{\gamma,r} \quad (121)$$

F. Second-moment based variable-density turbulence modeling

The present work addresses modeling within the Favre framework, specifically second-moment closures, where transport equations for second moments inevitably require closure of third moments.

Second-moment based closures solve for transport of averaged mass, momentum, energy and Favre stress $\tilde{R}_{ij} = \widetilde{u_i'' u_j''}$. In variable-density turbulence, additional second moments require evolution equations. A relevant modeling framework is the Besnard-Harlow-Rauenzahn (BHR) [2] modeling framework, in which the turbulent mass flux $\bar{\rho} a_k = \overline{\rho' u_k'}$ and the density-specific-volume covariance $b = \overline{\rho' v'}$ are additionally transported. Let us review the transport equations and the closures involved in second moment modeling of variable-density turbulence.

$$\text{Conserved scalar:} \quad (\bar{\rho} \tilde{C})_{,t} + (\bar{\rho} \tilde{C} \tilde{U}_k + \bar{\rho} \widetilde{c'' u_k''})_{,k} = 0 \quad (122)$$

$$\text{Momentum:} \quad (\bar{\rho} \tilde{U}_i)_{,t} + (\bar{\rho} \tilde{U}_i \tilde{U}_k + \bar{\rho} \tilde{R}_{ik} + \bar{P} \delta_{ik})_{,k} = 0 \quad (123)$$

$$\begin{aligned} \text{Total energy:} \quad & (\bar{\rho} (\tilde{\mathcal{E}} + \tilde{U}_k \tilde{U}_k / 2 + \tilde{R}_{kk} / 2))_{,t} + (\bar{\rho} (\tilde{\mathcal{E}} + \bar{P} + \tilde{U}_k \tilde{U}_k / 2 + \tilde{R}_{kk} / 2) \tilde{U}_j + \\ & \bar{\rho} \widetilde{e'' u_j''} + \bar{\rho} \tilde{R}_{jk} \tilde{U}_k + \bar{\rho} \widetilde{\rho u_k'' u_j''} / 2 - \bar{\rho} \bar{P} a_j + \overline{\rho' u_j'})_{,j} = 0 \end{aligned} \quad (124)$$

$$\text{Favre stress:} \quad (\bar{\rho} \tilde{R}_{ij})_{,t} + (\bar{\rho} \tilde{R}_{ij} \tilde{U}_k + (\bar{\rho} u_i'' u_j'' u_k'' + \overline{\rho' u_j' \delta_{ik}} + \overline{\rho' u_i' \delta_{jk}})_{,k} \quad (125)$$

$$+ \bar{\rho} (\tilde{R}_{jk} \tilde{U}_{i,k} + \tilde{R}_{ik} \tilde{U}_{j,k}) - (a_j \bar{P}_{,i} + a_i \bar{P}_{,j}) - (\overline{\rho' u_j'_{,i}} + \overline{\rho' u_i'_{,j}}) = 0 \quad (126)$$

$$\text{Scalar variance:} \quad (\bar{\rho} \widetilde{c'' c''})_{,t} + (\bar{\rho} \widetilde{c'' c''} \tilde{U}_k + \bar{\rho} \widetilde{c'' c'' u_k''})_{,k} = -2 \bar{\rho} \widetilde{c'' u_k''} \tilde{C}_{,k} \quad (127)$$

$$\text{Turbulent mass flux:} \quad (\bar{\rho} a_i)_{,t} + (\bar{\rho} a_i \tilde{U}_k)_{,k} = \overline{\rho v' p'_{,i}} + b \bar{P}_{,i} - \tilde{R}_{ik} \bar{\rho}_{,k} + \bar{\rho} (a_k a_i)_{,k} - \bar{\rho} a_k \bar{U}_{i,k} - \bar{\rho} (\overline{\rho' u_i' u_k' / \bar{\rho}})_{,k} \quad (128)$$

$$b = \overline{\rho' v'}: \quad (\bar{\rho} b)_{,t} + (\bar{\rho} b \tilde{U}_k)_{,k} = -2(b+1) a_k \bar{\rho}_{,k} + 2 \bar{\rho} a_k b_{,k} - \bar{\rho}^2 (\overline{\rho' v' u_k' / \bar{\rho}})_{,k} - 2 \bar{\rho}^2 \overline{v' v'} \quad (129)$$

The unclosed Favre moments are $\widetilde{c'' u_k''}$, $\widetilde{c'' c'' u_k''}$ and $\widetilde{u_i'' u_j'' u_k''}$. Unclosed turbulent fluxes in a_i and b equations are $\overline{\rho' u_i' u_k'}$ and $\overline{\rho' v' u_k'}$ respectively.

G. The Eulerian perspective on gradient transport for second moment turbulent flux

Gradient transport has previously also been derived from an Eulerian perspective [12] although with strong empirical assumptions. We review the Eulerian approach for passive scalar gradient transport and then derive results for the

third moment.

$$\overline{c'u'_j,t} + (\overline{c'u'_j} \overline{U}_k + \overline{c'u'_j u'_k},_k + \overline{u'_j u'_k} \overline{C}_{,j} + \overline{c'u'_k} \overline{U}_{,j} = -(\overline{p/\rho})' c',_j + (\nu_c \overline{u'_j c'},_k + \nu \overline{c' u'_j},_k) - (\nu_c + \nu) \overline{c',_k u'_j},_k \quad (130)$$

For steady-state, homogeneous turbulence, the balance between production and dissipation yields

$$\overline{u'_j u'_k} \overline{C}_{,j} = -(\nu_c + \nu) \overline{c',_k u'_j},_k \quad (131)$$

Next, the dissipation is assumed to be isotropic of the form $\overline{c',_k u'_j},_k \approx (\overline{c' u'_j})/\lambda_v^2$ such that the balance of production and dissipation results in

$$\overline{c' u'_j} = -\frac{\lambda_v^2}{\nu_c + \nu} \overline{u'_j u'_k} \overline{C}_{,j} \quad (132)$$

An outcome of this hypothesis is that the time-scale of relevance in the transport process gets associated with the viscous dissipation process. This is an inherent outcome of an Eulerian approach to turbulent transport.

Now, we use the approach above to derive gradient transport for the turbulent flux of the binary fluctuation $\rho' c'$. From an Eulerian perspective, the transport equation for the third moment can help in deriving an expression for the turbulent flux of the second moment.

$$\begin{aligned} (\overline{\rho' c' u'_j},_t + (\overline{\rho' c' u'_j} \overline{U}_{,k} + \overline{\rho' c' u'_j u'_k},_k = & -\overline{c' u'_j u'_k} \overline{\rho}_{,k} - \overline{\rho' u'_j u'_k} \overline{C}_{,k} - \overline{\rho' c' u'_k} \overline{U}_{j,k} \\ & - \overline{\rho' c' (u'_j u'_k)},_k - \overline{\rho' u'_j (c' u'_k)},_k - \overline{c' u'_j (\rho' u'_k)},_k \\ & - \overline{\rho' c' (p_{,j} / \rho)'} + (\nu_c \overline{\rho' u'_j c'},_k) + (\nu \overline{\rho' c' u'_j},_k) \\ & - \overline{\nu_c (\rho' u'_j),_k c',_k} - \overline{\nu (\rho' c'),_k u'_j},_k \end{aligned} \quad (133)$$

The isotropic dissipation assumption is of the form

$$\overline{\nu_c (\rho' u'_j),_k c',_k} \approx \nu_c \frac{\overline{\rho' c' u'_j}}{\lambda_v^2}, \quad \overline{\nu (\rho' c'),_k u'_j},_k \approx \nu \frac{\overline{\rho' c' u'_j}}{\lambda_v^2} \quad (134)$$

Assuming the mean shear term is negligible and the turbulence is homogeneous and stationary, the production-dissipation balance leads to the following expression for $\overline{\rho' c' u'_j}$

$$\overline{\rho' c' u'_j} = -\frac{\lambda_v^2}{\nu_c + \nu} (-\overline{c' u'_j u'_k} \overline{\rho}_{,k} - \overline{\rho' u'_j u'_k} \overline{C}_{,k} - \overline{\rho' c' (u'_j u'_k)},_k - \overline{\rho' u'_j (c' u'_k)},_k - \overline{c' u'_j (\rho' u'_k)},_k) \quad (135)$$

It is interesting that the Eulerian derivation for the second moment turbulent flux does not yield the gradient transport expressions employed in the literature, which only consists of second moment gradients. In contrast, the expression derived here also consists of first moment gradients. The similarity to Lagrangian gradient transport of the form in Eq. 141 is not a coincidence and is demonstrative of the transport physics accurately captured in the Lagrangian analysis.

H. Active-scalars and gradient transport

A passive scalar does not influence the velocity field. The mass density of the fluid, however affects the Lagrangian velocity field via inertia, and is therefore, active and not a passive scalar. This has two consequences for gradient transport.

- The Lagrangian moments involving the Lagrangian particle displacement vector are different in constant and variable-density flows. This is due to differential acceleration effects [24] in the presence of density variations.

$$\langle \xi_j v_k \rangle_{\text{CD}} = v_{\text{const}} \int_0^t \langle \xi_j p',_k \rangle d\tau \quad (136)$$

$$\langle \xi_j v_k \rangle_{\text{VD}} = \langle v \rangle \int_0^t \langle \xi_j p',_k \rangle d\tau + \int_0^t \langle \xi_j v' p',_k \rangle d\tau \quad (137)$$

where time-integration is carried out over a trajectory time-scale long enough such that $\langle \xi_j v_k(0) \rangle = 0$. Therefore, in contrast to constant-density turbulence, variable-density turbulence involves third moments involving the pressure-gradient and the specific volume.

- Dynamic effects on the active scalar flux are captured in gradient transport as unclosed initial-final point Lagrangian moments. For example, For a passive scalar c , in Eq. 11 we used

$$\langle c_0 v_k \rangle = \langle c'_0 v_k \rangle = 0 \quad (138)$$

However, for density, Eq. 138 does not necessarily hold. Instead,

$$\langle \rho_0 v_k \rangle = \langle \rho v_k \rangle = \langle \rho'_0 v_k \rangle \neq 0 \quad (139)$$

The term $\langle \rho v_k \rangle$ cannot be zero because the acceleration and consequently, the velocity in the Lagrangian trajectory depends on the mass-density $\rho(t; 0)$ associated with the Lagrangian particle. Therefore, gradient transport for turbulent mass flux yields

$$\langle \rho' v_k \rangle = -\langle \xi_j v_k \rangle \bar{\rho}_{,j} + \langle \rho'_0 v_k \rangle \quad (140)$$

and $\langle \rho'_0 v_k \rangle$ is an indicator of how well gradient transport approximates the turbulent mass flux. Similarly, Eq. 36 for binary scalar fluctuations will have additional unclosed terms for active scalars, such as density and those that are related to density via equation of state. As in Eq. 139, the moments of initial-point Eulerian fluctuations with final-point velocity can be non-zero in flows with pressure-gradient driven differential acceleration (see Appendix). In such a case, the non-zero moments that result from Eq. 36 are $\langle c'_{\alpha 0} c'_{\beta 0} v_k \rangle$, $\langle c'_{\alpha 0} \bar{C}_\beta^L v_k \rangle$ and $\langle c'_{\beta 0} \bar{C}_\alpha^L v_k \rangle$. These caveats hold for all gradient transport closures in variable-density flows, including the expressions in Eqs. 78-83.

Note: The terms that incorporate active scalar effects in Eq. 37 gradient transport are $\langle \rho'_0 c'_0 v_k \rangle$, $\langle \rho'_0 \bar{C}^L v_k \rangle$ and $\langle \bar{\rho}^L c'_0 v_k \rangle$. The equation may alternatively be expressed in a form where only a single unclosed term exists. By taking moments of Eq. 36 with the Lagrangian velocity $v_k(t; 0)$, we obtain

$$\langle \rho' c' v_k \rangle = -\langle c' \xi_i v_k \rangle \bar{\rho}_{,i} - \langle \rho' \xi_j v_k \rangle \bar{C}_{,j} - \langle \xi_i \xi_j v_k \rangle \bar{\rho}_{,i} \bar{C}_{,j} + \langle v_k \rho'_0 c'_0 \rangle \quad (141)$$

Here, the gradient transport expression also consists of terms linear in first moment gradients, and but the Lagrangian moments in such a case involve correlations with scalar fluctuations.

I. Numerical experiments

Two cases of numerical experiments are carried out to evaluate the relevance of the formal expressions to turbulent mixing zones. The first case demonstrates the quadratic dependence of scalar variance turbulent flux on the mean scalar gradient, as is predicted by formal gradient transport. The second case evaluates the turbulent mass flux component of the Favre scalar flux in buoyancy-driven turbulent mixing layers. For both cases, the data is reported for Cartesian grids with 256^3 points. This resolution is found to be sufficient for the quantities inferred from the simulations.

Case 1: Turbulent diffusion of passive scalar slab

The first numerical experiment involves mixing of a passive scalar slab in the presence of fine-grained isotropic turbulence. The scalar slab is introduced after the stationary turbulent flow-field has evolved over several large-eddy turnover times t_{LE} . Turbulent diffusion takes place in the presence of mean scalar gradient that weakens with time. From the expression in Eq. 45, we have for this case

$$\overline{c' c' u'_k} = \langle \xi_p \xi_q v_k \rangle (\bar{C}_{,p} \bar{C}_{,q}) \quad (142)$$

$$\overline{c' c' u'_2} = \langle \xi_2 \xi_2 v_2 \rangle (\bar{C}_{,2})^2 \quad (143)$$

The turbulence is stationary, homogeneous and isotropic and is forced at relatively high wavenumber range to ensure fine-grained turbulence with respect to the scalar gradient length-scale. The Taylor-scale Reynolds number is 250. The fields are evolved using a pseudospectral method and RK4 time integration.

Statistical averages are computed in the plane perpendicular to the direction of inhomogeneity x_2 . Therefore, the overbars and prime operators denote spatial averages in the context of this subsection.

In Fig 3(a), the instantaneous standard deviation σ of the moment $\langle c' c' u'_2 \rangle(x_2)$ is seen to scale as $(\bar{C}_{,2})^2$ (Eq. 45) post-transient. This is consistent with formal expression in Eq. 143 that predicts a quadratic dependence of the moment on the mean scalar gradient in stationary turbulence. The standard deviation (in x_2) is used here as a proxy for the magnitude of the third moment.

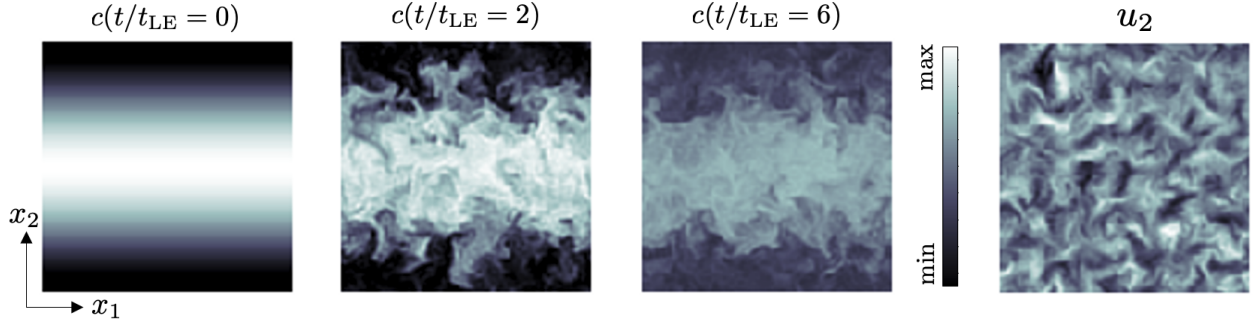


FIG. 2. Case 1: Scalar slab diffusing with time (left) and the instantaneous velocity component field of the stationary homogeneous isotropic turbulence (right).

Additionally, in Fig 3(a), we observe that the scalar variance turbulent flux magnitude scales as $\overline{c'c'}_{,2}$. This is predicted by the conventional gradient transport expression (Eq. 44). However, in Fig 3(b), we plot time-averaged normalized profiles of $\langle c'c'u'_2 \rangle$, $\overline{C}_{,2}$ and $\overline{c'c'}_{,2}$. The mean scalar gradient has two peaks, in the two mixing zones while the variance gradient has four peaks, two for each mixing zone. The magnitude envelope of $\overline{c'c'u'_2}$ can be seen to coincide with the mean gradient and not the variance gradient. Therefore, formal gradient transport can potentially provide more accurate estimates of second moment turbulent fluxes.

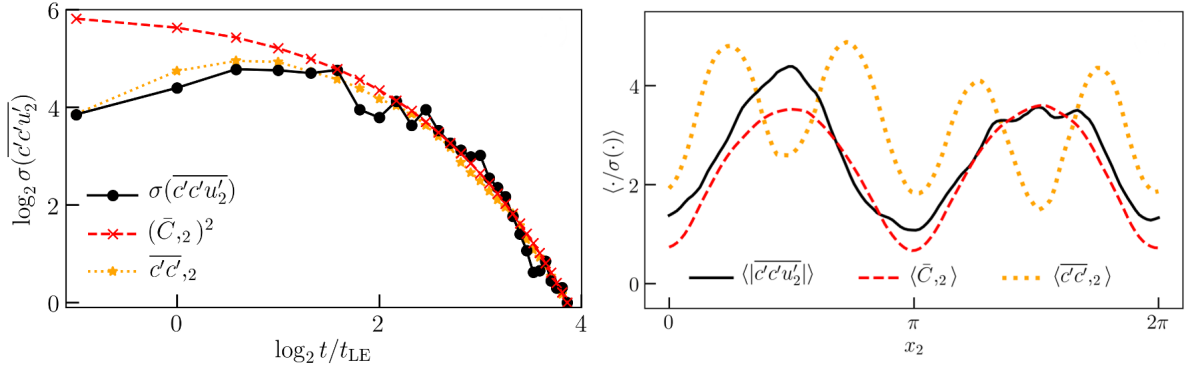


FIG. 3. (a) Third moment magnitude, mean and variance gradient as a function of time, and (b) Normalized profiles in x_2 , averaged between $4 \leq t/t_{LE} \leq 15$

Case 2: Rayleigh-Taylor turbulent mixing layer

Turbulent mass flux is an important dynamical variable in Rayleigh-Taylor unstable mixing layers. Therefore, simulations of incompressible buoyancy-driven mixing layers are carried out for three Atwood numbers $A = (\rho_2 - \rho_1)/(\rho_1 + \rho_2) = 0.5, 0.8$ and 0.9 to evaluate the mass-flux term. The details of the numerical method can be found in [1, 27]. The Reynolds number based on Taylor-microscale in the center of the mixing zone is approximately 75 for all three cases. The Froude number is fixed at 0.5.

The mass flux term $\overline{c''}(\langle \xi_2 v_2 \rangle \bar{\rho}_{,2} + \bar{\rho} a_2)$ (Eq. 85,86) is estimated using the approximation $\langle \xi_2 v_2 \rangle \approx -\overline{c'u'_2}/\overline{C}_{,2}$ and compared against the Favre scalar flux $\bar{\rho} c'' u''_2$. In Fig 4, the peak values of the fluxes in fully developed mixing zones are plotted. The values for the third moment $\overline{\rho'c'u'_2}$ are also plotted for comparison. The active scalar effects in the turbulent mass flux are seen to be negligible for moderate Atwood numbers but are important at higher Atwood numbers. As discussed in the main text, active scalar effects are implicitly neglected when gradient transport analysis is carried out with non-centered Favre fluctuations — the so-called argument by analogy. The formal expression allows explicit modeling of the mass flux effects in Favre fluxes.

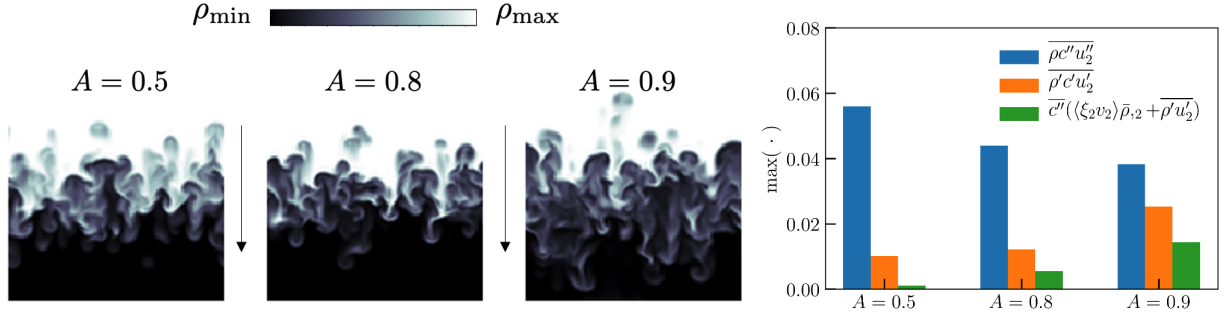


FIG. 4. Case 2. Buoyancy-driven turbulent mixing layers: (left) instantaneous density fields for three Atwood numbers, and (right) peak fluxes in the fully developed mixing zones. Arrows indicate direction of acceleration.

J. List of symbols and operators

$(\cdot)_{,j}$	Spatial gradient	$(\cdot)_0$	Initial point in Lagrangian trajectory
$\langle(\cdot)\rangle$	Lagrangian trajectory ensemble average	$(\cdot)^L$	Lagrangian fluctuation
$\overline{(\cdot)}$	Eulerian mean	$(\cdot)'$	Eulerian fluctuation
$\widetilde{(\cdot)}$	Favre mean	$(\cdot)''$	Favre fluctuation

TABLE I. Averaging and fluctuation operators

u_i	Eulerian velocity	v_i	Lagrangian velocity
ρ	Density	c_α	Scalar specie α
ξ_i	Lagrangian displacement vector	v	Specific volume
k	Specific kinetic energy	ϵ	Dissipation rate
R_{ij}	Turbulent stress	$\bar{\rho} a_i$	Turbulent mass flux
b	Density specific volume correlation	θ	Dilatation

TABLE II. Symbols for physical quantities

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