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# Reaction analogy based forcing for incompressible scalar turbulence

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We present a novel reaction analogy (RA) based forcing method for generating statistically stationary scalar fields in incompressible turbulence. The new method can produce more general scalar probability density functions (PDFs), for example quasi double-delta PDF, than current methods, while ensuring that scalar fields remain bounded, unlike existent forcing methodologies that can potentially violate naturally existing bounds. Such features are useful for generating initial fields in non-premixed combustion, inlet conditions for spatially developing flows, or for studying non-Gaussian scalar turbulence. The RA method mathematically models hypothetical chemical reactions that convert reactants in a mixed state back into its pure unmixed components. Various types of chemical reactions are formulated and the corresponding mathematical expressions derived such that the reaction term is smooth in scalar space and is consistent with mass conservation. For large values of the scalar forcing rate, the method produces statistically stationary quasi double-delta scalar PDFs. Quasi-uniform, Gaussian, and stretched exponential scalar statistics are recovered for smaller values of the scalar forcing rate. The shape of the scalar PDF can be further controlled by changing the stoichiometric coefficients of the reaction. The ability of the new method to produce fully developed passive scalar fields with quasi-Gaussian PDFs is also investigated, by exploring the convergence of the scalar variance spectrum to the Obukhov-Corrsin scaling and of the third order mixed structure function to the "four-thirds" Yaglom's law.

# I. INTRODUCTION

Direct numerical simulations (DNS) that employ forcing mechanisms have provided insights into the nature of velocity [e.g. 1–15] and scalar field turbulence [e.g. 15–18] by maximizing the inertial range of scales simulated on a given mesh or focusing on the response of the flow when only certain scales are excited. Simulations with a forcing term can also be used to estimate the numerical dissipation in non-DNS codes as done in Ref. [19], by following a suggestion from Ref. [9]. In addition, statistically stationary turbulent fields have been employed as inflow conditions for numerical simulations of spatially developing flows [20, 21].

In DNS studies of statistically stationary turbulence, various deterministic or random forcing terms usually mimic the energy supplied to the system by the natural shear production mechanism in the turbulent kinetic energy equation,  $\langle \mathbf{u}' \cdot \nabla \langle \mathbf{u} \rangle \cdot \mathbf{u}' \rangle$ , where  $\mathbf{u}'$  represents the velocity fluctuations and  $\nabla \langle \mathbf{u} \rangle$  is the mean velocity gradient. The velocity fluctuations can be replaced in this term with random noise or deterministic formulas, such that a specific form of the spectrum is obtained [16]. When the actual velocity fluctuations are used in the shear production term and  $\nabla \langle \mathbf{u} \rangle$  is constant, the forcing term becomes linear in velocity [2, 5, 9]. Typically, isotropic turbulence simulations are forced only at low wavenumbers. This maximizes the Reynolds number on a given mesh and ensures that the dynamics of the inertial range evolve naturally, without being influenced by the details of the forcing [22, 23].

Scalar turbulence has been studied in the past by analogous linear forcing methods, for example by mimicking the natural production term in the scalar variance transport equation corresponding to a constant mean scalar gradient [16] (referenced to below as IMG) or direct analogy with the linear velocity forcing term [18] (referenced to below as LS). The former method produces anisotropic scalar fields due to the directionality imposed by the mean scalar gradient, while the latter produces isotropic scalar fields. Neither method though guarantees any bounds for the scalar field itself. While this may be relevant to some practical problems, there are many problems where the scalar quantities have natural bounds (e.g. mass or molar fractions vary between 0 and 1).

In addition, most studies to date of statistically stationary scalar turbulence concern Gaussian or quasi-Gaussian distributions, even though, generally, Gaussianity is not a general feature of scalar turbulence. Different types of scalar PDFs have been reported from laboratory experiments and numerical simulations of passive scalars, which include stretched exponentials [24–26], pure Gaussian, and sub-Gaussian PDFs [17, 27]. Many practical problems naturally exhibit non-Gaussian scalar distributions, as is the case with multi-fluid flows such as scalar wakes, jets, or mixing layers, e.g. [28–30], buoyancy driven turbulence [31–34], and non-premixed combustion [35, 36].

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FIG. 1. Snapshots of the scalar field,  $\phi$ , produced by the proposed RA (with different target scalar dissipation rates), LS, and IMG forcing methods. The underlying turbulent field is the same for all four cases.

In this article, we propose a novel scalar forcing method based on a chemical reaction analogy (RA) which can produce more general scalar probability density functions (PDFs), for example quasi double-delta PDF, compared to current forcing methods that are limited to producing Gaussian or near Gaussian scalar PDFs. The new method also ensures the boundedness of the scalar field, in contrast to previous methods that can violate naturally existing bounds.

The RA method uses a hypothetical chemical reaction to convert the mixed fluid back into unmixed pure states. Reactants are identified based on a reaction analogy similar to that proposed in Ref. [33] to quantify the width of the Rayleigh-Taylor mixing layer and further generalized and discussed in Ref. [34]. Here, we consider even more general reaction rates together with their physical constraints to be used as forcing terms, and discuss the resulting statistically stationary scalar fields and their dependence on the forcing term parameters. The resulting forcing term is nonlinear and has similarities with several well-studied reaction-diffusion systems, as well as the cubic Ginzburg-Landau equation. There are also some analogies with the description of certain reacting flows using the mixture fraction as a progress variable [37]. None of these equations has been used to generate turbulent stationary scalar fields, and, as far as we could find, the form of the forcing term presented here is new; nevertheless, such connections may be worth studying in the future.

To demonstrate the potential of the method, we illustrate the scalar fields produced by various scalar forcing methods in figure 1. The RA method can produce both large concentrations of pure states (red and blue) or mixed states (white). In contrast, LS and IMG forcings only produce a large amount of mixed states.

# **II. THEORY AND FORMULATIONS**

We consider homogeneous isotropic turbulence governed by the incompressible constant density Navier-Stokes equations in a periodic box, subject to an explicit forcing term:

$$\nabla \cdot \boldsymbol{u} = 0, \quad \frac{\partial \boldsymbol{u}}{\partial t} + \boldsymbol{u} \cdot \nabla \boldsymbol{u} = -\nabla p + \nu \Delta \boldsymbol{u} + \boldsymbol{f}_{\boldsymbol{u}}, \tag{1}$$

where  $\boldsymbol{u}$  is the (zero mean) velocity vector, p is the pressure normalized by the (constant) density and the forcing,  $\boldsymbol{f}_{\boldsymbol{u}}$ , supplies energy at a constant rate to wavenumbers  $k = \|\boldsymbol{k}\| < 1.5$ . Maintaining a constant energy input rate has the advantage of being able to specify the Kolmogorov scale,  $\eta$ , and, thus, the degree at which the flow is resolved, at the onset [9].

The scalar field  $\phi$  obeys an advection diffusion equation:

$$\frac{\partial \phi}{\partial t} + \boldsymbol{u} \cdot \nabla \phi = \mathcal{D} \nabla^2 \phi + \left[ \underbrace{f(\boldsymbol{u})}_{\text{IMG}} \text{ or } \underbrace{f(\phi)}_{\text{LS or RA}} \right], \qquad (2)$$

where f is the scalar forcing term. The kinematic viscosity,  $\nu$ , and diffusion coefficient,  $\mathcal{D}$ , are constant.

Previous scalar forcings in Refs. [16] (denoted as IMG) and [15, 18] (denoted as LS) are linear and consist of a coefficient multiplied with either a velocity component,  $u_i$ , or the scalar field,  $\phi$ . The coefficient for the former is a constant mean scalar gradient while for the latter is instantaneously calculated to maintain a constant scalar variance.



FIG. 2. RA forcing terms for different stoichiometric coefficients versus  $\phi$ .

The RA method mathematically models chemical reactions occurring between hypothetical reactants identified in a mixed fluid state. Consider a partially mixed fluid state, in which the number densities are such that there are  $N_A$ molecules of species A and  $N_B$  molecules of species B per unit volume. The molar masses of species A and B are identical and the total number of molecules per unit volume is assumed to be a constant,  $N = N_A + N_B$ , which is required by incompressibility. We define the molar fractions for species A, B as  $X_A = N_A/N$ ,  $X_B = N_B/N$ . The scalar field  $\phi$  can then be related to the molar fractions of species A and B. If  $\phi \in [\phi_l, \phi_u]$ , then  $X_A = (\phi - \phi_l)/(\phi_u - \phi_l)$ and  $X_B = (\phi_u - \phi)/(\phi_u - \phi_l)$ , where  $\phi_l$  and  $\phi_u$  refer to the lower and upper bounds of the scalar field.

To formulate the reactions, we use ideas from a reaction analogy to describe mixing from Refs. [33, 34]. Consider hypothetical chemical species, the mixed state reactant, M, and the excess state reactant, E. This can be either fluid A, i.e.  $E_A$ , or fluid B, i.e.  $E_B$ , depending if the mixture is either A or B rich. The mixed state M is defined to contain an equal number of A and B molecules, while the excess reactant can be either A or B, such that the total equals the local concentrations of A and B. In regions where  $X_A < X_B$ , the number of mixed state molecules (consisting of equal number of A and B molecules) per unit volume is  $[M] = N_A$ . The remaining molecules of B, i.e.  $N - 2N_A$ , are present in the excess state so that the number of excess state molecules per unit volume is  $[E_B] = N - 2N_A = N_B - N_A$ . Similarly, when  $X_A > X_B$ , we have  $[M] = X_B$ ,  $[E_A] = N_A - N_B$ . Figure 2(a) illustrates the concentrations (or number densities) of M, E, A and B as a function of  $\phi$ .

A reaction between mixed and excess reactants can be represented by:

$$\begin{cases} mM + nE_B \to (2m+n)E_B & \text{when } X_A \le X_B \\ mM + nE_A \to (2m+n)E_A & \text{when } X_A > X_B, \end{cases}$$
(3)

where m and n are the stoichiometric coefficients. These reactions constitute demixing processes that transform a partially mixed state into the excess pure component. In terms of the scalar field  $\phi$ , the reactions transform the regions with below the average scalar values into the lower bound values and regions with above the average scalar values. Thus, the reactions counteract the effect of diffusion.

Using standard chemical kinetics formulas (e.g. [38]), the rate of increase of  $[E_{B,A}]$  due to reactions (3) between reactants M and  $E_{B,A}$  is  $2mK[M]^m[E_{B,A}]^n$  where K is the reaction rate constant. Expressing in terms of molar fractions, this becomes  $2mKX_A^m(X_B - X_A)^n$  when  $X_A < X_B$  and  $2mKX_B^m(X_A - X_B)^n$  when  $X_A > X_B$ . In terms of the scalar field, the forcing term in  $\dot{\phi}$  equation (after also transforming  $\dot{E}_{B,A}$  into  $\dot{\phi}$ ) further reduces to:

$$f_{\phi} = \begin{cases} -(\phi_u - \phi_l)mK(\frac{\phi_u + \phi_l - 2\phi}{\phi_u - \phi_l})^n (\frac{\phi - \phi_l}{\phi_u - \phi_l})^m; \ \phi \le \frac{\phi_u + \phi_l}{2} \\ (\phi_u - \phi_l)mK(\frac{2\phi - \phi_u - \phi_l}{\phi_u - \phi_l})^n (\frac{\phi_u - \phi}{\phi_u - \phi_l})^m; \ \phi > \frac{\phi_u + \phi_l}{2}. \end{cases}$$
(4)

For simplicity, we choose the scalar bounds as  $\phi_l = -1$  and  $\phi_u = 1$ . The forcing term can then be written in a compact form as

$$f_{\phi} = sign(\phi)f_c|\phi|^n (1-|\phi|)^m,\tag{5}$$

where  $f_c = 2mK/2^m$ . Different choices of the stochiometric coefficients result in unique mathematical expressions for  $f_{\phi}$ . For example, when m = 3, n = 1, the forcing acts less at large scalar magnitudes (figure 2 b). In comparison,

linear forcing, also shown in figure 2(b), acts stronger at larger scalar magnitudes. Simpler reaction expressions of the type  $A \to B$ ,  $X_B > X_A$ ;  $B \to A$ ,  $X_A > X_B$  could also be devised; however, they are generally discontinuous at  $\phi = 0$ . Alternately, more complex reactions could be devised for example by by changing the definition of the mixed state M.

There are several analogies between the forcing expression in (5) and well-known nonlinear reaction diffusion systems. For example, on each branch ( $\phi < 0$  or  $\phi > 0$ ) the n = 1, m = 1 formula is the same as Fisher's equation for spreading of advantageous allele within a given population [39]. Higher n, m values also lead to known equations on each branch, e.g. Newell-Whitehead-Segel equation describing Rayleigh-Benard convection or Zeldovich equation arising in combustion [39]. However, the reaction terms in these systems are not generally antisymmetric, which is a requirement for stationarity. For n = 1, m = 2, formula (5) reduces to  $\phi(1 - |\phi|)^2$ , which has some similarity with the reaction term in the cubic Ginzburg-Landau (GL) equation,  $\phi(1 - |\phi|^2)$ . The cubic GL equation arises naturally near critical phase transitions. If the linear term is positive (above the Hopf bifurcation, i.e. supercritical), the cubic term serves as a saturation to stop the growth of amplitude of the oscillations. If the linear term is negative (subcritical) and the cubic term is positive (excitation), the quintic term is needed to play the saturation role. The cubic GL equation is irreducible, so it is the simplest form that can describe such a bifurcation. There is also a parallel with combustion theory, in that an antisymmetric form of the reaction term can appear when the system is described by the mixture fraction as a progress variable for studying periodic reaction zones (e.g. [37]).

In summary, even though there are some similarities with several nonlinear reaction-diffusion systems, as far as we could find none of those systems has been used to generate stationary passive scalar turbulence with non-Gaussian PDFs and the particular form of our forcing term has not been studied before. Whether or not this form has practical relevance beyond our application could be an interesting topic, but it is outside the goals of our paper.

To eliminate the influence of small asymmetries in the initial scalar field, we further normalize the forcing expression. If this normalization is not performed, a particular reaction could be favored due to small numerical errors, transforming the entire scalar field into a trivial uniform state where either  $\phi = -1$  (or  $\phi_l$  in the general case) or  $\phi = 1$  (or  $\phi_u$  in the general case). To avoid this, the forcing is modified such that:

$$f_{\phi}^{*} = \begin{cases} f_{\phi}/\|f_{\phi}\|_{\phi \in [-1,0]} & \text{when } \phi \le 0\\ f_{\phi}/\|f_{\phi}\|_{\phi \in (0,1]} & \text{when } \phi > 0. \end{cases}$$
(6)

At stationarity, after taking the time average, the scalar variance equation becomes

$$(-\chi + f_c \langle \phi f_{\phi}^* \rangle)_{stationarity} = 0, \tag{7}$$

where  $\langle g \rangle$  represents the average (both over volume and time) of the quantity g and  $\chi$  is the scalar dissipation,  $\chi = \mathcal{D} \langle \nabla \phi \cdot \nabla \phi \rangle$ . The stationary value of the dissipation can be chosen *a priori* by imposing the forcing coefficient to be

$$f_c = \frac{\chi_{target}}{\langle \phi f_{\phi}^* \rangle},\tag{8}$$

where  $\chi_{target}$  denotes the preselected scalar dissipation value at stationarity. The volume averaged scalar dissipation fluctuates around the target value and its time average is very close to  $\chi_{target}$ . Indeed, the time variance of the volume averaged scalar dissipation displays very small values for all cases considered (figure 3). For the same velocity field and forcing parameters selected as described in section IIIB, the IMG and LS methods produce scalar dissipation variances of 0.1 and 0.013, respectively, which are at least an order of magnitude larger than those shown in figure 3 for the RA method. In principle, these small dissipation fluctuations could be further reduced by considering a modification of the forcing term of the type proposed in Ref. [14] for velocity forcing. Since the variability of the scalar dissipation is relatively small, we have not explored such modification.

Selecting the value of the scalar dissipation at the onset, as done for the velocity field in [9] to control the resolution of the simulation (see below), can also be used to estimate the numerical diffusion in non-DNS codes/simulations. Thus, at stationarity, relation (7) should still hold for non-DNS codes/simulations (e.g. in implicit large eddy simulations), with  $\chi$  now being the sum of the numerical and physical (if any) scalar dissipations. Then  $f_c$  can be used to estimate the numerical scalar dissipation.

The numerical results are obtained with a spectral version of the CFDNS code [40], which follows a standard, fully dealiased pseudo-spectral algorithm using a combination of truncation and phase shifting, for incompressible Navier-Stokes equations with a passive scalar [29]. To demonstrate the method, all results presented correspond to a Schmidt number,  $Sc = \nu/\mathcal{D} = 1$ . The velocity field is linearly forced as described above following [5, 9] and the target kinetic energy dissipation rate satisfies the resolution condition [9]



FIG. 3. The time variance over the stationary region of the volume averaged scalar dissipation normalized by  $\chi^2$  for different values of  $\chi_{target}$ .

$$\epsilon_{target} = \frac{\nu^3}{(\eta k_{max})_{target}} k_{max}^4,\tag{9}$$

where  $k_{max} = \frac{\sqrt{2}}{3}N$  is the maximum resolved wavenumber for a grid size  $N^3$ , and  $\eta$  is the Kolmogorov microscale.  $(\eta k_{max})_{target}$  needs to be large enough for the solution to be well resolved on a given grid. This is fixed at either 1.5 or 3.0, as specified below. For simplicity,  $\epsilon_{target} = 1.0$  for all cases. To maintain the required resolution,  $\nu$  and  $\mathcal{D}$  are changed appropriately to satisfy formula (9). Specifically, the simulations have been performed on 256<sup>3</sup>, 512<sup>3</sup>, and 1024<sup>3</sup> meshes, resulting in Taylor Reynolds numbers,  $Re_{\lambda}$ , ranging from 92 to 410. The turbulent kinetic energy at stationarity is  $k_t = \langle \mathbf{u} \cdot \mathbf{u} \rangle/2 \sim 3.0$ . The results are averaged over 7, 3, and 1.5 eddy turnover times for the three mesh levels, respectively. During stationarity, both the instantaneous values of kinetic energy,  $\epsilon = \nu \langle \nabla \boldsymbol{u} : \nabla \boldsymbol{u} \rangle$ , and scalar,  $\chi = \mathcal{D} \langle \nabla \phi \cdot \nabla \phi \rangle$ , dissipation rates are close to their target values,  $\epsilon_{target}$  and  $\chi_{target}$ .

Most of the results below are obtained with stochiometric coefficients m = 1 and n = 1, unless otherwise indicated. To increase maximum allowable  $\chi_{target}$ , the scalar bounds used in the forcing terms are smaller (i.e.  $\{-0.98, 0.98\}$ ) than the actual scalar bounds,  $\{-1, 1\}$ .

#### III. RESULTS

In this section, we present some sample results to highlight the properties of the new forcing method. First, the influence of the forcing parameters and resolution on the resulting scalar PDF are discussed, followed by a comparison with IMG and LS forcings in terms of scalar bounds, variance, PDF, and mechanical to scalar dissipation time scale ratio. The spectral behavior of the terms in the scalar variance equation for low and high values of  $\chi_{target}$  (or the forcing rate) is discussed next. Finally, we present results showing the convergence to the Obukhov-Corrsin spectral scaling for the scalar spectrum and to the 4/3 law for the mixed scalar-velocity third order structure function.

# A. The effects of forcing parameters on scalar PDF

We discuss the properties of the new RA forcing by examining how  $\chi_{target}$  and the stoichiometric coefficients affect the scalar PDF, using 256<sup>3</sup> simulations, with  $\eta k_{max} = 3.0$  ( $\Delta x \sim \eta$ ), corresponding to  $Re_{\lambda} \sim 92$ . This section also addresses the effect of resolution, using less resolved 256<sup>3</sup> simulations with  $\eta k_{max} = 1.5$  ( $\Delta x \sim 2\eta$ ), and  $Re_{\lambda} \sim 154$ , as well as 512<sup>3</sup> and 1024<sup>3</sup> simulations with  $\eta k_{max} = 3.0$  and  $Re_{\lambda} \sim 143$  and  $\sim 255$ , respectively.

Figure 4(a) illustrates the skewness,  $S = -\frac{\langle \phi^3 \rangle}{\langle \phi^2 \rangle^{3/2}}$ , and kurtosis,  $K = \frac{\langle \phi^4 \rangle}{\langle \phi^2 \rangle^2}$ , of the scalar PDFs generated by the RA forcing method for various target dissipation rates. Larger K values (i.e. more stretched PDFs) are produced for smaller  $\chi_{target}$ , while S remains negligible for all  $\chi_{target}$  values considered. Gaussian value of kurtosis is recovered by the method when  $\chi_{target} \sim 0.01$ . In terms of non-dimensional quantities, this value corresponds to  $\tau \sim 3.7$ , where the scalar and energy turnover time scale ratio is defined by



FIG. 4. (a) Skewness S (circles) and kurtosis (flatness) K (crosses) scalar PDF moments obtained for various target dissipation rates,  $\chi_{target}$ , ranging from 0.00046 to 0.932. The Gaussian kurtosis of 3.0 is indicated with dashed lines. (b) Stationary scalar PDFs for different  $\chi_{target}$  values.



FIG. 5. (a) Effects of stochiometric coefficients on the scalar PDF with  $\chi_{target} = 0.01$ . (b) Scalar PDFs obtained for  $\chi_{target}$  close to its largest  $\chi_{target}$  value supported on a 256<sup>3</sup> mesh with  $\eta k_{max} = 3.0$  ( $\chi_{target} = 0.932$ ) and  $\eta k_{max} = 1.5$  ( $\chi_{target} = 0.17$ ). For comparison, scalar PDFs from 512<sup>3</sup> and 1024<sup>3</sup> simulations with  $\chi_{target} = 0.932$  are also shown.

$$\tau = \frac{2k_t \,\chi}{\langle \phi^2 \rangle \epsilon}.\tag{10}$$

Figure 4(b) shows the PDF profiles during the stationary period for four values of  $\chi_{target}$ , 0.00046, 0.01, 0.129 and 0.932, corresponding to  $\tau$  values of 3.2, 3.7, 4.2, and 8.4, respectively. As  $\chi_{target}$  increases, the scalar field derivatives are larger, indicating the presence of more unmixed regions, with scalar values closer to the bounds. Thus, when  $\chi_{target} = 0.00046$ , the scalar PDF is more stretched than a Gaussian, with K = 3.7.  $\chi_{target} = 0.01$  yields a stationary quasi-Gaussian PDF, while  $\chi_{target} = 0.129$  results in a much flatter PDF (quasi-uniform), with K = 1.9. A further increase of  $\chi_{target}$  to 0.932 produces a quasi double-delta PDF with peaks near the forcing bounds and K = 1.2.

Figure 5(a) illustrates the scalar PDFs for the stochiometric coefficients m = 1, 2, 3 and n = 1, 2, 3, for  $\epsilon_{target} = 0.01$ . For m = 1, n = 1, the resulting scalar PDF is quasi-Gaussian, as explained above. When m = 1, n = 2, the PDF becomes a stretched exponential, with  $K \sim 13$ . The kurtosis continues to increase and reaches a value of  $\sim 30$  for m = 1, n = 3. The results are consistent with the forcing term dependence on the scalar values shown in figure 2. Thus, as the coefficient n is increased, the forcing acts more on the rich A or B mixtures and less near the fully mixed fluid, allowing the existence of more partially mixed fluid. This results in more elongated tails in the scalar PDF. On the contrary, when the coefficient m is increased, figure 2 shows that the forcing term acts stronger near the fully mixed fluid. This prevents the persistence of less mixed fluid, leaving a larger amount of fluid near the fully mixed value of  $\phi = 0.0$ . The resulting scalar PDFs have smaller kurtosis values, 2.4 and 2.3 for m = 2, 3, respectively (figure 5a).

For a given grid resolution, there is a maximum  $\chi_{target}$  beyond which the scalar fields are ill-resolved, as the scalar



FIG. 6. (a)  $L_{\infty}$  norm and (b)  $L_2$  norm of the scalar field using the RA, IMG, and LS methods. The x axis time is scaled with T, the eddy turnover time (~ 3.0) (c) Scalar PDFs produced by the RA, IMG and LS methods. (d) Mechanical to scalar dissipation time scale ratio comparison for the three forcing methods. The leftmost IMG and LS datapoints were obtained with mean scalar gradient of 0.1 and scalar variance matching the RA forcing with  $\chi_{target} = 0.01$ , respectively. The IMG and LS results are added to the plot based on their  $\chi$  values.

gradients increase with  $\chi_{target}$ . Similarly, for a fixed value of  $\chi_{target}$ , as the resolution (i.e.  $\eta k_{max}$ ) decreases, at some point the scalar gradients can no longer be represented on the mesh. Figure 5(b) illustrates the scalar PDFs corresponding to a scalar dissipation rate close to its maximum allowable value on a 256<sup>3</sup> mesh,  $\chi_{target} = 0.932$  for  $\eta k_{max} = 3.0$  ( $\Delta x \sim 2\eta$ ) and  $\chi_{target} = 0.17$  for  $\eta k_{max} = 1.5$  ( $\Delta x \sim \eta$ ). The results show that a bimodal (quasi double-delta) scalar PDF can only be obtained if the resolution is large enough to support the relatively large scalar gradients associated with this PDF. As the mesh size is increased, while maintaining  $\eta k_{max} = 3.0$  and  $\chi_{target} = 0.932$ , the double-delta scalar PDF gets larger peaks.

#### B. Comparison with previous methods

The ability to produce statistically steady quasi double-delta scalar PDFs distinguishes the RA method from any previous methods such as IMG [16] or LS [18]. In addition, the RA method has ability to keep the scalar field within presscribed bounds. Figure 6(a) illustrates the temporal evolution of the  $L_{\infty}$  norm of the scalar field generated by RA with two  $\chi_{target}$  values, IMG, and LS methods. The simulations use identical velocity fields with  $Re_{\lambda} \sim 92$  and  $\eta k_{max} = 3.0$  on 256<sup>3</sup> meshes. The RA method uses  $\chi_{target} = 0.01$ , for a quasi-Gaussian scalar PDF and  $\chi_{target} = 0.932$ , which gives a quasi double-delta scalar PDF. The mean scalar gradient used by the IMG method was chosen to be 1.0, while the forcing coefficient used by the LS method was chosen to match the scalar variance of the RA scalar with  $\chi_{target} = 0.932$ .

As figure 6(a) shows, the RA method keeps the scalar field within the specified bounds. For quasi double-delta scalar PDF ( $\chi_{target} = 0.932$ ), the scalar reaches its bounds, but the  $L_{\infty}$  scalar norm takes values slightly smaller

than the prescribed bounds for  $\chi_{target} = 0.01$ , as the reaction is not fast enough compared to molecular diffusion to guarantee the presence of pure fluids. On the other hand, for the IMG and LS methods, the scalar can take much larger values. For these forcing methods, the scalar extrema become larger and larger by increasing the values of the forcing parameters such as the scalar variance, scalar dissipation rate, or the imposed mean scalar gradient.

There are no a priori analytical estimates for the scalar variance obtained through the IMG method, while LS forcing maintains a certain scalar variance value. This is reflected in the evolution of the  $L_2$  norm shown in figure 6(b). The  $L_2$  norm results for the RA method with quasi double-delta PDF match the LS forcing results by design. As expected, for smaller values of  $\chi_{target}$  with all other parameters fixed, the results indicate smaller stationary values for the scalar variance, as the scalar PDF starts peaking at  $\phi = 0$ .

Figure 6c) compares the scalar PDFs obtained using the three methods. For  $\chi_{target} = 0.932$ , the RA and IMG results are close. By varying the mean scalar gradient for the IMG and forcing strength for the LS methods, the scalar PDF can depart slightly from a Gaussian (not shown), especially for the LS method where only certain parameters lead to a full Gaussian distribution, with a slightly stretched or compressed Gaussian being obtained in the general case.

The scalar dissipation rate is modeled in simpler mixing rate closures by assuming that the time scale ratio defined by (10) is constant [29, 41]. For a passive scalar, the time scale ratio is typically about 2, although significant variations can exist depending on the flow conditions [16, 29]. In combustion problems,  $\tau$  can be an order of magnitude larger than 2 [41]. Figure 6(d) shows the time scale ratio obtained for the three forcing methods. For the range of parameters considered, the IMG method produces  $\tau$  values close to 2, slightly increasing with the mean scalar gradient value. LS forcing results in  $\tau$  values close to 4, while the time scale ratio for RA forcing varies depending on  $\chi_{target}$ . For small  $\chi_{target}$  values,  $\tau \approx 4$ , similar to LS forcing results, but takes much larger values as the scalar distribution becomes bimodal (e.g.  $\tau \approx 8.4$  for  $\chi_{target} = 0.932$ ). For homogeneous isotropic turbulence, the time scale ratio is proportional to the ratio of the scalar to velocity Taylor microscales [42]. As the scalar PDF becomes bimodal, the scalar variations are restricted to a thin diffusion layer separating the pure fluid regions. In this case, the scalar gradients become large, increasing the values of  $\tau$ , offering a direct analogy with non-premixed combustion problems.

#### C. Forcing spectrum

Since the RA forcing is nonlinear, it is interesting to see how the forcing spectrum varies with the wavenumber. To illustrate the importance of the forcing term at different wavenumbers, we consider the spectral budget of the scalar variance transport equation:

$$\frac{\partial \langle \phi \phi^* \rangle}{\partial t} = \underbrace{\langle \hat{\phi} \hat{N}^* + \hat{\phi}^* \hat{N} \rangle}_{\text{Nonlinear transfer}} + \underbrace{\langle \hat{\phi} \hat{D}^* + \hat{\phi}^* \hat{D} \rangle}_{\text{Diffusion}} + \underbrace{\langle \hat{\phi} \hat{f}_{\phi}^* + \hat{\phi}^* \hat{f}_{\phi} \rangle}_{\text{Forcing}} \tag{11}$$

where

$$N = -\boldsymbol{u} \cdot \nabla \phi \tag{12}$$

$$D = \mathcal{D}\nabla^2\phi. \tag{13}$$

Here, the average,  $\langle \rangle$ , is over spherical shells and time (e.g. [43]), denotes the Fourier transform and \* is the complex conjugate. At stationarity, the time derivative on the left hand side of (11) vanishes. Figure 7 shows the terms on the right hand side of (11) from 1024<sup>3</sup> simulations with quasi-Gaussian ( $\chi_{target} = 0.01$ ) and quasi double-delta ( $\chi_{target} = 0.932$ ) PDFs. At low  $\chi_{target}$  values, RA forcing acts mostly at large scales, where it balances the transfer term, so that it leaves the small scalar scales largely unaffected (figure 7*a*). However, for large values of  $\chi_{target}$ , RA forcing can have a persistent influence at all wavenumbers (figure 7*b*).

The forcing term does not contain spatial derivatives, so it is expected to decrease faster than the transfer and dissipation terms and to become negligible at high enough wavenumbers. Indeed, that is the case with most of the RA forced simulations. For example, figure 7(c) shows that the forcing term increases in the dissipation range with the values of m and n, but remains more than an order of magnitude smaller than the dissipation term at small  $\chi_{target}$  values. Even for the quasi double-delta PDF cases, which have the largest coefficient in front of the forcing term, that term is smaller than the dissipation term in the dissipation range. Due to the lack of spatial derivatives, it is possible to invoke locality arguments similar to Ref. [44] to show that for any forcing coefficient, there is a large enough Reynolds number for which the influence of the forcing term becomes negligible at small scales. This is consistent with results shown in figure 7(c) for quasi double-delta scalar PDF ( $\chi_{target} = 0.932$ ), which indicate that the forcing term importance at small scales decreases with increasing the Reynolds number. In the next section, we explore the ability of RA forced simulations to recover fully developed high Reynolds number passive scalar dynamics.



FIG. 7. Absolute values of the spectra of the terms contributing to spectral scalar variance equation. (a)  $\chi_{target} = 0.01$ ,  $\eta k_{max} = 1.5$ ,  $Re_{\lambda} = 410$ , quasi-Gaussian scalar PDF, (b)  $\chi_{target} = 0.932$ ,  $\eta k_{max} = 3$ ,  $Re_{\lambda} = 255$ , quasi double-delta scalar PDF. (c) Forcing spectra normalized by the dissipation spectra in the dissipation range.

## D. Recovering passive scalar dynamics

The idea of universality of statistical laws for turbulent fluctuations has been central for fundamental turbulence research for the past several decades. In general, numerical [4, 12, 43] and experimental [45] evidence indicates that the scaling exponents of the higher order scalar structure functions depend on the injection mechanism, i.e. temporal forcing for the former and initial conditions for the latter. Thus, scalar scaling satisfying the universal equilibrium theory, if practically realizable, may only be achieved under rather special circumstances. In practical applications, for example in non-premixed combustion, the non-Gaussian statistics of the scalar fields may significantly increase the intermittency and modify even more the scaling behavior.

Here, we first explore the ability of the RA forced simulations to produce fully developed scalar spectra. To obtain high Reynolds number scalar statistics with a finite mesh, one usually forces the scalar field only at low wavenumbers to achieve a large separation of scales (e.g. [6, 43]). Therefore, we have also performed a simulation corresponding to



FIG. 8. Compensated scalar variance spectra,  $\chi^{-1} \epsilon^{1/3} k^{5/3} E_{\phi}(k)$ , versus the scaled wavenumber. Here, "filter" denotes a low wavenumber restriction of the RA forcing. The horizontal line corresponds to the Obukhov-Corrsin constant of 0.68.

 $\chi_{target} = 0.01$ , where the RA forcing is restricted to  $k \leq 1.5$ . Figure 8 shows that the compensated scalar variance spectra develop a plateau at low wavenumbers as a scale separation develops. The scalar spectrum and the Obukhov-Corrsin constant for the filtered RA forcing are very close to those obtained for similar Reynolds numbers using the low wavenumber scalar forcing in Ref. [43].

Next, we explore the ability of the RA forcing to recover the large Reynolds and Péclet numbers scaling of the mixed velocity scalar structure function (MSF). For stationary homogeneous isotropic velocity fields and homogeneous passive scalars, Yaglom's equation [43, 46] for the MSF derivative reads

$$\frac{\partial \langle (\Delta_r \phi)^2 \Delta_r u_r \rangle}{\partial \mathbf{r}} = -4\chi + 2\mathcal{D} \nabla_r^2 \langle (\Delta_r \phi)^2 \rangle + 2 \langle \Delta_r \phi \Delta_r f_\phi \rangle.$$
(14)

The scalar, forcing, and longitudinal velocity differences between two spatial points separated by the displacement  $\boldsymbol{r}$ , with  $\boldsymbol{r} = \|\boldsymbol{r}\|$ , are defined by  $\Delta_r \phi = \phi(\boldsymbol{x} + \boldsymbol{r}) - \phi(\boldsymbol{x})$ ,  $\Delta_r f_{\phi} = f_{\phi}(\boldsymbol{x} + \boldsymbol{r}) - f_{\phi}(\boldsymbol{x})$ , and  $\Delta_r u_r = (\boldsymbol{u}(\boldsymbol{x} + \boldsymbol{r}) - \boldsymbol{u}(\boldsymbol{x})) \cdot \boldsymbol{e}_r$ , where  $\boldsymbol{e}_r$  is a unit vector in the direction of displacement.

In regions much larger than Batchelor scale,  $\eta_B$ , the diffusion term becomes negligible. For scales much smaller than the energy containing scales, L, usually the forcing term contribution can also be neglected. After integrating over a ball of radius r, with  $\eta_B \ll r \ll L$  [43], Yaglom's equation predicts [4, 43, 47–49]

$$-\frac{\langle (\Delta_r \phi)^2 \Delta_r u_r \rangle}{\chi r} = \frac{4}{3}.$$
(15)

In general, when small  $\chi_{target}$  values are imposed using the RA method, MSF values are very close and they are also close to LS forcing results and slightly below the IMG forcing results with parameters described in section III B (figure 9a). Simulations with larger values of  $\chi_{target}$ , with quasi double-delta scalar PDF as the limiting case, consistently yield lower MSF values at all Reynolds numbers examined. In this case, the prefactor is large enough to prevent the forcing term to vanish after integrating Yaglom's equation. Nevertheless, the justification given in Ref. [43] is still valid, so  $\langle \Delta_r \phi \Delta_r f_{\phi} \rangle \rightarrow 0$  as  $r \rightarrow 0$ , and the forcing term contribution is expected to vanish at large enough Reynolds / Péclet numbers. Indeed, figure 9(a) shows that the MSF values become larger and seem to approach the 4/3 value as the Reynolds number is increased. However, full convergence to the 4/3 law may require much larger  $Re_{\lambda}$  values than those considered here. When the RA forcing is restricted to low wavenumbers and the scalar PDF is quasi-Gaussian, Yaglom's law is fully recovered (figure 9c). In this case, the results are consistent with previous forcing results using similar low wavenumber restrictions and Reynolds numbers [43]. In principle, one could minimize the Reynolds number needed to reach fully developed scalar statistics with a quasi double-delta PDF by using a combination of a low wavenumber restriction with a large  $\chi_{target}$  value but such that the simulations are still well resolved. This could be the subject of a future study.

# **IV. CONCLUSIONS**

We have presented a forcing method for producing stationary scalar fields in incompressible turbulence. The forcing term is constructed based on a hypothetical chemical reaction that transforms "mixed fluid" back into its "unmixed



FIG. 9. MSF values for (a)  $Re_{\lambda} = 92$ , with different forcing mechanisms; RA forcing corresponds to quasi-double-delta ( $\chi_{target} = 0.932$ ), quasi-Gaussian ( $\chi_{target} = 0.01$ ), and stretched ( $\chi_{target} = 0.00046$ ) scalar PDFs, (c) RA forcing for quasi-double-delta ( $\chi_{target} = 0.932$ ),  $Re_{\lambda} = 255$ , and quasi-Gaussian ( $\chi_{target} = 0.01$ ),  $Re_{\lambda} = 410$ , scalar PDFs. Here, "filter" denotes a low wavenumber restriction of the RA forcing.

components" or "pure states." The reaction form is chosen to ensure that the forcing term satisfies mass conservation, is smooth in the scalar space, and unbiased with respect to the two pure states. By construction, the scalar fields stay within predefined bounds, unlike previous methods that can violate naturally existing bounds, while also allowing more general scalar PDFs than previous methods. This method can also be extended to variable density and fully compressible flows. While there are some similarities with several well-known nonlinear reaction-diffusion systems, as far as we could find none of those systems have been used to generate stationary passive scalar turbulence with non-Gaussian PDFs and the particular form of our forcing term has not been studied before. Whether or not this form has practical relevance beyond our application, for example in relation to the cubic Ginzburg-Landau equation, could be an interesting topic for future studies.

By varying the target scalar dissipation, which controls the strength of the forcing term, scalar PDFs can be changed to cover a large range of kurtosis values, from stretched exponential, to quasi-Gaussian, approximately flat, and quasi double-delta PDF. Additional control on the shape of the scalar PDF can be exerted through the stoichiometric coefficients of the reactants (mixed and excess pure fluid) in the hypothetical reaction. As the  $\chi_{target}$  value increases and the scalar PDF approaches a bimodal, quasi double-delta shape, the time scale ratio of mechanical to scalar dissipation increases to large values, offering a direct analogy with non-premixed combustion problems.

Since the forcing term does not contain spatial derivatives, it is expected to decrease at large wavenumbers, compared to dissipation and nonlinear transfer terms in the spectral scalar variance equation. The results show that, while the forcing term increases at small scales with the stoichiometric coefficients (i.e. higher nonlinearities), for small values of the prefactor (i.e.  $\chi_{target}$ ), it remains much smaller than the dissipation term in the dissipation range. For large prefactor values, especially as a double-delta scalar PDF is approached, the forcing term (while still smaller than scalar dissipation) is no longer negligible in the dissipation range at  $Re_{\lambda} = 92$ . However, for all  $\chi_{target}$  values, the forcing term decrease at small scales with increasing  $Re_{\lambda}$ , so presumably, fully developed scalar statistics with quasi double-delta PDF can be obtained at large enough Reynolds numbers. When the forcing is restricted to low wavenumbers, the compensated scalar variance spectra develop a plateau, with an Obukhov-Corrsin constant similar to previous studies with low wavenumber scalar forcing. For all scalar PDFs, the third order scalar-velocity structure function (MSF) increases as the Reynolds number is increased. Simulations with quasi double-delta scalar PDF result in markedly lower MSF values than those with unimodal scalar PDFs.  $Re_{\lambda} = 410$  results with quasi-Gaussian scalar PDF and scalar forcing restricted to small wavenumbers fully recover Yaglom's 4/3 scaling and are consistent with previous results with low wavenumber IMG forcing. The convergence to 4/3 scaling is slower for simulations with large  $\chi_{target}$  values, especially quasi double-delta PDF, as the large prefactor of the forcing term requires much higher Reynolds number to achieve good separation of scales.

Previous studies indicate that scalar scaling following universal equilibrium theory may be obtained only under rather special circumstances, which may not be practically realizable. In many applications, scalar PDFs may be far from Gaussian (e.g. in non-premixed combustion), which could significantly change the intermittency behavior and scaling of the scalar fields. While we have not explored such scalings here, we hope that the new tool we have introduced could start shedding some light on the fundamental properties of scalar mixing closer to practical applications.

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