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A Rational Method of Determining Probability Distributions in Turbulent Reacting Flows

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Abstract

A method is presented for determining the joint probability distribution of scalar quantities in turbulent flows. Given a limited amount of statistical information – all mean values and second moments, say – the most likely distribution is shown to maximize the entropy, H. Consequently, the calculus of variations can be used to determine the most likely (i. e., maximum-entropy) distribution that is compatible with the given information. H is a function of the *a priori* probability which, it is shown, is uniform for passive scalars. In general, the *a priori* probability is a function of the reaction rates, and this functional dependence is determined.

Introduction

Local-mean properties in a turbulent reacting flow can be determined from the mean equations representing the conservation of mass, momentum, energy and chemical species concentration. In these equations, terms arise which are not directly related to the mean values and must be modelled, therefore, in order to provide a determinate set of equations. These terms can be considered in two groups, the first comprising velocity-velocity correlations (Reynolds stresses) and velocity-energy or velocity-species correlations (scalar fluxes). These correlations represent the transport of the quantity in question due to turbulent velocity fluctuations and are non-zero even in inert flows. Modelled transport equations for the Reynolds stresses have been proposed by Launder, Reece and Rodi [1] and Lumley and Kajeh-Nouri [2], while modelled scalar-flux equations are discussed by Launder [3]. Although these equations are not completely satisfactory there is no difficulty in principle in using this approach to determine the velocity correlations responsible for turbulent transport (at least in reasonably simple flows).

The second group of terms comprises the mean reaction rates of chemical species and, clearly, does not occur in inert flows. Only in the special case of a constant rate (i. e. isothermal) reaction can these terms be represented directly as correlations of the species concentrations and so, in general, an alternative statistical representation is required. In particular, for turbulent flames, the reaction rates are highly non-linear functions of the species concentrations; consequently, models that represent the mean rates as a truncated series of moments (see, for example, Borghi [4]) can only be justified for small temperature fluctuations. Few flames conform to this restriction. Thus, the previously-mentioned techniques for determining correlations cannot, by themselves, be used to determine the mean reaction rates.

If, instead of the mean equations, the transport equation for the joint probability distribution of the scalars (concentrations and enthalpy) is considered, then the reaction rates appear in closed form. That is, no modelling of the terms representing reaction is required. This observation led Dopazo and O'Brien [5] and Pope [6] to investigate the possibility of modelling and solving the equations. These investigations confirmed that the problem associated with the moment approach is avoided but other, most-likely superable problems arise. Even if it were not for these modelling difficulties, the strategy of solving the joint probability distribution function (pdf) equation is only viable for simple reactions in simple flows. This is because each scalar variable considered increases the dimensionality of joint-pdf by one, and so, for something as simple as a bunsen-burner flame, the solution of a four-dimensional integro-differential equation is required.

An approach which combines the relatively simple equations of the moment approach with the closed form of the reaction rates in the probability approach is to assume a shape for the joint-pdf based on the values of its moments. That is, transport equations are solved for the mean values and all second moments of the scalars and, from these, the joint-pdf is deduced. This immediately raises the question that this paper attempts to answer, namely: Given a limited amount of information (all first and second moments, for example) what is the most likely joint-pdf of the set of scalars?

The simplest case, and one that has received the attention of experimentalists and theoreticians, is that of a single passive contaminant. This can take the form of an inert trace species (Stanford and Libby [7]) or heat (Venkataramani, Tutu and Chevray [8]) and is of practical importance since an idealized diffusion flame can be characterized by an analogous quantity. The pdf of a passive scalar has been variously assumed to be a Gaussian (Hawthorne, Weddell and Hottel [9]), a beta-function distribution (Richardson, Howard and Smith [10], Rhodes [11]), a "clipped-Gaussian" (Lockwood and Naguib [12]), a double-delta function distribution (Bush and Fendell [13], Khalil, Spalding and Whitelaw [14]) and there have been several other suggestions (see Naguib [15]). Scalar quantities are usually bounded: for example, species concentrations, expressed as mass fractions, are bounded by zero and unity. Consequently, a distribution of infinite extent, such as a Gaussian, is inappropriate and the "clipped-Gaussian" is an *ad hoc* compensation for this defect. The other proposals mentioned are appropriate to bounded scalars but, notwithstanding empirical support for the beta-function distribution (Rhodes [11]), they all lack physical justification.

The beta-function distribution does appear to fit available data quite well and successful calculations of diffusion flames have been based on it, Jones [16]. In general, however, this or similar assumptions have three short-comings:

a) the distribution is only appropriate to a passive scalar and cannot, therefore, be generally used in reactive flows;

b) the assumptions cannot be simply extended to joint pdfs; and

c) the assumed distribution does not conform to any prescribed physical or statistical state.

Here, starting with a single passive scalar, the statistically-most-likely distribution is determined. The analysis is similar to that of Shannon [17] except that the assumption of uniform *a priori* probability is relaxed. The same approach has been employed in different contexts by Rumsey and Posner [18] and by Good [19]. The analysis is extended to an arbitrary number of scalars and in section 2 the joint-pdf appropriate to a set of reacting scalars is determined.

1. Non-Reactive Flows

The conservation equations for the scalar quantities considered are of the form,

$$\frac{\partial}{\partial t}(\rho\,\Phi_\alpha) + \frac{\partial}{\partial x_i}(\rho\,U_i\,\Phi_\alpha) = \frac{\partial}{\partial x_i}(\Gamma\frac{\partial\Phi_\alpha}{\partial x_i}) + \rho\,S_\alpha(\vec{\Phi})\,,\quad \alpha = 1, 2, 3\,\ldots. \tag{1}$$

Here Φ_α is one of the set of scalars $\vec{\Phi}$, ρ is the density, Γ the molecular diffusivity and $S_\alpha(\vec{\Phi})$ the reaction rate. For simplicity, the density is assumed to be constant but, provided density-weighted averages are employed, the result is applicable to variable density flows. In this section nonreactive systems ($S_\alpha = 0$) are considered, starting with a single scalar, Φ.

Statistical functions of Φ are defined in terms of ensemble averages,

$$\langle\Phi(\mathbf{x}, t)\rangle \equiv \lim_{N\to\infty}\frac{1}{N}\sum_{i=1}^{N}\Phi_{(i)}(\mathbf{x}, t)\,, \tag{2}$$

for example, where $\Phi_{(i)}(\mathbf{x}, t)$ represents the value of $\Phi(\mathbf{x}, t)$ is the ith of N realizations. The probability distribution function is formally defined by way of the Dirac delta function as,

$$p(\psi; \mathbf{x}, t) \equiv \langle\delta(\Phi(\mathbf{x}, t) - \psi)\rangle\,, \tag{3}$$

so that $p(\psi)d\psi$ is the probability of Φ being in the range $\psi < \Phi < \psi + d\psi$. Alternatively, p can be defined as the limit of a histogram which can be constructed as follows: consider the space ψ corresponding to realizable values of Φ. If Φ is a bounded variable, $0 < \Phi < 1$ say, then ψ is also defined for $0 < \psi < 1$: if any value of Φ is allowable, then ψ is of infinite extent. This space is divided into J intervals of width $\Delta\psi$, with $\psi_{j+1} = \psi_j + \Delta\psi$. Now, for each interval a number n_j is defined by the number of realizations of Φ within the interval,

$$\psi_j < \Phi_{(i)}(\mathbf{x}, t) < \psi_j + \Delta\psi\,. \tag{4}$$

Thus, the set of numbers $n_1, n_2 \ldots n_j \ldots n_J$ can be regarded as the histogram associated with the values of Φ_i (i = 1, N) and, evidently,

$$\sum_{j=1}^{J} n_j = N . \tag{5}$$

In the limit of small $\Delta\psi$ and large N, the histogram and the pdf coincide giving the alternative expression,

$$p(\psi_j) = \lim_{\substack{\Delta\psi N \to \infty \\ \Delta\psi \to 0}} \left(\frac{n_j}{N\Delta\psi}\right) . \tag{6}$$

For a given flow with given boundary and initial conditions, Φ and consequently $p(\psi)$ can, in principle, be determined from eq. (1) and the velocity equations. The question here, however, is statistical rather than deterministic: Given only a limited amount of statistical information ($\langle\Phi\rangle$ and $\langle\Phi'^2\rangle$, say), what is the most likely pdf, $p(\psi)$? The relationship (6) between the histogram and the pdf allows this question to be rephrased as: Given a limited amount of statistical information about $\Phi_{(i)}(x, t)$ (i = 1 ... N), what is the most likely associated set of numbers n_j? With no *a priori* knowledge of Φ, a given realization is equally likely to be within any interval and so equally likely to contribute of any n_j*). The most likely values of n_j are, therefore, those that can be achieved in the greatest number of ways, W. To give an extreme example, there is only one way in which the numbers $n_k = N$ and $n_j = 0$ ($j \neq k$) can occur; namely for each realization to be within the same range. The distribution, $n_k = N - 1$, $n_{k+1} = 1$, $n_j = 0$ ($j \neq k, j \neq k + 1$) can be realized in N ways, that is with each $\Phi_{(i)}$ but one being within the same range and with any one of the N realizations being in the adjacent interval. In other words, the second distribution is N-times more likely than the first since it can be achieved in N, as opposed to one, ways.

To generalise this procedure, we wish to calculate the number of different ways a given set of numbers can be formed. This problem arises in an analogous context in statistical mechanics (Tolman [20], p. 76, Eyring et al. [21], p. 86) and the result is that the number of ways is,

$$W = N! \prod_{j=1}^{J} (1/n_j!) . \tag{7}$$

The most likely set of numbers, n_j, and the most likely associated histogram is, therefore, that for which W given by equation (7) is a maximum.

The number of ways W may be seen to be a function of N and it is also a function of $\Delta\psi$. In particular, as $N\Delta\psi$ and $1/\Delta\psi$ tend to infinity, so also does W. Consequently,

*) This is the assumption of uniform *a priori* probability. It is shown later that this assumption is justified for passive scalars. For reactive scalars the assumption is not valid and is avoided, therefore.

in order to transfer the above result to pdfs by taking these limits, a function of W, N and $\Delta\psi$ is required that tends to a finite limit. Such a function is

$$H'(n, \Delta\psi, W) \equiv \ell n(\Delta\psi\, W^{1/N}/\Phi^*) \,, \tag{8}$$

where Φ^* is any characteristic measure of Φ which is included to render the argument of the logarithm non-dimensional (the natural choice being $\Phi' \equiv \langle\Phi'^2\rangle^{1/2}$). For given N and $\Delta\psi$, H' is an increasing function of W, and so maximising H' is equivalent to maximising W.

An alternative expression for H' in terms of N, $\Delta\psi$ and n_j can be obtained by applying Stirlings formula (for large n_j) to equation (7),

$$\ell n\, W = N\, \ell n\, N - \sum_j n_j\, \ell n\, n_j \,. \tag{9}$$

Then, from this and from the definition of H', it follows,

$$\begin{aligned} H'(N, \Delta\psi, n_j) &= \frac{1}{N}(N\, \ell n\, N - \sum_j n_j\, \ell n\, n_j) - \ell n \frac{\Phi^*}{\Delta\psi} \\ &= -\sum_j \frac{n_j}{N}(\ell n\, n_j + \ell n(\Phi^*/N\Delta\psi)) \\ &= -\sum_j \frac{n_j}{N\Delta\psi}\, \ell n(\frac{n_j\,\Phi^*}{N\Delta\psi})\, \Delta\psi \,. \end{aligned} \tag{10}$$

Now from (10) it is evident that H, the limit of H', is given by,

$$\begin{aligned} H &\equiv \lim_{\substack{\Delta\psi N \to \infty \\ \Delta\psi \to 0}} H'(N, \Delta\psi, n_j) \\ &= -\int p(\psi)\, \ell n(p(\psi)\, \Phi^*)\, d\psi \,, \end{aligned} \tag{11}$$

since the limit of $n_j/N\Delta\psi$ is $p(\psi_j)$. Thus, the most likely pdf is that for which H given by equation (11) is a maximum. H is refered to as the entropy of the distribution.

The maximisation of H is, of course, subject to available information: if, for example, the first two moments, $\langle\Phi\rangle$ and $\langle\Phi'^2\rangle$, are known, then the most likely pdf is that for which H is a maximum and,

$$\int p(\psi)\, d\psi = 1 \,, \tag{12}$$

$$\int \psi p(\psi)\, d\psi = \langle\Phi\rangle \,, \tag{13}$$

$$\int (\psi - \langle\Phi\rangle)^2\, p(\psi)\, d\psi = \langle\Phi'^2\rangle \,. \tag{14}$$

Using the calculus of variations, the required pdf can be shown to be,

$$p(\psi) = \exp(A + B\psi + C\psi^2)\,, \tag{15}$$

where the coefficients A, B and C are determined from eqs. (12–14). If ϕ is unbounded, evaluating the coefficients leads to,

$$p(\psi) = (2\pi\,\langle\phi'^2\rangle)^{-\frac{1}{2}} \exp\{-\frac{1}{2}(\psi - \langle\Phi\rangle)^2/\langle\Phi'^2\rangle\}\,, \tag{16}$$

which, clearly, is a Gaussian distribution.

For bounded distributions, the coefficients cannot be expressed as explicit functions of $\langle\Phi\rangle$ and $\langle\Phi'^2\rangle$, but eqs. (12–15) can readily be solved by numerical means. Figure 1 shows the resulting pdfs of the temperature at three different locations in a heated axi-symmetric jet. The maximum-entropy distribution is shown as a full line, the beta-function distribution as a broken line and the points are taken from the recent experimental data of Moneib [22].

Figure 1a corresponds to the edge of the jet (r/D = 3.6), figure 1c to the centre-line (r/D = 0) and figure 1b to a location in between (r/D = 2.6). It may be seen that at the edge of the jet the pdf is concentrated close to the lower bound where there is a spike. On the other hand, on the centre-line the distribution is unaffected by either bound and, consequently, the maximum-H distribution is Gaussian. For both these positions, there is good quantitative agreement between the measurements and the two predicted distributions.

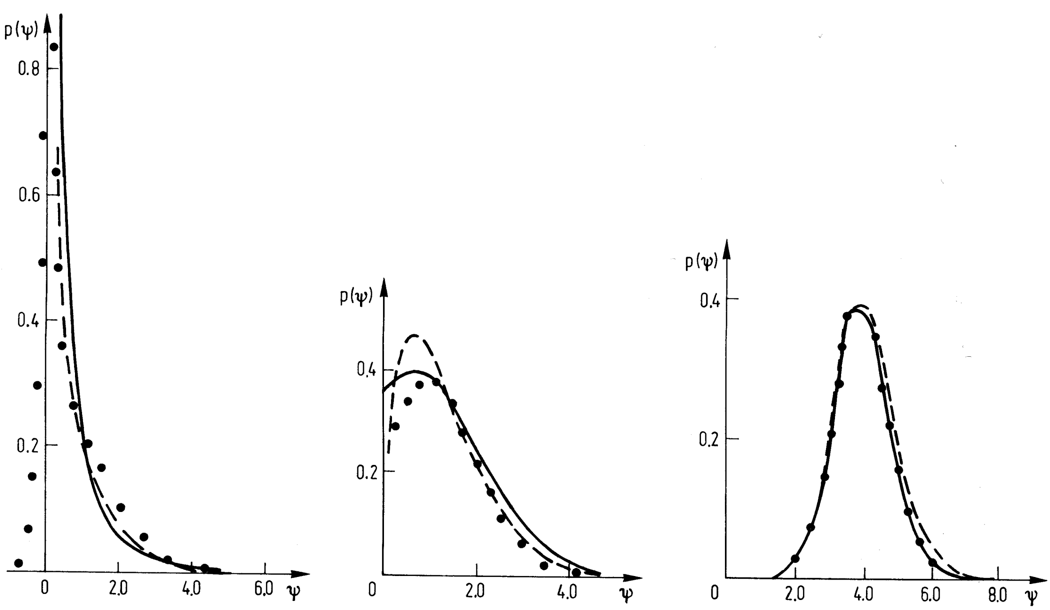

Fig. 1: $p(\psi)$ against ψ in an axisymmetric heated jet at x/D = 22.0; $\psi \equiv (T - T_0)/T'$.
● Data of Moneib (1977) —— maximum-H distribution – – – beta-function distribution

At $r/D = 2.6$, the distribution is clearly affected by the lower bound but there is no spike. In this instance, the discrepancy between the measurement and both predicted distributions is more evident. The beta-function distribution over-estimates the maximum probability by about 25%, while the maximum-H distribution predicts this quantity to within 5%, but over-estimates the probability near the lower bound. Considering that the predicted distributions are based on only two moments, the agreement at all three stations is as good as can be hoped for.

Attention is now turned to the joint-pdf of an arbitrary number (m) of scalars, $\vec{\Phi}$. The joint-pdf can be defined either in terms of delta functions (analogous to eq. (3)) or as the limit of a histogram (analogous to eq. (6)). In both cases the definition is such that $p(\vec{\psi}; x, t)\, d\vec{\psi}$ represents the probability in an m-dimensional element of $\vec{\psi}$-space, $d\psi_1\, d\psi_2 \ldots d\psi_m$. The previous analysis can be repeated for $p(\vec{\psi})$ instead of $p(\psi)$ simply by considering an m-dimensional histogram with interval $\Delta\vec{\psi}$. Then with $\vec{\Phi}^*$ as the product of the characteristic measures of each Φ_α ($\alpha = 1, m$), an equivalent to eq. (11) is obtained,

$$H = -\int p(\vec{\psi})\, \ell n(p(\vec{\psi})\, \vec{\Phi}^*)\, d\vec{\psi}\ . \tag{17}$$

Thus, the most likely joint-pdf of a set of scalars is that for which the entropy H given by eq. (17), is a maximum.

If all first and second moments are known, then the maximum-entropy distribution is,

$$p(\vec{\psi}) = \exp(A + B_\alpha\, \psi_\alpha + C_{\alpha\beta}\, \psi_\alpha\, \psi_\beta)\ , \tag{18}$$

where summation is implied over repeated sufficies, and the coefficient A, B_α and $C_{\alpha\beta}$ can be evaluated from,

$$\int p(\vec{\psi})\, d\vec{\psi} = 1\ , \tag{19}$$

$$\int \psi_\alpha\, p(\vec{\psi})\, d\vec{\psi} = \langle\Phi_\alpha\rangle\ , \tag{20}$$

$$\int (\psi_\alpha - \langle\Phi_\alpha\rangle)\, (\psi_\beta - \langle\Phi_\beta\rangle)\, p(\vec{\psi})\, d\vec{\psi} = \langle\Phi'_\alpha\, \Phi'_\beta\rangle. \tag{21}$$

For unbounded variables this produces the m-dimensional Gaussian distribution,

$$p(\vec{\psi}) = (2\pi)^{-m/2} |\langle\Phi'_\alpha\, \Phi'_\beta\rangle^{-1}|^{\frac{1}{2}} \exp\{-\tfrac{1}{2}\, \langle\Phi'_\alpha\, \Phi'_\beta\rangle^{-1}\, (\psi_\alpha - \langle\Phi_\alpha\rangle)\, (\psi_\beta - \langle\Phi_\beta\rangle)\}\ , \tag{22}$$

where $\langle\Phi'_\alpha\, \Phi'_\beta\rangle^{-1}$ represents the inverse of the matrix of second moments of the fluctuations $\Phi'_\alpha = \Phi_\alpha - \langle\Phi_\alpha\rangle$ and $|\langle\Phi'_\alpha\, \Phi'_\beta\rangle^{-1}|$ is the respective determinant. Inevitably, for bounded scalars, eqs. (18–21) can only be solved numerically. It should be noted that, in this process, the bounds are imposed simply by performing the integration in eqs. (19–21) over the bound region. Consequently, irregular bounds pose no problem.

2. Reactive flows

In the last section it was shown that, with only a limited amount of statistical information, the most likely pdf or joint-pdf is that for which H is a maximum. The single assumption on which this result rests is that, *a priori* (in the absence of any statistical information), $\vec{\Phi}$ is equally likely to be in any region of $\vec{\psi}$-space. The *a priori* probability $q(\vec{\psi})$ can be defined so that the probability of $\vec{\Phi}$ being in the range $\vec{\psi} < \vec{\Phi} < \vec{\psi} + d\vec{\psi}$ (compared with some reference range $\vec{\psi}_r < \vec{\Phi} < \vec{\psi}_r + d\vec{\psi}$) is $q(\vec{\psi})\, d\vec{\psi}$.

The scalar quantities under consideration are not random variables but are determined by eq. (1) and similar equations for the velocity field. These equations can be used, therefore, to assess the *a priori* probability. For passive scalars, eq. (1) becomes

$$\frac{\partial}{\partial t}(\rho \Phi_\alpha) + \frac{\partial}{\partial x_i}(\rho\, U_i\, \Phi_\alpha) = \frac{\partial}{\partial x_i}(\Gamma \frac{\partial \Phi_\alpha}{\partial x_i}), \qquad \alpha = 1, 2, 3 \ldots \tag{23}$$

and defining

$$\Phi'_\alpha \equiv \Phi_\alpha + C_\alpha, \quad \psi'_\alpha \equiv \psi_\alpha + C_\alpha \tag{24}$$

where C_α are constants, it follows,

$$\frac{\partial}{\partial t}(\rho \Phi'_\alpha) + \frac{\partial}{\partial x_i}(\rho\, U_i\, \Phi'_\alpha) = \frac{\partial}{\partial x_i}(\Gamma \frac{\partial \Phi'_\alpha}{\partial x_i}) \tag{25}$$

Now, with $q(\vec{\psi})$ as the *a priori* probability of $\vec{\Phi}$ (given by eq. (23)), since (23) and (25) are indistinguishable, the *a priori* probability of $\vec{\Phi}'$ must be the same function: that is,

$$q(\vec{\psi}) = q(\vec{\psi}') = q(\vec{\psi} + \mathbf{C})\,. \tag{26}$$

And, since the constants $\mathbf{C}$ can be chosen arbitrarily, (26) can only be satisfied if q is uniform. Thus, for passive scalars, (1) indicates that uniform *a priori* probability is the only possible non-contradictory assumption.

For reacting scalars the same argument cannot be applied directly because of the reaction rate term. Instead we proceed in three stages: first, the definition of entropy appropriate to non-uniform *a priori* probability is deduced; second, the functional form of $q(\vec{\psi})$ is obtained; and finally, the specific form of $q(\vec{\psi})$ for the present situation is given.

For an arbitrary transform space $\vec{\psi}^*$, $p^*(\vec{\psi}^*)$ is related to $p(\vec{\psi})$ by

$$p^*(\vec{\psi}^*)\, d\vec{\psi}^* = p(\vec{\psi})\, d\vec{\psi}\,, \tag{27}$$

or

$$p^*(\vec{\psi}^*) = p(\vec{\psi}) \left| \frac{\partial \vec{\psi}^*}{\partial \vec{\psi}} \right|^{-1}, \tag{28}$$

where $|\partial\vec{\psi}^*/\partial\vec{\psi}|$ is the determinant of the Jacobian of the transformation. The same transformation applies to $q(\vec{\psi})$. Consequently $\vec{\psi}^*$ can be chosen as a space of uniform *a priori* probability. That is

$$\alpha = \text{constant} = q^*(\vec{\psi}^*) = q(\vec{\psi})\left|\frac{\partial\vec{\psi}^*}{\partial\vec{\psi}}\right|^{-1} . \tag{29}$$

Since in $\vec{\psi}^*$-space the *a priori* probability is uniform, the entropy is given by (17)

$$H = -\int p^*(\vec{\psi}^*) \ln(p^*(\vec{\psi}^*)\,\vec{\Phi}^*)\, d\vec{\psi}^* \tag{30}$$

and, transforming this integral back to $\vec{\psi}$ space yields

$$H = -\int p(\vec{\psi}) \ln(p(\vec{\psi})/q(\vec{\psi}))\, d\vec{\psi} \quad ^{\dagger)} \tag{31}$$

where, for simplicity, the constant α has been taken as $1/\vec{\Phi}^*$. Thus the entropy of a distribution with *a priori* probability $q(\vec{\psi})$ is given by eq. (31), and the most-likely distribution maximizes this "entropy".

As in the case of passive scalars, the form of the *a priori* probability can be determined by examining the behavior of $q(\vec{\psi})$ under linear transformations. The details of this examination are given by Pope [23].

The only function of $\vec{\psi}$ that appears in the transport equation (1) is the reaction rate. It is to be expected therefore that $q(\vec{\psi})$ is a function of $S_\alpha(\vec{\psi})$ only. However, in order to confirm this expectation, it is supposed that $q(\vec{\psi})$ may be an intrinsic function of $\vec{\psi}$ as well: such an explicit dependence upon $\vec{\psi}$ proves to be inadmissible. In addition to $\vec{\psi}$ and $S_\alpha(\vec{\psi})$, a turbulent time scale τ and the correlation $C_{\alpha\beta} \equiv \langle\Phi'_\alpha \Phi'_\beta\rangle$ — none of which is a function of $\vec{\psi}$ – are introduced for normalization. Thus, $q(\vec{\psi})$ can be written as an unknown functional, Q',

$$q(\vec{\psi}) = Q'(\vec{\psi}, S_\alpha(\vec{\psi}), \tau, C_{\alpha\beta}) . \tag{32}$$

The *a priori* probability in the linear-transformed $\vec{\psi}$-space (denoted by an asterisk) is given by

$$q^*(\vec{\psi}^*) = q(\vec{\psi})|C|^{1/2}/|C^*|^{1/2} . \tag{33}$$

Here $|C|$ is the determinant of $C_{\alpha\beta}$. Now, since the functional form of q should be independent of the transformation, it follows:

$$|C|^{1/2}\, Q'(\vec{\psi}, S_\alpha(\vec{\psi}), \tau, C_{\alpha\beta}) = |C^*|^{1/2}\, Q'(\vec{\psi}^*, S^*_\alpha(\vec{\psi}^*), \tau, C^*_{\alpha\beta}) . \tag{34}$$

Consequently, eq. (32) can be written as

$$q(\vec{\psi}) = |C|^{-1/2}\, Q(I_1, I_2 \ldots I_n) , \tag{35}$$

$^{\dagger)}$ This quantity sometimes is called the "loss of information" or "gain of entropy".

where Q is an unknown functional and $I_1, I_2 \ldots I_n$ are all the nondimensional invariant functions that can be formed from the arguments of Q'. There is just one such invariant:

$$X(\vec{\psi}) \equiv \tau [C^{-1}_{\alpha\beta} S_\alpha(\vec{\psi}) S_\beta(\vec{\psi})]^{1/2} . \tag{36}$$

Thus,

$$q(\vec{\psi}) = |C|^{-1/2} Q(X(\vec{\psi})) . \tag{37}$$

Equation (37) indicates that $q(\vec{\psi})$ is a function of the single invariant $X(\vec{\psi})$ which can be regarded as the ratio of the turbulent time scale to the reaction time scale. The functional Q is unknown. However, for inert flows, $X(\vec{\psi})$ is zero and hence, irrespective of Q, $q(\vec{\psi})$ is a constant, confirming the previous result.

A physical argument is now employed to determine $q(\vec{\psi})$ for a *single* reactive scalar. All other things being equal, the probability of finding a moving point in a region of space is inversely proportional to its speed. Reaction and molecular mixing can be thought of as transport in ψ-space, with respective speeds of $|S(\psi)|$ and Φ'/τ. The interpretation of $|S(\psi)|$ as the speed of transport in ψ-space due to reaction is clear since, by definition, in the absence of all other influences,

$$\frac{d\Phi}{dt} = S(\Phi) . \tag{38}$$

Similarly, the speed associated with molecular mixing is defined as the rate of change of Φ' in the absence of all other influences:

$$\frac{\Phi'}{\tau} = -\frac{d\Phi'}{dt} = \frac{\Gamma}{\Phi'\rho} \langle \frac{\partial\Phi}{\partial x_i} \frac{\partial\Phi}{\partial x_i} \rangle . \tag{39}$$

This equation serves to define τ. With the *a priori* probability being inversely proportional to the sum of these speeds, $q(\psi)$ is given by

$$q(\psi) = \tau^{-1}(\Phi'/\tau + |S(\psi)|)^{-1} \tag{40}$$

(τ^{-1} is a normalisation factor). Or, dividing through by Φ'/τ,

$$q(\psi) = (\Phi')^{-1}(1 + \tau|S(\psi)|/\Phi')^{-1} . \tag{41}$$

This expression for $q(\psi)$ has been obtained from physical arguments. Is it, one may ask, consistent with the form of $q(\vec{\psi})$ deduced from the transformation properties (eq. (37)), and can it be extended to more than one scalar? Both questions can be answered in the affirmative since the term $\tau|S(\psi)|/\Phi'$ is nothing but the invariant $X(\psi)$. Thus the general expression for $q(\vec{\psi})$ consistent with equation (41) is

$$q(\vec{\psi}) = |C|^{-1/2}/(1 + X(\vec{\psi})) . \tag{42}$$

3. Concluding remarks

It has been shown that the statistically-most-likely distribution maximizes the entropy

$$H \equiv - \int p(\vec{\psi}) \ln(p(\vec{\psi})/q(\vec{\psi}))\, d\vec{\psi} \tag{43}$$

subject to available constraints. The *a priori* probability $q(\vec{\psi})$ is a function of the single invariant

$$X(\vec{\psi}) \equiv \tau[\langle \Phi'_\alpha \Phi'_\beta \rangle^{-1} S_\alpha(\vec{\psi}) S_\beta(\vec{\psi})]^{1/2} \tag{44}$$

and the specific form suggested is

$$q(\vec{\psi}) = |\langle \Phi'_\alpha \Phi'_\beta \rangle|^{-1/2}/(1 + X(\vec{\psi}))\,. \tag{45}$$

If the available constraints are a knowledge of all first and second moments, the maximum-entropy distribution is

$$p(\vec{\psi}) = q(\vec{\psi}) \exp(A_0 + A_\alpha \psi_\alpha + B_{\alpha\beta} \psi_\alpha \psi_\beta)\,. \tag{46}$$

The principal result is that the most likely joint-pdf is that for which H given by (43) is a maximum. Strictly, a distinction should be made between the "most likely" and the "mean of all likely" distributions. However, in this instance, as in the statistical mechanics analogue, the most likely distribution is overwhelmingly so, making the distinction unnecessary.

Since the maximum-H distribution has been determined from physical arguments, it would be surprising if it violated any constraints generally applicable to pdfs. Indeed it does not. $p(\vec{\psi})$ is guaranteed to be non-negative because the calculus of variations gives an expression of the form of equation (46): that is, $p(\vec{\psi})$ is equal to $q(\vec{\psi})$ multiplied by the exponential of a function, both of which are non-negative quantities. In addition, since $q(\vec{\psi})$ is zero outside the bounds of a bounded distribution, so also is $p(\vec{\psi})$. It is also required of an assumed distribution that it be defined for all realizable values of the moments. Thus too is guaranteed. H is defined for any realizable distribution and hence, for any realizable set of moments, at least one value of H exists. The existence of one value of H is sufficient for the maximum-H distribution to be defined. A more rigorous argument with the same conclusion is given by Rumsey and Posner [18].

Finally we note that the assumption that $p(\vec{\psi})$ maximises H (subject to *all* available information) is the best possible assumption. H is a measure of information: therefore the maximum-H distribution contains a minimum of information. Any other assumed distribution contains additional (and therefore spurious) information.

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