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# Lagrangian modelling of turbulent diffusion, buoyancy and chemical processes

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## ABSTRACT

Linear Lagrangian equations for turbulent motion and buoyancy are shown to be consistent with the Eulerian budget equations for the mean wind and potential temperature fields and their coupled variances. These equations take reference to a locally isotropic dissipation according to Kolmogorov theory and a return-to isotropy pressure redistribution according to Rotta. They permit a selfconsistent calculation of the flow in dependence on three dimensionless flow numbers. These flow numbers are the Prandtl number under neutral stratification Pr, the critical gradient Richardson number $Ri_c$ and a new introduced number $Ri_0$. The timescale of turbulent motion normalized to the vertical sheared horizontal mean wind is calculated within this approach in dependence on the gradient Richardson number. The calculation of means fields can be included into the solution algorithm. Nonlinear equations can be derived using different concepts. They provide two relations between Pr, $Ri_c$ and $Ri_0$ for homogeneous turbulence. Using these relations different measurements provide in dependence on Pr in a very good agreement the same critical Richardson number of $Ri_c = 3 / 10$. Taking this value as a third relation one obtains Pr = 1 and $Ri_0 = 1 / 10$.

## THE LAGRANGIAN APPROACH

In the Lagrangian approach a turbulent flow is regarded as a whole of fluid particles each having a constant mass. In correspondence with hydrodynamics equations have to be derived for the motion of these particles and their properties like for instance a potential temperature or a chemical composition. In this way, the flow can be calculated matched on inhomogeneous terrain and around obstacles, which may be related with considerably problems in other approaches. Correspondingly, this Lagrangian approach is very convenient for describing turbulent diffusion in complex flows and near sources [1]. Whereas concepts are given for the description of particles dynamics, the inclusion of buoyancy effects and chemical reactions was in discussion over a long time. As shown by Heinz and Schaller [2, 3] linear equations for particle motion and buoyancy can be derived which are consistent with the Eulerian budget equations for the mean wind and potential temperature fields and their coupled variances. These equations permit a selfconsistent calculation of the flow in dependence on three dimensionless flow numbers. Chemical transformations can be included into this description. Using different concepts nonlinear equations can be derived, which permit the calculation of these flow numbers [4].

## LINEAR THEORY

Lagrangian stochastic equations can be used as approximations of the hydrodynamic equations for high-Reynolds number turbulent flows. These equations for fluid particle motion and particle potential temperature have to be derived in correspondence with hydrodynamic theory. This can be guaranteed up to second-order comparing the transport equations for the mean values and variances of the considered quantities [2]. Here, the variance equations are taken in the approximations of Kolmogorov and Rotta, that means a locally isotropic dissipation and a return-to isotropy are assumed. The coefficients appearing in the Lagrangian equations are calculated in terms of a turbulent timescale $\tau$ and closure parameters $k_1$, $k_3$ and $k_4$ arising from the above assumptions. The turbulent timescale can be related with gradients of the mean wind and potential temperature fields by a rescaling procedure, and the closure parameters $k_1$, $k_3$ and $k_4$ can be related with dimensionless flow numbers [3]. These equations are selfconsistent, because the calculation of mean quantities and variances can be included into the solution algorithm. As only remaining parameters three dimensionless flow numbers appear, the Prandtl number under neutral stratification Pr, the critical gradient Richardson number $Ri_c$, and a new introduced flow number $Ri_0$.
Neglecting chemical reactions, each particle is characterized at the time t by its position $\mathbf{x}_L(t)$, velocity $\mathbf{U}_L(t)$ (vectors with components $x_L^I(t)$ and $U_L^I(t)$, where I = 1, 2, 3 and the subscript L denotes a Lagrangian quantity) and potential temperature $\Theta_L(t)$. Combining particle velocity $\mathbf{U}_L(t)$ and $\Theta_L(t)$ to the 4-dimensional vector $\mathbf{Z}_L(t) = (\mathbf{U}_L(t), \Theta_L(t))$, these equations read

$$\frac{d}{dt} x_L{}^I(t) = Z_L{}^I(t), \qquad (1a)$$

$$\frac{d}{dt} Z_L{}^i(t) = \langle a^i \rangle + G^{ij}(Z_L{}^j - \langle Z_E{}^j \rangle) + b^{ij} \frac{dW^j}{dt}, \qquad (1b)$$

where the small superscripts run from 1 to 4 in difference to capitals and summation over repeated superscripts is assumed. The first two terms in (1b) give the systematic particle motion with unknown coefficients $\langle a^i \rangle$ and $G^{ij}$, where the ensemble average is denoted by $\langle \cdots \rangle$. The ensemble averages of Eulerian quantities (subscript E) are estimated at fixed positions $\mathbf{x}$ which are replaced by $\mathbf{x} = \mathbf{x}_L(t)$ in the equations. The last term of (1b) describes the

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influence of a stochastic force, characterized by the white noise $dW^j / dt$ and an intensity matrix b with elements $b^{ij}$. Here, $dW^j / dt$ is a Gaussian process having a vanishing mean and uncorrelated values to different times, $\langle dW^i / dt \rangle = 0$ and $\langle dW^i / dt\,(t) \cdot dW^j / dt\,(t') \rangle = \delta_{ij}\, \delta(t - t')$, $\delta_{ij}$ denotes the Kronecker delta and $\delta(t - t')$ the delta function. Instead of considering the equations (1a-b) for the stochastic transport of particles and their changing properties, the equivalent Fokker-Planck equation can be considered for the probability density to find given values of particle properties at given locations and times. The Lagrangian joint mass density function will be denoted by $F_L$. This function is similar to the corresponding probability density function, but normalized to the mean concentration $\langle c(\mathbf{x}, t) \rangle$ of observed particles,

$$\int d\mathbf{Z}\, F_L(\mathbf{Z}, \mathbf{x}, t) = \langle c(\mathbf{x}, t) \rangle. \tag{2}$$

The transport of $F_L$ can be derived by different methods and is given in correspondence with (1a-b) by the equation

$$\frac{\partial F_L}{\partial t} + \frac{\partial}{\partial x^I} Z^I F_L = -\frac{\partial}{\partial Z^i}\left[\langle a^i \rangle + G^{ij}(Z^j - \langle Z_E{}^j \rangle)\right] F_L + \frac{\partial^2}{\partial Z^i \partial Z^j} B^{ij} F_L, \tag{3}$$

where $B^{ij} = 1/2\, b^{ik} b^{kj}$ is introduced. Assuming a state-independent and locally isotropic dissipation according to Kolmogorov theory, $B^{ij}$ is given by

$$B = \frac{1}{4\tau}\begin{pmatrix} C_0 q^2 & 0 & 0 & 0 \\ 0 & C_0 q^2 & 0 & 0 \\ 0 & 0 & C_0 q^2 & 0 \\ 0 & 0 & 0 & C_1 \langle (Z_E{}^4 - \langle Z_E{}^4 \rangle)^2 \rangle \end{pmatrix}, \tag{4}$$

where $C_0$ and $C_1$ are unknown universal constants, $q^2$ is twice the turbulent kinetic energy, that means $q^2 = \langle (Z_E{}^I - \langle Z_E{}^I \rangle)(Z_E{}^I - \langle Z_E{}^I \rangle) \rangle$, and $\tau$ is a turbulent timescale. By $F_L$ the statistical properties of an ensemble of observed particles (that means distinguished in some way, for instance emitted by a source) are determined at a single point. Considering the motion of all fluid particles, the corresponding mass density function is denoted by the (Eulerian) mass density function F, normalized to the averaged fluid density $\rho$,

$$\int d\mathbf{Z}\, F(\mathbf{Z}, \mathbf{x}, t) = \langle \rho(\mathbf{x}, t) \rangle. \tag{5}$$

This mass density has to fulfill the transport equation (3), too. This relation of F with the unknown coefficients $\langle a^i \rangle$ and $G^{ij}$ can be used for deriving consistency constraints between these coefficients and Eulerian means and variances of the velocity and potential temperature fields. Multiplying (3) with $Z^i$ and integrating over $\mathbf{Z}$, the transport equations for the mean values of the wind and potential temperature fields can be derived, so that $\langle a^i \rangle$ is determined by

$$\frac{D\langle Z_E^{\ i}\rangle}{Dt} + \frac{\partial V^{iL}}{\partial x^L} = \langle a^i \rangle, \tag{6}$$

where the abbreviation $D / Dt = \partial / \partial t + \partial / \partial x^K \cdot \langle Z_E^{\ K}\rangle$ is used and the matrix of second moments of the coupled wind and potential temperature field is written by

$$V = \begin{pmatrix} \langle u^1u^1\rangle & \langle u^1u^2\rangle & \langle u^1u^3\rangle & \langle u^1\theta\rangle \\ \langle u^2u^1\rangle & \langle u^2u^2\rangle & \langle u^2u^3\rangle & \langle u^2\theta\rangle \\ \langle u^3u^1\rangle & \langle u^3u^2\rangle & \langle u^3u^3\rangle & \langle u^3\theta\rangle \\ \langle \theta u^1\rangle & \langle \theta u^2\rangle & \langle \theta u^3\rangle & \langle \theta^2\rangle \end{pmatrix}, \tag{7}$$

with $z^k = Z_E^{\ k} - \langle Z_E^{\ k}\rangle$ for the fluctuations. Accordingly, by multiplication of (3) with $Z^i\ Z^j$ and integration over **Z**, the transport equations for the second moments can be derived which read

$$\frac{DV^{ij}}{Dt} + I(Z_E^{\ i}, Z_E^{\ j}) = G^{ik}V^{kj} + G^{jk}V^{ki} + \frac{C_0 q^2}{2\tau}\delta_{ij} - \frac{C_0 q^2 - C_1 V^{44}}{2\tau}\delta_{i4}\delta_{j4}, \tag{8}$$

where a source of loss or gain of potential temperature is neglected for simplicity and the operator $I(X_E, Y_E)$ is used ($X_E$, $Y_E$ are any Eulerian quantities and $x = X_E - \langle X_E\rangle$ and $y = Y_E - \langle Y_E\rangle$ are their fluctuations) with

$$I(X_E, Y_E) = \frac{\partial}{\partial x^K}\langle z^K xy\rangle + \langle z^K x\rangle \frac{\partial \langle Y_E\rangle}{\partial x^K} + \langle z^K y\rangle \frac{\partial \langle X_E\rangle}{\partial x^K}.$$

These equations (6) and (8) had been compared by Heinz and Schaller [2] with the corresponding Eulerian budget equation of first- and second-order. Using the Boussinesq approximation and the incompressibility constraint, $\partial Z_E^{\ K} / \partial x^K = 0$, the conservation equations of momentum and potential temperature read

$$\frac{\tilde{D} Z_E^{\ i}}{Dt} = \nu \frac{\partial^2 Z_E^{\ i}}{\partial x^K \partial x^K} + (\alpha - \nu)\frac{\partial^2 Z_E^{\ 4}}{\partial x^K \partial x^K}\delta_{i4} - \langle \rho\rangle^{-1}\frac{\partial p}{\partial x^K}\delta_{Ki} -$$

$$- g\left(1 - \beta\left(Z_E^{\ 4} - \langle Z_E^{\ 4}\rangle\right)\right)\delta_{i3}, \tag{9}$$

where $\tilde{D} / Dt = \partial / \partial t + \partial / \partial x^K \cdot Z_E^{\ K}$, $\nu$ is the kinematic viscosity, $\alpha$ the coefficient of molecular heat transfer, $\beta$ the thermal expansion coefficient, p the pressure and g the acceleration due to gravity. Consequently, $\langle a^i\rangle$ is determined by the averaged right-hand side of (9),

$$\langle a^i\rangle = \nu \frac{\partial^2 \langle Z_E^{\ i}\rangle}{\partial x^K \partial x^K} + (\alpha - \nu)\frac{\partial^2 \langle Z_E^{\ 4}\rangle}{\partial x^K \partial x^K}\delta_{i4} - \langle \rho\rangle^{-1}\frac{\partial \langle p\rangle}{\partial x^K}\delta_{Ki} - g\delta_{i3}. \tag{10}$$

Assuming a locally isotropic dissipation according to Kolmogorov and a return-to isotropy pressure redistribution according to Rotta, transport equations of second-order can be derived from the conservation equations leading to

$$\frac{DV^{ij}}{Dt}+I(Z_E{}^i,Z_E{}^j)=\left\{-\frac{k_1}{4\tau}\delta_{ik}+\frac{k_1-k_3}{2\tau}\delta_{i4}\delta_{k4}+\beta g\delta_{i3}\delta_{k4}\right\}V^{kj}+$$

$$+\left\{-\frac{k_1}{4\tau}\delta_{jk}+\frac{k_1-k_3}{2\tau}\delta_{j4}\delta_{k4}+\beta g\delta_{j3}\delta_{k4}\right\}V^{ki}+k_2q^2\frac{\partial\langle Z_E{}^L\rangle}{\partial x^K}\left[\delta_{Li}\delta_{Kj}+\delta_{Lj}\delta_{Ki}\right]+$$

$$+\frac{q^2}{2\tau}\cdot\frac{k_1-2}{3}\delta_{ij}-\frac{(k_1-2)/3\cdot q^2-(2k_3-2k_4-k_1)V^{44}}{2\tau}\delta_{i4}\delta_{j4}, \qquad (11)$$

where the closure parameters $k_1$, $k_2$, $k_3$ and $k_4$ are introduced. The equations (11) are consistent with the derived transport equations (8) for all variances $V^{ij}$, if $k_2 = 0$, $C_0 = (k_1 - 2) / 3$, $C_1 = 2k_3 - 2k_4 - k_1$ and in particular the unknown coeffient matrix G is chosen by

$$G^{ij}=-\frac{k_1}{4\tau}\delta_{ij}+\frac{k_1-k_3}{2\tau}\delta_{i4}\delta_{j4}+\beta g\delta_{i3}\delta_{j4}. \qquad (12)$$

Hence, the coefficients $\langle a^i\rangle$, $G^{ij}$ and $b^{ij}$ are given by the turbulent timescale $\tau$, the first moments $\langle Z_E{}^i\rangle$, the pressure gradient $\partial\langle p\rangle/\partial x^K\ \delta_{Ki}$ and the closure parameters $k_1$, $k_3$ and $k_4$.

## TIMESCALE AND FLOW NUMBERS

Investigating the local solutions of (11), where all gradients of variances and the third moments are neglected, the turbulent timescale $\tau$ can be calculated in dependence on the gradients of the mean fields [2], and the closure parameters $k_1$, $k_3$ and $k_4$ can be related with three dimensionless flow numbers [3]. Considering a vertical sheared horizontal wind field only, with the gradient Richardson number $Ri = \beta g\ \partial\langle Z_E{}^4\rangle / \partial x^3 / [(\partial\langle Z_E{}^1\rangle / \partial x^3)^2 + (\partial\langle Z_E{}^2\rangle / \partial x^3)^2]$ this relation for the timescale normalized to the vertical sheared horizontal wind $T = \tau\ [(\partial\langle Z_E{}^1\rangle / \partial x^3)^2 + (\partial\langle Z_E{}^2\rangle / \partial x^3)^2]^{1/2}$ reads

$$\frac{8}{k_1{}^2}\frac{k_1-2}{3}T^2Ri^*\left[Ri^*-Ri_c{}^*\right]=-\left[\left(1+Pr^*\right)Ri^*-Pr^*\right]-$$

$$-\sqrt{\left[\left(1+Pr^*\right)Ri^*-Pr^*\right]^2-4Ri^*Pr^*\left[Ri^*-Ri_c{}^*\right]}, \qquad (13)$$

where the abbreviations $Ri_c{}^* = Ri_c / Ri_0$ as well as $Pr^* = Pr / Ri_0 \cdot (k_1 - 2) / 3$ are used. This relation is valid for $Ri < Ri_c$ and $Ri_c < Pr\ (k_1 - 2) / 3 < (k_1 + 7) / 6 + [(k_1 + 7)^2 / 36 + Ri_c]^{1/2}$. Here, the abbreviations

$$Ri_c = (k_1 - 2)\cdot(k_3 - k_4)/(4k_4 + 3k_1 + k_1k_4,$$
$$Pr = k_3 / k_1,$$
$$Ri_0 = Ri_c \cdot Pr/(k_3/k_4 - 1)$$

are used. Orientating the $x^1$-axis into the mean horizontal wind direction, the turbulent Prandtl number $Pr_t$ is defined by $Pr_t = (V^{13}\, \partial\langle Z_E^4\rangle / \partial x^3) / (V^{34}\, \partial\langle Z_E^1\rangle / \partial x^3)$. It can be shown that Pr denotes the Prandtl number under neutral stratification, so that this number can be estimated by the limit $\partial\langle Z_E^4\rangle / \partial x^3 \to 0$. The number $Ri_c$ can be interpreted as critical Richardson number determining the transition from laminar to turbulent motion, because the timescale relation is valid only for $Ri < Ri_c$. The number $Ri_0$ was new introduced. Studying consistency conditions of local solutions of the second-order equations it can be seen, that this number limits the range of validity of the timescale relation to $Ri \approx -Ri_0$ [3]. This flow number is a global gradient Richardson number characterizing a slightly unstable stratification. By these results closed equations are given depending only on the three flow numbers, because the calculation of the mean wind and potential temperature fields can be included into the solution algorithm.

## CHEMICAL TRANSFORMATIONS

The changing of composition is desribed by a mixing model proposed by Pope [5], avoiding problems related with the application of a stochastic differential equation for this process. According to that, with probability $1-N\,\Delta t / \tau_m$ in a time interval $\Delta t$ the compositions of all stochastic particles $n = 1, N$ do not change, but with probability $N\,\Delta t / \tau_m$, the compositions of a pair of particles changes. Therefore, for two particles (selected at random and denoted by p and q) their values $\mathbf{M}_L^{(p)}$ and $\mathbf{M}_L^{(q)}$ are replaced by the common mean. This model can be written in the following way:

- with probabiliy $1-N\,\Delta t / \tau_m$,

$$\mathbf{M}_L^{(n)}(t+dt) = \mathbf{M}_L^{(n)}(t) + (\langle \mathbf{a}_m \rangle + \mathbf{S}_m)\Delta t, \quad n = 1, N, \tag{14a}$$

- with probability $N\,\Delta t / \tau_m$,

$$\mathbf{M}_L^{(n)}(t+dt) = \frac{1}{2}\left(\mathbf{M}_L^{(p)}(t) + \mathbf{M}_L^{(q)}(t)\right) + (\langle \mathbf{a}_m \rangle + \mathbf{S}_m)\Delta t, \quad n = p, q \tag{14b}$$

$$\mathbf{M}_L^{(n)}(t+dt) = \mathbf{M}_L^{(n)}(t) + (\langle \mathbf{a}_m \rangle + \mathbf{S}_m)\Delta t, \quad n \neq p \text{ and } n \neq q, \tag{14c}$$

$\tau_m$ being the mass fraction timescale to be determined and $\mathbf{a}_m$ and $\mathbf{S}_m$ are vectors with elements $a_m^\alpha$, $S_m^\alpha$, where $\alpha = 1, k$ runs for the different mass fractions, which have to be determined or given as source for loss and gain of substance.

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Let us assume in formal correspondence with the above closure assumptions leading to (11)

$$2 \cdot \lambda \cdot \left\langle \frac{\partial m_\alpha}{\partial x^k} \frac{\partial m_\beta}{\partial x^k} \right\rangle = k_4^* \tau^{-1} \left\langle m_\alpha m_\beta \right\rangle,$$

where $m_\alpha$ denotes a mass fraction fluctuation, $\lambda$ is the coefficent of molecular composition transfer and $k_4^*$ is a closure parameter taking on the role of $k_4$. For the timescale of mixing $\tau_m$ one obtains

$$\tau_m = \frac{\tau}{k_4^*}, \tag{15}$$

if it is assumed that the transport of composition can be considered as similar to that of the other scalar quantity, that means the potential temperature.

## NONLINEAR THEORY

Nonlinear equations can be obtained using concepts of the theory of stochastic processes and of nonequilibrium statistical mechanics, where the potential temperature is included to describe buoyancy effects [4]. Within the former one approach nonlinear Markovian equations can be derived in dependence on the distribution function of turbulent fluctuations. Assuming a locally Gaussian distribution of wind and potential temperature fluctuations, an equation can be derived which is quadratic in the fluctuations. The limits of applicability of this equation can be assessed investigating the influence of non-Gaussianity [6]. On the other hand, using the second one approach a more general equations including non-Markovian effects can be derived. Studying these equations for homogeneous turbulence for the $C_0$ and $C_1$ the relations $C_0 = 2 / 3$, and $C_1 = 2\ k_4$ can be found. Hence, using the above relations of $C_0$ and $C_1$ with $k_1$, $k_3$ and $k_4$, these parameters can be calculated for instance in dependence on $k_3$, or the flow numbers in dependence on Pr. In doing so it is found that all measured values of $Ri_c$ are very near

$$Ri_c = \frac{3}{10}, \tag{16a}$$

where the deviations are smaller that 6%. Taking this value into account, the flow numbers can be estimated for homogeneous turbulence to be

$$Pr = 1, \tag{16b}$$

$$Ri_0 = \frac{1}{10}. \tag{16c}$$

These values can be used to calculate the second-order closure parameters $k_1$, $k_3$ and $k_4$, for which many different values had been found in dependence on the determination method.

## CONCLUSIONS

Closed linear equations for particle motion and potential temperature are given depending only on three dimensionless flow number. For these the values (16a-c) are found for homogeneous turbulence. More general nonlinear equations can be derived as described above. These equations can be applied to calculate turbulent flow and dispersion of substances over inhomogeneous terrain, near sources and obstacles. The asymmetry of the distribution of turbulent fluctuations as well as of the dispersion coefficient matrix can be investigated.

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