



Modelling turbulence and turbulent dispersion in the stratified atmospheric boundary layer by PDF methods

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Abstract

A new model concept is presented for the description of turbulence and transport of reactive substances in the stratified atmospheric boundary layer. All the flow field and the transport of substances are described by the motion of fluid particles and change of their properties as composition or temperature. The essential advantage of this concept over other methods consists in the simulation of mixing processes of particle properties and the exact treatment of arbitrary complicated reaction schemes. These are the main stumbling blocks in conventional methods and may be the reason for errors of several orders of magnitude in concentration calculations. Essential features of this approach are illustrated and computational aspects are discussed.

1 Introduction

The estimation of emissions of pollutants into the atmosphere, their spatial distribution and concentration change due to chemical transformations requires accurate models for these processes in order to develop efficient strategies for the environmental protection. One of the key problems of such models is the description of turbulence-chemistry interactions.¹⁻³ Pollutants are often released



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at different locations, such that reactions cannot occur. This un-mixedness is often neglected in conventional Eulerian atmospheric chemistry models, which may result in serious overestimates of reaction (as illustrated below) and consequently in concentration calculations with errors of several orders of magnitude.⁴ The consideration of the mixedness of substances in Eulerian transport equations for substance concentrations requires the incorporation of the cross-correlations of concentration fluctuations. However, their calculation complicates the description of transport and chemistry considerably and is related to different additional assumptions as e.g. on higher-order chemistry terms.^{5,6}

Lagrangian particle models have been used in the last decade as an universal tool for the description of the transport of reactive substances. The model equations can be solved simply also for complex flows, details of emissions can be taken into account and these models provide statistical information, which is important for the estimation of the frequency of very high concentration values. But the essential advantage of Lagrangian models over other methods is the ability to handle reaction exactly,⁷ no averaged reaction rates have to be calculated. On the other side, these conventional Lagrangian models are suffered from the fact, that information about the surrounding flow is required as input for such models. This is related to problems to describe the interaction (the substance mixing) between particles which transport an emitted substance and particles of the ambient flow.

The development of advanced Lagrangian models requires both the description of substance transport and turbulent flow simulation, that means the motion (and change of properties) of all the fluid particles of the flow has to be simulated. This concept needs Lagrangian particle equations in accord with budget equations of turbulence. This statistical approach describes the one-point probability density function (PDF) of turbulent fluctuations (therefore such models are often called Lagrangian PDF methods), such that all the moments of this PDF are determined. By choosing suited coefficients of the Lagrangian equations, the budgets up to second-order determined by Lagrangian stochastic models can be made exactly consistent with Eulerian budget equations. Turbulence equations for buoyant flows can be also reflected, if a particle temperature is incorporated in the Lagrangian framework.⁸ This approach provides an important contribution to the design of Lagrangian models for complex flows, because they are derived in accord with the extensively studied empirical Eulerian turbulence equations. But even more important, this approach provides a new method for accurate simulations of mixing and reaction, where the latter process is described without any approximation.

The realizability of this concept and the performance of such a model are demonstrated here. At first, the main features of this approach as well as the applied model are presented in the next section. It is worth emphasizing that this concept is developed for inhomogeneous turbulent flow with shear and

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stratification, such that real atmospheric boundary layer processes can be described. Some results are shown on the simulation of developing turbulence as well as transport and mixing of two reactive scalars. This is in particular done, in order to demonstrate the differences in handling chemical reactions exactly (by the model presented here) and by usually applied approximations. The segregation parameter (the covariance of tracers normalized to their mean values) plays here an important role, which is calculated in dependence on the reaction rate. Finally, computational aspects of the realization of such simulations are discussed.

2 The model

The particle motion is considered in a two-dimensional area within a rectangular grid with non-uniform spacing in both coordinates x (the boundary layer length) and z (the boundary layer height). Particle inflow is through the west boundary, then the particles move in the x -direction over the surface (south boundary) and outflow is through the east boundary. The initial properties of the particles have to be given at the west boundary, that means e.g. the mean values and variances of their velocities, temperatures and frequencies. The same values are assumed for these quantities along the x -direction at the initial time.

The particles move from west to east, where reflection conditions at the upper (north) and lower (south) boundaries are chosen, such that the variances of the velocity field remain constant at these boundaries. The particles have a position \mathbf{x}_L and velocity \mathbf{U}_L (two-dimensional vectors, where the subscript L refers to a Lagrangian quantity), a (potential) particle temperature Θ_L and a composition vector Φ_L . The particle velocities and temperatures change then according to (i runs from from 1 to 2)

$$\frac{d}{dt} \mathbf{x}_L^i(t) = \mathbf{U}_L^i(t), \quad (1a)$$

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

$$\frac{d}{dt} \Theta_L(t) = \langle a_\theta \rangle + G_\theta^j (U_L^j - \langle U_E^j \rangle) + G_\theta (\Theta_L - \langle \Theta_E \rangle) + b_\theta \frac{dW}{dt}, \quad (1c)$$

where dW^j/dt is a Gaussian process with vanishing mean values $\langle dW^j/dt \rangle = 0$ and uncorrelated values at different times, $\langle dW^i/dt(t) \cdot dW^j/dt(t') \rangle = \delta_{ij} \delta(t - t')$. In these equations, δ_{ij} is the Kronecker delta, $\delta(t - t')$ is the delta function and $\langle \dots \rangle$ denotes the ensemble average. The Eulerian quantities (index E) are estimated at fixed positions, which are replaced in the Lagrangian equations by the actual particle position.

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The particles move in a way by these equations, such that Eulerian budget equations of turbulence are fulfilled for the mean values and variances of the velocity and temperature fields.⁸ These constraints determine the coefficients in the Lagrangian equations (1a-c). The drift terms $\langle a^i \rangle$ and $\langle a_\theta \rangle$ depends then upon the spatial gradients of the mean velocity, temperature and pressure fields, and all the other coefficients G_{ij}^j , G_θ^j , G_θ , b^{ij} , b_θ are estimated in terms of the mean Eulerian flow frequency ω . This quantity is calculated simultaneously with the solution of (1a-c) in accord with shear and stratification. All the details to these equations can be found elsewhere.⁸ The mixing of composition can be described by any model, where e.g. the simple IEM-model was applied for the simulations considered below.⁷ The initial particle properties have to be given as input information and the model provides then the mean values of the velocity, temperature and concentration fields as well as all the variances and covariances of these fields, that means in particular the calculation of concentration fluctuations is involved in these simulations.

3 Turbulence and turbulent dispersion of reactive substances

The performance of the model described in the previous section is illustrated now by results obtained for the development of initially isotropic turbulence and transport, mixing and reaction of two substances. These simulations are performed for a two-dimensional boundary layer by using a grid of 21 grid points in both directions corresponding with 100 m height and 400 m length. The spacing is equal in the z-direction (the height of the boundary layer), and the distance of x-grid points grows gradually from left (inflow) to right (outflow).

The development of initially isotropic turbulence is considered in a neutrally stratified homogeneous shear flow in order to demonstrate the performance of the model. The vertical shear of the mean horizontal velocity (U is simply written for $\langle U_E^1 \rangle$) at the west boundary is proposed as $dU / dz = 10 \text{ s}^{-1}$, and the turbulence is assumed to be isotropic at this inflow boundary. The initial frequency is set to be equal to the vertical shear dU / dz of the mean horizontal velocity. The simulation of the development of turbulence shows very similar features as observed in corresponding large-eddy simulations (LES).⁹ Two components of the tensor of anisotropy $A^{11} = \langle u^2 \rangle - 1/3$ and $A^{13} = \langle uw \rangle$ taken at the half boundary layer height are presented in Figure 1, where u and w are the fluctuations of the horizontal and vertical velocity, respectively. We see, the stationary values of these components are in a good agreement with the theoretical values of this model, which are also presented. The overprediction of the component A^{11} is related to the finite number of particles (see section 4) applied for these simulations.

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The transport, mixing and reaction of two tracers are then calculated in this flow field as an example for the simulation of reactive substance transport. This is done in accord with simulations and experiments of Komori et al.¹⁰⁻¹² They performed experiments in water tunnel with a square cross section and a turbulence generation grid installed at the entrance to the test section. Two reactants A and B were introduced into the device consisting of two sections separated by a thin plate. The plate was removed at the initial time, and the reactants are pushed through the test section. According to these experiments, the two reactants are released here (in concentrations of $10 \text{ g} / \text{m}^2$) as composition of particles in the upper and the lower half of the considered boundary layer. A second-order reaction between these compounds is assumed,

$$\frac{dA}{dt} = \frac{dB}{dt} = -\frac{dP}{dt} = -kAB, \quad (2)$$

where P gives the product of this reaction and k is the reaction rate.

The change of concentration C (normalized to the initial concentration) of that tracer released in the lower boundary layer is shown in Figure 2 along the mean boundary layer height for different values of k in $\text{m}^2 / (\text{gs})$. As it can be seen in this curve, the effect of reaction for k = 2 and 10 is found to be not very different due to the limiting effect of mixing. These results agree rather good with the experimental results of Komori et al. for reactive (with an infinitely high reaction rate) as well as for non-reactive flows,^{11, 12} which are also presented in Figure 2. This comparison confirms at least qualitatively the calculated results. A quantitative assessment is complicated by the fact, that the turbulence is produced differently. The measurements of Komori et al. were performed for decaying grid-turbulence without shear, whereas here homogeneous shear flow is considered. However, apart from the differences in the turbulence production, it seems that mixing and transport of tracers are described in a very similar way. The effect of different relatively high reaction rates is also found to be limited by the mixing of compounds under these circumstances in the model of Komori et al.¹⁰

The difference in the handling of nonlinear reaction rates by the approach presented here and by conventional methods is described in Figure 3. Closure of reaction terms is required in all the Eulerian models,¹³ which may cause serious errors for the mean non-linear reaction rates. Exactly this is demonstrated in Figure 3. The tracer transport is calculated in the same way as done before, where the instantaneous reaction rates in (2) are replaced now by the mean rates. We see, that this results in serious overpredictions of the reaction effect. This model predicts e.g. a disintegration of the tracer for k = 10, whereas the tracer concentration reaches a constant level in the simulations with instantaneous reaction rates as given in Figure 2.



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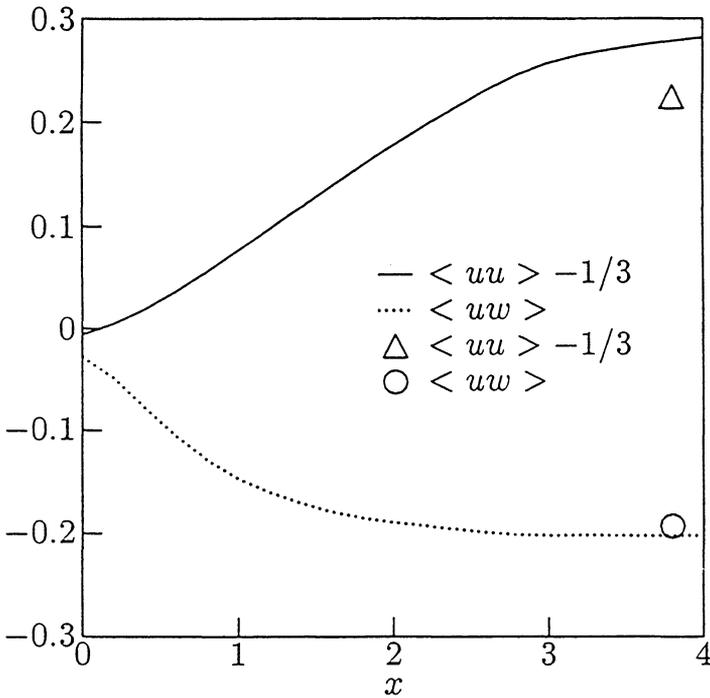


Figure 1. The development of initially isotropic turbulence is shown for two elements of the anisotropy tensor. The triangle and the circle depict the corresponding theoretical values of this model. The horizontal coordinate x gives the distance with an unit of 100 m.

Such mixing effects can be taken into account by the cross-correlation between fluctuating concentrations of A and B (normalized by the mean concentrations)

$$S = \frac{\langle (A - \langle A \rangle)(B - \langle B \rangle) \rangle}{\langle A \rangle \langle B \rangle} = \frac{\langle AB \rangle}{\langle A \rangle \langle B \rangle} - 1 \quad (3)$$

which is commonly expressed as the segregation or 'un-mixedness' parameter S . This quantity varies between -1 and zero and indicates the effect of mixing on the mean chemical reaction rate. This quantity is often neglected in studies of chemical processes in the atmosphere, which may lead to considerable errors. This is revealed by the transport equation for the mean concentration of the reactant A, which reads

$$\frac{\partial \langle A \rangle}{\partial t} + \nabla \cdot (\langle U_E \rangle \langle A \rangle) = \nabla \cdot (K \cdot \nabla \langle A \rangle) - k \langle A \rangle \langle B \rangle (1 + S), \quad (4)$$

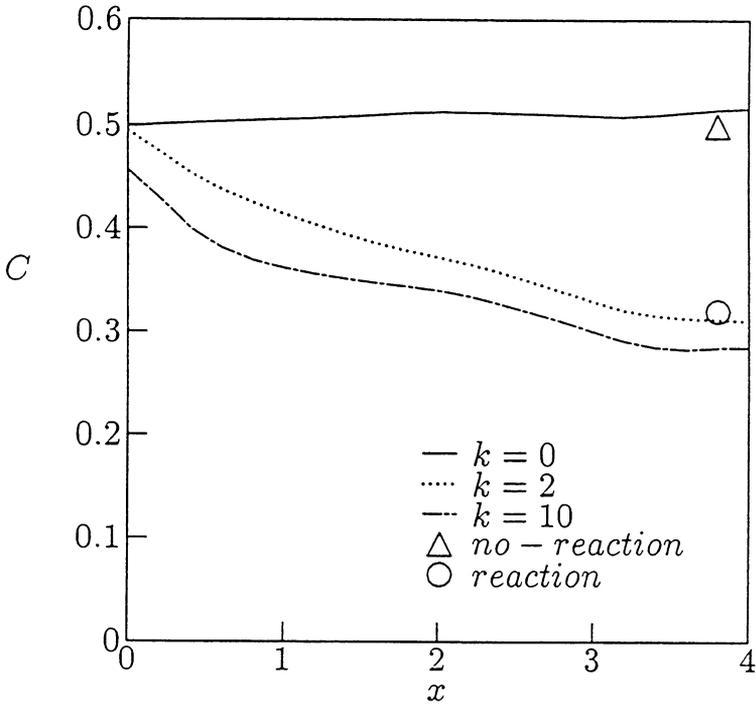


Figure 2. The concentration (normalized to the initial value and calculated at the middle boundary layer height) of the reactant released in the lower half of the boundary layer. The values of k represent different values for the reaction rate (in $\text{m}^2 / (\text{g s})$). The triangle and the circle show measured stationary values for this concentration without and with reaction, respectively.^{11, 12}

where K is the eddy diffusivity tensor and ∇ is the Laplace operator. It may be seen, that the neglect of values of S near -1 leads to a remarkable overestimate of reaction as it can be observed in Figure 3.

The segregation parameter for the tracer transport simulations with instantaneous reaction rates is shown in Figure 4. These calculations were performed with vanishing initial cross-correlation, such that S is near zero (the deviation arises from the averaging procedure) at the inflow region. The effect of reaction does correspond with the experimental result of Komori et al.,^{11, 12} that means this parameter becomes smaller due to the decay of A and B . However, there are quantitatively differences in the results. Without reaction, stationary values of about $S = -0.5$ were found by Komori et al., and with reaction they found $S = -1$ for an infinitely high reaction rate. These differences may be related to the differences in the turbulence production.



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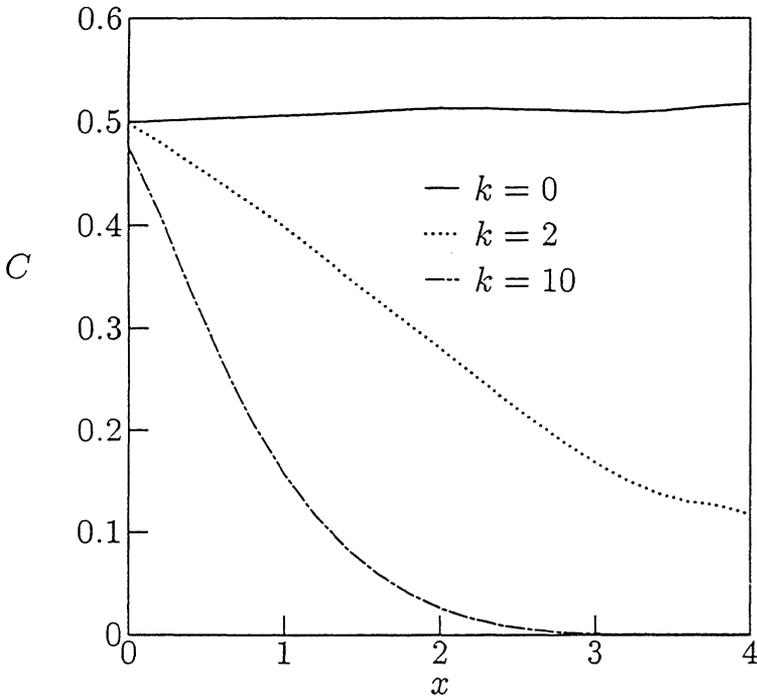


Figure 3. The concentration C calculated as in Figure 2, but here with mean concentration values in the reaction term instead of the instantaneous ones.

4 Computational aspects

The simulation of the motion and change of properties of all the particles of a turbulent reactive flow seems to increase the computational effort compared with conventional methods. However, this is not found for these simulations. Additionally, the presented concept benefits from the ability of Lagrangian models to scale nearly linearly on multiprocessor computers. The program may run independently at each processor apart from the calculation of mean values, which are obtained as sum over all the processor results. However, this amount of communication between processors is relatively small and accounts only a few percent of the simulation time.

These simulations were performed at the Cray T3E of Delft University of Technology. The calculation of the motion of about 100000 particles at each processor requires with 8 processors approximately one hour, if 200 time steps are performed.

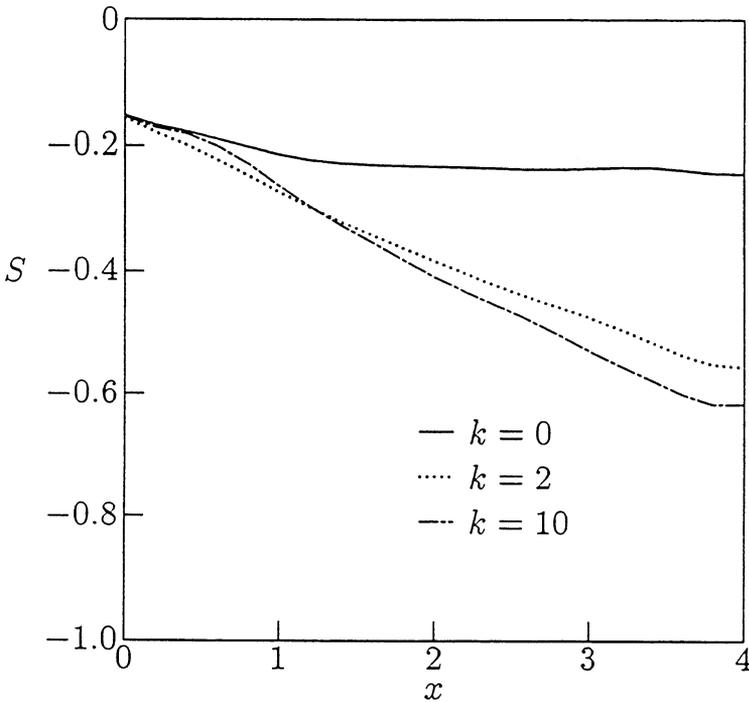


Figure 4. The segregation parameter for the simulations of tracer transport shown in Figure 2. The values of k represent different values for the reaction rate (in $\text{m}^2/(\text{g s})$) and the unit of the distance x is 100 m.

5 Concluding remarks

It is demonstrated, that the here presented concept for the simulation of turbulence, mixing and reaction of substances offers new chances for the development of accurate and efficient simulation models for these processes in the atmospheric boundary layer. This approach can be applied under real conditions, that means shear and stratification are taken into account. The considered examples provide results for the turbulence in accord with LES. The simulation of tracer transport and reaction agrees well with experimental results as demonstrated with respect to the data of Komori et al. In particular it is illustrated, that the consideration of instantaneous reaction rates may be related to a remarkable gain in the accuracy of results. Further investigations on mixing and reaction in stratified flows are in preparation.



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