In this chapter we give a short overview of the Eulerian techniques for modelling turbulent flows, transport and chemical reactions. We first present the basic Eulerian equations describing the velocity and concentration changes in incompressible flows. Next, we discuss the computational problems which arise while solving these basic equations numerically. Different models have been developed to overcome these difficulties, the time and volume averaging techniques are summarised in Sections 2.3 and 2.4. We end this chapter with a discussion of the drawbacks of the Eulerian approach for modelling turbulent transport in reactive turbulent flows.

2.1 Basic equations

Let us consider the reactive species $A$ and $B$ released in a turbulent flow. Species $A$ and $B$ react to form $C$ according to the isothermal, irreversible chemical reaction $A + B \rightarrow C$. The concentration of each chemical compound $\alpha$ (\(\alpha = A, B, C\)) denoted by $c_{\alpha}$ must, at each time, satisfy the material mass balance taken over a volume element. This means that the concentration of species $A$ must satisfy the equation

$$\frac{\partial c_A}{\partial t} + \nabla \cdot (u c_A) = \nabla \cdot (D \nabla c_A) - k c_A c_B + S_{A}(\mathbf{x}, t).$$

(2.1)

The advective flow velocity is represented by $u$, where $u_i$ is the component in $x_i$ direction (\(i = 1, 2, 3\)) and $D$ describes molecular diffusion processes and is often given by a single diffusion constant. The second term on the right-hand side represents the chemistry process and $S_{A}$ denotes the source of species $A$ at location $\mathbf{x} = (x, y, z)$ and time $t$. Similar equations hold for the concentrations of species $B$ and $C$.

We assume that the irreversible chemical reaction does not have any effect on the turbulent flow itself and neither does the presence of pollutants (the concentrations are dilute). This implies that the reaction rate and the molecular diffusivities are constant. Furthermore, we assume that the flow is incompressible, thus
the following scalar equation holds

$$\nabla \cdot \mathbf{u} = 0$$  \hspace{1cm} (2.2)

The flow velocity $\mathbf{u}$ satisfies the incompressible Navier-Stokes equations, which read

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\frac{1}{\rho} \nabla p + \nu \nabla^2 \mathbf{u}$$  \hspace{1cm} (2.3)

where $\rho$ is the constant density, $p$ denotes the pressure and $\nu$ is the constant kinematic viscosity. The values of $\rho$ and $\nu$ are prescribed by the flow. The seven equations (2.1)–(2.3) form a complete set in the unknowns $c_\alpha$ ($\alpha = A, B, C$), $p$ and $\mathbf{u}$ and can in principle be solved with suitable initial- and boundary conditions. We refer to (Garratt, 1992; McComb, 1990) for a more detailed presentation of the equations of motion.

2.2 Direct numerical simulation

In Direct Numerical Simulations (DNS), the incompressible Navier-Stokes and the transport-reaction equations are solved numerically using for instance pseudo-spectral techniques (Riley et al., 1986; Timmermans and van de Vosse, 1993), finite-difference (Vreugdenhil, 1993) or finite-element (Segal, 1993) methods.

A turbulent flow consists of many superposed eddies of differing sizes. The smallest eddies are in the order of the Kolmogorov length scale ($\approx 1$ mm) and the largest ones, in the atmospheric boundary layer may be as large as 1 km. The corresponding Reynolds number in the atmosphere is in the order of $10^7$. If we want to resolve all scales of motion down to the smallest eddies in three dimensional atmospheric turbulence, we need in each spatial direction at least $10^6$ grid-points (Garratt, 1992). Thus in the order of $10^{18}$ grid-points are needed to make a simulation of the atmospheric boundary layer. Storing one variable for so many points requires about $10^{18}$ bytes. Even with todays parallel computers it is impossible to store such a huge amount of data.

Direct numerical simulation of the Navier-Stokes equations is computationally only feasible for moderate Reynolds numbers, in the order of $10^2$ (Riley et al., 1986; Wissink, 1995). Alternatively, less computationally demanding models have been developed. These models are based on filtering techniques and fall into two categories; the ones based on time averaging called the Reynolds Average Navier-Stokes (RANS) models and those based on volume averaging; the Large Eddy Simulations (LES).
2.3 Reynolds averaged simulation

In Reynolds averaged models all variables are decomposed into a mean and a fluctuation. For instance the Reynolds decomposition of the velocity $u$ is represented by $u = \overline{u} + u'$, where $\overline{u}$ represents the ensemble averaged component or mean velocity. The vector $u'$ denotes the fluctuation and is often called the stochastic or turbulent component (Panofsky and Dutton, 1984, p.33 and 34). Like $u$, the concentration can be decomposed in a mean and a fluctuating part. Substituting these Reynolds decompositions into the transport-reaction equation (2.1) of the concentration of constituent $A$ and averaging leads to

$$\frac{\partial \overline{c_A}}{\partial t} + \nabla \cdot (\overline{u} \overline{c_A}) = - \nabla \cdot (u' \overline{c_A}) + \nabla \cdot (D \nabla \overline{c_A}) - k (\overline{c_A} \overline{c_B} + \overline{c_A} \overline{c_C}) + S_A(x,t).$$

Note that $\overline{c_A} = 0$. Similar Reynolds averaged equations can be derived for the concentrations of $B$ and $C$ and the Navier-Stokes equations. The equations contain terms like the mean $\overline{c_A}$ but also terms like $u' \overline{c_A}$ and $\overline{c_A} \overline{c_B}$ which cannot be rewritten in terms of the mean variables $\overline{u}, \overline{c_A}$ or $\overline{c_B}$, and do not vanish. The resulting system of equations has more unknowns than equations; hence no solution for the mean concentrations and the mean velocity can be given unless additional assumptions are made. This problem is called the turbulence closure problem.

2.3.1 First order closure models

In mixing-length theory, often called K-theory, the following assumptions are made to close the Reynolds averaged equations (Seinfeld, 1975). First, we assume that there is a $3 \times 3$ symmetric matrix $\mathcal{K}$ called the eddy-diffusivity tensor such that

$$(u' c'_\alpha)_j = - \sum_{k=1}^{3} \mathcal{K}_{jk} \frac{\partial \overline{c_\alpha}}{\partial x_k}$$

for $j = 1, 2, 3$ and $\alpha = A, B, C$. If the coordinate axes coincide with the principal axes of the eddy diffusion then $\mathcal{K}$ is diagonal with elements

$$(u' c'_\alpha)_j = -\mathcal{K}_{jj} \frac{\partial \overline{c_\alpha}}{\partial x_j}$$

for $j = 1, 2, 3$. In the atmosphere the eddy diffusivities are typically in the order of $0.1-100 \ m^2 / s$ (Chock and Winkler, 1994a). Secondly, we assume that molecular diffusion is negligible compared to turbulent transport, thus

$$\nabla \cdot (D \nabla \overline{c_A}) << \nabla \cdot (u' c'_A).$$
Furthermore in most practical applications, the effect of concentration fluctuations on the rate of reaction is neglected:

\[ k \left( \overline{c_A c_B} + \overline{c_A'} \overline{c_B'} \right) \approx k \overline{c_A c_B}, \]

assuming that the concentration fluctuations \( \overline{c_A'} \overline{c_B'} \) are negligible compared to the mean product \( \overline{c_A c_B} \).

These assumptions lead to a closed advection-diffusion-reaction equation

\[ \frac{\partial \overline{c_A}}{\partial t} + \nabla \cdot (\overline{u} \overline{c_A}) = \nabla \cdot \left( \kappa \nabla \overline{c_A} \right) - k \overline{c_A c_B} + S_A(x, t). \]

For the concentrations of \( B \) and \( C \) similar equations hold.

The Reynolds averaged Navier-Stokes equations read

\[ \frac{\partial \overline{u}}{\partial t} + \overline{u} \cdot \nabla \overline{u} = -\frac{1}{\rho} \nabla \overline{p} + \nabla \cdot \left( \nu \nabla \overline{u} + \tau^R \right) \]

where \( \overline{p} = \rho \) and \( \rho' = 0 \) since the flow is incompressible. The elements of the Reynolds stress tensor \( \tau^R \) read \( \tau^R_{ij} = \overline{u_i' u_j'}. \) The averaged Navier-Stokes equations are closed by parametrisation of the Reynolds stresses. For instance in the K-\( \epsilon \) models (Wissink, 1995) the Reynolds stresses are approximated by

\[ \tau^R_{ij} = -\frac{2}{3} K \delta_{ij} + \nu_T \left( \frac{\partial \overline{u}_i}{\partial x_j} + \frac{\partial \overline{u}_j}{\partial x_i} \right) \]

where \( K \) is the turbulent kinetic energy and \( \nu_T \) represents an eddy viscosity.

### 2.3.2 Second order closure models

In first order closure models the contribution of fluxes, variances and covariances are parameterised. In second order closure models one formulates conservation equations for these second moments. The rate of change in time of \( \overline{u_i' c_A'} \), for example, is expressed in terms of the mean production, the buoyant production, the turbulent transport, the pressure covariance and the chemistry. We may formulate differential equations for all second moments. There are many different techniques to solve these second order equations by parameterisation of the third moments. For more information on second order closures we refer to (Deardorff, 1978; Garratt, 1992; Verver et al., 1997). The system of equations which model the process has become much larger and more complex. Solving such systems numerically requires considerably more computer power than the first order closure models.
2.4 Large eddy simulation

Like in RANS models, in large eddy simulations, the velocity field is not completely resolved. In these models a spatial filter is used to separate the large and the small-scales of turbulence. If we denote the large-scale components by a hat-symbol, the transport-reaction equation for the concentration of species \( A \) for example can be written as

\[
\frac{\partial \hat{c}_A}{\partial t} + \nabla \cdot (\hat{\mathbf{u}} \hat{c}_A) = \nabla \cdot (D \nabla \hat{c}_A) - k \hat{c}_A \hat{c}_B + S_A(\mathbf{x}, t) - \nabla \cdot (\hat{\mathbf{u}} \hat{c}_A) - k(\hat{c}_A \hat{c}_B - \hat{c}_A \hat{c}_B)
\]  

(2.4)

The filtered mean velocity is obtained from averaging over space (Wissink, 1995),

\[ \hat{u}_i = \int_{\mathcal{V}} u_i(x') \phi(x - x') dx' \]

where \( \mathcal{V} \) denotes the total flow area and \( \phi \) is a filter function (for example a Gaussian kernel). The filtered Navier-Stokes equations read

\[
\frac{\partial \hat{\mathbf{u}}}{\partial t} + \hat{\mathbf{u}} \cdot \nabla \hat{\mathbf{u}} = -\frac{1}{\rho} \nabla \hat{p} + \nabla \cdot (\nu \nabla \hat{\mathbf{u}} + \tau^S)
\]

where \( \tau^S \) denotes the subgrid stresses \( \tau_{ij}^S = -(\hat{u}_i \hat{u}_j - \hat{u}_i \hat{u}_j) \).

In the derivation of equation (2.4) and the Navier-Stokes equations, we assumed that in high Reynolds-number flows the small scales of turbulence are nearly isotropic and independent of the geometry. Only the equations comprising the large scales are solved. The small-scale components or subgrid stresses are parametrised. The last two terms on the right hand-side of equation (2.4) (shown on a separate line) describe the effect of small scales on the advection and chemistry of the system. In large eddy simulations the subgrid stresses are parameterised by subgrid-scale models. In the last few years, a lot of progress has been made in developing good subgrid-scale models. We name the Smagorinsky model (Germano, 1992) and a model closely related to Reynolds averaging (Beets and de Arellano, 1996) where a subgrid scale eddy diffusivity \( D_t \) is introduced

\[ \hat{\mathbf{u}} \nabla \hat{c}_A - \hat{\mathbf{u}} \hat{c}_A = -D_t \nabla \hat{c}_A. \]

A general overview of subgrid scale approximations can be found in (Ferziger, 1993; Mason, 1994). The computational and memory requirements of large eddy simulations are still so high that only recently high performance computers became powerful enough to simulate simple turbulent flows using large eddy techniques (Schumann, 1989).

Like in Reynolds average methods, in large eddy simulations the chemistry part of the equations is often modelled by neglecting the contribution of the term
In (Beets and de Arellano, 1996; Beets et al., 1996) techniques are proposed to approximate the transport equations for the concentration fluctuations, \( c_{AB} \). The results from these improved models demonstrate that neglecting the contribution of \( c_{AB} \) may lead to large errors in the concentration predictions in atmospheric applications.

### 2.5 Limitations of the Eulerian methods

The Eulerian approach for modelling turbulent transport and chemistry is perhaps the best known and most used technique to describe the spread of species in turbulent flows. The Reynolds averaged simulations can be readily solved numerically and give satisfactory results for the long range transport. Only DNS is capable to solve the full range of length scales, but they are only computationally feasible for small Reynolds numbers. The filtering methods, RANS and LES approximate the small spatial or temporal scales. They assume that the spatial and temporal variations in the source and velocity scales are large with respect to the parametrised scales (Seinfeld, 1975). Therefore these average models do not correctly describe the turbulent transport of species released from point sources. Furthermore, the differential equations are numerically solved by discretisation in space, such that a point source is spread out over a grid-cell. In (Deardorff, 1978) it is also shown that higher order Reynolds average simulations are inherently incapable of predicting the turbulent transport of multiple plume sources. For small times, the Lagrangian statistical theory is more suitable for describing the spread of a plume.

The averaging techniques RANS and LES make parametrisation of the small-time scales and the subgrid-scales, hence no information on the internal structure of the turbulent flow is available. The problem of first order closure models lies in the determination of the eddy diffusivity. The approximations are made empirically and do not lead to robust models which give correct solutions for all turbulent flows. In LES the prescription of boundary conditions has a significant influence on the solution. See (Garratt, 1992) for a more detailed discussion of the advantages and drawbacks of RANS and LES for modelling turbulent transport.

We have seen that the first order models are restricted to the mean (averaged over time or space) concentrations and model the chemistry process by the product of the mean concentrations. However, the rate at which two species react is affected by the rate at which the turbulent transport mixes them together and hence depends on the concentration fluctuations. From experiments (Komori and Ueda, 1984; Saetran et al., 1989) we have to conclude that the fluctuations around the mean may be as large as the mean concentration itself. Accordingly, first order models do not correctly describe the chemical processes in turbulent flows because they neglect the concentration fluctuations.

In the next chapters, we investigate the feasibility of using Lagrangian statistical models to describe the turbulent transport of passive tracers and reactive constituents.