Population Balance Modelling in Turbulent Reacting Flows

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Reacting flows with particle formation

Several problems involve formation of particles via a physical or chemical process taking place in a flow:

- Soot formation
- Nanoparticle production
- Aerosol condensation
- Precipitation

Challenges in modelling particle formation in turbulent flows

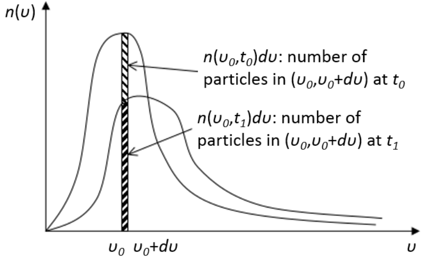
Apart from the known problems in turbulent reacting flows (chemical kinetics, turbulence, turbulence-chemistry interaction) the following issues appear in turbulent flows with particle formation:

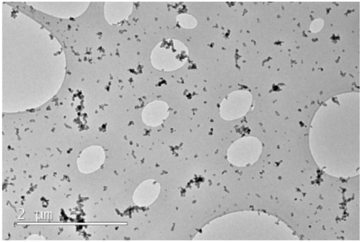
- Aerosol dynamics
    - Nucleation
    - Surface processes (growth or shrinkage)
    - Inter-particle processes (coagulation, aggregation)
    - Breakage
- Aerosol-turbulence interaction
    - Unknown correlations arising from non-linear interactions between particles, species and temperature

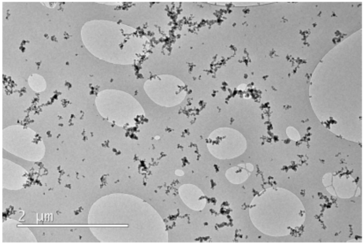
The particle size distribution

Importance of particle size distribution:

- Growth and oxidation rates depend on surface area
- Aggregation rate can be size-dependent
- Prediction of the PSD is increasingly important for new regualtions







Population Balance Equation (PBE)

Define the continuous particle size distribution as density $n(v)$, i.e. the number of particles of size v per unit size.

Let Y_α be the concentrations of chemical species.

Continuous PBE (nucleation, growth and aggregation)

$$\frac{\partial n(v,t)}{\partial t} + \frac{\partial}{\partial v}(G(v,Y_\alpha)n(v,t)) = B(Y_\alpha)\delta(v-v_0)$$
$$+\frac{1}{2}\int_0^v \beta(v-w,w)n(v-w,t)n(w,t)dw - \int_0^\infty \beta(v,w)n(v,t)n(w,t)dw$$

Physical and chemical processes included in the PBE above:

- Particle formation (nucleation)
- Continuous size change (growth, oxidation)
- Coagulation/aggregation

Solution of the PBE

- Complex integro-differential equation
- Methods for solution:
  - Analytical and similarity solutions
  - Monte Carlo methods
  - Method of moments and variants
  - Discretisation methods
- Discretisation methods do not require closure assumptions and predict the PSD
- Main challenges in discretisation methods:
  - Distribution can vary over several orders of magnitude, nucleation is localised
  - Coagulation is an integral term
  - Conservation of moments in coagulation
  - Growth/oxidation term is first-order derivative - possibility of sharp fronts

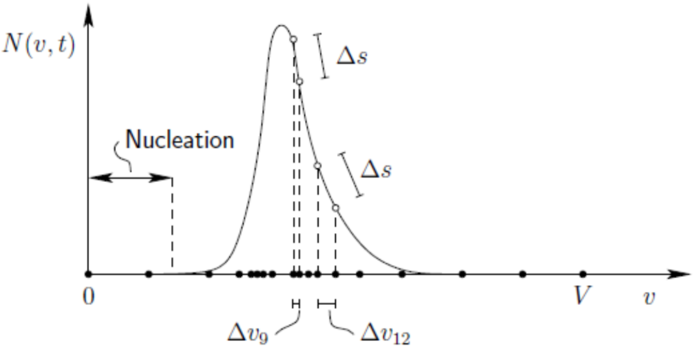
Adaptive grid

We introduce a *coordinate transformation*

$$(\tau, t) \mapsto v = \bar{v}(\tau, t)$$

Kinematic constraints:

- Avoid node depletion
- Control grid stretching

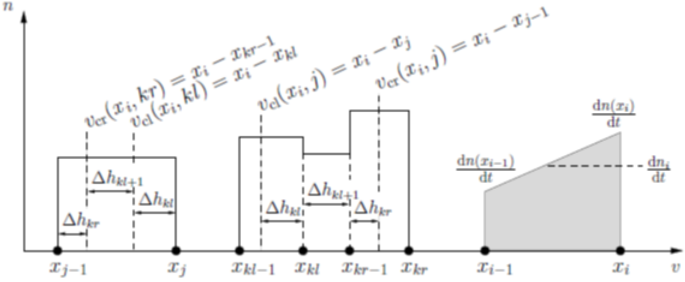


Conservative finite volume method for coagulation

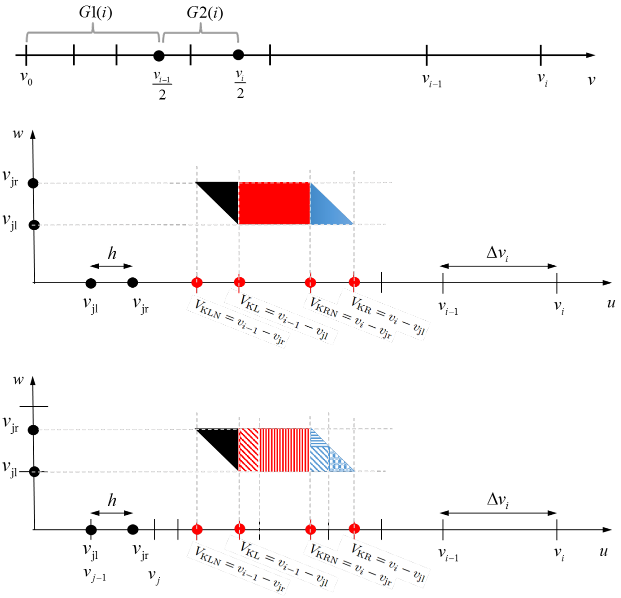
Finite volume formulation for the coagulation terms:

$$\frac{dn_i}{dt} = \frac{1}{\delta v_i}\left(\int_{v_{i-1}}^{v_i} dv \int_{v_{nuc}}^{\frac{v}{2}} \beta(v-w,w)n(v-w)n(w)dw - \int_{v_{i-1}}^{v_i} n(v)dv \int_{v_{nuc}}^{v_{max}} \beta(v,w)n(w)dw\right)$$

Error in conservation of the moments:



Geometric approach for performing the double integration while conserving the first moment:



Spatially distributed PBE

The PBE must be augmented to account for:

- Convective transport in physical space
- Thermophoresis - u_i^t, depending on T, μ and $\frac{dT}{dx_i}$
- Particle diffusion (much smaller than species' diffusion)

Spatially distributed continuous PBE

$$
\begin{aligned}
\frac{\partial n(\upsilon, x_i, t)}{\partial t} + \frac{\partial\left[(u_i + u_i^t) n(\upsilon, x_i, t)\right]}{\partial x_i} + \frac{\partial (G(\upsilon, Y_\alpha(x_i, t)) n(\upsilon, x_i, t))}{\partial \upsilon} \\
= B(Y_\alpha(x_i, t)) \delta(\upsilon - \upsilon_0) + \frac{\partial}{\partial x_i}\left(D_p \frac{\partial n(\upsilon, x_i, t)}{\partial x_i}\right) \\
+ \frac{1}{2}\int_0^\upsilon \beta(\upsilon - w, w) n(\upsilon - w, x_i, t) n(w, x_i, t) dw \\
- \int_0^\infty \beta(\upsilon, w) n(\upsilon, x_i, t) n(w, x_i, t) dw
\end{aligned}
$$

Closure problem for particle formation in turbulent flow

In a RANS or LES simulation of a turbulent flow with particle formation, unclosed terms result from the following:

- Type 1 Correlation between reactive scalars (nucleation and growth rates)
- Type 2 Correlation between reactive scalars and number density (growth)
- Type 3 Correlation between number densities (aggregation)

To obtain closure for the turbulent PBE, we develop the Population Balance - Probability Density Function (PBE-PDF) approach.

Consider the joint pdf of reactive scalars and number densities at different sizes, as obtained by a discretised PBE:

$$Y_\alpha(\mathbf{x},t), N_i(\mathbf{x},t) \rightarrow f(\mathbf{y},\mathbf{n};\mathbf{x},t)$$

The PBE-PDF equation

- A transport equation for the evolution of the pdf can be derived
- Terms arising from turbulence-aerosol dynamics interaction appear in closed form

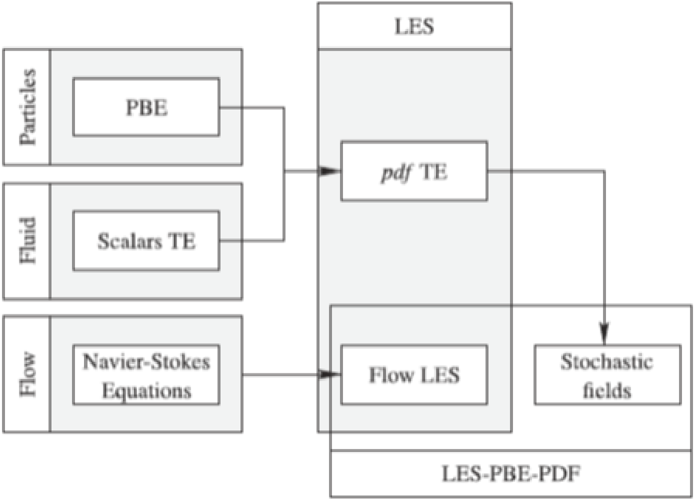
PBE-PDF equation

$$
\begin{aligned}
&\overline{\rho}\frac{\partial \tilde{f}}{\partial t} + \overline{\rho}\tilde{u}_j \frac{\partial \tilde{f}}{\partial x_j} + \frac{\partial}{\partial x_j}\left(\overline{\rho}\langle u_j'' | \mathbf{y}, \mathbf{n}\rangle \tilde{f}\right) \\
&\qquad = -\frac{\partial}{\partial y_\alpha}\left(\left\langle \overline{\rho} D_\alpha \frac{\partial^2 y_\alpha}{\partial x_i^2} \middle| \mathbf{y}, \mathbf{n} \right\rangle \tilde{f}\right) - \frac{\partial}{\partial y_\alpha}(\overline{\rho}\dot{\omega}_\alpha(\mathbf{y})\tilde{f}) \\
&+ \frac{\partial}{\partial n_i}(\overline{\rho} G_i(\mathbf{y}, \mathbf{n})\tilde{f}) - \frac{\partial}{\partial n_0}(\overline{\rho} B(\mathbf{y})\tilde{f}) - \frac{\partial}{\partial n_i}(\overline{\rho} A_{b,i}(\mathbf{n})\tilde{f}) + \frac{\partial}{\partial n_i}(\overline{\rho} A_{d,i}(\mathbf{n})\tilde{f})
\end{aligned}
$$

The PBE-PDF method in LES

- The PBE-PDF method can be extended to LES by defining a subgrid joint scalar-number density pdf
  - Can be interpreted as the probability of having a certain composition and PSD at the subgrid scales
- An LES-PBE-PDF equation can be derived for the subgrid pdf
  - Closed form for the reaction and PBE terms
  - Modelling is needed for turbulent transport and micromixing, but approach is less sensitive to the models
- LES-stochastic field
  - LES used to obtain the velocity field
  - Ensemble of fields represents the PBE-PDF

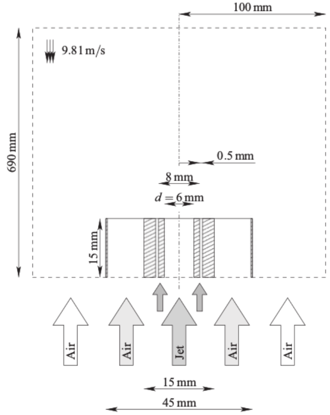
Structure of LES-PBE-PDF code



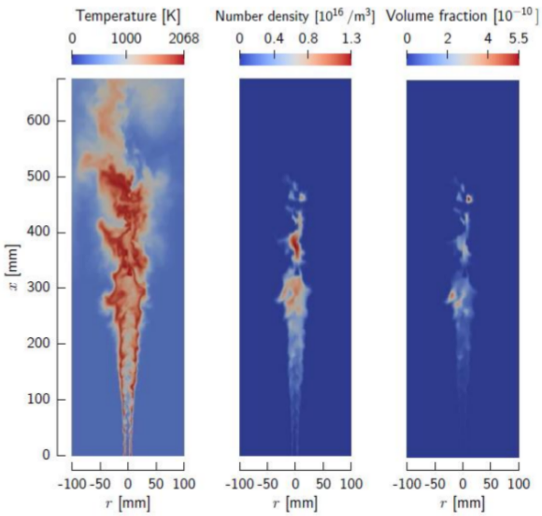
Case study: LES-PBE-PDF simulation of Delft flame III

(Sewerin and Rigopoulos, Combust. Flame 2018, vol. 189, pp. 62-76)

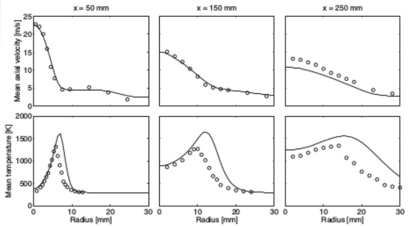
- Turbulent non-premixed natural gas ($CH_4 + N_2$) - air flame
- Gas-phase chemistry: GRI 1.2
- PBE discretisation: explicit adaptive grid (30 nodes)
- Turbulent-aerosol dynamics interaction: PBE-PDF
- LES grid: 672x70x36
- Stochastic field method (8 fields)
- CFD code: BOFFIN



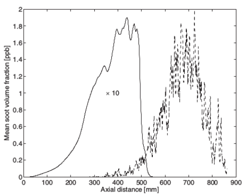
Experimental setup



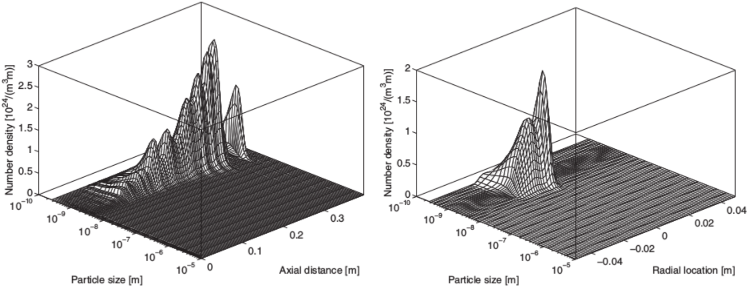
Soot volume fraction radial profiles at 15 mm and 50 mm



Velocity and temperature radial profiles



Axial profile of soot volume fraction



Evolution of PSD

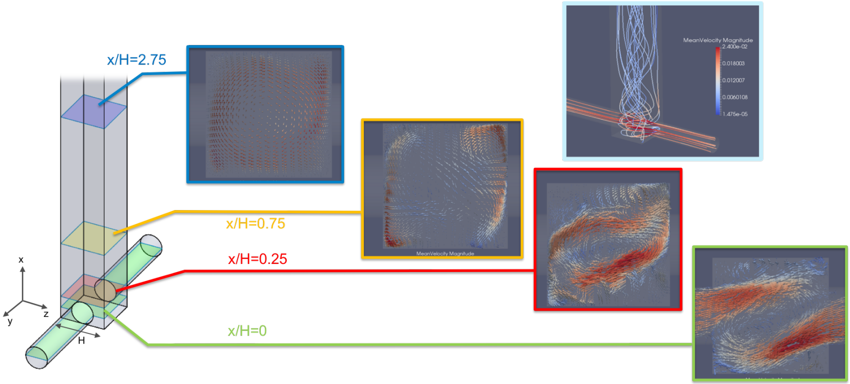
Average runtimes for advancing the LES-PBE-PDF model by one time step ($\Delta t = 1.2 \cdot 10^{-6}$ s) on 4 nodes (96 MPI processes) of a Cray XC30 Supercomputer (ARCHER UK).

Physical process	**Average runtime (s)**
Scalar convection/diffusion	6.89
Mixing	1.017
Gas-phase chemistry	10.85
PBE	9.11
Flow field	1.94
All processes	33.54

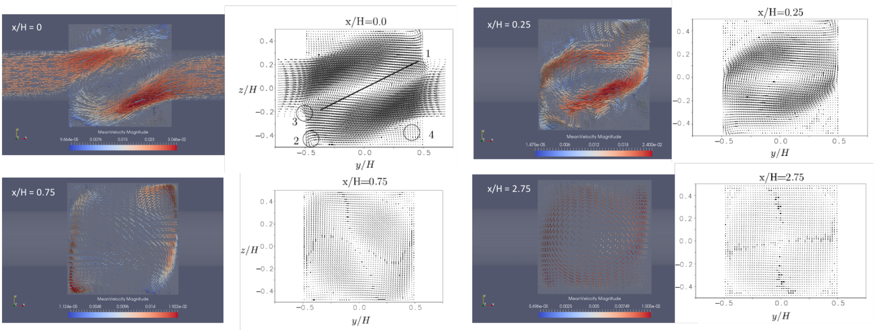
Hence the discretised PBE occupies only a modest proportion of the CPU time per time step.

Precipitation in liquid flows with PBE and DNS

- Precipitation of $BaSO_4$ in a T-mixer
- Preliminary DNS results
- Currently coupling PBE and DNS (code PANTARHEI)



Comparison with PIV results (F. Schwertfirm, J. Gradl, H. Schwarzer, W. Peukert and M. Manhart, Int. J. Heat Fluid Flow 2007, vol. 28, pp. 1429-1442)



Conclusions

- Methods have been developed for efficient and accurate solution of the PBE
- The PBE-PDF method provides closure to the unclosed terms in the turbulent PBE in RANS or LES
- The stochastic field method allows numerical solution of the LES-PBE-PDF
- The extra CPU time for the PBE is affordable
- Future work:
    - Investigation of PBE-PDF with DNS
    - Turbulent reacting flows with particle formation in the liquid phase (e.g. precipitation)