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MMC-LES modelling of droplet nucleation and growth in turbulent jets

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HIGHLIGHTS

- Sparse-Lagrangian particle method for particle nucleation and growth.
- Sparse particle and standard (dense) particle methods yield near to identical results.
- New particle method captures the correct trends when varying the precursor concentration.
- The interactions between turbulence and nucleation can modify the averaged nucleation rates by up to 250%.

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ABSTRACT

An Eulerian large-eddy simulation (LES) is coupled with a sparse-Lagrangian particle method to solve the population balance equation for aersol nucleation and growth in turbulent flows. We use the LES for the solution of the filtered velocity and mixing fields while the particle method provides one-point statistics of the gaseous species and the condensed phase such that the non-linear aerosol nucleation and growth terms appear naturally in closed form. A sparse particle implementation requires additional localisation in a reference space, and this localisation is realised here by employing the generalised multiple mapping conditioning (MMC) mixing model. The complete model, called MMC-LES, is validated by comparison with experimental data from nucleation studies in a turbulent, hot, nitrogen jet laden with dibutylphthalate (DBP) that condenses during mixing with a coflow of cold air. Acceptable agreement is found between the MMC-LES predictions and the experimental data. The average DBP droplet sizes are well predicted, and predictions of the total droplet number and the dependencies of droplet statistics on precursor concentrations are satisfactory. A comparison of the sparse particle method with results from conventional (dense) Monte Carlo-LES simulations demonstrates the capabilities of MMC-LES to predict aerosol nucleation and growth at relatively low computational cost. An additional quantitative analysis of the interactions between the turbulence and the non-linear nucleation source and growth terms shows that sub-grid effects must not be neglected and interactions between turbulence and nucleation can modify averaged nucleation rates by more than 250%.

1. Introduction

The formation of aerosols or particulate matter from the gas phase is widespread in both nature and industry. Examples include condensation leading to the formation of raindrops, the formation of soot and flame synthesis of industrial commodities such as carbon black, titania, silica and specialised metals and metal oxides. Existing experimental databases provide some understanding of the physics of aerosol dynamics and their resulting characteristics (Hu et al., 2003; Echavarria et al., 2011; Gupta et al., 2011), but do not usually provide all necessary correlations between hydrodynamic and thermodynamic quantities needed for model development. This is particularly true for small scale turbulence effects on precursor chemistry, droplet nucleation and growth. As a consequence, although there has been some success in modelling the evolution of aerosol particle size distributions (PSD) in laminar flame reactors (Echavarria et al., 2011; Tsantilis et al., 2002; Dang and Swihart, 2009), turbulent flame synthesis modelling efforts have been less successful.

The spatio-temporal evolution of the PSD is governed by the population balance equation (PBE). In devising methods to solve the PBE, we distinguish between sectional methods (Friedlander, 2000), moment methods (Pratsinis, 1988) and direct Monte Carlo

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methods (Kruis et al., 2000, 2012). Moment methods, of which there are many varieties, mathematically reduce the PSD to its moments. Typically only the first two or three moments are required leading to a method with low computational cost. In sectional methods the PSD is discretized into sections representing discrete particulate sizes. Implementation may be conceptually quite simple as the governing equations for gaseous species and particulates are solved in the same way and the transition between the phases is uncomplicated. Direct Monte Carlo methods use a large number of stochastic particles to reconstruct the statistics of the aerosol interactions including direct modelling of collisions and agglomeration. The reader is referred to Rigopoulos (2010) for a comprehensive review of the different flavours of PBE methods in reactive flows.

Independent of the PBE solution method, some specific closure problems arise in turbulent environments and convincing (and universal) modelling strategies for accurate predictions are not yet well established. Temporally and/or spatially averaged conservation equations can be derived, but the key challenge for turbulent flows is the closure of the averaged nucleation and growth terms due to their highly non-linear dependence on instantaneous and local thermodynamic properties such as supersaturation and surface tension. It is acknowledged that simplified approaches which neglect turbulence-chemistry-aerosol interactions (e.g. the *perfectly stirred reactor* model) may suffice for some flow conditions (Bhatt and Lindstedt, 2009; Xiong and Pratsinis, 1991). However, most applications of practical interest feature distinctly inhomogeneous conditions, such as turbulent jets, and modelling of the large and small (i.e. subgrid) scale inhomogeneities is of paramount importance for accurate predictions.

In the context of large eddy simulations (LES) of the turbulent flow field, the most common methods for the solution of the PSD are the different flavours of the method of moments (Marchisio and Fox, 2013). When the moment method for the PSD is combined with flamelet models for the gaseous reactive species, predictions of soot formation are good (Mueller and Pitsch, 2013; Chittipotula et al., 2011), albeit uncertainties with respect to the parameterisation of the flamelets for both kinetically fast gaseous species and kinetically slow aerosol species remain. So far, only a few flame synthesis studies have combined LES (Loeffler et al., 2011; Garrick and Wang, 2011) with the sectional method. These have all neglected turbulence interactions with aerosol nucleation and growth despite clear evidence from direct numerical simulation (DNS) studies (Das and Garrick, 2010) that such contributions are typically not small. The synthesis of aerosols in turbulent flames involves coupled, three-way interactions between turbulence, chemistry and aerosol dynamics. It is convenient in fundamental studies to eliminate one of these aspects allowing for the other interactions to be analysed in isolation. As the present paper focuses on model closures for turbulence-aerosol interactions non-reacting systems with a condensating species present themselves as ideal test cases. Droplet nucleation and growth of dibutylphthalate (DBP) in a turbulent mixing layer constitutes such a case, and the recent DNS by Zhou et al. (2014) confirms the value of investigating turbulence-aerosol interactions in isolation. Two key observations were made: (1) instantaneous droplet number density strongly correlates with the instantaneous gaseous DBP mass fraction; and (2) number density does not show a unique dependence on the mixture fraction due to aerosol transport in mixture fraction space. From the first observation we conclude that turbulent correlations between nucleation and gaseous species concentrations cannot be neglected when modelling the nucleation rate. From the second observation we conclude that particulate evolution cannot be solely based on the state of the mean mixing field.

From the above discussion, it is apparent that additional fundamental research into the model closures for averaged or filtered nucleation and growth terms is needed. To address this issue, the modelling community is putting considerable effort into modelling the Lesniewski and Friedlander (1998) and Lesniewski (1997) experimental cases of DBP at varying concentrations issuing from turbulent round jets, which are also studied in this paper. Most relevant for the current work are the studies by Veroli and Rigopoulos (2011), Garmory and Mastorakos (2008) and more recent studies that primarily focused on the modelling of the nucleation rates (Zhou and Chan, 2011, 2014; Pesmazoglou et al., 2014). Veroli and Rigopoulos (2011) employed the Monte Carlo PDF form of the PBE method combined with a RANS solution of the turbulent flow field. They highlighted the importance of the averaging effects on supersaturation and thus liquid droplet inception. The shapes of the droplet size distributions (DSD's) were predicted reasonably well but nucleation rates were noticeably under-predicted. Garmory and Mastorakos (2008) used a stochastic fields PDF-PBE approach to solve the first three moments of the DSD. They investigated the effect of different surface tension models on nucleation rates and achieved much improved agreement of the predicted zeroth moment with measurements reported in Lesniewski (1997). Zhou and Chan (2011, 2014) and Pesmazoglou et al. (2014) investigated the accurate implementation and closure of nucleation in an LES context. Although nucleation rates were predicted with satisfactory accuracy, the approaches neglected surface growth and the back-coupling of condensation rates on gaseous DBP concentrations, which can become important for cases with higher DBP concentrations.

The present work on DPB nucleation and growth complements the modelling reviewed above with extension to a highly computationally efficient formulation of the PDF-PBE approach in an LES framework. A hybrid scheme is used, involving an Eulerian solver for the LES of the turbulent velocity field and a Monte Carlo stochastic particle method for the joint PDF of the mass fractions of gaseous species and the size-dependent number density of the liquid DBP droplets. While Monte Carlo PDF methods traditionally require stochastic particle resolutions of $\mathcal{O}(10)$ to $\mathcal{O}(100)$ per RANS or LES grid cell (the so called dense, or intensive, particle method), here the simulation is performed by a sparse particle method requiring far fewer stochastic particles than LES grid cells. This sparse particle method, called MMC-LES, is well established for turbulent combustion modelling (Cleary et al., 2009) and was recently applied to the synthesis of silica nanoparticulates in a direct numerical simulated mixing layer (Vo et al., 2017a). The objective here is to apply MMC-LES for the first time to droplet nucleation and growth in a turbulent lab-scale flow. Additionally, analysis is performed to identify the influence of turbulence on aerosol inception and growth and to quantify potential errors associated with inaccurate modelling of the interactions of turbulence with the instantaneous thermophysical state. We also test the sensitivity of the predicted droplet characteristics toward different formulations of the growth rate.

This paper is structured as follows: the governing equations for droplet dynamics, nucleation and growth are presented in Section 2. Section 3 introduces the MMC-LES concept for the solution of these governing equations plus the equations for the conservation of the gas phase species, mass and momentum. The experimental configuration and the numerical setup are introduced in Sections 4 and 5, respectively. Section 6 presents results on droplet statistics for a reference case and cases with varying DBP loadings. The interactions of turbulence and nucleation and growth at the LES subgrid scales are analysed in Section 7, before conclusions are drawn in Section 8.