# POPULATION BALANCE OF PARTICLES IN FLOWS

The population balance methodology provides a powerful framework for studying polydisperse entities such as aerosols, crystals and bubbles. This self-contained and accessible book explains how this theoretical framework can be employed across a wide range of scientific, engineering and environmental problems. The methodology is explained step by step, showing readers how to use these techniques by formulating the population balance problem, choosing models and implementing appropriate solution methods. Particular focus is given to the coupling of the population balance with fluid mechanics and computational fluid dynamics (CFD), in both laminar and turbulent flows. Applications of the population balance methodology are explored in case studies including nanoparticle synthesis, soot formation and crystallisation, and sample open-source code is provided. This book will be valuable to researchers across a range of disciplines including chemical and mechanical engineering, physics and environmental science and can be used as a resource for advanced undergraduate and graduate courses.

STELIOS RIGOPOULOS is Reader in Thermofluids at Imperial College London. He has conducted research on the population balance methodology and its applications for over 20 years, receiving a Royal Society University Research Fellowship for his work. His group has pioneered methods for solving the population balance and coupling it with fluid dynamics, including turbulent flow, with applications that included aerosols, crystallisation, soot formation and nanoparticle synthesis.

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## From Aerosols to Crystallisation

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Dedicated to my parents, Vasileios and
Ermioni, and to my wife, Hsing-fen, with gratitude.

# Contents

# Preface

### Aims and Content of the Book

The population balance is a methodology applicable to a wide range of problems of scientific, engineering and environmental interest. These include atmospheric aerosols, nanoparticle synthesis, crystallisation, cloud formation, sprays, bubble flow, granulation, biochemical engineering, polymerisation and astrophysics. In all of these problems, it is desired to predict the distribution of properties (such as particle size) of a population of entities such as aerosol particles, colloids and droplets. The interest in these properties may arise from a need to tailor them to an application (as in the case of functional nanoparticles), to mitigate their environmental impact (as for a harmful aerosol such as soot) or to build models for a process for which these properties are of importance (such as a surface reaction involving the particle surface area). The evolution of the properties is shaped by a number of processes such as aggregation, growth and transport, and the population balance is an approach for building models that link the evolution of the desired distribution to these processes.

The objective of this book is to present the population balance as a general methodology, to explain the details of its deployment and to show how it can be applied via a number of case studies. In particular, the book discusses the formulation of population balance models, the physical and chemical models that act as constitutive equations, the solution methods available for the population balance equation (PBE) and, finally, the coupling of PBE with fluid dynamics, which includes the effect of turbulence and the implementation of the PBE in computational fluid dynamics (CFD) codes. The methods, which are presented in the first five chapters of this book, are demonstrated in the final chapter via a number of case studies drawn from the research of the author's group, in problems of nanoparticle synthesis, soot formation and crystallisation. All of these feature a spatially dependent population balance and show the potential of the coupled fluid dynamics–population balance approach.

The methodologies employed in the various fields where the population balance is applied are often not described under the term 'population balance'. For example, the term General Dynamic Equation is usually employed in aerosol science, while in other fields, the name 'Smoluchowski equation' is used in honour of one of the pioneering figures in this field. One of the main objectives in the present book is to describe these methodologies under a unified perspective. The term 'population balance' has been chosen both because of its increasing use across several fields and of its descriptive nature.

One of the main developments in the field of population balance modelling during the last two decades has been the coupling of the PBE with fluid dynamics. The developments in CFD have enabled the emergence of coupled CFD-PBE simulations with a variety of methods. One of the aims of the present book is to communicate these developments, with special attention paid to turbulent flow, which raises unique modelling challenges.

While the formulation is presented from a unified perspective, the particular models will be motivated by the case studies in the last chapter. This is necessitated by the very wide range of problems that can be approached by the population balance methodology, each of which requires its own set of physical, chemical and sometimes biological models to be incorporated. It is hoped that workers in other fields will find relevant material in the discussion of the formulations and solution methods, which are quite general.

The book's approach is sufficiently general to render it relevant to workers from several disciplines including chemical engineering, mechanical engineering, environmental engineering, physics and chemistry, while some parts cover in more detail focussed subject areas such as aerosol science and crystallisation. The book has been written to serve a dual purpose, as a textbook for advanced undergraduate and graduate students, as well as a research monograph.

## Structure of the Book

The content of the book is organised in the following way.

In Chapter 1, the population balance is presented as a general methodology, along with its range of applications. Several of the main concepts and terminology that will be used throughout the book are introduced. A brief discussion of preliminary concepts, such as distributions and their properties, are also included.

Chapter 2 presents a unifying framework for a hierarchy of population balance formulations. These include spatially homogeneous and inhomogeneous, one-dimensional and multidimensional or coupled (multiple) PBEs, as well as simplified monodisperse models. The coupling of PBE with CFD is also discussed.

The physical and chemical models that act as constitutive equations are discussed in Chapter 3. Emphasis is, as mentioned, on the models used in the case studies in this book. The inverse problem, that is, the determination of kinetic parameters from experiments, is also discussed.

Chapter 4 discusses the solution methods that have been developed for the various forms of the PBE. At first, the methods are described in the context of the spatially homogeneous PBE. The coupling of the PBE with fluid dynamics and CFD is addressed in the later sections.

Chapter 5 discusses certain unique theoretical and computational problems that are introduced into population balance modelling by the presence of turbulence.

Finally, in Chapter 6, the theory and methodology presented in the previous chapters is applied to three case studies, chosen from the research of the author's group. The topics are silica nanoparticle synthesis in a laminar flame, soot formation in laminar and turbulent flames and precipitation in turbulent flow. They all involve coupling of PBE and fluid dynamics, thus bringing together the book's main themes.

## Use of the Book for Courses

Parts of the book are intended to function as material for courses on population balance. Such a course could be offered at an advanced undergraduate or graduate level. The knowledge required is a typical undergraduate science or engineering background, including mathematics (elements of calculus and differential equations) and elementary fluid dynamics. All methods shown in the book are developed from scratch. Overlap is avoided in topics that have been covered in detail elsewhere, but several appendices are provided in some cases.

The material that could form the basis for a course consists mainly of Chapters 1–3 and parts of Chapter 4. Chapters 1 sets the stage, while Chapter 2 describes the various formulations of the methodology. The instructor may focus on parts of Chapter 3, depending on the focus of the course. The range and coverage of solution methods in Chapter 4 is probably too detailed for a course, but an overview and a few elementary ones (such as the solution of the Smoluchowski equation) would be needed to give the student a complete idea of how to formulate and solve a problem with the population balance methodology.

## Accompanying Code

A suite of codes implementing some of the methods discussed in this book can be found at a link provided in the online resources for the book:

https://www.cambridge.org/9781316512579#resources

The codes are in Fortran 90 and are open source.

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Both the book itself and the research that led to it would not have been made possible without the support of my parents, Vasilios and Ermioni, my sister, Maria, and my wife, Hsing-fen, particularly in the difficult COVID-19 years during which much of the book was written.

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More Information

# Notation

The following is a brief description of the mathematical notations that will be employed in the present book. All symbols are defined when first encountered.

**Cartesian Tensor Notation**

- Employed for equations written in Cartesian coordinates.
- Indices (1,2,3) refer to the three Cartesian coordinates $(x, y, z)$, respectively.
- A free index (i.e. one that is not repeated) indicates the set of all components of a vector or a matrix, or a system of equations for all values of the index, for example:

$$u_i = \begin{bmatrix} u_1 \\ u_2 \\ u_3 \end{bmatrix}, \quad u'_i u'_j = \begin{bmatrix} u'_1 u'_1 & u'_1 u'_2 & u'_1 u'_3 \\ u'_2 u'_1 & u'_2 u'_2 & u'_2 u'_3 \\ u'_3 u'_1 & u'_3 u'_2 & u'_3 u'_3 \end{bmatrix}.$$

- An index repeated twice within the same term indicates summation over all values of that index, for example:

$$\frac{\partial u_i}{\partial x_i} = \frac{\partial u_1}{\partial x_1} + \frac{\partial u_2}{\partial x_2} + \frac{\partial u_3}{\partial x_3}.$$

  Each index does not appear more than twice within the same term.

**Boldface Notation**

- Used when we need to refer to an entire vector with a single symbol, usually inside a function, for example:

$$u_i(\mathbf{x}, t) = u_i(x_1, x_2, x_3, t).$$

### Component Notation

- Used when we wish to distinguish between the coordinates and emphasise their physical meaning, for example:

$$\frac{dp}{dz} = -\rho g.$$

  Also used when employing non-Cartesian coordinates.
- The Cartesian components of the velocity vector are indicated by $(u, v, w)$ for the $(x, y, z)$ coordinates, respectively.