Simulations of flame generated particles

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1 Physical Background

2 Homogeneous Problems

3 Spatially Inhomogeneous Problems

4 Model Comparison
Particles are Everywhere

- Soot from diesel engines.
- Ice crystals in clouds.
- Laundry powder.
- Paint and sunblock.
- Inhaled medicines.
- Circumstellar discs.
- Reactor catalysts.
- Metal oxide sensors.

We want to understand and/or control particle formation and development.
Coagulation is non-linear and non-local

(= Aggregation, Agglomeration, Coalescence, Flocculation)

Particle formation

- Microscopic particles built up from molecules
- Masses can cover 8 orders of magnitude
- Fractal structures develop

Important applications require a study of several internal coordinates

- Sooty flames
- Diesel engines
- Gas turbines
- Ice clouds
- Pharmaceutical crystallisation
- Speciality chemical synthesis
- Washing powder
- Carbon nanotube synthesis
\[
\frac{\partial}{\partial t}c(t, x, y) + \nabla_x (u(x)c(t, x, y)) \\
= I(x, y) + c(t, x, y - \delta) \beta(x, y - \delta) - c(t, x, y) \beta(x, y) \\
+ \iint_{y_1, y_2 \in \mathcal{Y}} \ K(x, y_1, x, y_2) c(t, x, y_1) c(t, x, y_2) \, dy_1 \, dy_2 \\
- c(t, x, y) \int_{y_1 \in \mathcal{Y}} K(x, y, x, y_1) c(t, x, y_1) \, dy_1
\]

- Boundary and initial conditions.
- Coagulation is non-local (in \(\mathcal{Y}\)) and non-linear.
Initial Value Problem

\[
\frac{d}{dt} \int_\mathcal{Y} \phi(y) c(t, y) dy = \int_\mathcal{Y} \phi(y) I(y) dy + \int_\mathcal{Y} \left[ \phi(y + \delta) - \phi(y) \right] \beta(y) c(t, y) dy
\]
\[
+ \int_{\mathcal{Y} \times \mathcal{Y}} \left[ \phi(y_1 + y_2) - \phi(y_1) - \phi(y_2) \right] c(t, y_1) c(t, y_2) K(y_1, y_2) dy_1 dy_2
\]

Markov jump process (Marcus-Lushnikov processes) with particles of weight \(1/n\) given by:

- Inception at rate \(nI(\mathcal{Y})\) with distribution of new particle \(I(\cdot)/I(\mathcal{Y})\),
- surface reaction for each particle \(y \mapsto y + \delta\) at rate \(\beta(y)\),
- coagulation for each pair \(y_1, y_2\) forming \(y_1 + y_2\) at rate \(K(y_1, y_2)/n\).
Initial Value Problem

\[ \frac{d}{dt} \int_Y \phi(y) c(t, y) dy = \int_Y \phi(y) I(y) dy + \int_Y \left[ \phi(y + \delta) - \phi(y) \right] \beta(y) c(t, y) dy \]

\[ + \int_{Y \times Y} \left[ \phi(y_1 + y_2) - \phi(y_1) - \phi(y_2) \right] c(t, y_1) c(t, y_2) K(y_1, y_2) dy_1 dy_2 \]

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- Inception at rate $nI(Y)$ with distribution of new particle $I(\cdot)/I(Y)$,
- surface reaction for each particle $y \mapsto y + \delta$ at rate $\beta(y)$,
- coagulation for each pair $y_1, y_2$ forming $y_1 + y_2$ at rate $K(y_1, y_2)/n$.

These processes are moderately well understood:

- Convergence and approximation properties.
- Fluctuations.
- Computational accelerations (No MLMC).
- Interesting applications:
Development of Silica Particles

With thanks to W. J. Menz & M. Kraft, University of Cambridge

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Initial Value Population Balance Problems

- Systems all spatially homogeneous (Lagrangian transformation).

- Theoretical results for constant coefficients
  - Boltzmann setting: Wagner 92,
  - Coagulation: Jeon 98, Norris 99,
  - Famous review by Aldous 99,
  - More general interactions: Eibeck & Wagner 03, Kolokoltsov book 10,
  - Convergence rate: e.g. Cepeda & Fournier 11.

- Numerical success even when coefficients are functions of time.

Challenge 1
Find an effective Monte Carlo method for the original boundary value problem.

Challenge 2
Find an efficient way to estimate the effects of parameter uncertainty.

Application is the study of different models for particle (irregular aggregate) transport.

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**Challenge 1**

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**Challenge 2**

Find an efficient way to estimate the effects of parameter uncertainty.

Application is the study of different models for particle (irregular aggregate) transport.
Spatial grid, $\mathcal{X} = \bigcup_{j=1}^{J} \mathcal{X}_j$.

Reuse homogeneous particle system inside each cell (almost).

My work focuses on exit boundaries.

For numerical purposes split transport and reaction terms.

Tightness of process laws.

Weak characterisation of limit trajectories.

Functional SLLN.

Central Limit Theorem.
First test problem

Parameters

- \( \mathcal{X} = [0, 0.2] \)
- \( \mathcal{Y} = \mathbb{R}_+^3, y = (red, green, blue) \)
- \( K \equiv 10^{-5} \)
- \( u \equiv 1 \)
- \( I(dx, dy) = 10^8 \left( \delta_{(1,0,0)}(dy) + \delta_{(0,1,0)}(dy) + \delta_{(0,0,1)}(dy) \right) dx \)

Steady state moments

\[
m_k(x) := \int_{\mathcal{Y}} \text{mass}(y)^k c(x, y) dy
\]

- \( m_0(x) = \sqrt{\frac{2 \times 3 \times 10^8}{10^{-5}}} \tanh \left( \sqrt{\frac{3 \times 10^8 \times 10^{-5}}{2}} \frac{x}{1} \right) \)

- higher order moments are polynomials in \( x \).

Numerical results imply there is an \( x \)-differentiable steady state, which is reached quite quickly.
Particle concentration: $m_0(x) / 10^6 \text{ m}^{-3}$

$n = 64 \quad \Delta x = 4 \times 10^{-3}$
$n = 1024 \quad \Delta x = 4 \times 10^{-3}$
$n = 1024 \quad \Delta x = 1 \times 10^{-3}$
$n = \infty \quad \Delta x = 0$
Second mass moment $m_2(x)/10^{-39} \text{kg}^2\text{m}^{-3}$

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\( x / \text{m} \)

\( 0.00 \quad 0.05 \quad 0.10 \quad 0.15 \quad 0.20 \)
Relative Variance of the Mass Distribution

\[ \sqrt{n \times \text{var}(m_1(x))} / m_1(x) \]

- Constant coagulation kernel
Recall: Coagulation of $y_1$ and $y_2$ at rate $K(y_1, y_2) / 2n \Delta x$.

Does the new particle start at $x_1$ or $x_2$ or ... ?

What about mass and momentum conservation?
Recall: Coagulation of $y_1$ and $y_2$ at rate $K(y_1, y_2)/2n\Delta x$.

Does the new particle start at $x_1$ or $x_2$ or ...?

What about mass and momentum conservation?
Introduce weights, proportional to the number of physical particles represented.

- Both computational particles remain.
- Only one computational particle changes, jump rate doubles.
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Mass and momentum conserved.
Second Test Problem

- Weighted method shown with green symbols.
- All methods show approximately $O\left(\frac{1}{\sqrt{n}}\right)$ noise.
Confidence Intervals

- Demonstrated a working BVP method.
- Burn-in and then average results over time.
- I do not know good coupling methods for parameter sensitivity studies.
- Minimising confidence interval widths will be important.

What I have:
- Central limit theorem with explicit variance ($\sigma^2$) for simulation noise.

What I still need:
- Mean reversion strength / Decorrelation time / Mixing time ($\theta$).

Simplest Ansatz:
- Ornstein-Uhlenbeck process for an observed quantity $Y_t$ (average particles concentration for positions between 0.175 and 0.2).

$$dY_t = \theta(m - Y_t)\,dt + \sigma\,dB_t$$

Is this a reasonable model? (Some theoretical results in this direction.)
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  $$dY_t = \theta (m - Y_t) \, dt + \sigma \, dB_t$$
- Is this a reasonable model? (Some theoretical results in this direction.)
Observe a functional at $t_i$ with spacing $\Delta t$ as $Y_{t_i}$, then (O-U assumption)

$$Y_{t_{i+1}} = Y_{t_i}e^{-\theta \Delta t} + m \left(1 - e^{-\theta \Delta t}\right) + \sigma \sqrt{\frac{1 - e^{-2\theta \Delta t}}{2\theta}} Z_i$$

where $Z_i$ are iid $N(0, 1)$.

Linear regression for $e^{-\theta \Delta t}$ and $m \left(1 - e^{-\theta \Delta t}\right)$.

$e^{-\theta \Delta t} = 0.786 \pm 0.008$

$m = 7.75 \pm 0.28$

How good is the assumption of normally distributed noise?
Quartile Plot for Regression Residuals

Straight line shows normal distribution matching 1st and 3rd quartiles of data.
Recall samples $Y_i$ at times $t_i$, $i = 1, \ldots, i_{\text{samp}}$ with spacing $\Delta t$.

Estimate mean as

$$\frac{1}{i_{\text{samp}}} \sum_{i=1}^{i_{\text{samp}}} Y_i.$$

Simulation cost is “burn-in” + $i_{\text{samp}} \Delta t$.

Variance of estimator is

$$\text{var}\left(\frac{1}{i_{\text{samp}}} \sum_{i=1}^{i_{\text{samp}}} Y_i\right) = \frac{\sigma^2}{2i_{\text{samp}} \theta} \left(1 + \frac{2e^{-\theta \Delta t}}{1 - e^{-\theta \Delta t}}\right) + O\left(\frac{1}{i_{\text{samp}}^2}\right).$$

For fixed cost, variance is monotone decreasing, but bounded away from 0 in $i_{\text{samp}} \propto 1/\Delta t$.

Practical considerations will intervene before $i_{\text{samp}}$ gets too big / $\Delta t$ too small.
Transport Model Uncertainty for Soot Particles

- Appel-Bockhorn-Frenklach soot model with spherical particles.
- Laminar premixed flame.
- Different soot particle transport models (increasing complexity)
  - advection,
  - advection with thermophoresis adjustment,
  - advection and diffusion.
- Uniform spatial grid (\(\Delta x\)).
- Splitting time (\(\Delta t\)).
- Max particles per cell (\(n\)).
- Pre-calculated chemical conditions (including (\(u\)) taken from an old study courtesy of Jasdeep Singh.
Small difference in peak height:
No diffusion: 
$(2.86 \pm 0.02) \times 10^{17}$
random diffusion:
$(2.76 \pm 0.02) \times 10^{17}$
thermophoretic drift:
$(2.76 \pm 0.01) \times 10^{17}$.

Possible to quantitatively distinguish diffusive effects.
Better coupling methods are needed.

Some progress is possible with careful analysis.

Thank you