Determining Cluster-Cluster Aggregation Kernels Using Inverse Methods

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Introduction

- Many processes in nature and industry involve large scale coalescence or aggregation of mass clusters.

$$\mathbf{A_m} + \mathbf{A_n} \xrightarrow{\mathbf{K(m,n)}} \mathbf{A_{m+n}}$$

- Such processes can be accurately modelled by a mean-field approximation (MFA) if spatial correlations are absent or can be factored out.
- As there are few analytic solutions to the MFA, determination of the aggregation rate function, or kernel, **K**, of the MFA is often via a mixture of experimentation, simulation, and experienced guesswork.
- Existing inverse methods for aggregation processes may rely on further assumptions about **K**, and these assumptions might not hold.
- Also, it might not be feasible to carefully control natural processes.

Aims

- In this research we seek to determine the extent to which key inverse methods can be held to apply when the key assumptions above cannot be guaranteed for an observed aggregation process.
- Specifically, we seek to determine the extent to which the existence of noise and spatial correlations in an aggregation process can be 'folded back' into an estimation of a kernel function that could then be used as a phenomenological surrogate within the MFA model.
- Such a phenomenological model might then prove useful for enhancing the accuracy of some models of larger processes that include aggregation processes.

The Smoluchowski Coalescence Equation

- Without noise or injected mass, the MFA for a pure aggregation process is the Smoluchowski Coalescence Equation (SCE):

$$\frac{\partial N(m,t)}{\partial t} = \frac{1}{2}\int_0^m K(m-m_1, m_1)N(m-m_1,t)N(m_1,t)dm_1 - \int_0^\infty K(m,m_1)N(m_1,t)N(m,t)dm_1$$

- To model a system with finite maximum mass size, noise, mass injection and fragmentation, additional terms are needed. Fragmentation might need to be modelled using a separate kernel, and the SCE might need to be written in a stochastic form.

The Scaling Hypothesis

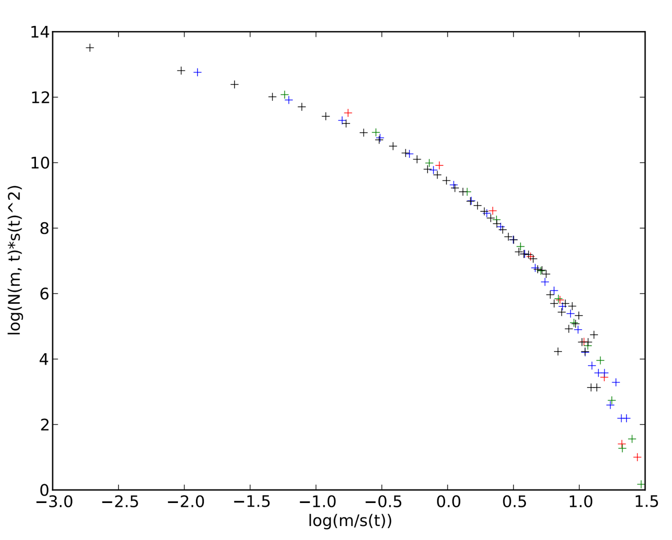
- Analytic solutions to the SCE are often only possible because of assuming that the Scaling Hypothesis (SH) holds[2]. The SH asserts that all solutions that start from reasonable initial conditions eventually approach a scale invariant solution at large times.
- A solution is of scaling form given a function of the typical mass, $\mathbf{s(t)}$ such that:

$$N(m,t) = \frac{C}{s(t)^2}\Phi\left(\frac{m}{s(t)}\right)$$

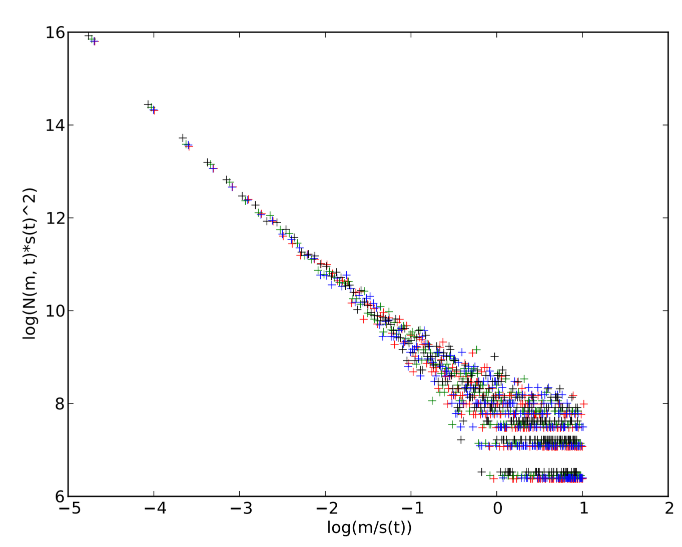
- If scaling holds, then this property can be exploited in inverse methods. However, there are some aggregation processes where neither scaling nor mean-field assumptions hold.

Generating Fiducial Data

- In order to test the accuracy and reliability of suitable inverse methods we have created a Monte Carlo simulation to integrate the SCE, tested with kernel functions for which the analytic solutions of the SCE are known. The simulation can then be used to generate data from more general kernels.



Decay problem scaled data collapse for mass distributions with $K(x,y) = (xy)^{1/4}$.



Scaled data collapse for the stationary distribution with injection, with $K(x,y) = \frac{1}{2}(x^{1/4} + y^{1/4})$

Inverse Methods

- At their heart, most inverse methods work by 'least-squares' minimizing the square of the normed error of a linear mapping, $\mathbf{y} = \mathbf{Ak}$, until a suitable fit $\mathbf{k}$ for the observed data $\mathbf{y}$ is found, if this is feasible.
- However, inverse problems are often ill-posed.
- **Regularisation**
  - Ill-posedness can be overcome to some degree by adding a regularising term to that penalises solutions with large fluctuations.

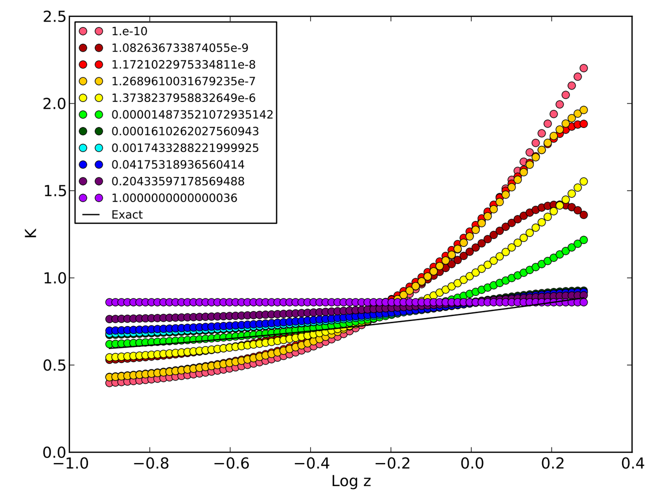
$$J(k) = \min_k \quad \|Ak - y\|_1^2 + \lambda_{reg}\|k\|_2^2$$

  - As a first step, we implemented this approach as used specifically for the derivation of aggregation kernels[3] when the SH applies.
  - However, the recommended regularisation did not work, and experimentation led us to use[1]:

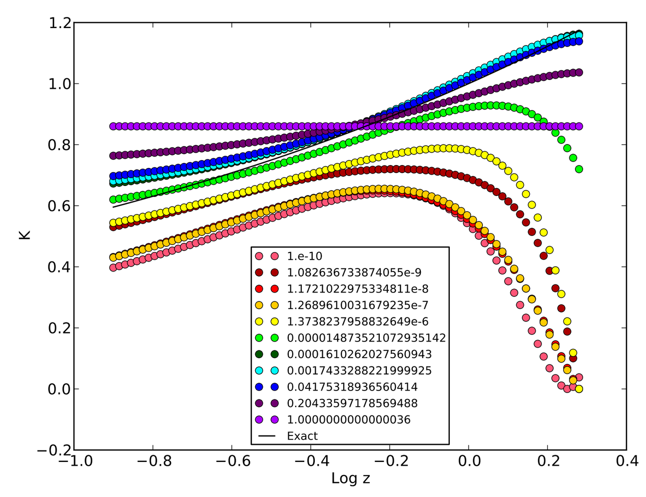
$$\lambda_{reg} w(k) = \lambda_{reg} \log\left(\prod_i (|k_i| + 1)_i\right)$$

Results

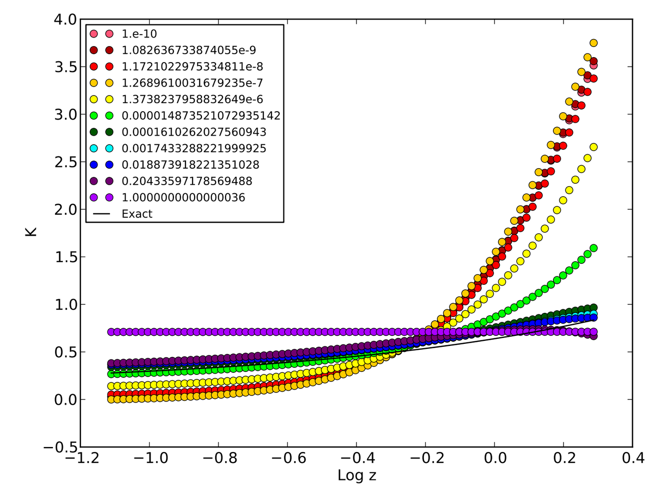
- Decay Problem: For suitable values of the regularisation parameter, λ_{reg}, we retrieved good approximations to input kernels $K(x,y) = \frac{1}{2}(x^\xi + y^\xi)$, $\xi \in \{0.0, 0.25, 0.50, 0.75, 1.0\}$ from simulated data with some inherent noise. The most interesting cases are shown below. (Black lines represent the known input kernels.)



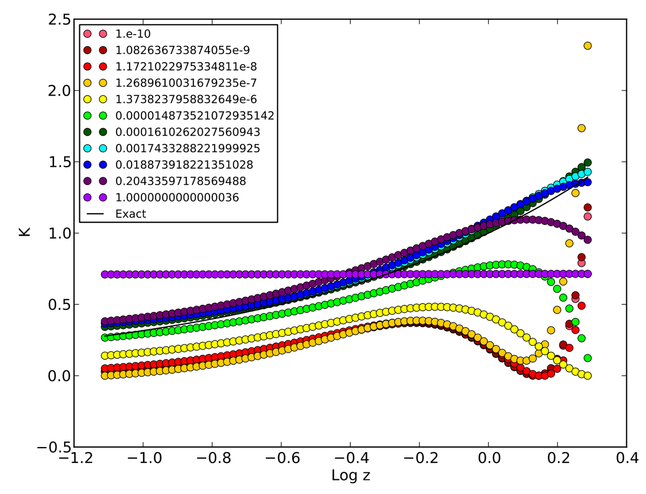
Kernel edge estimates recovered for the case $\xi = 0.25$.



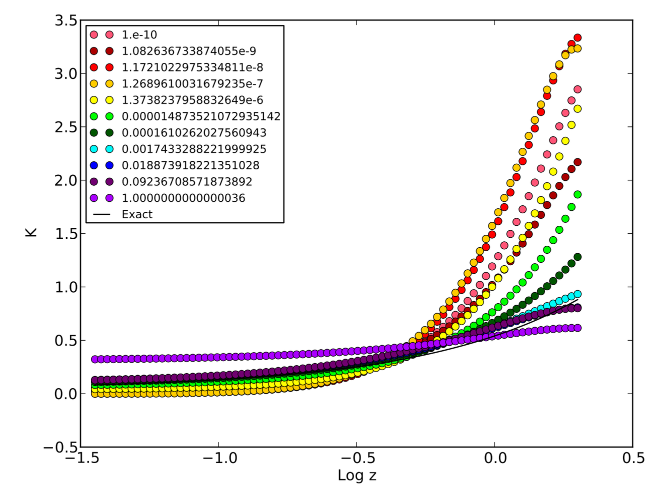
Kernel diagonal estimates recovered for the case $\xi = 0.25$.



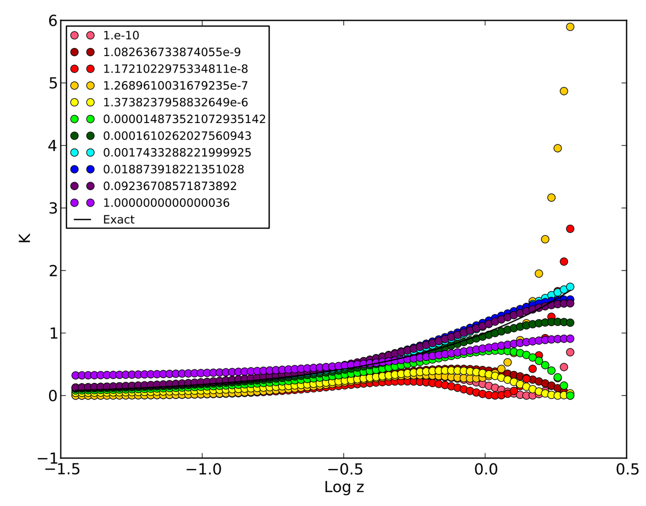
Kernel edge estimates recovered for the case $\xi = 0.50$.



Kernel diagonal estimates recovered for the case $\xi = 0.50$.



Kernel edge estimates recovered for the case $\xi = 0.75$.



Kernel diagonal estimates recovered for the case $\xi = 0.75$.

Further Work

- More study of the use of orthogonal function representations.
- Work-in-progress on a more powerful time-dependent approach.
- Application to stationary and non-scaling distributions.

References

[1] Colm Connaughton and Peter P. Jones. Some Remarks on the Inverse Smoluchowski Problem for Cluster-Cluster Aggregation. In *J. Phys.: Conf Ser.*, number 012005 in 333, 2011.

[2] F. Leyvraz. Scaling theory and exactly solved models in the kinetics of irreversible aggregation. *Phys. Reports*, 383(2):95–212, 2003.

[3] H. Wright and D. Ramkrishna. Solutions of Inverse Problems in Population Balances - I. Aggregation Kinetics. *Computers Chem. Eng.*, 16(12):1019–1038, 1992.