

Determining Cluster-Cluster Aggregation Rate Kernels Using Inverse Methods

A Brief Non-Mathematical Research Introduction
for the Uninitiated

Version 0.2

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20-6-2009

To introduce our research into the inverse problem in cluster-cluster aggregation dynamics, first let's ask, what is an inverse problem? It's usually contrasted with a 'forward' problem. In a forward problem, we have investigated a physical phenomenon that we think we have a good theoretical model for, and have obtained experimental data. The task is to implement the model, compare the data that it produces with the experimental data, and to explain the differences, if any. With an inverse problem typically there is only a rough idea of what the model should be for some given phenomenon, and values of key parameters in the model are unknown. Given experimental data for the real phenomenon, the inverse problem is the attempt to determine the values of the key model parameters from the data. If the data is sufficiently complete this task is relatively easy. But if the data is sparse or contains 'noise', then matters become more difficult.

Now, what is cluster-cluster aggregation? Imagine some oil mixed with water; two liquids that are immiscible in the same container. Also imagine that the mixture has been thoroughly stirred so that there are many, very small globules of oil, of roughly equal size, evenly distributed throughout the water. Then we leave the mixture alone, and allow the currents within the fluid to bring oil globules together here and there as time passes. Typically, when two oil globules come into contact with each other they will merge to form a larger globule. Mass is conserved in this merger process, so the mass of the large globule is equal to the sum of the masses of the two contributing globules.

We might also imagine that the process of merger between globules could happen (statistically, averaging over a large number of mergers) at a rate dependent on the magnitudes of two contributing masses. For example, if there is a very large globule in a sea of very small globules then we might expect mergers between the large globule and small globules to happen at a faster rate than mergers between small globules, simply because the large globule occupies more space and constitutes a bigger target as the mixture swirls. However, in another completely different mixture, mergers between large and small globules might be less likely, and hence happen at a slower rate, because of chemical, electrostatic, or hydrodynamic interferences.

In other aggregation processes the masses might also have geometric structure, like grains of sand, molecules or pollen, and that instead of merging into a bigger blob, they aggregate to form larger clusters with spatial structure. Clusters can, of course, aggregate with other clusters, and this is what distinguishes the process of cluster-cluster aggregation (CCA) from, for example, diffusion limited aggregation (DLA). (In DLA a single large cluster is formed by the accumulation of lone small masses over time - e.g. water molecules joining a snowflake under freezing conditions.) Although clusters can have spatial structure, in physics we can deliberately overlook this should we choose to if we can argue that in some huge mixture all clusters look sufficiently alike. That is, we might deliberately make some averaging approximation to simplify our models.

Research in cluster-cluster aggregation has progressed for a long time using a model based on the Smoluchowski Coagulation Equation (SCE). The effectiveness of the SCE relies on a number of assumptions that are, in effect, averaging approximations: well-mixed, very large systems; spatial uniformity of processes; mixtures sufficiently dilute that only pairwise interactions between particles are likely. This allows tractable discussion of (average) merger rates in terms of the amounts of specific mass sizes present in the mixture. That is, the SCE states that the average number of masses (per unit of volume) of mass M present at some time t is a function of the rates at which other pairs of masses present can clump together to form a mass of size M , minus the rates at which masses of size M might join with other masses. We can think of these two rates as being like forces tugging in opposite directions on an abstract particle, its position on the real number line representing the number of masses of size M that are present at some time. In physics, particles in fields experience forces - our abstract particle is in a field created by the presence of any other particles. Hence the SCE is considered to be a member of the class of so-called mean-field approximations: it is valid only if we can average over a sufficiently large number of interactions to be able to talk sensibly about the forces on our abstract particle.

An advantage of the SCE is that it is the simplest effective description of an aggregating system. However, there are also some snags with the SCE. One is that many real systems are not vast and internally they exhibit (statistical) spatial heterogeneities - for example if large masses hoover up all smaller masses in their immediate neighbourhood creating 'moats' of empty liquid around them - so the averaging can break down. Another snag is that the SCE has only been mathematically solved for a small number of simple merger-rate functions. (Merger-rate functions are called kernels, in the technical jargon). This means that we can only find out the results of arbitrary kernels through computer simulations, and we have no good formulaic way of predicting which kernel would describe which real system adequately well. Working backwards in order to infer kernels from experimental data might allow us greater insight into both of these problems.