

# Statistical inference for disordered sphere packings\*

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**Abstract:** This paper gives an overview of statistical inference for disordered sphere packing processes. These processes are used extensively in physics and engineering in order to represent the internal structure of composite materials, packed bed reactors, and powders at rest, and are used as initial arrangements of grains in the study of avalanches and other problems involving powders in motion. Packing processes are spatial processes which are neither stationary nor ergodic. Classical spatial statistical models and procedures cannot be applied to these processes, but alternative models and procedures can be developed based on ideas from statistical physics.

Most of the development of models and statistics for sphere packings has been undertaken by scientists and engineers. This review summarizes their results from an inferential perspective.

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## 1. Introduction

Disordered sphere packing processes are a widely-used class of spatial stochastic processes for which few effective inferential methods are available. Realizations of these processes are used to model many composite and granular materials in physics and engineering, but rarely is any assessment of model fit undertaken. Standard methods from spatial statistical inference are of no use in these assessments, but strategies for model assessment can be constructed from statistical tools developed in the fields of application.

Almost all models and descriptive statistics for packings have arisen from applications of packings to problems in science and engineering. This review gathers these achievements and organizes them into a coherent inferential program. It begins with a presentation of the major uses of sphere packings in science and engineering. Next, the difficulties associated with defining and formally representing a packing process are discussed. The physical models and computer programs used to simulate packings are outlined, and the importance of experimental work is emphasized. Many types of descriptive statistics are then

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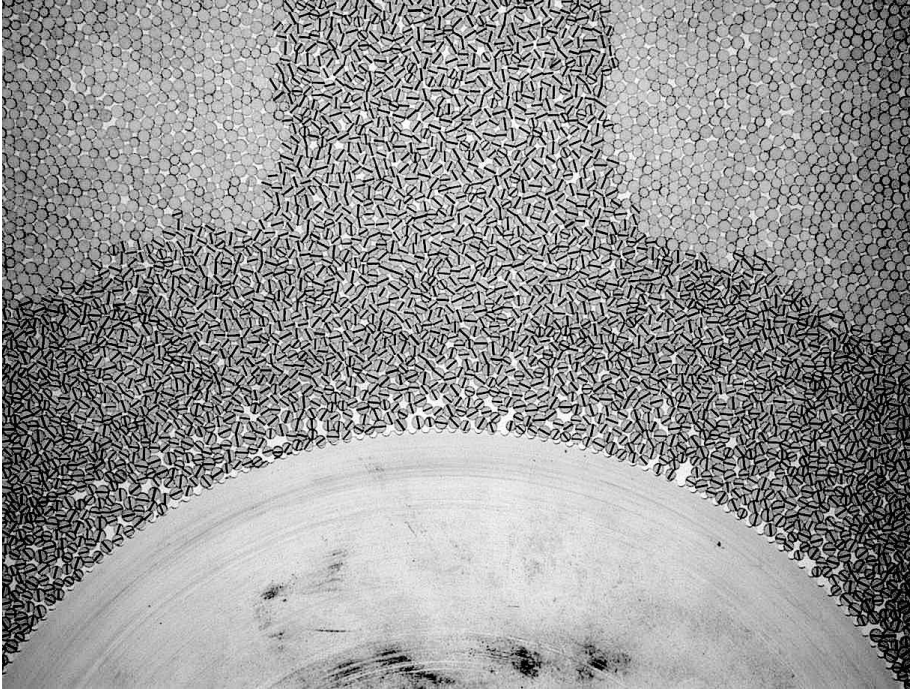


FIG 1. Digital image of plastic discs in a Couette shear apparatus. The discs in the upper half of the image are packed. Image provided by B. Utter, James Madison University.

presented and compared. The review ends with a discussion of how model assessment can be undertaken, given the nature of the models and the descriptive statistics available.

### 1.1. Terminology

Objects of many different shapes can be packed, but this review will focus on packings of spheres. In packings of objects, the spherical objects are never exactly spherical. When necessary, distinctions will be made between spherical objects and the ideal spheres which summarize their shape. Packings of spherical objects will be referred to as physical packings, to distinguish them from their idealizations. A physical packing process will be an established procedure for forming a packing using a particular set of physical objects. Physical packing processes will be assumed to generate packings by halting the flow of spherical objects with some form of confining surface. Physical packings produced by fixing each sphere into place, one at a time, will not be considered.

It will generally be assumed that the spheres in packings are of equal diameter (monodisperse) rather than being of many different sizes (polydisperse). Spheres in  $\mathbb{R}^2$  will be termed discs, spheres in  $\mathbb{R}^3$  will be termed spheres, and in higher dimensions they will be termed hyperspheres. In the applied literature, planar

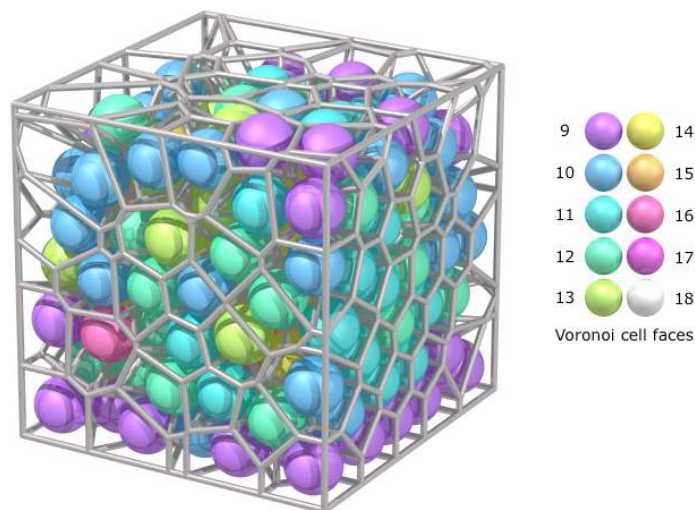


FIG 2. Three dimensional packing of spheres within a hard-sided cube. The Voronoi tessellation of the packing and the number of faces for each cell are also shown. Image provided by C.H. Rycroft by means of Voro++ imaging software [1].

packings of discs are sometimes referred to as packings of rods. This is misleading, since it assumes absolute rigidity for any physical rods being modelled.

A point lattice is a regular structure of points. Examples include the cubic point lattices in space and in the plane, and the face-centered cubic packing and hexagonal close packing in space. Packings will be ordered if their sphere centres form a subset of a point lattice. The study of ordered packings in high dimensional spaces yields interesting abstract mathematical results [2–4] which can be used in the development of efficient codes [5]. Specialized packings can also be used in the study of discrete analytical functions [6]. Few of these results have any application to the study of disordered packings in space or in the plane. Packings will be assumed to be disordered and not based on a point lattice unless otherwise stated.

### 1.2. Examples of use

Sphere packings were first used in attempts to explain the patterns seen in X-ray diffraction studies of liquids and non-crystalline solids [7, 8]. Bernal [9] and Scott [10] attempted to model macroscopically what an amorphous mass of densely arranged atoms might look like, using physical packings of monosized steel spheres as their model. More recent work [11] has suggested that this approach to modelling may not be feasible, since packings of steel spheres are formed by the action of a different set of forces from those found at the atomic scale.

Sphere packings are used extensively in the modelling of granular materials. A granular material consists of a collection of solid particles surrounded by a

gas or a liquid. Granular materials can be made to flow when acted upon by a force. Examples of granular flow include the flow of particles from hoppers, avalanches and mudslides, soil liquification, and soil erosion by wind and water. The complex interaction between grains during flow makes it impossible to model granular flow by means of conventional methods from statistical physics [12–14]. Spheres are often used to represent grains, and sphere packings are used as models for powders at rest. Simulations of powder flow often require a sphere packing as an initial state.

Sphere packings can be used in the study of composite materials. In the study of metal sintering [15], a packing can represent the initial state of a metal powder before it is compressed and heated to form a solid metal part. Packings can be used as models for some colloids and for concrete. A colloid is a mixture of two immiscible liquids, in which the dispersed liquid may form spherical inclusions within the other. For some colloids, the arrangement of inclusions may be modelled by a sphere packing [16]. Concrete is a composite of rocks of many different shapes and sizes held together by a matrix of cement [17]. Sphere-packing-based models for concrete are highly idealized, but may be the only feasible way to abstractly model such a complex material [18].

Packings can be used as models for porous structures. A packed bed reactor is a large steel vessel filled with solid catalyst-impregnated particles. Liquid reactants flow in at one end of the vessel, and the product emerges from the opposite end. Design of these reactors requires being able to model both the reaction in the pore spaces around the particles and the transfer of heat through the particles. Sphere packings are the simplest form of particle packing that can be used to model the internal structure of these reactors [19].

When a packing is used in modelling a porous structure, a granular flow, or a composite material, it is being used as part of a model for phenomena that take place on a larger scale. If a powder at rest is poured, the absence of any continuum model for the flow makes it necessary to simulate the flow by modelling the motion of individual spheres. The physical models for motion of individual spheres and their interactions are simpler than the continuum models, and so the right choice of model for individual sphere motion will result in the right behaviour for the flow as a whole.

## 2. Defining a packing process

To define a sphere packing process, it is necessary to define what is meant by a configuration of packed spheres. This requires first being able to define a packing of physical objects, and then finding an abstract definition that can be used as the basis for a formal probability model.

### 2.1. Physical packings of spherical objects

A physical packing of spheres is a collection of spherical objects which are acted upon by a collection of forces which are in balance. No spheres are in motion

with respect to each other, nor are they in motion with respect to the containing surface. No motion can take place unless additional unbalanced forces are applied. Each sphere is in contact with containing surfaces and other spheres. These contacts deform the objects and the container, and forces associated with these deformations are in equilibrium with the force of gravity and other forces. Composite materials with spherical inclusions are formed by a set of forces acting on spheres surrounded by the matrix phase in liquid form.

Physical packings are generally constructed by directing a granular flow into a container or onto a surface. The granular flow is highly dependent on the exact arrangement of the spheres within it and on the interactions between spheres as the packing forms. Because the exact arrangement of spheres can never be known and the results of interaction cannot be predicted or observed, the process of forming a packing can be viewed as sampling a single packing at random from the collection of all packings that could be generated by the physical packing process.

While entire packings occur randomly with each repetition of the construction process, the internal structure of each packing is not random. It is not possible to change the position of any one sphere without exerting an unbalanced force on the packing. If a single sphere from the interior of a packing were omitted from the list of all sphere locations in a packing, its location could be easily determined. The difficulties in describing any one packing arise from there being no obvious way to summarize important aspects of its disordered structure.

## *2.2. Mathematical representation of a sphere packing*

In order to carry out traditional probabilistic modelling, it is necessary to construct a probability measure over a sample space of packings. It is not possible to usefully define either a sample space or a measure for packing processes, on account of the limitations of existing mathematical models.

A packing process is a random set process [20, 21], since each realization of the packing can be considered to be a random closed subset of  $\mathbb{R}^d$  for some  $d \in \{2, 3, \dots\}$ . The random set process sample space  $\Omega_0$  contains all closed subsets of  $\mathbb{R}^d$  and so is too large and lacking in structure to be useful. Since each packing can be represented as a union of closed sets of simple structure, it is possible to construct a much simpler sample space than  $\Omega_0$ .

The spherical objects that comprise a physical packing are not ideal spheres before they are packed, and their deformed aspherical shapes cannot be observed once they are packed. If the spherical objects are very close to being spherical and deform very little, then it is reasonable to summarize the structure using only the sphere centroid coordinates.

To construct a sample space for the packing process based on sphere centroids, it is necessary to isolate the essential shape of the packing. The shape consists of every aspect of the arrangement of the spheres that is invariant under rotations, reflections, and translations of the spheres, their container, and the balanced forces. Once these are accounted for, each packing can define a point

in  $\mathbb{R}^{dn}$  by using the first  $d$  coordinates to represent the coordinates of the first sphere location in  $\mathbb{R}^d$ , the next  $d$  coordinates to represent the location of the second sphere, and so on. The sample space  $\Omega_1$  is defined by the collection of all such points. Since the labelling of the spheres is arbitrary, each shape will be represented by  $n!$  distinct points in  $\mathbb{R}^{dn}$ .

The structure of  $\Omega_1$  is complex, since it is a subset of a very high dimensional space. The complexity can be reduced by identifying packing shapes that have topologically similar contact networks. The contact network of  $n$  packed spheres is defined to be the graph whose edges consist of all line segments connecting centroids of spheres in contact, together with the line segments connecting each sphere centroid to points where the sphere is in contact with a boundary. In the case of physical spheres the contact will be a region, but the centroid of that region can be used to define the edge.

Graph isomorphisms can be used to establish equivalence classes among the contact networks. Two contact networks are graph isomorphic iff there is a bijective map from the vertices of one contact network onto the other that preserves edges. Small perturbations in sphere shape, orientation, and sphere distortion under load can yield many different packings with graph isomorphic contact networks. The sample space  $\Omega_1$  will consist of many small clumps, each consisting of packings with isomorphic contact networks. If the equivalence classes are defined by graphs which share a common subgraph, then some clumps associated with these classes will overlap. If one particular labelling of spheres were chosen, the lack of any natural labelling system for the spheres would result in many of these intersections being lost.

The sample space  $\Omega_1$  is still too complex to be of practical use. The extent of the clumps associated with each equivalence class of contact networks cannot be determined, on account of the inability to observe very small distortions and irregularities of physical sphere shape within the packed structure. Since the number of equivalence classes must be finite, it is possible to define a discrete sample space  $\Omega_2$  consisting of the graphs that define the equivalence classes. If every element of  $\Omega_2$  could be identified, then it may be possible to estimate or derive a discrete probability measure for the packing process.

It is not possible to explore and analyze  $\Omega_2$  by means of theoretical arguments if the number of spheres exceeds 10. For smaller numbers of spheres, it is possible to use theory to isolate the arrangement of spheres which most efficiently covers a unit square [22]. For more than 10 spheres, the densest packing in  $\Omega_2$  can only be found by experimentation on physical or simulated packings [23–25]. Experiments of either type could never be guaranteed to include elements of every equivalence class, particularly when a very large number of distinct equivalence classes exist. Identification of distinct networks is also complicated by the need to determine if near-neighbouring discs are in physical contact. Experimental studies have shown that spheres can be very close without being in contact [9, 26].

Experiments involving simulation models require that the model sample from the same sample space as does the physical process. If the models are based on ideal spheres, this may not happen if some contact networks found in physi-

cal systems cannot be constructed from ideal spheres. This problem might be avoided by incorporating much more complex models of the initial shapes of spheres, but modelling their shapes after distortion under load would be impossible using current continuum models [27, 28]. These difficulties can also be avoided by assuming that each sphere of radius  $r$  has the shape of an ideal sphere but is composed of a hard centre of radius  $r - \delta$  and soft deformable outer shell of thickness  $\delta$ .

The probability measure on  $\Omega_2$  is determined by the dynamics of the physical process which produces the packing. Since it is not possible to represent this process formally, it is not possible to derive this distribution. It is also not feasible to estimate the probabilities of individual equivalence classes, since the number of possible classes increases rapidly with the number of spheres. Families of distribution functions are of no use, since there is no obvious simple and low-dimensional method of coordinatizing the equivalence classes. The tradition in physics is to assume that there is a uniform distribution over  $\Omega_2$ . This assumption greatly simplifies the mathematical analysis of the packing process, and it serves as the basis of the hard-core Gibbs model [29, 30]. Experimental investigation of packings of 7 discs suggests that the assumption of uniform probability on  $\Omega_2$  is not valid for packing processes [31].

The lack of formal probability models for packing processes presents several major challenges to using them as part of scientific or engineering applications. First, the absence of a simple, natural, formal model such as the Gibbs process in statistical physics rules out any purely formal asymptotic analyses of packing processes. Second, likelihood-based inference and Bayesian inference for packing processes is impossible. Third, any model for a packing can only be evaluated by comparing an independent random sample of realizations of the model with a sample of observations of the physical packing process of interest.

### 3. Stochastic models for packings

There are two possible alternatives to formal models for packing processes. Either the model can be another physical process for packing different spherical objects, or it can be a computer program that assembles ideal or soft-shelled spheres into packed arrangements.

#### 3.1. Physical models for packings

A physical model for a packing of objects is a different physical system which is easier to replicate and study than the original physical packing process. Physical models are useful for investigating packings of soft objects and objects of irregular shape [32] when these objects are too difficult to simulate.

An early use of physical models involved attempts to model the molecular structure of liquids [7]. Scott [10] and Bernal [8, 9] both used packings of monosized steel spheres as models for atomic structure. Monosized steel spheres were

placed into rubber balloons and bound with rubber bands to maximize packing density and minimize the effects of gravity on packing structure. In later experiments, paint was added to the balloon and allowed to set. The agglomerated mass was dissected carefully in order to identify which spheres were in contact, and physical models of the contact network were constructed out of wire. More elaborate measurement methods were developed [33], but interest in this approach soon waned on account of the amount of time and effort required to obtain results.

Physical packings are also distinguished by the material that fills the voids around the spheres. A physical process that packs spheres in room temperature air is distinct from one that packs spheres surrounded by warm liquid wax or paint. To make low-density packings, spheres have been packed in a fluid of equivalent density [16]. It is also possible to make a colloid which is very much like a packing by combining two immiscible fluids, one of which forms near-spherical inclusions in the other [34].

Physical packing processes for discs can also be developed. These could be as simple as swirling a tea tray full of thick coins and then inclining the tray until the coins come to rest. Processes of this kind using monosized discs produce realizations which are very close to hexagonal lattice packings, and so more complicated methods are needed to obtain more disordered realizations. Non-overlapping discs can be placed at random locations on a slightly stretched sheet of rubber. The tension in the sheet is released, forcing the discs together into a smaller area. The positions of the discs are recorded, the sheet is retensioned, the discs are placed back on the sheet, and the tension is released again. By cycling through this procedure, a disordered disc packing can be generated [35].

Interest in physical packings has been revived as new remote sensing methods for 3-dimensional packings have been developed. X-ray tomography [36–39] and magnetic resonance imaging [40] can be used to map the interiors of very large packings. Confocal microscopy has been used to study both sphere packings [41, 42] and colloidal packings [34].

### 3.1.1. *What can be learned from physical packings*

Physical packing processes can demonstrate properties which frustrate intuition-based analyses of their structure. Experimental examination of physical packings is necessary to fully appreciate the complexities of their structure that may need to be captured by simulation models.

First, there is no single well-defined packed state for spheres. Different packing processes can have very different sample spaces, and that sample space can change greatly over the duration of the packing process. Experiments with monosized spheres poured into containers showed that the density of the initial configuration of the spheres could be increased by rodding the packing or by shaking it vertically [10]. There appeared to be a well-defined limit to the density achievable by these methods, which resulted in the proposal that there was a well-defined and stable physical state termed a random close packing of



spheres. This definition has been challenged [43], especially after experiments demonstrated that random close packings could be made to evolve into denser packings with near-ordered structure [44, 45].

Experimental investigation of monodisperse packings suggested that disordered packings were not the densest possible arrangements of spheres in space. For dimensions  $2 \leq d \leq 8$ , the densest lattice packings can be identified [5]. In the plane, it can be analytically proven that the hexagonal close packing is denser than any other packing [46]. In three dimensions, a computer-based proof has established that the body-centred and face-centred packings are denser than all other three-dimensional packings [47].

Polydisperse packings are far more disordered than monodisperse packings. One way of disrupting the near-order of physical packings of monosized discs is to randomly introduce a small amount of size variation into the discs. This strategy was used to prevent crystallization in the packing shown in Figure 1. Physical packings with a fixed sphere size distribution can be very difficult to assemble, since the smaller spheres often segregate and accumulate at the base of the packing. These difficulties also occur in simulated polydisperse sphere packings [48, 49].

### 3.2. General comments on simulation models

Simulation models are large and complex computer programs. To save computing time, assumptions are often made which can limit the ability of these models to represent physical packing processes.

#### 3.2.1. Randomness

Most algorithms for packing generation are used to produce a single large packing for use in the study of a scientific or engineering problem. Although repeated experiments show an inherent variability in experimental outcomes, it is very unusual to see any attempt to capture this variability in a simulation model.

Uncertainty about model outcomes arises from the sensitivity of the dynamics of physical packing processes to initial and boundary conditions. The sensitivity can be modelled by choosing initial particle positions from a realization of a Poisson process, a hard-core Markov spatial process, or from a packing process. Initial and boundary conditions are seldom discussed in detail in the applied literature, and models for the initial arrangements of spheres are not generally fit to data in any formal way.

#### 3.2.2. Boundaries

Most physical packings are formed by a confining surface. The surface may be completely closed and hold every sphere into a rigid position, it may be open and allow the packing to have a free boundary, or it may be a plane orthogonal

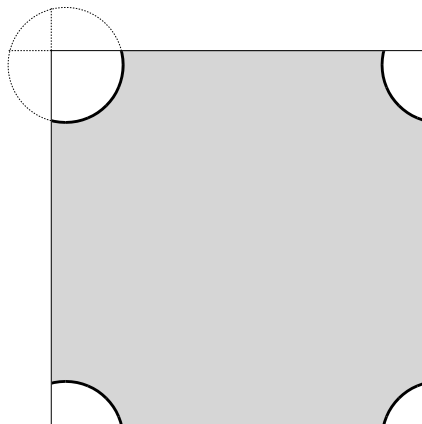


FIG 3. When periodic boundary conditions are imposed, spheres at the edges of the packing appear at several distant locations within the window. A disc near a corner could appear in four separate locations, while a sphere could appear in as many as eight.

to the action of the unbalanced force. Simulations omit confining surfaces in order to avoid difficulties in coding or to simulate packing structures far from any confining surfaces.

It is possible to avoid using any boundaries. Spheres can be packed on the surface of a hypersphere of one dimension higher [50], but this can introduce unwanted effects from hypersphere surface curvature unless many relatively small spheres are used. Packings can also be assembled by simulating a force acting towards a central point. It is not clear that a packing simulated by a central force would represent any packing found in nature.

Confining surfaces can be avoided by imposing periodic boundary conditions. Consider a disc packing in the plane and a rectangle  $W$ . If the discs are monosized with diameter  $\delta$ , then any disc whose centre lies within  $\delta/2$  of a side of  $W$  has the part of the disc outside of  $W$  appear on the opposite side of  $W$  (see Figure 3). Since this can occur on any edge, the window  $W$  is toroidal and the plane is tiled by copies of  $W$  containing a disc packing. If any effects of the boundary on packing structure vanish after some distance  $k\delta$  from the boundary, then the interior of a realization may be indistinguishable from that of a stationary packing. Efforts have been made to determine a value for  $k$  [51, 52], but these efforts are often based on a single statistic and may not reflect every effect of periodic boundaries on the structure of a realization.

Periodic boundary conditions may be used within confining boundaries in order to save computational effort. Within cylindrical confining boundaries, it is possible to pack four periodic cells [53]. This symmetrical structure may have a strong effect on the distribution of statistics calculated from the packing.

### 3.2.3. Packings of non-spherical objects

Most simulation models pack monodisperse spheres, since the code for these programs is relatively easy to write. It is dangerous to assume that these models can represent packings of more complicated physical objects, unless experimental evidence can confirm that they do.

Modelling packings of non-spherical objects can be very difficult. While a sphere can be fully specified by its centre and radius, a reasonable simulation of a small rock will require a long list of numbers to specify the polynomial surfaces that are spliced together to produce a crude representation of its shape. Determining contacts between asymmetrical objects is also very difficult, although it can be simplified by modelling the object as a collection of adhering spheres [54]. The shapes of ceramic rings have been approximated by assemblies of triangles [55] as part of a simulation of packed beds. Elliptical objects can be easily simulated, and can be shown to pack more densely than spheres in simulations [56–58] and in physical experiments [59].

### 3.3. Ballistic algorithms

Ballistic algorithms are the simplest algorithms which can be used to produce configurations of packed spheres or discs. They were the first developed and are relatively simple to code, but they are not based on the physics of packing formation. Packings generated by these algorithms are less dense than those generated by physical packing processes [60]. Ballistic models must be used with caution, as there is no reason to believe that they have the same sample spaces as do physical processes.

The first ballistic algorithms were developed by Vold [61, 62] to model the formation of flocs. Spheres are dropped at random locations onto a surface. They stop falling either when they hit that surface or another sphere. If they hit another sphere, then they either are locked in place with probability  $p$ , or are rolled down the packed spheres until they reach a stable position. With  $p = 0$ , the algorithm can produce realizations of loose packings. The path chosen for each sphere superficially mimics the path of a falling physical sphere, but the path is inconsistent with the dynamics of a physical sphere impacting onto an existing packing.

Vold also developed a packing algorithm for rods composed of a set of  $k$  spheres in contact with their centres arranged along a line [64], and developed a central version of her algorithm [63]. In central ballistic packing algorithms, spheres are packed as if gravity were acting towards a single point rather than towards a flat surface. This results in packings which are isotropic, but which cannot be produced by any physical packing process. Later high-density central packing simulators [65, 66] produce packings with estimated volume fractions between 0.61 and 0.62, slightly below those seen in random close packings. In central disc-packing algorithms, near-ordered packings are avoided by seeding the realization with a non-triangular configuration of discs [67].

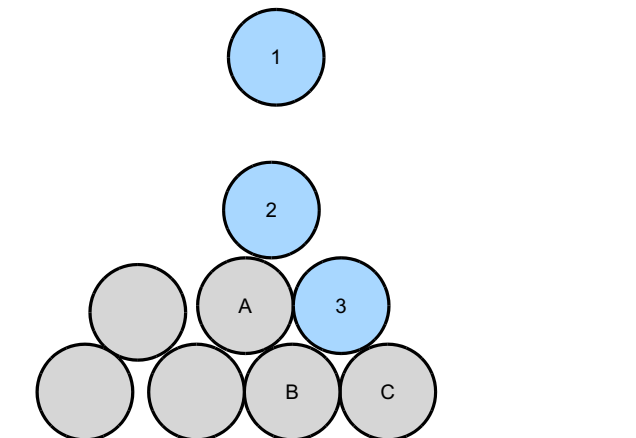


FIG 4. In a ballistic packing algorithm, the blue sphere starts at a random location (1) above the already-dropped gray spheres. It drops to position (2), and then rolls down the side of sphere A until it comes to rest against sphere B. If this is not a stable position, it rolls along sphere B until it is stabilized by resting against sphere C. The rolling process is purely geometric, and does not represent any physical process.

Visscher and Bolsterli [68] developed a non-central algorithm which fills a box with an impenetrable base but periodic boundary conditions on its sides. A sphere is dropped at a random location, and falls vertically until it contacts another sphere. Then, it follows the shortest path along the surface of the already-placed spheres until it comes to rest in a gravitationally stable position (Fig. 4). Each sphere drop is repeated  $k$  times, and the final position chosen is the position that is closest to the base. The algorithm produces realizations with estimated volume fraction of 0.582 in  $\mathbb{R}^3$ , which is less dense than a shaken physical packing. In the planar version of this algorithm, the initial spheres dropped have a slightly different size in order to prevent the formation of near-ordered realizations.

The Visscher-Bolsterli algorithm has been improved in various ways. A central version has been developed [69], with the intention of producing packings that contain arching structures. Arching structures arise when several adjacent spheres form a vault-like structure around a void of unusually large size. Non-periodic boundaries have been added to the algorithm, so that the spheres fill a box in space [70] or in the plane [71], or fill a cylinder [72–74]. Irregular shapes can be packed within periodic boundaries [55, 75].

The Visscher-Bolsterli algorithm can be modified to pack spheres of different sizes. When the largest and smallest spheres are greatly different in size, the smaller spheres tend to accumulate at the base of the packing unless they are allowed to randomly stick at mechanically unstable positions [48]. More complicated deposition rules have been devised to avoid size segregation in the plane [76, 77].

The density of physical packings can be increased by shaking them. The density of ballistic packings can be increased by subjecting the initial packing

to rearrangements which superficially imitate shaking. A simple rearrangement algorithm orders the initially packed spheres by height, then redeposits them at the same planar location using the Visscher-Bolsterli placement rules [78, 79]. This algorithm was used to study segregation by size after shaking. In a shaking algorithm for monosized spheres, each sphere in the packing is displaced upwards by a small Normally distributed perturbation, and is then subjected to many small three-dimensional Normal perturbations which are allowed if no collisions occur. Once the number of collisions reaches a set threshold, the packing is collapsed from the bottom using Visscher-Bolsterli placement rules. This algorithm can increase the estimated mean volume fraction from 0.581 to 0.590, but still yields a loose packing [80]. If the initial vertical displacements are too large, then the rearrangement can simplify the contact network and decrease the volume fraction. Rearrangement methods can also be applied to packings of irregularly shaped objects in a container with hard boundaries [81].

### 3.4. *Rearrangement algorithms*

Algorithms which rearrange the points in point patterns were the first to achieve volume fractions similar to those seen in random close packings. These programs begin with a realization of a Poisson or a regular lattice point pattern. The points in the pattern are subjected to deterministic or random translations which eventually produce a sphere configuration close to that of a packing. These methods do not attempt to replicate the dynamic interactions between spheres which occur during the formation of a physical packing.

The Jodrey-Tory algorithm [82–84] was the first simulation algorithm to produce realizations with estimated volume fractions similar to those for dense physical packings. It is initialized by a configuration of  $n$  points in the interior of a rectangular prism with periodic boundaries. A sphere of unit radius is attached to each point. In the first stage of the process, the radius of each sphere shrinks by 0.0001 units. The distance between each sphere and its nearest neighbour is found, and the closest pair defining overlapping spheres is identified. This pair are moved apart along a line joining the points until the spheres no longer overlap. When the distance between the closest two spheres drops below a threshold, all spheres have their diameter increased by 0.0002 units, and the process repeats. After 2000 cycles, a second routine of shrinking and translation removes all remaining overlaps. Once the initial configuration of points has been chosen, the algorithm is entirely deterministic. There are also versions of the algorithm which pack ellipsoids [85] and discs of two different radii [86].

The Jodrey-Tory algorithm can be initialized with one of its own previous outcomes. If this is done many times, then the configuration becomes denser and less disordered [87]. In experiments, packings subjected to shearing forces can also become much denser and less disordered [45], but this occurs by physical mechanisms that are not represented within the Jodrey-Tory algorithm.

Other deterministic rearrangement procedures use more complicated rearrangement rules which may depend on all neighbours [88] or on fixing the con-

tact network early in the rearrangement [89]. The latter strategy can be used to maximize disorder in planar disc packings [90].

Rearrangement algorithms can also be inspired by statistical physics models for the motion of ideal gas molecules. These models do not represent the physics of the condensation of a gas. Lubachevsky and Stillinger [91, 92] begin with points uniformly distributed within a region. Random velocities are assigned to each point, and each point begins to grow a disc at a constant rate. When discs collide, the collision is elastic and momentum and energy are conserved. After a few thousand iterations of disc expansion, the discs form a nearly packed arrangement. There is a three-dimensional version of this algorithm [93] and a version which can pack ellipsoids [94].

Rearrangement algorithms can also be written with non-periodic boundaries. They can pack spheres on the surface of a large sphere in  $\mathbb{R}^4$ , avoiding any boundary effects [50]. They can be used as the basis of programs to estimate the most efficient packing of a small hard-bounded region by a fixed number of spheres [24]. When combined with elements of ballistic algorithms, they can be used to pack cylinders [95].

### 3.5. Dynamic algorithms

Neither ballistic nor rearrangement algorithms are based on the physics of packing formation. Dynamic algorithms seek to model the trajectories and changes in momentum of individual grains as they form a packing.

To form a packing, flowing grains must expend their kinetic energy. This happens through the deformation of the packed grains and their container, but also through frictional contacts, through damage due to erosion and fracturing, through drag forces on the surrounding fluid, and through the actions of other forces. Few of these processes can be directly observed.

The classical hard-sphere simulation models used in molecular dynamics simulations [96] are unsatisfactory for granular flow. Inelastic hard-sphere collisions in classical models cause un-physical clumping [97]. The discrete element method (DEM) model of Cundall and Strack [98] avoids this problem by replacing the hard-sphere inelastic collision with an idealized inelastic collision between spheres with soft shells. Normal and tangential frictional forces during collision are modelled by the actions of a spring and a dashpot, which represent the deformation upon contact and the force of restitution which opposes the deformation (see Figure 5). The first DEM models for granular flow also included the effects of gravity and of van Der Waal's forces [99], but were unable to generate packings as dense as physical packings unless the frictional effects were eliminated.

Hundreds of DEM models for various aspects of granular flow have been constructed [100]. These models are based on a balance between a set of forces which depend on the material involved, the size of the particles, and the fluid through which they are flowing. Some forces, such as the electrostatic force and gravity, can be modelled in a single straightforward way. For frictional and drag

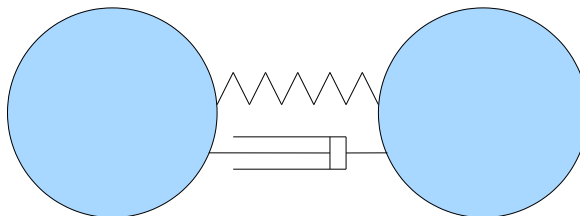


FIG 5. Inelastic collisions between spheres in a DEM model are represented by a spring and a dashpot. The dashpot imposes the deceleration associated with the initial collision and deformation, while the spring represents the force of restitution that restores the original shape.

forces, many different empirical and theoretically-based force models may be available [101]. Since the particle-fluid and interparticle interactions cannot be observed during granular flow, these DEM models are speculative. Without a thorough and objective way of assessing these models, there is no way to tell of if they properly represent the physics of granular flow.

#### 4. Descriptive statistics

Sample spaces for packing processes are subsets of very high dimensional spaces. To be able to recognize common aspects of realizations from the same process, it is necessary to find statistics which can summarize these aspects within individual realizations.

Most classical spatial statistics are designed for use on realizations of spatial processes which are both stationary and ergodic (SE). Packing processes cannot be assumed to be SE, but statistics intended for use on realizations of SE processes can be used to summarize some aspects of packings.

##### 4.1. Random set statistics

Sphere packing processes are examples of random closed sets. A random set  $\Phi$  can be formally represented by a collection of random indicator functions

$$\begin{aligned} I_{\Phi}(x) &= 1 \text{ if } x \in \Phi \\ &= 0 \text{ if } x \notin \Phi \end{aligned}$$

for all  $x \in \mathbb{R}^d$ . For the remainder of this section,  $\Phi$  will be assumed to be a packing process.

There is no formal expression for how the  $I_{\Phi}(x)$  are related to each other, but aspects of this dependence can be summarized by statistics intended to estimate the moments of SE random sets. The  $p^{\text{th}}$  moment  $M_p$  of  $\Phi$  is defined over the collection of  $p$  points  $\{x_1, \dots, x_p\} \in \mathbb{R}^d$  as

$$\begin{aligned} M_p(x_1, \dots, x_p) &= E[I_{\Phi}(x_1) \dots I_{\Phi}(x_p)] \\ &= Pr[x_1 \in \Phi \text{ and } x_2 \in \Phi \text{ and } \dots x_p \in \Phi]. \end{aligned}$$

These probabilities are defined as the limiting values of the relative frequency of their defining events over a large random sample of independent realizations of the packing process. In the physics literature, they are known as  $k$ -point correlation functions or as the  $k$ -point probability functions.

The expected value of the volume of fraction of spheres in a realization is a function of  $M_1(x)$ . To define a volume fraction for a realization, it is necessary to define a region  $A$  in which the packing lies. If the packing is rigidly enclosed by a boundary, then  $A$  would be the interior of the bounded region. If the boundary took the form of a cup or a surface, it would be necessary to create a realization-specific region based on the convex hull of the realization or some similar structure. For the packing process  $\Phi$ , the volume fraction for a realization is

$$\widehat{\phi}_1 = \frac{|\Phi \cap A|}{|A|}$$

where  $|\cdot|$  indicates volume. If a random set is SE and  $A$  is non-random,  $E[\widehat{\phi}_1] = m_1$ , where  $m_1$  is the common value of  $M_1(x)$  at all  $x \in \mathbb{R}^d$ . If  $\Phi$  is not SE and  $A$  is not random, then  $\widehat{\phi}_1$  estimates

$$\frac{1}{|A|} \int_A M_1(t) dt,$$

where  $t \in \mathbb{R}^d$ .

In the physics literature on packings, it is often assumed that the packing is large, that boundary effects are insignificant, and that the packing process is SE. There is no distinction made between a parameter of the packing process, the expected value of the parameter estimate, and the observed value of the parameter estimate from a single large sample. For the volume fraction, the implicit assumption is that  $\widehat{\phi} = E[\widehat{\phi}_1] = m_1$ . Assumptions of this kind are also made for other descriptive statistics for realizations.

Estimators for higher moments of SE random sets can be used as descriptive statistics for random sets. If a random set is SE, its reduced second moment for a vector  $x \in \mathbb{R}^d$  is

$$m_2(x) = M_2(0, x) = E[I_\Phi(0)I_\Phi(x)] = Pr[0 \in \Phi \text{ and } x \in \Phi]$$

which can be estimated by

$$\widehat{\phi}_2(x) = \frac{|\widehat{\Phi} \cap A \cap \widehat{\Phi}_x \cap A_x|}{|A \cap A_x|}$$

where  $A_t = \{x \in \mathbb{R}^d : x + t \in A\}$ . Third- and fourth-moment-based estimators can also be defined.

The most commonly used descriptive statistic for packings is  $\widehat{\phi}_1$ . It showed that the earliest simulation algorithms were simulating looser packings than random close packings. Later simulation algorithms were judged by their ability to attain estimated volume fractions close to those of a random close packing [83].



The volume fraction cannot be used alone for characterizing packing processes, since it reveals nothing about interactions between spheres. Local estimates of  $\widehat{\phi}_1$  can be used to describe differences in structure within non-stationary sphere packings [55, 102].

The statistic  $\widehat{\phi}_2(x)$  is rarely used. Instead, point process statistics are used to describe sphere interactions. For monodisperse sphere packings, there is a close relationship between  $\widehat{\phi}_2(x)$  and the pair correlation function for the sphere centres [103].

Moment statistics can be calculated for transformed random sets. If the spheres grow at a constant rate until they fill space,  $\widehat{\phi}_1$  and the Euler-Poincaré coefficient can be plotted as a function of the degree of expansion [104, 105].

The complement of the packing is its void structure. In an SE random set, this structure can be described by estimates of the spherical contact distribution function  $S(r)$ , which is also known as the pore size distribution function [80, 89]. Given an arbitrary point  $x$  in the complement of an SE random set,  $S(r)$  is the probability that the nearest point in the random set to  $x$  lies within a distance  $r$  of  $x$ . It can be estimated by finding the distance to the nearest point in the random set from many locations in the void and using these distances to construct an empirical cumulative distribution function. It has been used to compare physical packings with simulated packings and to investigate the applicability to packings of theoretical approximations for  $S(r)$  which arise in statistical mechanics [106].

#### 4.2. Point process statistics

When the packed objects are spherical, the centres of the spheres form a realization of a point process which summarizes many important aspects of the structure of the packing. As in the case of random set statistics, many point process statistics were originally defined for use with SE point processes.

The basic point process statistics are estimators of functions of the Palm measure and of the first and second moment measures of SE point processes [107]. It is difficult to estimate statistics based on the third moment measure, but statistics based on the number of  $r$ -close triples in a realization have been constructed [108].

The intensity  $\lambda$ , defined for a SE process to be the mean number of points per unit volume, is seldom estimated since  $\lambda$  is a constant multiple of  $\widehat{\phi}_1$  for monosized sphere packings. Local estimates of intensity have been used to investigate the internal structure of large physical packings [36].

The  $K$ -function  $K(r)$  is the second reduced moment function of a SE point process. The quantity  $\lambda K(r)$  is the expected number of points in a disc of radius  $r$  about a point in a realization. The central point of the disc is excluded from the count. The pair correlation function  $g(r)$  is defined by

$$g(r) = \frac{1}{db_d r^{d-1}} \frac{\partial K(r)}{\partial r},$$

where  $b_d$  is the dimension of the unit ball in  $\mathbb{R}^d$ . The pair correlation function may be also be referred to as the radial distribution function, although that name is also applied to the quantity  $RDF(r) = \lambda db_d r^{d-1} g(r)$ . Statistics based on  $g(r)$  are often used in the physics literature to describe point interactions within realizations.

Estimates of  $g(r)$  for isotropic SE point processes are constructed as follows. A sequence of concentric spherical shells differing in radius by  $\delta > 0$  are constructed around each observed point. At distances  $k\delta$ , the total number of points between the shells of radius  $(k-1)\delta$  and  $k\delta$  are averaged over all observed points and then divided by the volume of the shell at distance  $k\delta$ . These values are then presented as a histogram, which is an estimate of  $g(r)$ . For isotropic SE point patterns it is necessary to make edge corrections to estimates of  $g(r)$  [109], since the shells centered at points near the edge of the observation window will extend beyond the window edge. These corrections could be omitted, provided that every window has exactly the same size, shape, and position when applied to the realizations. If corrections are not applied, the best choice is to only use shells which lie entirely within the window.

An estimate of  $g(r)$  for a large simulated sphere packing is shown in Figure 6. This type of plot has been used to show the effects on packing structure of increasing the relative size of the Van Der Waals force in a DEM model [110].

Other statistics can be used to describe point interactions in realizations. If  $x$  is any observed point in a realization of an isotropic SE process, then the nearest-neighbour function  $D(r)$  at  $r$  is the probability that the nearest point in the realization to  $x$  is within a distance  $r$  of  $x$ . If  $x$  is taken to be an arbitrary point in space instead of an observed point, then the empty space function  $H_s(r)$  at  $r$  is the probability that the nearest point in the realization to  $x$  is within a distance  $r$  of  $x$ . For the Poisson process, these two probabilities are identical. For monosized sphere packings,  $D(r)$  is certain to be a delta function located at the diameter of a sphere. More informative are estimated distributions for distances to the  $k$ -nearest neighbour [90], where  $k$  is larger than the minimum number of spheres required to lock a particular sphere into a mechanically stable position.

### 4.3. Statistics based on triangulations and tessellations

Statistics based on triangulations and tessellations of sphere centres were initially applied to packings by physicists who were using physical packings as models for the molecular structure of liquids and amorphous solids [9, 26]. Near neighbours can be clearly defined as spheres whose centres are connected by a triangulation edge, and the contact network of the packing is a subgraph of the triangulation. The tessellation is the dual graph of the triangulation and is a simplified description of the void structure of the packing.

The Delaunay triangulation and the Voronoi tessellation are generally used as bases for statistics. The tessellation is constructed by finding all points in  $\mathbb{R}^d$  which are equidistant between sphere centres. Euclidean distance is used in the construction, but other distance measures can be used to generate different

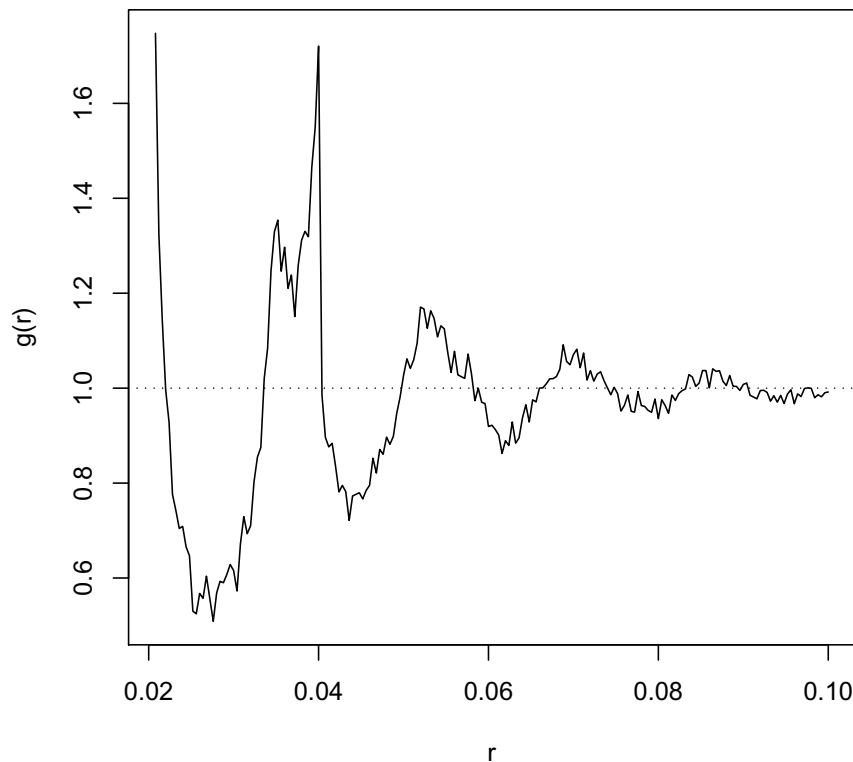


FIG 6. Plot of the estimated pair correlation function  $g(r)$  for the centres of spheres in a simulated packing of 3456 monosized spheres of diameter 0.02 [111]. The curve has a singularity at one sphere diameter, and has local maxima at  $\sqrt{3}$  and 2 sphere diameters. The function is asymptotic to 1 at large distances.

tessellation structures [112]. The triangulation is the dual of the tessellation, generated by joining pairs of sphere centres which define a tessellation edge. Figure 8 shows both the triangulation and tessellation generated by the centres of the spheres in Figure 7. Figure 2 shows a tessellation skeletonizing the void space around packed spheres.

The simplest statistics that can be extracted from triangulations and tessellations are lists of characteristics for each cell. For both types of cells, the area, the perimeter, the largest and smallest angles, and the longest and shortest sides can be found. The coordination number of each sphere is defined to be the number of its triangulation edges that belong to the contact network.

Statistics are calculated for each cell and then summarized either by a list of summary statistics (mean, standard deviation, minimum, and maximum) or by a histogram. In the physics literature, the histograms are often referred to as plots of the distribution of the statistic. This statement is misleading, since the histogram is summarizing a list of local features of a single observation unless the process is SE. Figure 9 shows the histogram for the cell volumes of

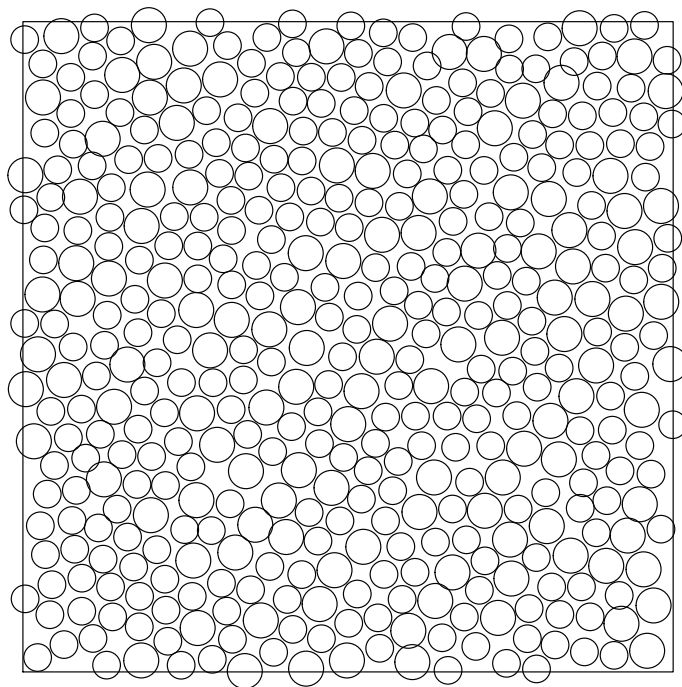


FIG 7. A representation of the arrangement of packed discs in a subset of the packing seen in Fig. 1. Measurement error associated with the image capture process makes it impossible to determine which pairs of spheres are in contact.

the triangulation shown in Figure 8. Gamma density functions have been used to summarize the shape of histograms for standardized [113] and unstandardized [90] Voronoi cell volumes. Voronoi cell volumes have been proven to be Gamma distributed for tessellations generated by homogeneous Poisson point processes [114].

Tessellation statistics have been used to investigate the differences between realizations of physical and simulated packings. Plots can be made of mean coordination number versus  $\widehat{\phi}_1$  [36, 115], or of the standard deviations of Voronoi cell face area and volume against  $\widehat{\phi}_1$  [116]. Comparisons of histograms of coordination numbers are not powerful enough to clearly distinguish realizations from different simulation algorithms [60].

Measurement errors in position observations and disc size observations can prevent clear identification of pairs of contacting discs. When these errors occur, the contact network cannot be accurately constructed. Figure 7 shows part of the packing from Figure 1 after reconstruction from a digital image. While the topological structure of its associated triangulation (Fig. 8) can be captured, accurate extraction of the contact network is impossible.

There are often a small number of spheres within the interior of a physical packing which are not rigidly held in place by other spheres. Triangulation-

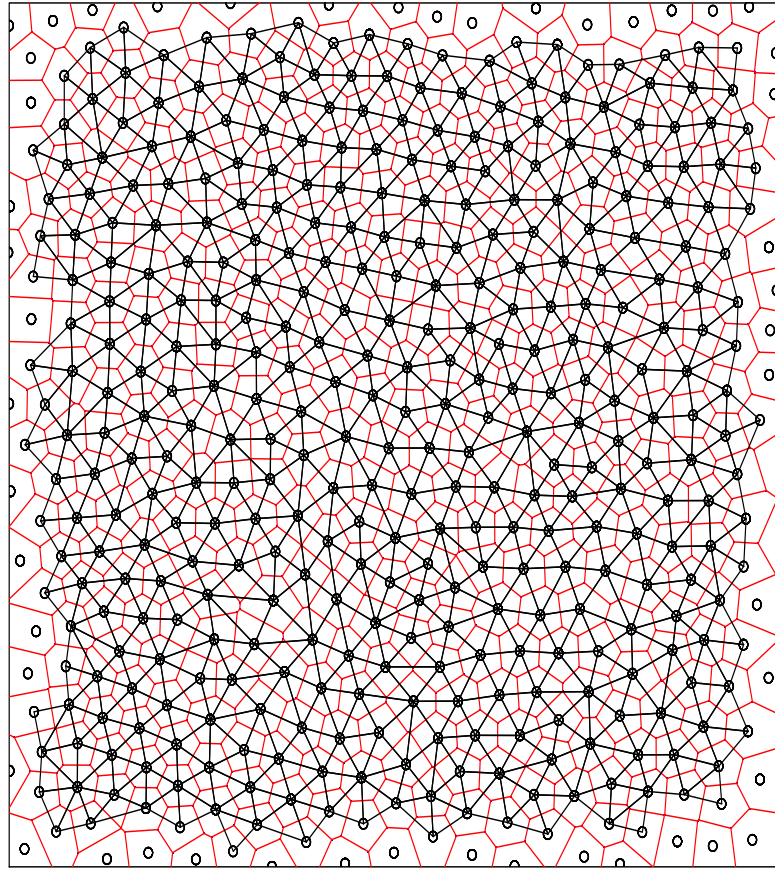


FIG 8. Voronoi tessellation (red) and Delaunay triangulation (black) for the centres of the spheres in Fig. 7. The triangulation connects each sphere centre with its near neighbours and the tessellation summarizes the void structure around the discs.

based statistics can also be used to identify these spheres, which are known as rattlers. The fraction of rattlers in a packing can be used as a measure of packing efficiency [88]. Identification of rattlers is complicated by uncertainty as to whether or not neighbouring spheres are in contact.

More elaborate statistics can be developed from the tessellation and the triangulation. The local density of a packing can be defined as the ratio of the sphere volume to the volume of its Voronoi cell [36]. The escape fraction statistic is the empirical cumulative distribution function of the diameter of the largest sphere that could escape from each tessellation cell through the gaps between neighbouring spheres. It has been shown to distinguish between physical packings of different volume fraction [36]. A topological density has been defined which is based on a notion of topological distance [117, 118]. This distance is defined by choosing a sphere, and then identifying a sequence of shells radiating out from

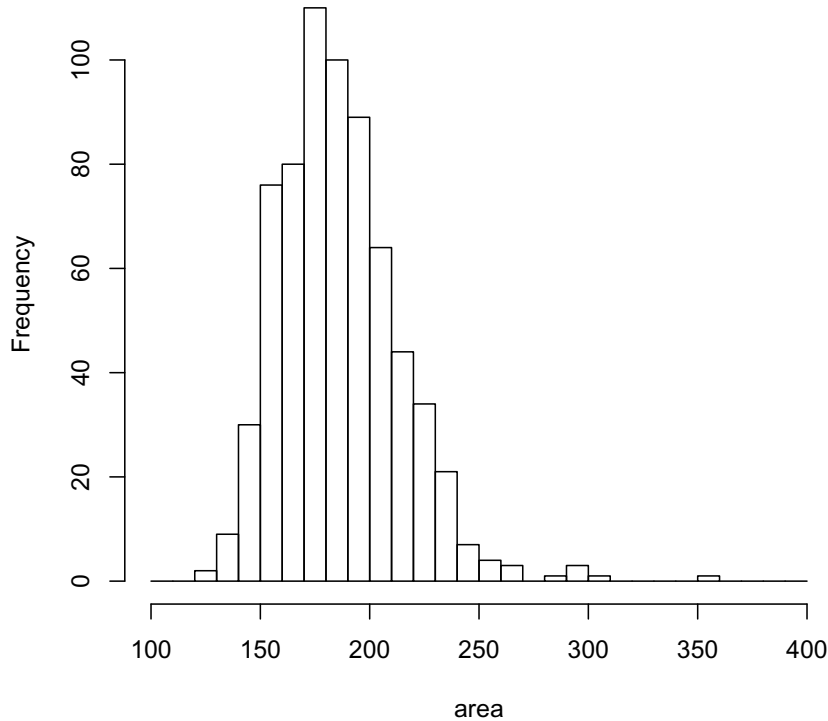


FIG 9. Histogram of cell volumes from the triangulation in Figure 8. The shape of the cell volume histogram can be summarized by a translated Gamma density with  $\alpha = 3.95$  and a scale parameter of 13.92. This gamma density does not represent the distribution of any clearly defined random variable, but instead summarizes information contained in a long list of highly dependent local measurements taken on a single observed packing.

it. The first shell contains only those spheres which touch the central sphere, the second contains all spheres in contact with the first shell but not contained within the first shell, and so on. The density is defined as the leading coefficient of the quadratic fit of the number of spheres in each shell to the shell number. The smallest values of the topological density are those of point lattices.

#### 4.4. Statistics based on local order

When physical packings were first proposed as models for the molecular structure of liquids, researchers sought to determine whether or not packings possessed some form of local order [7]. This local order would take the form of small subunits with near-lattice structure, combined in a complicated way to produce the general disorder of the packing.

The presence of locally-ordered structures within point patterns may not be obvious to the eye. Materials have been found in nature which are quasi-crystalline [119, 120]. The arrangement of atoms in these materials would appear

disordered to the eye, but x-ray diffraction reveals that their structure can be modelled by a projection into  $\mathbb{R}^3$  of a higher dimensional point lattice.

It is necessary to distinguish between topological and geometric concepts of order. A packing is topologically ordered if the sphere centres can be continuously translated so as to transform its Delaunay triangulation into a point lattice without breaking any bonds [121]. In a planar packing, topological defects of the lattice structure can be easily identified and counted [122].

A packing is geometrically disordered if its Delaunay triangulation differs topologically from that of a point lattice. Statistics which quantify local order within geometric disorder are based on local measurements which identify the presence of point lattice structure. Statistics can be derived from the locations of contact points on individual spheres, expressed in spherical coordinates. The fourth- and sixth-order spherical harmonic functions can be evaluated for the contact points on each sphere, and then averaged either over the individual spheres or over many spheres. The fourth-order harmonics have non-zero averages in the presence of local cubic lattice structure, while the sixth-order harmonics have non-zero averages in the presence of local hexagonal close-packed lattice structure. Averages over single spheres have been used to study the structure of very large physical packings [36]. Averages over many spheres were originally developed to study the emergence of crystallization in simulated liquids [123, 124], and improved averages have been used to compare realizations of simulated sphere packings [43]. Averages over many spheres based on the sixth harmonic were used to describe the reduction in disorder observed in simulated sphere packings seen in long runs of the Jodrey-Tory packing simulation algorithm [125]. Two different many-sphere averages of the sixth harmonic have been used to compare realizations from three different rearrangement models [126]. Neither average was powerful enough to distinguish between realizations from different models.

Statistics based on spherical harmonics cannot identify ordered structure that is found within small clusters of neighbouring spheres. Statistics which reveal this type of ordered structure can be constructed using the side lengths of Delaunay simplices, which are tetrahedra formed by four Delaunay triangles which share common edges [127]. In studies of the crystallization of packings over long runs of the Jodrey-Tory algorithm, measures of tetrahedrality and quadrotahedrality were found to be more powerful at tracking changes than were the averages of spherical harmonics [87].

#### *4.5. Statistics based on models for physical properties*

Mathematical models for physical phenomena can be applied out of physical context to yield new statistics. Suppose that a specimen of composite material has the structure of a sphere packing. The inclusion phase is represented by the spheres, while the matrix phase is represented by the void around the spheres. If the two phases have different physical properties, then the bulk physical properties of the composite are found by solving a set of differential equations that use

the packing structure as a boundary condition. These bulk properties describe the structure of the packing in a very different way from moment-based and traditional tessellation-based statistics.

#### 4.5.1. Statistics based on frictionless flow

In frictionless flow, a fluid experiences no internal resistance due to shear. Heat flow by means of conduction and the flow of electricity are examples. Models for frictionless flow are relatively simple to construct.

For electrical flow, the packing and its complement can be considered to be two materials with differing electrical resistivity. On the surface of the packing, two disjoint sets of spheres can be considered to be connected to electrodes of infinite conductance. When a unit DC potential is placed between these electrodes, the potential can be calculated at all points within the packing and its complement. From this, a bulk resistance for the composite can be calculated. This resistance defines a mean distance across the packing [128], whose form is determined both by the relative resistivity of the two phases and by the disordered structure of the packing.

For a composite modelled by a disordered packing, the bulk resistance is difficult to calculate. There are no exact methods, and numerical methods require that the packing be discretized very crudely. If equal resistance is assigned to all of the edges of the contact network, then the potential at all vertices of the circuit can be found easily by means of the properties of random walks through the network [129]. The bulk resistance is a weighted average of potentials at the electrode regions. If the network lies in the plane, the potentials can be plotted and used as a diagnostic tool. A plot of the current along each edge, as calculated from potential differences between the defining vertices, is more useful than the potential as a descriptor of structure. Bulk resistance has been used to develop a test for the presence of anisotropy in sphere packings [130].

Models for heat conduction can also be used. The spheres in a packing can be expanded in order to generate contact surfaces between neighbouring spheres. A plot of the bulk heat conductance of a packing as a function of the degree of spherical expansion has been used to distinguish between packings generated by different models [131].

#### 4.5.2. Statistics based on shearing flows

If a fluid flows through a fixed packing, it develops internal frictional losses which depend on the void structure of the packing. If the packing itself is made to flow as a powder, then its flow is strongly affected by frictional losses arising from collisions between spheres. These losses greatly complicate the modeling of the flows.

If the packing is considered to be fixed, the flow of a fluid through its complement can be modeled. All fluid flow is assumed to be laminar, since modeling turbulent flow through a complex structure is impractical. Major simplifications



are required in order to be able to calculate bulk flow properties. The complement of the packing can be represented by a piping network whose structure is determined from the Voronoi tessellation, and pipe resistances can be assigned on the basis of local void geometry [19]. The velocity profile and pressure gradient of the simulated flow can be used as statistics. If the diffusion of contaminants through the flow is modeled [132], many different statistics related to contaminant flux and concentration can be calculated.

If the packing itself flows, this flow can take place in a vacuum, in air, in a liquid, or in both air and liquid. These flows can be modeled using DEM models which represent compaction processes that do not induce particle fracture [133, 134], flow in mixers and drums [135–137], and flow during avalanches [138, 139]. Flow in mutiaxial [53, 140, 141], and shear test apparatus [142, 143] can also be simulated. The most useful statistics would emerge from models for flows in which the spheres are nearly packed and the triangulation changes gradually over time. Flows of fluidized packings would be less useful, since the triangulation would be rearranged by the fluidization process. A plot of the number of sliding contacts between spheres as a function of the total number of contacts during simulated flow can distinguish between realizations of different packing models [144].

#### 4.5.3. Thermodynamic statistics

Given a packing of spheres in a region with periodic boundaries, each sphere can be very slightly shrunken in place. The shrunken spheres can be acted upon by a pre-specified potential, and the motion of the particles under that potential can be tracked. Algorithms for simulating these motions have been developed as part of thermodynamic models for hard-sphere atoms [145–147], and also for non-spherical shapes [58]. Once these algorithms have been run through many time steps, statistics representing pressure, temperature, and other thermodynamic quantities can be calculated.

#### 4.6. Graphical methods

Graphs and images can be used to reveal internal structure that can be used as the basis of new statistics.

A simple plot of the discs in a realization can reveal the near-ordered arrangements of planar packings of monosized discs. The Delaunay triangulation can be plotted on top of the discs, and triangles can be coloured to identify triples of discs in mutual contact [93]. Edges of the triangulation which are associated with defects can be coloured to illustrate the overall structure of those defects in large packings [71].

To produce images of the arrangements of spheres, it is necessary to use ray tracing to induce shadows on sphere surfaces. Colours can also be used to identify spheres with unusual local properties (see Fig. 2). Specialized software such as the Voro++ package is needed to produce these plots [1].

#### 4.7. Other statistics

Descriptive statistics for packings of  $n$  spheres must summarize features of a subset of a very high dimensional space. There is no theory that establishes that the statistics listed in the previous sections are sufficient to accomplish this. To find additional statistics, it is possible to examine unusual experimental results, model possible mechanisms to explain these results, and then develop statistics based on these models.

Agitation of granular materials can cause size segregation [148, 149]. While intuition suggests that large dense objects should settle while small light objects would rise, the opposite can happen in mixtures of nuts containing large and dense Brazil nuts [49]. Simulated sphere packings have been used to investigate the paradox [150, 151]. These simulation experiments could be used as sources of new statistics for polydisperse packings.

#### 4.8. Smoothly defined and structurally defined statistics

Many descriptive statistics for disordered packings are lacking in power and are difficult to interpret on account of how they are defined. Suppose that a statistic is based on measurements taken at many different locations within a single packing. This statistic will be *smoothly defined* if it assigns equal importance to each of the measurements on which the statistic is based.

To estimate the 2-point correlation function  $\widehat{\phi}_2(r)$  for a disc packing, a fine regular lattice of  $N$  observation points can be imposed over the packing. At each observation point, a value of 1 is assigned if the point and its translate by a vector of length  $r$  are both in a disc. Otherwise, the value 0 is assigned. This procedure would be repeated for  $k$  different equally-spaced orientations of a vector of length  $r$  at each observation point. The estimate is constructed by taking the average of the  $Nk$  values. While the statistic does contain information about the internal dependence structure of the packing, its construction mixes local measurements of dependence with no regard for the overall disordered arrangement of the spheres.

Other smoothly defined statistics can be constructed from the Delaunay triangulation or the Voronoi tessellation. When a histogram is made of all triangle areas from a triangulation, all information about how the triangles fit together is lost. The same is true for averages of triangle areas, their standard deviations, and their extreme values.

Statistics based on physical properties differ from smoothly averaging statistics in that their construction depends on the disordered arrangement of the spheres alone. Bulk electrical resistance can be thought of as a distance across a network through which current flows. This distance is based on a weighted average of all the possible paths that could be taken across the network, with the weights being chosen to be consistent with the laws of electricity and the paths being defined by the disordered structure of the network. Any statistic based on path properties of a random walk or on the properties associated with

mechanical deformation of the packing would also be dependent on the entire packing structure. The form of these statistics are *structurally determined* by the disordered arrangement of the spheres.

Statistics which are smoothly defined are ideal for use with the Poisson process, where there is no interaction between the points in a realization. For a point process defined by the centres of packed spheres, smoothly defined statistics are often estimated on a length scale which is much smaller than that of the individual sphere diameters. Information about sphere shape becomes confounded with interaction effects. This confounding makes the value of the statistic difficult to interpret in terms of features of the realization which can be seen. When physics-based statistics are used on packings, all of the information is captured on the same length scale as the spheres. There is a vast literature associated with electrical, granular-flow, and fluid-flow properties which relates those properties to easily-visualized physical phenomena.

#### 4.9. Asymptotics

Finding the exact distribution of point process statistics is very difficult or impossible, except in the case of the Poisson process. Conventional asymptotic methods can be used to find between-realization distributions for some statistics when a large number of packings are observed. It is also possible to take thermodynamic limits by increasing the number of spheres in the packing on which the statistic is calculated.

Laws of large numbers based on thermodynamic limits are of great importance in materials science and physics. If a composite can be modeled by a packing with the spheres and the void having different electrical resistances, then sufficiently large specimens will have the same bulk resistance regardless of the structure of the packing. In the case of ideal gases, equilibrium statistical thermodynamics [152–154] derives limit laws from probabilistic models of atomic behaviour. The limiting quantities, experienced as pressure, temperature, volume, and entropy, show no signs of variability arising from the dynamic chaos present at the atomic level.

The proofs of central limit theorems and laws of large numbers in thermodynamic limits require that the dependence of one sphere's location on that of another declines sufficiently fast as the distance between the spheres increases. For a ballistic deposition model, limit theorems have been proven for statistics describing the time evolution of the packing height as the area over which spheres are deposited increases [155]. Attempts have been made to construct traditional statistical physics models for packings [156, 157]. These attempts are based on the assumption that all packed configurations are equally likely to occur, which is contradicted by experimental evidence [31].

Thermodynamic limits are not necessarily useful in all applications. If a tube has a diameter of 15 *cm*, packing it with 1 *cm* diameter plastic spheres is much different from packing it with plastic spheres having a diameter of 10  $\mu\text{m}$ . Interatomic and electrostatic forces will cause packings of very small spheres to

have a much different structure than packings whose formation is governed by frictional forces alone.

The mathematical intractability of probabilistic limit laws for disordered spatial patterns has resulted in the development of alternative approaches to finding thermodynamic limits for statistics. If a composite is modeled by a lattice packing of spheres, then methods from potential theory can be used to get very good approximations for the bulk electrical conductance [158, 159] based on the conductance of individual spheres. When the packing is disordered, these methods cannot be used on account of lack of symmetry. Homogenization [160, 161] can be used to find the limiting behaviour of statistics defined by differential equations which have spatially varying boundary conditions. Often, these results are presented as bounds between which the limiting value of the statistic must fall. While the mathematical arguments behind homogenization represent an elegant use of the calculus of variations, it is unclear that the underlying assumptions for these arguments can be satisfied by any real material. In one case, a mathematical argument has been constructed to establish that a structure exists which has a particular homogenization limit for bulk electrical resistance [162].

## 5. Inference

The central unsolved problem in statistical inference for packing processes is how to assess the adequacy of a fitted model. If this assessment is undertaken carelessly or not at all, there is no basis for assuming that the realizations of the model represent any aspect of reality.

Packing process models can be fit by means of the method of minimum contrast [163]. Parameter estimates are chosen by minimizing the difference between a statistic calculated from the model and a statistic calculated from the data. Often, the statistic chosen is the  $K$ -function. This method ensures that there is some resemblance between the model output and the data, but it says nothing about how far that resemblance can be extended. Since the  $K$ -function lacks the power to distinguish point patterns which can be distinguished by eye [164], there is no reason to believe that a fit based on it will result in a good fit for other statistics calculated from the packing.

It is not possible to avoid assessing a model for a packing process by claiming that the model is derived from physical first principles. Physical models may form packings by a different mechanism than does the physical packing process that the model is supposed to represent. Ballistic and re-arrangement models construct packings by methods which have no connection to physical processes. DEM models depend on assumed models for unobserved interactions between spheres that drive packing formation.

The type of model assessment required depends on the purpose of the model. The physical property of interest for the packing must be summarized by a response statistic, ideally one which is univariate. If the model is needed solely to make predictions and the fitted model reliably gives highly accurate predictions in all of the circumstances where it is needed, then no further model assessment

may be necessary. If the model is needed to explain how the response emerges from the detailed structure of the packing, if the fitted model predictions are not accurate, or if a highly robust prediction model is needed, then it is necessary to engage in an extensive assessment of the fitted model.

Assessment of fitted models is only possible if a significant amount of the variability in the response statistic can be explained by a small number of descriptive statistics. If response variability is explained by many different mechanisms that require dozens of statistics to describe, then it is unlikely that any assessment will be possible, or that any useful model will be constructible. The extremely high dimension and complexity of the sample space implies that there is no guarantee that a low-dimensional collection of explanatory descriptive statistics will exist.

Finding the explanatory statistics which explain response variability is the single most difficult problem in assessing the models. It is analogous to establishing that the properties of an ideal gas at equilibrium can be described primarily by its temperature, its volume, its pressure, and the number of atoms. No theory has been developed to establish how many explanatory statistics are needed to explain a response, nor is there any clear way to find these statistics. They need to be discovered, as the ideal gas law was discovered, by doing many experiments and devising new summarizations of the important aspects of the experimental results. Instrumentation must be available to ensure that the statistics can be accurately measured, especially in the case of three-dimensional packings.

Since the model will not be able to exactly reproduce packings observed in experiments, values of the explanatory statistics from one observed packing cannot be compared with statistics from a single simulated packing. Instead, model evaluation must be based on comparing the joint distributions of the explanatory statistics arising from the model and arising from the data. If a comprehensive collection of explanatory variables has been found and there is no strong evidence that the two joint distributions differ, then the model can be considered to represent the observed physical packing process.

Some progress has been made towards the development of methods for model assessment in packings. Various methods have been developed for describing deviations from complete spatial randomness in stationary point patterns [165], but it is not clear if these methods will be useful for distinguishing between different types of non-Poisson disorder. Residuals for point processes have been developed [166] which can be used to detect spatial trend and interpoint interaction effects. Physicists have used descriptive statistics to quantify differences between simulation models for packings [167–169] and between different types of physical packings [36]. Descriptive statistics have also been used to quantify the effects of changing parameters in DEM models [170].

## 6. Conclusions

Sphere packing processes challenge basic working assumptions used in both spatial statistical inference and in the fields of application.

There are no formal models for packing processes, and so Bayesian and likelihood-based inference are impossible. Physical models and simulation algorithms are the only models available, and they cannot be analyzed using formal mathematical arguments. All forms of inference must be based on comparing a random sample from the model with a random sample from an experiment. No useful information can be found from a single large realization, since the processes being studied are neither stationary nor ergodic nor isotropic. In any comparison of a single large observation with a single packing from a model, there will be no way of knowing which attributes of the packings are typical and which arise from between-realization variability.

The internal disorder within a realization cannot be represented by a random model, as is the case with Markov spatial processes. Instead, the between-sample variance in response statistics must be explained by descriptive statistics which summarize aspects of the disorder within each realization. Existing descriptive statistics are mostly constructed for stationary and isotropic processes, and so are unlikely to be able to fully capture the effects of within-realization inhomogeneity. New statistics will be required to for particular responses, and general theories for the physical properties of packings will only emerge if common sets of descriptive statistics can be used to explain many different responses in many different physical systems.

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