

Peer Review Information

Journal: Nature Computational Science

Manuscript Title: Identification and analysis of 3D-pores in packed particulate materials

Corresponding author name(s): Professor Tatiana Segura

Editorial Notes:

Transferred manuscripts This manuscript has been previously reviewed at another journal that is not operating a transparent peer review scheme. This document only contains reviewer comments, rebuttal and decision letters for versions considered at Nature Computational Science.

Reviewer Comments & Decisions:

Decision Letter, initial version:
--

Date: 23rd June 23 05:01:33

Last Sent: 23rd June 23 05:01:33

Triggered By: Jie Pan

From: jie.pan@us.nature.com

To: tatiana.segura@duke.edu

BCC: jie.pan@us.nature.com

Subject: Decision on Nature Computational Science manuscript NATCOMPUTSCI-23-0461-T

Message: ** Please ensure you delete the link to your author homepage in this e-mail if you wish to forward it to your co-authors. **

Dear Professor Segura,

Your manuscript "The empty space of packed particulate materials" has now been seen by 4 referees, whose comments are appended below. You will see that while they find your work of interest, they have raised points that need to be addressed before we can make a decision on publication.

The referees' reports seem to be quite clear. Naturally, we will need you to address

all of the points raised.

While we ask you to address all of the points raised, the following points need to be substantially worked on:

- Please provide benchmark results to demonstrate the benefits. When discussing your results, please avoid overhypes.
- Please follow our referees' suggestion to better discuss the literature, especially those missed references suggested by referees.
- Please carefully address our referees' comments about the code and datasets. Following the suggestion from one of the referees, we recommend to use open-source programming languages, such as python, for implementing your computational framework.
- Please address various technical issues raised by our referees, including clarification and justification about the technical points/assumptions/choice of parameters, robustness of the framework, additional statistical tests, etc.
- The author Peter Cheng is from a company. Please update the competing interest statement accordingly in your revised manuscript.

Please use the following link to submit your revised manuscript and a point-by-point response to the referees' comments (which should be in a separate document to any cover letter):

[REDACTED]

** This url links to your confidential homepage and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this e-mail to co-authors, please delete this link to your homepage first. **

To aid in the review process, we would appreciate it if you could also provide a copy of your manuscript files that indicates your revisions by making use of Track Changes or similar mark-up tools. Please also ensure that all correspondence is marked with your Nature Computational Science reference number in the subject line.

In addition, please make sure to upload a Word Document or LaTeX version of your text, to assist us in the editorial stage.

If you have any issues when updating your Code Ocean capsule during the revision process, please email the Code Ocean support team Cc'ing me.

To improve transparency in authorship, we request that all authors identified as 'corresponding author' on published papers create and link their Open Researcher and Contributor Identifier (ORCID) with their account on the Manuscript Tracking System (MTS), prior to acceptance. ORCID helps the scientific community achieve unambiguous attribution of all scholarly contributions. You can create and link your ORCID from the home page of the MTS by clicking on 'Modify my Springer Nature account'. For more information please visit <http://www.springernature.com/orcid>.

We hope to receive your revised paper within three weeks. If you cannot send it within this time, please let us know.

We look forward to hearing from you soon.

Best regards,

Jie Pan, Ph.D.
Senior Editor
Nature Computational Science

Reviewers comments:

Reviewer #1 (Remarks to the Author):

This manuscript presents a voxel-based method to identify pores in particle packing. In addition, voxels that are equidistant to several surrounding particles are identified, which form "ridges" and "peaks" based on the number of surrounding particles. To my knowledge, pores in particle packing have been studied a lot. The commonly used method based on Voronoi and Delaunay, which are duals, can already identify various pore properties. According to the definition, the "ridges" and "peaks" proposed in this work are corresponding to the Voronoi cell edge and vertex. They are not new concepts.

In addition, several methods are already proposed to perform Voronoi analysis for particle packing, including non-spherical particles. Note these methods consider particles as geometry bodies but not points, and some methods also use voxels. Essentially these methods give similar results according to the same mathematical definition.

The results obtained from the method are neither new nor significant. The equations are not numbered, so I have to cite the page number and line number below:
page 9, line 216. This equation may be new, but it just shows the relationship between pore number and packing fraction. This relationship seemingly is not very meaningful, and it depends on the definition of a pore.
page 9, line 224. This equation is the data fitting for the median pore volume and packing fraction. These two parameters both describe the void in a packing. It is not strange they have such a simple relationship.
page 11, line 268. This equation is the data fitting for pore volume and the number of surrounding particles. Similar data can be found in the previous studies on the relationships between different Voronoi and Delaunay properties.
In the literature, there are many studies on the relationships between Delaunay/Voronoi properties and packing fraction. Some are based on theoretical analysis. Comparably, this work does not provide new insights.

Reviewer #2 (Remarks to the Author):

The authors have developed an application (LOVAMAP) for the characterization of void space in assemblies of granular particles. To the reviewer's knowledge and evaluation, the work is thorough, novel, and internally consistent. The paper is well-

written, figures are informative and very information-dense, and appropriate supplementary information is provided to allow the reader to reproduce much of the authors' work. The topic is likely of interest to the Journal's readers.

This manuscript is likely suitable for publication, but I suggest that the items listed below should be considered and addressed, as appropriate, before a final decision can be made. There are a number of instances where additional clarity would be welcome, and in some cases, I have questions about technical details of the work. Robustness in the face of several variables and asymptotic behavior are not always well-demonstrated. Specific comments and questions are detailed below.

****General****

1. On L410 of SI, the authors state that $1 \mu\text{m}^3 = 1 \text{ pL}$. This is incorrect: $1 \mu\text{m}^3 = 10^{-18} \text{ m}^3 = 10^{-15} \text{ L} = 1 \text{ fL}$. Is this a typo isolated to this line? Or is this unit glitch pervasive through the manuscript?
2. Given that specimen size remains constant at $(600 \mu\text{m})^3$ while particle sizes and shapes change, can all assemblies be considered as REV's? If the sizes of the assemblies were scaled up by an order of magnitude (for constant particle size), would results or interpretations change?
3. Some of the comparative metrics used by the authors are somewhat confusing in that they are absolute -- e.g., # of pores. Wouldn't these quantities be better expressed in a normalized fashion?
4. Pursuant to (2) and (3) above, I suspect that I might be missing something obvious, but it would be helpful if the authors were more explicit about when and how they are normalizing some of their measurements. Are there other cases where clarity could be achieved through normalization?
5. The authors provide few details about the physics of specimen assembly, particularly for the reader unfamiliar with the proprietary Houdini software. Contact mechanics, particle properties, damping, time scaling, inertial number, and convergence criteria (among others) will all have a significant influence on the equilibrated state of the particle assemblies. How sensitive are the authors' findings to these metrics?
6. Line 16 -- "significant leap forward" is subjective at best and hyperbolic at worst. Suggest removing or toning down.
7. Line 187 -- "state of the art" is subjective and difficult to quantify or substantiate. Suggest removing.
8. Is LOVAMAP an acronym? For what?
9. EDF16 -- if assembly size is constant, aren't (a) and (c) just proxies for number of particles? That is, aren't all three figures effectively equivalent?
10. The authors have elected to develop LOVAMAP using proprietary software, MATLAB. Why not use something open source like Python or Julia? Is there specific functionality required by LOVAMAP that is only available in MATLAB? Will the application run on Octave? (I appreciate that this question is largely philosophical, but I am genuinely curious about functionality.)
11. The authors have done a commendable job in studying and reporting the existing literature on the topic of void space characterization. Given the breadth and depth of publications on this topic, an exhaustive review is not possible. That said, there are several notable products that the authors may want to consider, including several existing open source applications for pore space characterization. Some of these are identified below.

Existing Software

1. PoreSpy: <https://porespy.org/index.html>
2. PorousMediaAnalysis: <https://github.com/VasiliBaranov/PorousMediaAnalysis>
- 3a. plug im!: <https://www.plugin.fr/>
- 3b. Pore Network Partitioning plugin: <https://www.plugin.fr/plugin/104>

Other References

- Al-Raoush, R. 2007. "Microstructure characterization of granular materials." *Physica A: Statistical Mechanics and its Applications*, 377 (2): 545–558.
<https://doi.org/10.1016/j.physa.2006.11.090>.
- Dong, H., and M. J. Blunt. 2009. "Pore-network extraction from micro-computerized-tomography images." *Phys. Rev. E*, 80 (3): 036307.
<https://doi.org/10.1103/PhysRevE.80.036307>.
- Hormann, K., V. Baranau, D. Hlushkou, A. Höltzel, and U. Tallarek. 2016. "Topological analysis of non-granular, disordered porous media: determination of pore connectivity, pore coordination, and geometric tortuosity in physically reconstructed silica monoliths." *New J. Chem.*, 40 (5): 4187–4199.
<https://doi.org/10.1039/C5NJ02814K>.
- Jiang, Z., K. Wu, G. Couples, M. I. J. Van Dijke, K. S. Sorbie, and J. Ma. 2007. "Efficient extraction of networks from three-dimensional porous media." *Water Resour. Res.*, 43 (12). <https://doi.org/10.1029/2006WR005780>.

****Perfect Packing (SC and HCP)****

1. Computation of the porosity of each packing for comparison with the closed-form solution might be a better (or, an additional) demonstration of performance because it includes both internal (3D) and boundary (2D) pores. Presumably, LOVAMAP would identify a boundary at the equator of the exterior spheres, so the geometry would be nice and clean.
2. EDF4 -- the metric being quantified here [$P\# = f(\%100 \text{ um particles})$] is not very intuitive for me. Why was this metric chosen? Why does a good linear fit imply that calculations are accurate?
3. EDF4 -- there appears to be a shift in pore space topology between 50 um and 60 um small spheres (i.e., the character of the linear fits changes from $y = O(1e-4)*x + O(1e-2)$ to $y = O(1e-5)*x + O(1e-3)$). Why? What is happening physically?

****Random packing of non-spherical and non-rigid monodisperse particles****

1. EDF7 -- the ordinary one-way ANOVA test assumes that (a) data are normally distributed; and (b) all populations have the same standard deviation. The test is somewhat robust in the face of violations of these assumptions, but only if the populations are large and similarly-sized, which is not the case for these data. Could a better statistical test have been selected?
2. Line 202, et al. -- "ellipsoid" is generally meant to imply "scalene ellipsoid" wherein the lengths of the three semi-axes are different. The authors are considering prolate spheroids ($a > b = c$) where "spheroid" indicates that two of the semi-axes are of equal length. Suggest changing the terminology to reflect this.
3. Line 214 -- suggest specifying that # of pores is per unit volume.
4. Fig. 2J -- if I perform a sample calculation using $\delta = 100 \text{ um}$ and $\phi = 0.38$, my result is $\gamma = 4.5$, which does not appear consistent with Fig. 2J. What am I missing?

****Plotting against the number of particles surrounding a 3D-pore****

1. Fig. 4F -- why is the maximum number of particles surrounding a pore changing with the particle size? For example, why does it go down from 50 μm to 60 μm and then back up from 60 μm to 70 μm ? Could these values be specimen size dependent?

2. Lines 280-281 -- this statement seems somewhat speculative. All of the particles have surface curvature. Ellipsoid packings are known to have higher random-close-packed solid fractions than spheres, but only by a few percent (say, 3-5%). The manner in which the elastic contact model is implemented for the ellipsoids is equally important, but no details on this are provided. Correct porting of Hertz-Mindlin contact mechanics from spheres to ellipsoids is nontrivial and often overlooked (i.e., in many cases, the spherical H-M formulation is applied directly to ellipsoidal particles, which is not strictly correct). Numerous discrete element method (DEM) simulations comparing packing of ellipsoids and spheres have been published. The authors are encouraged to consult the work of Shiwei Zhao, Tang-Tat Ng, and Matthew Kuhn, all of whom have published extensively in this area.

Google Scholar Pages

Shiwei Zhao: <https://scholar.google.com/citations?user=YIpVjMUAAAAJ&hl=en>

Tang-Tat Ng: <https://scholar.google.com/citations?user=dIA6SAYAAAAJ&hl=en>

Matthew Kuhn: <https://scholar.google.com/citations?user=Sz6W67YAAAAJ&hl=en>

Other References

Ouadfel, H., and L. Rothenburg. 2001. "'Stress-force-fabric' relationship for assemblies of ellipsoids." *Mechanics of Materials*, 33 (4): 201-221.
[https://doi.org/10.1016/S0167-6636\(00\)00057-0](https://doi.org/10.1016/S0167-6636(00)00057-0).

****3D-pore volume distributions****

1. These results bring to mind previous work on informational (Jaynes, Shannon) entropy. I invite the authors to consider some of the existing literature on packing entropy in granular materials, which is directly related to this discussion.

Entropy References

Brown, C. B. 1978. "The Use of Maximum Entropy in the Characterization of Granular Media." *Proceedings of the U.S.-Japan Seminar on Continuum-Mechanical and Statistical Approaches in the Mechanics of Granular Materials*, 98-108. Sendai, Japan: Tokyo: Gakujutsu Bunken Fukyukai.

Brown, C. B. 1993. "Entropy methods in the constitutive modeling of granular material." *Powders & Grains 93 : Proceedings of the Second International Conference on Micromechanics of Granular Media*, C. Thornton, ed. Birmingham, UK: A.A. Balkema.

Brown, C. B. 2000. "Entropy and Granular Materials: Model." *J. Eng. Mech.*, 126 (6): 599-604. [https://doi.org/10.1061/\(ASCE\)0733-9399\(2000\)126:6\(599\)](https://doi.org/10.1061/(ASCE)0733-9399(2000)126:6(599)).

Jowitt, P. W., and J. Munro. 1975. "The influence of void distribution and entropy on the engineering properties of granular media." *Proceedings of the 2nd International Conference on Applications of Statistics and Probability in Soil and Structural Engineering*, E. Schultze, ed., 365-385. Aachen, F.R.G.: Deutsche Gesellschaft für Erd- und Grundbau.

Kanatani, K. 1981. "The use of entropy in the description of granular materials."

Powder Technology, 30 (2): 217–223. [https://doi.org/10.1016/0032-5910\(81\)80015-0](https://doi.org/10.1016/0032-5910(81)80015-0).

Kuhn, M. R. 2014. "Dense granular flow at the critical state: maximum entropy and topological disorder." Granular Matter, 16 (4): 499–508.

<https://doi.org/10.1007/s10035-014-0496-2>.

McNamara, S., P. Richard, S. K. de Richter, G. Le Caër, and R. Delannay. 2009.

"Measurement of granular entropy." Phys. Rev. E, 80 (3): 031301.

<https://doi.org/10.1103/PhysRevE.80.031301>.

Reviewer #2 (Remarks on code availability):

The authors state the following:

Raw datasets can be downloaded at

<https://seguralab.duke.edu/what/highlights/lovamap>. The copyrights of the LOVAMAP software are owned by Duke University. To request a license to this software, please contact the Digital Innovations department at the Duke Office for Translation & Commercialization.

I visited the provided URL but did not see a way to download any of the 'raw datasets'. I did not request a license for this software from the Digital Innovations department at the Duke Office for Translation & Commercialization.

Reviewer #3 (Remarks to the Author):

In this work the authors developed techniques to characterize different aspects of the void space associated with different particle packings. While I find the techniques to be useful for the study of particle packings, in particular the improvement on the extraction of the medial axis for the void space, I think that there are major issues that need to be addressed before this paper can be recommended for publication, as detailed below.

1) The authors claim that their approach is a significant leap forward from previous methods used for void space analysis, which in my opinion is not well justified. There are a number of previous works on void statistics of particle packings that the authors fail to mention:

C. E. Maher, F. H. Stillinger, and S. Torquato, Characterization of void space, large-scale structure, and transport properties of maximally random jammed packings of superballs, Physical Review Materials, 6 025603 (2022).

C. E. Zachary, Y. Jiao, and S. Torquato. Hyperuniformity, Quasi-Long-Range Correlations, and Void Space Constraints in Maximally Random Jammed Particle Packings. II. Anisotropy in Particle Shape, Physical Review E, 83, 051309 (2011).

C. E. Zachary, Y. Jiao, and S. Torquato. Hyperuniformity, Quasi-Long-Range Correlations, and Void Space Constraints in Maximally Random Jammed Particle Packings. I. Polydisperse Spheres, Physical Review E, 83, 051308 (2011).

C. E. Zachary and S. Torquato. Anomalous Local Coordination, Density Fluctuations,

and Void Statistics in Disordered Hyperuniform Many-Particle Ground States, Physical Review E, 83, 051133 (2011).

I would like to see the authors put their results in the context of these previous works, and highlight the differences between their approach and those void statistics used in these previous works. It is not clear to me whether their approach is really a big improvement over existing void statistics.

2) It is well known in the physics community that the properties of particle packings and the associated void spaces, are dependent on the particle size and shape distributions, packing fraction, generation procedures (e.g., equilibrium vs nonequilibrium), and boundary conditions. It is not clear to me whether many statements in this work are as generally applicable to particle packings as the authors have claimed. Just as one example (there are many others), the authors claim that "our pore segmentation reveals that spheres, rods, and nuggets of equivalent volume produce statistically similar numbers of 3D-pores, while ellipsoids produce statistically more." It is not clear to me whether this statement will hold if the packing fraction changes, or a different procedure is used to generate the particle packings. The authors should clearly identify the conditions under which their results apply.

Reviewer #3 (Remarks on code availability):

The code appears usable and useful for the community.

Reviewer #4 (Remarks to the Author):

The manuscript by Riley et al. provides a model capable of extracting the local shape of the porosity in packed granular media. The algorithm used here is based on estimation of particle-particle distance that allows a better reconstruction of the space between particles compared with watershed or erosion type algorithm according to the authors. The manuscript provides a brief explanation on the construction of the pore phase using the LOVOMAP and then showcase its use by quantifying the number and size distribution of porosity in various particles packing. The rest of the manuscript demonstrates the use of the algorithm to obtain different quantities on pore structures form by different packing. The actual code is not provided and doesn't seem to be freely available, but the strategy is detailed in the supplementary document. I have a few general comments to discuss some aspect of the resulting data, but this seems like a new way to look at particle assemblies as far as I know. More importantly, I could see this applied in various situation ranging from surface active phenomenon (catalysis) to some more materials processing related issue (pore/defect evolution during heat treatment/sintering).

- The algorithm is dependent on the correct numbering and identification of the initial particles. Is the identification made in the first step of the volume rastering using already available particles identification from the assembly formation or does it give the particle identification number based on a separation distance or any other criteria? That's a significant difference with data that could be obtained experimentally in which the particles can be touching/not differentiate. In that sense the initial labelling would be key to obtain the correct pore structure and thus a first step using watershed in 3D for instance would be needed to obtain a correct labelling, making this technique reliable only for clean/simulated data.

- Material fabrication is highly impacted by pore shape, for instance material strength/toughness dictated by pore shape and sharpness, sintering of powder guided by the surface curvature evolution over temperature. Is it possible to extract local curvature from these models?
- The criteria to put a 'door' or not, i.e. connecting the pore spaces together or not seems relatively arbitrary. In terms of properties, for instance anything depending on percolation or mass transport in these pore space, two pores connected would be counted as disconnected using the criteria given. I agree that the criterion is well explained, however I am missing a rationale for choosing such a criterion in the first place since some of these pores are connected. This in turn have strong implications on the data extracted later, so it should be explained or if it is arbitrary it should be presented as this.
- How the equations for pore size/volume chosen in figure 2? It seems arbitrary to have two different ones but maybe it was simply the formula shape that gave the best fit? On that note, are the x axis of the graphs in figure 2 J and K particles diameter directly (in μm for instance) or the fitting formula value as it seems to be the case from the formatting.
- Could this work with irregular particles as well (not just roughness on the surface but actually rough, multifaceted shape)?

Author Rebuttal to Initial comments

Dear Professor Segura,

Your manuscript "The empty space of packed particulate materials" has now been seen by 4 referees, whose comments are appended below. You will see that while they find your work of interest, they have raised points that need to be addressed before we can make a decision on publication.

The referees' reports seem to be quite clear. Naturally, we will need you to address all of the points raised.

While we ask you to address all of the points raised, the following points need to be substantially worked on:

- Please provide benchmark results to demonstrate the benefits. When discussing your results, please avoid overhypes.

To provide benchmark results, we compare our 3D-pore segmentation approach (LOVAMAP) against both 2-D pore segmentation and over-segmentation in 3-D, in which *each* local center point of the void space defines the center of a pore. 2-D pore segmentation comparisons are discussed in Fig. 2A-B and Extended Data Fig. 9. For over-segmentation, we highlight the difference in approach for both interior pores (Fig. 1K), as well as exit/entrance pores (Fig. 1L). Data comparisons are discussed in Extended Data Fig. 12 and Fig. 5B. Our algorithms for determining medial axis landmarks were not developed as extensions of prior art by which to compare results. Regarding performance, we did not benchmark code performance because runtimes were reasonable.

- Please follow our referees' suggestion to better discuss the literature, especially those missed references suggested by referees.

We have thoroughly reviewed references suggested by reviewers and have updated our references and text accordingly.

- Please carefully address our referees' comments about the code and datasets. Following the suggestion from one of the referees, we recommend to use open-source programming languages, such as python, for implementing your computational framework.

We have addressed reviewer comments about code and data availability. As is known in the academic community, Matlab is commonly used for research and prototyping. Matlab is freely available to Duke students and was the preferred choice of language for the student on this manuscript. Portions of the code are written in C++, and we have plans to transition the code entirely to a non-proprietary language.

- Please address various technical issues raised by our referees, including clarification and justification about the technical points/assumptions/choice of parameters, robustness of the framework, additional statistical tests, etc.

Below we have addressed all reviewer comments to the best of our ability. In response to reviewer comments and concerns, we have updated text and figures.

- The author Peter Cheng is from a company. Please update the competing interest statement accordingly in your revised manuscript.

Our Competing Interest statement has been updated to reflect this.

Please use the following link to submit your revised manuscript and a point-by-point response to the referees' comments (which should be in a separate document to any cover letter):

To aid in the review process, we would appreciate it if you could also provide a copy of your manuscript files that indicates your revisions by making use of Track Changes or similar mark-up tools. Please also ensure that all correspondence is marked with your Nature Computational Science reference number in the subject line.

In addition, please make sure to upload a Word Document or LaTeX version of your text, to assist us in the editorial stage.

If you have any issues when updating your Code Ocean capsule during the revision process, please email the Code Ocean support team Cc'ing me.

To improve transparency in authorship, we request that all authors identified as 'corresponding author' on published papers create and link their Open Researcher and Contributor Identifier (ORCID) with their account on the Manuscript Tracking System (MTS), prior to acceptance. ORCID helps the scientific community achieve unambiguous attribution of all scholarly contributions. You can create and link your ORCID from the home page of the MTS by clicking on 'Modify my Springer Nature account'. For more information please visit please visit www.springernature.com/orcid.

We hope to receive your revised paper within three weeks. If you cannot send it within this time, please let us know.

We look forward to hearing from you soon.

Best regards,

Jie Pan, Ph.D.
Senior Editor
Nature Computational Science

Reviewers comments:

Reviewer #1 (Remarks to the Author):

This manuscript presents a voxel-based method to identify pores in particle packing. In addition, voxels that are equidistant to several surrounding particles are identified, which form "ridges" and "peaks" based on the number of surrounding particles. To my knowledge, pores in particle packing have been studied a lot. The commonly used method based on Voronoi and Delaunay, which are duals, can already identify various pore properties. According to the definition, the "ridges" and "peaks" proposed in this work are corresponding to the Voronoi cell edge and vertex. They are not new concepts.

In addition, several methods are already proposed to perform Voronoi analysis for particle packing, including non-spherical particles. Note these methods consider particles as geometry bodies but not points, and some methods also use voxels. Essentially these methods give similar results according to the same mathematical definition.

We appreciate Reviewer #1's concern with the novelty of our work. In the Background of our manuscript, we discuss the uses of Voronoi and Delaunay tessellation in particle and void space analysis. The vast majority of prior work focuses on particle properties, which is illustrated by Reviewer #1's insight that 'ridges' and 'peaks' correspond to edges and vertices of Voronoi cells that surround *particles* (geometry bodies). Performing packed particle analysis using a Voronoi tessellation on particles will fragment the void space, thereby inhibiting the study of 3D-pores,

which is the goal of our work. We are specifically interested in segmenting the void space of packed particles into 3D-pores because we view 3D-pores as the particulate-void-space-counterpart to particles; we work with cells that explore the void pockets of packed particles, so rather than particles, we are interested in pores. For works that use Delaunay triangulation to segment void space, our Background discusses the shortcomings, including the reliance on thresholding if particle contact points are used.

We acknowledge that 2D-ridges, 1D-ridges, and peaks of the medial axis are not novel concepts; however, we are reporting on the novelty of our methodology for accurately identifying these landmarks – specifically, using our ‘Medial Axis by Particle Configuration (MAPC)’ plus ‘set of smallest loops’ approach to identify where each and every 1D-ridge lies in the void space, irrespective of the resolution of the grid. Our approach uses the particle configuration alone, which also allows us to identify unique 1D-ridges used for further pore analysis.

The results obtained from the method are neither new nor significant. The equations are not numbered, so I have to cite the page number and line number below:

page 9, line 216. This equation may be new, but it just shows the relationship between pore number and packing fraction. This relationship seemingly is not very meaningful, and it depends on the definition of a pore.

We appreciate Reviewer #1’s recommendation to number our equations – the text has been updated accordingly. The equation referenced here (now labeled 1) corresponds to Figure 2J, which relates pore number with particle size (δ) and packing fraction (Φ) through a transformation (δe^Φ). The motivation for deriving this equation is the belief that particle size and packing fraction have the largest influence on the number of pores (per volume) in a packing. An example of the usefulness of such an equation is in the design of granular biomaterials for the production of brain tissue organoids. Biomedical engineers use packed microgels to culture organoid-precursors in the void space. Researchers may use our equation to approximate the number of pores in their granular material (since average particle diameter and packing fraction are easily-obtainable metrics), which would correspond to the number of distinct regions available for neurosphere (organoid-precursor) growth (i.e., the upper bound for number of organoids capable of being produced).

page 9, line 224. This equation is the data fitting for the median pore volume and packing fraction. These two parameters both describe the void in a packing. It is not strange they have such a simple relationship.

The equation referenced here (now labeled 2) corresponds to Figure 2K, which relates median pore volume with particle size (δ) and packing fraction (Φ) through a transformation ($\delta\sqrt{\Phi}$). We believe the simplicity of both transformations in Equations (1) and (2) is elegant, and the simplicity of the curves (i.e., quadratic equations) supports the accuracy of our segmentation approach since a complicated relationship would be unexpected. To continue with the organoid example used above, Equation 2 would be useful for predicting and thereby tuning the size of neurospheres (organoid-precursor), since neurospheres roughly grow to the size of the void pocket (unpublished results).

page 11, line 268. This equation is the data fitting for pore volume and the number of surrounding particles. Similar data can be found in the previous studies on the relationships between different Voronoi and Delaunay properties.

In the literature, there are many studies on the relationships between Delaunay/Voronoi properties and packing fraction. Some are based on theoretical analysis. Comparably, this work does not provide new insights.

The equation referenced here (now labeled 3) corresponds to Figure 4F. To address the prior works, it would be helpful if Reviewer #1 could be more specific about the ‘relationships between...Voronoi and Delaunay properties,’ since ‘Voronoi and Delaunay properties’ do not correspond to 3D-pores in our minds.

To our knowledge, no previous work has accurately segmented 3D-pores across a range of particle packing types (including soft particles), showcased an extensive visual library of 3D-pores, and reported the interesting analysis that we present in our manuscript, such as finding consistent 3D-pore trends among different particle packings and deriving an explanation for the unique shape of the pore volume distribution seen for packed spheres (namely, distinct pore

types as defined by the number of surrounding particles). We believe the work will be valuable and interesting to multiple fields that work with particulate material.

Reviewer #2 (Remarks to the Author):

The authors have developed an application (LOVAMAP) for the characterization of void space in assemblies of granular particles. To the reviewer's knowledge and evaluation, the work is thorough, novel, and internally consistent. The paper is well-written, figures are informative and very information-dense, and appropriate supplementary information is provided to allow the reader to reproduce much of the authors' work. The topic is likely of interest to the Journal's readers.

This manuscript is likely suitable for publication, but I suggest that the items listed below should be considered and addressed, as appropriate, before a final decision can be made. There are a number of instances where additional clarity would be welcome, and in some cases, I have questions about technical details of the work. Robustness in the face of several variables and asymptotic behavior are not always well-demonstrated. Specific comments and questions are detailed below.

We thank Reviewer #2 for their thorough comments that have undoubtedly strengthened the manuscript and improved clarity throughout.

****General****

1. On L410 of SI, the authors state that $1 \mu\text{m}^3 = 1 \text{ pL}$. This is incorrect: $1 \mu\text{m}^3 = 10^{-18} \text{ m}^3 = 10^{-15} \text{ L} = 1 \text{ fL}$. Is this a typo isolated to this line? Or is this unit glitch pervasive through the manuscript?

This is an isolated typo! We thank Reviewer #2 for catching this. The text has been updated to say “1000 pL = $1 \mu\text{m}^3$ ”.

2. Given that specimen size remains constant at $(600 \mu\text{m})^3$ while particle sizes and shapes

change, can all assemblies be considered as REV's? If the sizes of the assemblies were scaled up by an order of magnitude (for constant particle size), would results or interpretations change?

Our simulated assemblies can indeed be considered REV's since the scale is arbitrary. If the container size were scaled up (while holding particle size and mesh size (dx) constant), we would expect the same results/interpretations, e.g., the same distribution shapes for pore volume or the same relative relationships for # 3D-pores per pL.

3. Some of the comparative metrics used by the authors are somewhat confusing in that they are absolute -- e.g., # of pores. Wouldn't these quantities be better expressed in a normalized fashion?

(addressing Q3 & Q4 beneath Question 4)

4. Pursuant to (2) and (3) above, I suspect that I might be missing something obvious, but it would be helpful if the authors were more explicit about when and how they are normalizing some of their measurements. Are there other cases where clarity could be achieved through normalization?

Review #2 has brought to our attention that we did not consistently mention throughout the text that graphs reporting the total number of 3D-pores are normalized by the container size. For clarity, we have updated the main text (excerpt below) to explicitly describe which descriptor data has been normalized, as well as refer to "number of 3D-pores per pL" throughout the text. We agree that this normalization makes the data more comparable across different assemblies and will likely also be more useful for readers looking to use our results to predict porosity within their own structures.

Updated text:

When reporting 3D-pore data, we normalize the total number of 3D-pores by container volume, while pore descriptors that do not depend on container size, such as pore volume, are reported as is.

5. The authors provide few details about the physics of specimen assembly, particularly for the reader unfamiliar with the proprietary Houdini software. Contact mechanics, particle properties, damping, time scaling, inertial number, and convergence criteria (among others) will all have a significant influence on the equilibrated state of the particle assemblies. How sensitive are the authors' findings to these metrics?

Unfortunately, since Houdini is proprietary, we are unaware of precisely how the physics solver works. One would have to reach out to SideFX to inquire about the details. However, Houdini is the gold standard in the film and animation industry for physics-based simulation (and much more), which gives us confidence in the software. We have updated the text to include an explicit mention of using “default” parameter values for all parameters not otherwise stated. For someone who is familiar with Houdini, they will be able to reproduce our simulations with the information provided in this manuscript. Houdini offers a free version of their software, which we used in this paper.

Reviewer #2 brings up an interesting idea for a paper that focuses on the sensitivity of particle configuration (and therefore LOVAMAP results) to simulation parameters in Houdini. We have counted at least 24 parameters associated with the rigid body solver in Houdini, and we anticipate more for the finite element solver (soft bodies). A comprehensive sensitivity analysis would warrant a stand-alone study. However, we were also curious about parameter sensitivity, so we chose to explore container shape and friction in Extended Data. We present this data and analysis in ED 9 and 10.

6. Line 16 -- "significant leap forward" is subjective at best and hyperbolic at worst. Suggest removing or toning down.

We appreciate this suggestion, which raised a flag for another reviewer as well. We have updated the text to say, “Our approach is an improvement upon previous methods that anchor onto simplifying assumptions about local tetrahedral packing or that over-segment the space.”

7. Line 187 -- "state of the art" is subjective and difficult to quantify or substantiate. Suggest removing.

We agree the phrase ‘state-of-the-art’ stood out, so the line has been updated to state a fact: “We next study random close packing of spheres by generating particle domains using SideFX Houdini, an industry-standard physics simulation software.”

8. Is LOVAMAP an acronym? For what?

We are not intending for LOVAMAP to be presented as an acronym for anything. Our logo for the LOVAMAP software was designed with “LOVAMAP” in all caps, so we had kept this formatting in the text. While we have become accustomed to LOVAMAP in all caps, we are open to “Lovamap” if Reviewer #2 believes this will clear confusion.

9. EDF16 -- if assembly size is constant, aren't (a) and (c) just proxies for number of particles? That is, aren't all three figures effectively equivalent?

Yes, we are presenting the same runtime data for three different (related) parameters to show how runtime scales with each (e.g., in case one reader thinks in particle diameter while another thinks in particle count). To clear up confusion, we have updated the Figure title to say “Runtimes for packed particle analysis of monodisperse spheres using LOVAMAP.”

10. The authors have elected to develop LOVAMAP using proprietary software, MATLAB. Why not use something open source like Python or Julia? Is there specific functionality required by LOVAMAP that is only available in MATLAB? Will the application run on Octave? (I appreciate that this question is largely philosophical, but I am genuinely curious about functionality.)

As mentioned above in response to the Editor, MATLAB is freely available to Duke students and was the preferred choice of language for development since MATLAB is commonly used for research and prototyping in academia. Portions of the code are written in C++, and we have plans to transition the code entirely to a non-proprietary language. The current version of the code will likely not run in Octave because 1) our ‘set of smallest loops’ code (for finding 1D-ridges) is written in C++, so new code would likely be required for calling C++ in Octave, and 2) we use seven toolboxes (MATLAB or otherwise) listed below. According to octave.org, “Octave syntax is largely compatible with MATLAB.” The exceptions to this compatibility may or may not lie in our dependencies, which contain code that we do not own.

- [Image Processing Toolbox](#)
- [Statistics and Machine Learning Toolbox](#)
- [SLM - Shape Language Modeling](#)
- [circlefit3d - fit circle to three points in 3d space](#)
- [Inhull](#)
- [Generate maximally perceptually-distinct colors](#)
- [GetFullPath](#)

11. The authors have done a commendable job in studying and reporting the existing literature on the topic of void space characterization. Given the breadth and depth of publications on this topic, an exhaustive review is not possible. That said, there are several notable products that the authors may want to consider, including several existing open source applications for pore space characterization. Some of these are identified below.

Existing Software

1. PoreSpy: <https://porespy.org/index.html>
2. PorousMediaAnalysis: <https://github.com/VasiliBaranov/PorousMediaAnalysis>
- 3a. plug im!: <https://www.plugin.fr/>
- 3b. Pore Network Partitioning plugin: <https://www.plugin.fr/plugin/104>

These are fantastic resources, and we appreciate the references. The base approach for segmentation in these products appears to be morphological reconstruction and watershed,

which can accommodate non-particulate spaces. These products are therefore more generally applicable for pore analysis, making them important tools for the community. For particulate material specifically, we offer LOVAMAP as an option for improved accuracy in pore identification because the approach uses the particle configuration as a basis for segmentation. While the present manuscript does not cover a wide range of void space descriptors, we will use these resources as inspiration when developing additional metrics. We are choosing to limit our references to research articles that discuss methodologies (that are implemented in the above products); however, if Reviewer #2 feels software should be referenced as well, we are happy to oblige.

Other References

Al-Raoush, R. 2007. “Microstructure characterization of granular materials.” *Physica A: Statistical Mechanics and its Applications*, 377 (2): 545–558. <https://doi.org/10.1016/j.physa.2006.11.090>.

Al-Raoush 2007 primarily focuses on particle metrics, which makes this manuscript a fantastic resource for 3D particle segmentation. We have added the citation to our background referencing particle-focused work, as well as void-space analysis that considers the Voronoi tessellation pod surrounding each particle.

Dong, H., and M. J. Blunt. 2009. “Pore-network extraction from micro-computerized-tomography images.” *Phys. Rev. E*, 80 (3): 036307. <https://doi.org/10.1103/PhysRevE.80.036307>.

This citation has been added to our discussion of prior methods that use maximally inscribed balls. The benefit here is that the authors are able to analyze micro-CT-derived images of sandstone (Fig. 6), where individual particles are not necessarily delineated. They also go on to analyze fluid flow through the void space, which we did not cover. Dong and Blunt 2009 is a very interesting manuscript that is relevant and distinctly different from our work. We have updated our Conclusion to address Future Directions that explore fluid flow analysis.

Hormann, K., V. Baranau, D. Hlushkou, A. Höltzel, and U. Tallarek. 2016. “Topological analysis

of non-granular, disordered porous media: determination of pore connectivity, pore coordination, and geometric tortuosity in physically reconstructed silica monoliths.” New J. Chem., 40 (5): 4187–4199. <https://doi.org/10.1039/C5NJ02814K>.

Hormann et al. 2016 present important work on non-granular, porous media, which is not analyzable by LOVAMAP. This is a more recent paper in the space that we had not previously read, so we appreciate the referral. While we have come across a number of manuscripts that cover segmentation of non-particulate porous media, we chose to limit the focus of our references to particulate material in hopes of narrowing the scope of prior work covered in our background. We have, however, updated our Conclusion to reiterate the particulate requirement of LOVAMAP :

Updated text:

Our segmentation approach becomes less appropriate if the scale of the void-space-of-interest lies within particles – for example, if particles are large shards of trabecular bone. We have explored, in theory, a future update to the code that extends void space analysis to non-particulate matter; however, to maintain the accuracy that we achieve with our MAPC approach, rather than distinct particles, distinct ‘surfaces’ would need to be identified – where every change in curvature (in 3D) would define a different surface of the solid material. As one can imagine, this is a challenging but possible endeavor. Lastly, since LOVAMAP was specifically developed for particulate material, analyzing experimental data derived from imaging systems will require particle segmentation as the first step, which is a well understood segmentation problem.

Jiang, Z., K. Wu, G. Couples, M. I. J. Van Dijke, K. S. Sorbie, and J. Ma. 2007. “Efficient extraction of networks from three-dimensional porous media.” Water Resour. Res., 43 (12). <https://doi.org/10.1029/2006WR005780>.

Similar to Hormann et al. 2016, Jiang et al. 2007 implement an algorithm that can apply to non-particulate, porous media, which is a different target than LOVAMAP. The manuscript by Jiang et al. does a fantastic job covering terminology and reviewing background, and similar to Hormann et al., they apply real images of sandstone to study flow and permeability through the

void space. For future applications of our work, we will take inspiration from metrics used in Jiang et al., such as one of their schemes to compute a ‘shape factor.’

****Perfect Packing (SC and HCP)****

1. Computation of the porosity of each packing for comparison with the closed-form solution might be a better (or, an additional) demonstration of performance because it includes both internal (3D) and boundary (2D) pores. Presumably, LOVAMAP would identify a boundary at the equator of the exterior spheres, so the geometry would be nice and clean.

Apologies for the confusion! We indeed compared our results to closed-form solutions, which are reported in Extended Data Fig. 4a. We were unable to derive an expression for the more complicated shape of the larger pore in hexagonal packing. The wording of the caption may have been unclear, so the text has been updated below:

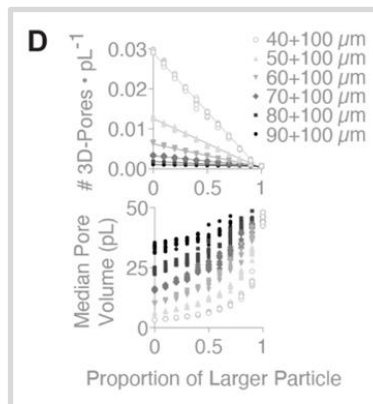
Updated caption:

For square packing of spheres, we report the closed-form expression for 3D-pore volume scaled to picoliters, then use this expression to compute the root mean squared error normalized by the mean (NRMSE) for LOVAMAP outputs. For hexagonal packing, we derive an expression that closely approximates 3D-pore volume of the smaller pore and report NRMSE for LOVAMAP outputs; for the larger pore, we fit a third degree polynomial to LOVAMAP outputs using least squares (reporting R-squared). For all equations, x = particle diameter of sphere (μm), y = 3D-pore volume (pL).

2. EDF4 -- the metric being quantified here [$P\# = f(\%100 \text{ um particles})$] is not very intuitive for me. Why was this metric chosen? Why does a good linear fit imply that calculations are accurate?

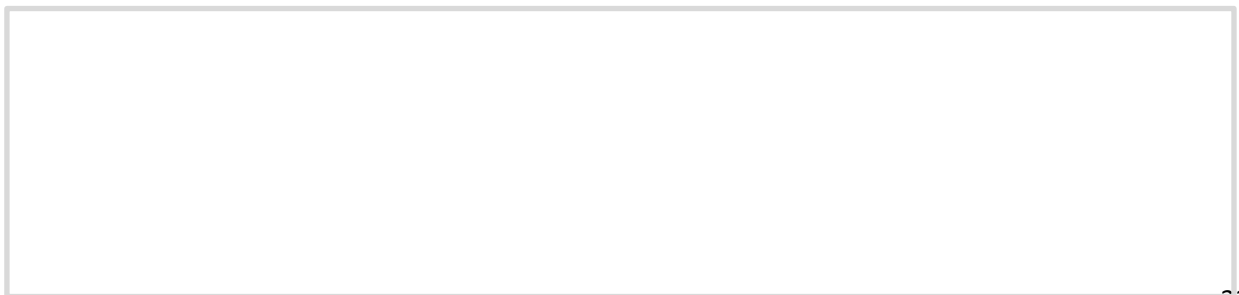
To improve clarity, the x-axis label in Fig. 2D (which corresponds to the data in EDF4b) has been updated to ‘Proportion of Larger Particle’ with values now ranging between 0 and 1 (copied below). The data corresponds to packings comprising 2 particle species at different proportions,

and we chose to plot by the proportion of the 100 μm diameter (larger) particle. A good linear fit need not imply accuracy (in fact, we were not necessarily expecting a linear fit, but it is an interesting finding); however, we report equations of the best fit lines so the scientific community may use them to predict the number of 3D-pores given the proportions of populations in their bidisperse sample.



3. EDF4 -- there appears to be a shift in pore space topology between 50 μm and 60 μm small spheres (i.e., the character of the linear fits changes from $y=O(1e-4)*x+O(1e-2)$ to $y=O(1e-5)*x+O(1e-3)$). Why? What is happening physically?

To address this question, we filled in the remaining mixtures (80 + 100 μm and 90 + 100 μm) (EDFig. 4b copied below) and plotted the slope values as a function of smaller-particle size to show that the data follows an exponential curve (EDFig. 4c copied below). The curve itself is not necessarily anticipated, but the good fit ($R^2 = 0.9995$) suggests that we are seeing an expected phenomena. This data has been added to Extended Data Fig. 4.

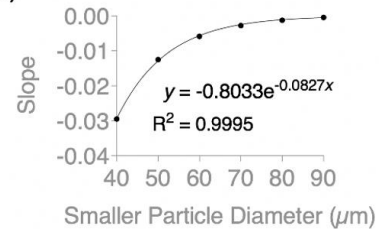


b)

3D-pores · pL⁻¹

40 & 100:	$y = (-2.949\text{e-}2)*x + 0.02902$	R ² : 0.9960
50 & 100:	$y = (-1.247\text{e-}2)*x + 0.01261$	R ² : 0.9938
60 & 100:	$y = (-5.826\text{e-}3)*x + 0.006271$	R ² : 0.9871
70 & 100:	$y = (-2.748\text{e-}3)*x + 0.003345$	R ² : 0.9836
80 & 100:	$y = (-1.223\text{e-}3)*x + 0.001876$	R ² : 0.9502
90 & 100:	$y = (-4.115\text{e-}4)*x + 0.001105$	R ² : 0.7084

c)



****Random packing of non-spherical and non-rigid monodisperse particles****

1. EDF7 -- the ordinary one-way ANOVA test assumes that (a) data are normally distributed; and (b) all populations have the same standard deviation. The test is somewhat robust in the face of violations of these assumptions, but only if the populations are large and similarly-sized, which is not the case for these data. Could a better statistical test have been selected?

We had made the assumption that our data is normally distributed with comparable SD because data are simulated replicates; however, Reviewer #2 raises an important point that our sample sizes are small. Normality tests in Prism 9 show that all descriptors (i.e., VVF, Normalized Internal Pore Volume, # Particles, Total # Pores, # Surface Pores, and # Interior Pores) for all particle groups (i.e., spheres, ellipsoids, rods, and nuggets) pass the Shapiro-Wilk test for normality, with 3 exceptions: # Interior Pores of nuggets, and VVF of ellipsoids and nuggets. Remaining normality tests indicate that sample size is too small for many non-spherical groups. Bartlett's test to determine if SD is significantly different shows no difference across descriptors, with the exception of Normalized Internal Pore Volume and # Particles. Rather than tailor our statistical tests for these exception, we have replotted results using Welch's ANOVA test in Prism 9. We chose Welch's ANOVA over the nonparametric Kruskal-Wallis test per recommendation in Liu H., 2015 (<https://doi.org/10.25772/BWFP-YE95>) given our dataset. Text and figures have been updated accordingly.

2. Line 202, et al. -- "ellipsoid" is generally meant to imply "scalene ellipsoid" wherein the lengths of the three semi-axes are different. The authors are considering prolate spheroids ($a > b = c$) where "spheroid" indicates that two of the semi-axes are of equal length. Suggest changing the terminology to reflect this.

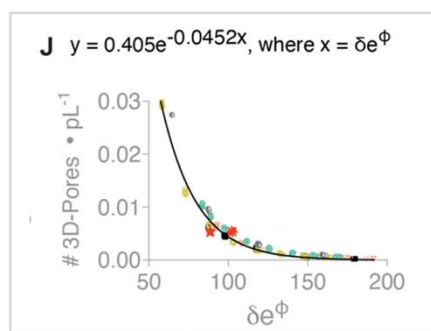
We thank Reviewer #2 for raising this point – we value precise terminology. Text in the main manuscript, as well as Extended Data figures and Supplementary Material, have been updated to specify that our ellipsoids are prolate spheroids. We will continue to generally refer to the group as ‘Ellipsoids’ to stay consistent with previous manuscripts (Zhao, S., Evans, T.M. & Zhou, X. Three-dimensional Voronoi analysis of monodisperse ellipsoids during triaxial shear. *Powder Technology* 323, 323-336 (2018); Donev A., Cisse I., Sachs D., Variano E.A., Stillinger F.H., Connelly R., Torquato S., Chaikin P.M. Jammed Disordered Packings Using Ellipsoids. *Science* 303: 990-993 (2004), etc.), which also specify whether their ellipsoids are prolate or oblate spheroids.

3. Line 214 -- suggest specifying that # of pores is per unit volume.

This is a great suggestion. We have updated the text accordingly.

4. Fig. 2J -- if I perform a sample calculation using $\delta=100$ μm and $\phi=0.38$, my result is $y=4.5$, which does not appear consistent with Fig. 2J. What am I missing?

Thank you for providing these sample values. Our work is shown below next to a copy of Fig. 2J:



$$\begin{aligned}
 \delta &= 100 \\
 \Phi &= 0.38 \\
 x &= \delta e^{\Phi} \\
 &= 100e^{0.38} \\
 &= 146.228459 \\
 y &= 0.405e^{-0.0452x} \\
 &= 0.405e^{(-0.0452 \cdot 146.228)} \\
 &= 0.00054573
 \end{aligned}$$

****Plotting against the number of particles surrounding a 3D-pore****

1. Fig. 4F -- why is the maximum number of particles surrounding a pore changing with the particle size? For example, why does it go down from 50 μm to 60 μm and then back up from 60 μm to 70 μm ? Could these values be specimen size dependent?

This data showcases the randomness of the samples being generated. Plots are showing cumulative pores from $N = 10$ particle domains each (this point has been clarified in the text). A pore surrounded by a large number of particles is a very rare event and becomes more rare as particle size increases relative to a fixed container size (i.e., particle count and 3D-pore count decreases). For example, the second plot in Fig. F showing 50 μm diam domains contains $\sim 28,000$ datapoints (pores), and yet only 4 pores lie above 25 surrounding particles. Differences in the max number of surrounding particles among groups are related to random chance and the shape of the curves. We believe the lack of larger pores (with more surrounding particles) in our 60 μm diam plot indicates that the rare event never occurred in the 10 domains sampled.

2. Lines 280-281 -- this statement seems somewhat speculative. All of the particles have surface curvature. Ellipsoid packings are known to have higher random-close-packed solid fractions than spheres, but only by a few percent (say, 3-5%). The manner in which the elastic contact model is implemented for the ellipsoids is equally important, but no details on this are provided. Correct porting of Hertz-Mindlin contact mechanics from spheres to ellipsoids is nontrivial and often overlooked (i.e., in many cases, the spherical H-M formulation is applied directly to ellipsoidal particles, which is not strictly correct). Numerous discrete element method (DEM) simulations comparing packing of ellipsoids and spheres have been published. The authors are encouraged to consult the work of Shiwei Zhao, Tang-Tat Ng, and Matthew Kuhn, all of whom have published extensively in this area.

Google Scholar Pages

Shiwei Zhao: <https://scholar.google.com/citations?user=YIpVjMUAAAAJ&hl=en>

Tang-Tat Ng: <https://scholar.google.com/citations?user=dIA6SAYAAAAJ&hl=en>

Matthew Kuhn: <https://scholar.google.com/citations?user=Sz6W67YAAAAJ&hl=en>

Other References

Ouadfel, H., and L. Rothenburg. 2001. "Stress-force-fabric' relationship for assemblies of ellipsoids." *Mechanics of Materials*, 33 (4): 201-221. <https://doi.org/10.1016/S0167->

[6636\(00\)00057-0](#).

Reviewer #2's previous comment brought to our attention that a lack of large pores (surrounded by more particles) in ellipsoid packings relative to all other groups could be due to random chance. Therefore, the conclusions discussed in Lines 280-281 (referenced in this comment) have been removed. Instead, we highlight the clearer observation that rods and nuggets produce larger pores surrounded by more particles. The text has been updated to:

We also compare 3D-pore volume for each particle domain category in **Figure 4A-E**, for which we observe differences in data trends across plots (**Fig. 4H**). Relative to the other groups, rods and nuggets appear more likely to produce large pores surrounded by many particles. As seen in the last column of **Fig. 4D and E**, the straight and/or flat nature of rods and nuggets allows for settling into plate- or log-like stacking, which increases local particle density. However, it is difficult to draw accurate conclusions to explain the aforementioned trend.

Regarding simulations, Houdini is a proprietary software, so we can only make an educated guess about its algorithms. That being said, Houdini's rigid body solver is likely a classic rigid body solver and not a DEM. We are unfamiliar with DEM solvers. While we acknowledge the fact that there are important things to consider in a DEM solver, since we chose to use Houdini, these points did not arise for us.

****3D-pore volume distributions****

1. These results bring to mind previous work on informational (Jaynes, Shannon) entropy. I invite the authors to consider some of the existing literature on packing entropy in granular materials, which is directly related to this discussion.

Entropy References

Brown, C. B. 1978. "The Use of Maximum Entropy in the Characterization of Granular Media." Proceedings of the U.S.-Japan Seminar on Continuum-Mechanical and Statistical Approaches in the Mechanics of Granular Materials, 98–108. Sendai, Japan: Tokyo: Gakujutsu Bunken Fukyukai.

Brown, C. B. 1993. "Entropy methods in the constitutive modeling of granular material."

Powders & Grains 93 : Proceedings of the Second International Conference on Micromechanics of Granular Media, C. Thornton, ed. Birmingham, UK: A.A. Balkema.

Brown, C. B. 2000. "Entropy and Granular Materials: Model." J. Eng. Mech., 126 (6): 599–604. [https://doi.org/10.1061/\(ASCE\)0733-9399\(2000\)126:6\(599\)](https://doi.org/10.1061/(ASCE)0733-9399(2000)126:6(599)).

Jowitt, P. W., and J. Munro. 1975. "The influence of void distribution and entropy on the engineering properties of granular media." Proceedings of the 2nd International Conference on Applications of Statistics and Probability in Soil and Structural Engineering, E. Schultze, ed., 365–385. Aachen, F.R.G.: Deutsche Gesellschaft für Erd- und Grundbau.

Kanatani, K. 1981. "The use of entropy in the description of granular materials." Powder Technology, 30 (2): 217–223. [https://doi.org/10.1016/0032-5910\(81\)80015-0](https://doi.org/10.1016/0032-5910(81)80015-0).

Kuhn, M. R. 2014. "Dense granular flow at the critical state: maximum entropy and topological disorder." Granular Matter, 16 (4): 499–508. <https://doi.org/10.1007/s10035-014-0496-2>.

McNamara, S., P. Richard, S. K. de Richter, G. Le Caër, and R. Delannay. 2009. "Measurement of granular entropy." Phys. Rev. E, 80 (3): 031301. <https://doi.org/10.1103/PhysRevE.80.031301>.

We thank Reviewer #2 for the references! Coincidentally, we are using the Shannon entropy score extensively in a follow-up manuscript to this work. The references provided are valuable and appreciated.

Reviewer #2 (Remarks on code availability):

The authors state the following:

Raw datasets can be downloaded at <https://seguralab.duke.edu/what/highlights/lovamap>. The copyrights of the LOVAMAP software are owned by Duke University. To request a license to this software, please contact the Digital Innovations department at the Duke Office for Translation & Commercialization.

I visited the provided URL but did not see a way to download any of the 'raw datasets'. I did not request a license for this software from the Digital Innovations department at the Duke Office for Translation & Commercialization.

Apologies for the confusion – the raw datasets are provided for free after users register with our site (found at the bottom of the webpage in the link provided in the comment above). The intention is to know who is accessing our data. For the purpose of manuscript review, we hope Code Ocean is sufficient, since registering with our site will violate anonymity.

Reviewer #3 (Remarks to the Author):

In this work the authors developed techniques to characterize different aspects of the void space associated with different particle packings. While I find the techniques to be useful for the study of particle packings, in particular the improvement on the extraction of the medial axis for the void space, I think that there are major issues that need to be addressed before this paper can be recommended for publication, as detailed below.

1) The authors claim that their approach is a significant leap forward from previous methods used for void space analysis, which in my opinion is not well justified. There are a number of previous works on void statistics of particle packings that the authors fail to mention:

C. E. Maher, F. H. Stillinger, and S. Torquato, Characterization of void space, large-scale structure, and transport properties of maximally random jammed packings of superballs, *Physical Review Materials*, 6 025603 (2022).

C. E. Zachary, Y. Jiao, and S. Torquato. Hyperuniformity, Quasi-Long-Range Correlations, and Void Space Constraints in Maximally Random Jammed Particle Packings. II. Anisotropy in Particle Shape, *Physical Review E*, 83, 051309 (2011).

C. E. Zachary, Y. Jiao, and S. Torquato. Hyperuniformity, Quasi-Long-Range Correlations, and Void Space Constraints in Maximally Random Jammed Particle Packings. I. Polydisperse Spheres, *Physical Review E*, 83, 051308 (2011).

C. E. Zachary and S. Torquato. Anomalous Local Coordination, Density Fluctuations, and Void Statistics in Disordered Hyperuniform Many-Particle Ground States, *Physical Review E*, 83, 051133 (2011).

I would like to see the authors put their results in the context of these previous works, and

highlight the differences between their approach and those void statistics used in these previous works. It is not clear to me whether their approach is really a big improvement over existing void statistics.

Professor Torquato is one of the most influential contributors to the particulate matter space, so we appreciate the list of publications by which to compare our work. Reviewer #2 also had issue with our use of the phrase ‘significant leap forward,’ which we now recognize was inappropriate. We have updated the text to say, “Our approach is an improvement upon previous methods that anchor onto simplifying assumptions about local tetrahedral packing or that over-segment the space.”

A distinction of our work is the identification and analysis of 3D-pores, which are the natural open pockets of void space within packed particles. This is in contrast to metrics that focus on particle properties, such as mean width and average contact number in **Maier, et al. 2022** and **Zachary, et al. Part II. 2011**. This is also in contrast to void space metrics that view the void space as a single entity, as is the case with the pore-size probability density function, $P(\delta)$ defined in **Zachary, et al. Part I. 2011** and **Maier, et al. 2022**. Pore size in these works by **Torquato et al.** refers to globally sampling Euclidean distance transform (EDT) values of the void space with no notion of 3D-pore segmentation. We are specifically interested in segmenting the void space of packed particles into 3D-pores because we view 3D-pores as the particulate-void-space-counterpart to particles, which is a newer perspective for viewing and analyzing void space. This approach has revealed interesting findings that we report in our manuscript.

Zachary, et al. Part I. 2011 studies 3- and 4-particle loops of binary disk packings and reports the void area normalized by the area of the smaller circle (*the paper states “normalized by the volume of a small particle,” but this appears to be a typo*). These particle loops are indeed examples of 2-D versions of 3D-pores. Our work extends this analysis by identifying and analyzing particle ‘loops’ in 3-D (i.e., 3D-pores) for a variety of packings (e.g., monodisperse, polydisperse, non-spherical, soft-body, etc.) and reporting 3D-pore volume, among other metrics. In addition, while **Zachary, et al.** mention that “loops with more [than 4] particles are rare in the packings,” we have found in our simulations of 3-D packings (and 3-D experimental data not

included in this manuscript) that it is common to find 3D-pores surrounded by many particles (see **Figure 4** of the present manuscript). In the current manuscript, we do not claim that our packings are in the MRJ state; however, we have updated our outputs by including a table that lists $1 - \Phi$ for all particle domains studied in our manuscript (see **Extended Data Table 1**), as well as particle contact information for all particle domains (see Code Ocean ‘results’ or Data Availability), in order to provide additional information that can be used to assess jamming.

The intention of the present work is to highlight our novel approach for segmenting 3D-pores that is robust for a variety of packed particle types. We then delve into 3D-pore analysis using a small set of pore descriptors (e.g., total # of 3D-pores, pore volume, longest length) in order to demonstrate accuracy and usefulness of the methodology. We anticipate that new patterns revealed by our analyses are of interest to the field. We chose to focus on a limited number of descriptors in order to keep the content focused (covering too many descriptors became overwhelming for readers). For this reason, we do not expand into phenomenon such as flow or mass transport through the void space (discussed in **Maier, et al. 2022** and **Torquato 2020**). **Maier, et al. 2022**, **Zachary, et al. Part I. 2011**, and **Zachary, et al. Part II. 2011** discuss particle volume fraction, Φ , (i.e., void volume fraction, $1 - \Phi$); however, since we are analyzing 3D-pores, we do not discuss $1 - \Phi$ in this paper. We have published a manuscript that discusses $1 - \Phi$ at length (Riley, et al. *Small* 2023). A deeper dive into 3D-pore and non-3D-pore void space analysis is planned for future manuscripts, which include some similar metrics seen in works by **Torquato et al.** For example, we compute a ‘mean local thickness’ of 3D-pores that is adapted from Hildebrand T and Rueggsegger P, 1997 (a variation on mean width applied to particles in **Maier, et al. 2022**), and we extract information termed ‘regions data’ that is conceptually similar to $F(\delta)$ and $H_v(r)$ (“void nearest-neighbor probability density function” in **Torquato 2020**).

Regarding superballs discussed in **Maier, et al. 2022**, we can certainly simulate domains comprising superballs with ranging p to highlight the difference between the results in **Maier, et al.** and our work. Our approach to generate rigid particle packings differs from **Maier, et al.** in that we use a rigid-body dynamics solver, a physics-based method for simulating the motion of rigid bodies. By tweaking simulation parameters, we can likely produce superball packings in MRJ without running into instability issues; however, this is something to be investigated. Rather than simulate new superball domains, we could instead analyze the exact superball

domains generated in **Maher, et al. 2022** if they can be provided. However, the scope of this work seems fitting for a follow-up manuscript to **Maher, et al. 2022**, which is an exciting prospect that we are happy to explore. For such a follow-up study, our analysis would cover our full range of descriptors that extend beyond the few discussed in the current manuscript. The goal would be to study the void space of superballs from the perspective of 3D-pores to draw conclusions about porosity that expand upon findings in **Maher, et al. 2022** like “pore sizes in...superball packings tend to become smaller...as ϕ increases” and “the maximum pore sizes in these packings are...bounded and less than the semiaxes of the superballs.”

Lastly, aside from our plots and data, a strength of the present work is 3D-pore visualization. We strive to make pore analysis attainable to the masses by drawing readers *into* the pores with our 3D renders. We showcase the variation in pore types across a variety of packed particle structures. In addition, we include analysis of deformable particles, which expands beyond traditional rigid particles and is highly valuable to the growing field of granular biomaterials.

2) It is well known in the physics community that the properties of particle packings and the associated void spaces, are dependent on the particle size and shape distributions, packing fraction, generation procedures (e.g., equilibrium vs nonequilibrium), and boundary conditions. It is not clear to me whether many statements in this work are as generally applicable to particle packings as the authors have claimed. Just as one example (there are many others), the authors claim that “our pore segmentation reveals that spheres, rods, and nuggets of equivalent volume produce statistically similar numbers of 3D-pores, while ellipsoids produce statistically more.” It is not clear to me whether this statement will hold if the packing fraction changes, or a different procedure is used to generate the particle packings. The authors should clearly identify the conditions under which their results apply.

To address these concerns, we have updated the main text to clarify our conditions, and we added the underlined portion to line 264: “A user working with similar particle types under similar conditions would only need to know δ and Φ of their system.” In addition, we have included new experiments and analysis that explore particle packing properties of container shape and particle friction (presented in Extended Data 9 and 10). We continue to see the same trends across our particle-shape groups. A complete sensitivity analysis for particle packing

properties would make for an interesting and useful follow-up study.

Reviewer #3 (Remarks on code availability):

The code appears usable and useful for the community.

Reviewer #4 (Remarks to the Author):

The manuscript by Riley et al. provides a model capable of extracting the local shape of the porosity in packed granular media. The algorithm used here is based on estimation of particle-particle distance that allows a better reconstruction of the space between particles compared with watershed or erosion type algorithm according to the authors. The manuscript provides a brief explanation on the construction of the pore phase using the LOVOMAP and then showcase its use by quantifying the number and size distribution of porosity in various particles packing. The rest of the manuscript demonstrates the use of the algorithm to obtain different quantities on pore structures form by different packing. The actual code is not provided and doesn't seem to be freely available, but the strategy is detailed in the supplementary document. I have a few general comments to discuss some aspect of the resulting data, but this seems like a new way to look at particle assemblies as far as I know. More importantly, I could see this applied in various situation ranging from surface active phenomenon (catalysis) to some more materials processing related issue (pore/defect evolution during heat treatment/sintering).

We appreciate the insights and constructive feedback from Reviewer #4. Below we attempt to clear confusion by addressing all points raised, including updates to the text and figures.

- The algorithm is dependent on the correct numbering and identification of the initial particles. Is the identification made in the first step of the volume rastering using already available particles identification from the assembly formation or does it give the particle identification number based on a separation distance or any other criteria? That's a significant difference with data that could be obtained experimentally in which the particles can be touching/not differentiate. In that sense the initial labelling would be key to obtain the correct pore structure

and thus a first step using watershed in 3D for instance would be needed to obtain a correct labelling, making this technique reliable only for clean/simulated data.

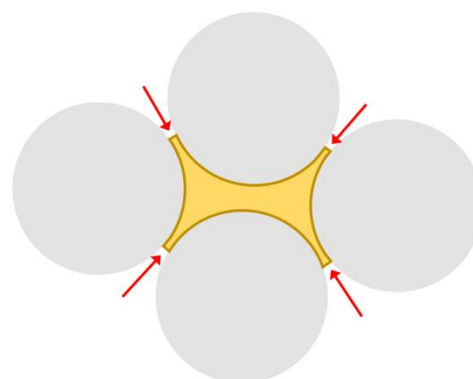
Reviewer #4 brings up a very important point. LOVAMAP was designed for granular material, so correct identification of individual particles is necessary. This is straightforward for simulated data. However, as with all uses of ‘real’ experimental data, the more clean the data, the more accurate the outputs. The granular field is familiar with thresholding images for particle or void space analysis, but Reviewer #4 is correct that the requirement for particle segmentation introduces an additional opportunity for inaccuracy and user bias. Luckily, while dependent upon image quality, mesh size, etc., particle segmentation is a relatively simpler segmentation problem, especially for spherical or convex particles, which is implemented in countless papers and software (both open source and licensed). In addition, the surge of machine learning algorithms makes us confident that accurate particle segmentation is achievable. For this purpose, we are currently adapting ML algorithms and software that already exist and are readily available to the public. Furthermore, while more accurate particle segmentation will lead to more accurate LOVAMAP results, if two touching particles are unable to be resolved, the code will consider the touching particles as one and continue with segmentation (see the response to the last comment from Reviewer #4). To process experimental data with LOVAMAP (not yet published), we have been using a watershed-based code to accomplish 3D-particle segmentation that is reported in our 2023 *Small* manuscript: <https://doi.org/10.1002/sml.202303466>. We have updated our Conclusion to address this comment:

Updated text:

Lastly, since LOVAMAP was specifically developed for particulate material, analyzing experimental data derived from imaging systems will require particle segmentation as the first step, which is a well-understood segmentation problem.

- Material fabrication is highly impacted by pore shape, for instance material strength/toughness dictated by pore shape and sharpness, sintering of powder guided by the surface curvature evolution over temperature. Is it possible to extract local curvature from these models?

The short answer is, yes – if one has a definition for curvature, we can use LOVAMAP to compute the metric. For curvature in particular, we have not yet derived an overly-satisfying metric in 3-D (although we are exploring known options, e.g., Minkowski tensors, etc.). At the moment, we can identify the number of ‘vertices’ (or ‘spinous tips’) of a 3D-pore, which alludes to curvature. To the right, we are showing a 2-D schematic of a pore (yellow) with its vertices indicated by red arrows. This data will be presented in follow-up papers along with many other metrics. As a biomaterials lab, we are very motivated to explore the relationship between particle properties and void space curvature since cells respond to surface curvature.



- The criteria to put a ‘door’ or not, i.e. connecting the pore spaces together or not seems relatively arbitrary. In terms of properties, for instance anything depending on percolation or mass transport in these pore space, two pores connected would be counted as disconnected using the criteria given. I agree that the criterion is well explained, however I am missing a rationale for choosing such a criterion in the first place since some of these pores are connected. This in turn have strong implications on the data extracted later, so it should be explained or if it is arbitrary it should be presented as this.

To reduce our word count in the main text, we had moved our rationale for our ‘door’ criteria to Supplementary Information, with the phrase “(see SI Methods for rationale)” following the sentence that describes our criteria. But we can see how this was too subtle, so to alert readers in a clearer way, we have added the title of the subsection to the phrase. It now reads: “(see SI Methods “Clustering peaks that belong to the same open space” for rationale).” In addition, we have added a secondary explanation for our rationale (with schematic) in SI to supplement the first explanation and provide more clarity. The updated text in SI is copied at the end of our response. If Reviewer #4 believes either of these explanations should be moved to the main text, we are happy to do so.

Regarding Reviewer #4's comment, **"In terms of properties, for instance anything depending on percolation or mass transport in these pore space, two pores connected would be counted as disconnected using the criteria given,"** we would like to clarify our goal and approach in hopes of addressing the concern. The goal is to segment 3D-pores that represent the open pockets of space inherent to the system, so this goal is entirely reliant on the geometry of the void space and independent of phenomena that are being studied, such as mass transport. To accomplish this goal, we are identifying the peaks (local center points) of the void space, which are again entirely reliant on the geometry of the system. Our criteria for associating peaks that are part of the same open space relies on the value of the Euclidean distance transform at each peak, as well as the relative distance between peaks. This criteria should be independent of structure properties other than geometry (assuming an unchanging system at equilibrium). We agree that there are unlimited options for criteria to accomplish such a segmentation; however, we settled on this approach because 1) the approach feels natural and intuitive, 2) rather than relying on a user-inputted parameter (which would introduce bias), the approach entirely relies on the geometry of the system, 3) we visually see good results, and 4) others in the field have used the same idea with good results (citations included in the main text). We have added the first point to the main text to motivate the choice (points 2 and 4 were already mentioned).

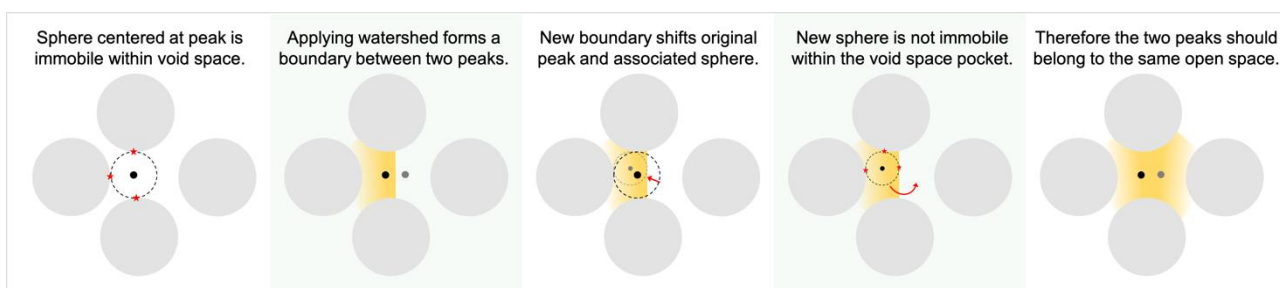
Updated text:

Now that we have an accurate representation of peaks connected by 1D-ridges, we aim to cluster peaks that belong to the same open space. This is the first step toward removing doors in order to accurately segment the void space into natural pockets. To help visualize our methodology, envision spheres centered at each peak with a radius equal to the peak's EDT value. For each pair of peaks, we check whether the physical distance between the sphere centers (peaks) is less than the sum of their radii (EDT values). Namely, if $d < r_1 + r_2$, where d is the Euclidean distance between p_1 and p_2 , and r_1 and r_2 are the radii of p_1 and p_2 , respectively, then p_1 and p_2 belong to the same open space (**Figure 1E**). The peaks associated with overlapping spheres are then considered 'connected,' and this data is stored in a sparse peak-adjacency matrix.

The rationale for this approach can also be explained using the analogy of rooms and doors. To start, all peaks represent the center of distinct rooms, all 1D-ridges that connect peaks represent the hallways connecting them, and the minimum EDT along 1D-ridges represents the location of a door that separates each room. By definition, a peak point, p , is a distance, r , from multiple walls (particles) in the room. For p to be the sole center of the room, a

sphere centered at p with radius r should not extend through any doors. If it does, the door is too massive for the space and should be knocked down to form one giant room.

An alternative explanation for our rationale goes as follows: for a given peak, p , consider the sphere, s_p , centered at the peak whose radius is equal to the peak's EDT value. By definition, this sphere is touching at least four boundary points (particles) and is therefore 'fixed' in place. If we were to use the naïve approach for segmentation, where the watershed algorithm is applied to each peak, consider a scenario where a watershed-boundary bisects s_p . This would occur when two peaks are closer to each other than they are to their nearest particle boundaries. If we segment the void space at this watershed-boundary, the original peak, p , no longer represents a local center point, and a new peak, p_{new} , must be identified that fits within the boundaries of the particles and the watershed-boundary. However, the sphere, $s_{p_{new}}$, centered at p_{new} is not immobile in the pocket of space – if you imagine shaking the system, $s_{p_{new}}$ would rattle around because it is small enough to pass through the watershed-boundary. When these scenarios occur, we argue that the peaks belong to the same open space. A 2D schematic of this explanation is shown below (see **Fig. 1G** for a schematic of the alternative scenario).

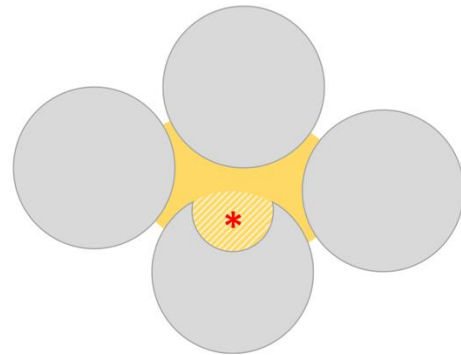


- How the equations for pore size/volume chosen in figure 2? It seems arbitrary to have two different ones but maybe it was simply the formula shape that gave the best fit? On that note, are the x axis of the graphs in figure 2 J and K particles diameter directly (in μm for instance) or the fitting formula value as it seems to be the case from the formatting.

Yes, our approach was to identify the equation that gave the best fit line for each dataset. The x-axes in Figs. 2J,K refer to the fitting formula values. We have updated the plots in Figs. 2J,K to remove the “x =” in the x-axis labels in hopes that this more standard notation will clear up any confusion.

- Could this work with irregular particles as well (not just roughness on the surface but actually rough, multifaceted shape)?

The code itself is robust for multifaceted shapes in that it is able to accomplish segmentation. However, for non-convex shapes, void space that exists within the convex hull of the particle will always be associated with the nearest pore that lies outside of the convex hull. To demonstrate with a 2D schematic, I am showing a collection of four gray particles, where the bottom particle is non-convex. The red asterisk indicates a void space region within the bottom particle that lies within the convex hull of the particle. This region (dashed yellow) will be considered part of the pore shown in solid yellow that lies immediately adjacent to the dashed yellow.



The code was specifically developed for granular material, so the segmentation approach becomes less appropriate if the scale of the void-space-of-interest lies within particles – for example, if the ‘particles’ were large shards of trabecular bone. We have explored, in theory, a future update to the code that extends the void space analysis to non-granular material; however, to maintain the accuracy that we can accomplish with our MAPC approach, rather than distinct particles, we would have to identify distinct ‘surfaces’ – where every change in curvature (in 3D) would define a different surface of the solid material. As one can imagine, this is a challenging endeavor and would require substantial code change. We have updated our Conclusion to include this text:

Updated text:

Our segmentation approach becomes less appropriate if the scale of the void-space-of-interest lies within particles – for example, if particles are large shards of trabecular bone. We have explored, in theory, a future update to the code that extends void space analysis to non-particulate matter; however, to maintain the accuracy that we achieve with our MAPC approach, rather than distinct particles, distinct ‘surfaces’ would need to be identified – where every change in curvature (in 3D) would define a different surface of the solid material. As one can imagine, this is a challenging but possible endeavor.

Decision Letter, first revision:

Date: 12th September 23 23:55:02

Last Sent: 12th September 23 23:55:02

Triggered By: Jie Pan

From: jie.pan@us.nature.com

To: tatiana.segura@duke.edu

CC: computacionalscience@nature.com

BCC: jie.pan@us.nature.com

Subject: AIP Decision on Manuscript NATCOMPUTSCI-23-0461A

Message: Our ref: NATCOMPUTSCI-23-0461A

12th September 2023

Dear Dr. Segura,

Thank you for submitting your revised manuscript "The empty space of packed particulate materials" (NATCOMPUTSCI-23-0461A). It has now been seen by the original referees and their comments are below. The reviewers find that the paper has improved in revision, and therefore we'll be happy in principle to publish it in Nature Computational Science, pending minor revisions to satisfy the referees' final requests and to comply with our editorial and formatting guidelines.

We are now performing detailed checks on your paper and will send you a checklist detailing our editorial and formatting requirements in about 10 business days. Please do not upload the final materials and make any revisions until you receive this additional information from us.

TRANSPARENT PEER REVIEW

Nature Computational Science offers a transparent peer review option for original research manuscripts. We encourage increased transparency in peer review by publishing the reviewer comments, author rebuttal letters and editorial decision letters if the authors agree. Such peer review material is made available as a supplementary peer review file. **Please remember to choose, using the manuscript system, whether or not you want to participate in transparent**

peer review.

Please note: we allow redactions to authors' rebuttal and reviewer comments in the interest of confidentiality. If you are concerned about the release of confidential data, please let us know specifically what information you would like to have removed. Please note that we cannot incorporate redactions for any other reasons. Reviewer names will be published in the peer review files if the reviewer signed the comments to authors, or if reviewers explicitly agree to release their name. For more information, please refer to our [FAQ page](https://www.nature.com/documents/nr-transparent-peer-review.pdf).

Thank you again for your interest in Nature Computational Science. Please do not hesitate to contact me if you have any questions.

Sincerely,

Jie Pan, Ph.D.
Senior Editor
Nature Computational Science

ORCID

IMPORTANT: Non-corresponding authors do not have to link their ORCIDs but are encouraged to do so. Please note that it will not be possible to add/modify ORCIDs at proof. Thus, please let your co-authors know that if they wish to have their ORCID added to the paper they must follow the procedure described in the following link prior to acceptance: <https://www.springernature.com/gp/researchers/orcid/orcid-for-nature-research>

Reviewer #2 (Remarks to the Author):

The authors have done a nice job replying to reviewer comments in general, and to my own, specifically. I find the manuscript to be much improved as a result of the authors' continued efforts. I have two remaining comments that I believe the authors should consider, but I view the manuscript as very much moving in the right direction.

EDF4: Why is R^2 for the 90/100 mixture significantly lower than for the other mixtures? Random chance? A boundary case where the model begins to unravel? What would happen at 95/100? 100/105? What does the transition from 80/100 ($R^2=0.95$) to 90/100 ($R^2=0.71$) look like? Does R^2 decrease gradually? Or, is there a sharp drop off at some point?

Particle gradation: The authors have elected to focus on materials that are quite poorly graded (i.e., uniform). The minimum ratio of $d_{\text{small}}/d_{\text{large}}$ appears to be 0.4. In addition, only monodisperse and bidisperse assemblies of particles have been considered. Referring now specifically to spheres, it is well-known that packing geometry is quite simplistic for monodisperse assemblies. Indeed, for monodisperse assemblies it has long been known (Filep 1937) that there are a finite (and countable) number of microstates accessible in the system. (It has been shown numerically that the same is likely true for bidisperse assemblies, but no closed-form solution exists.) In a monodisperse assembly of frictionless particles a minimum of

$cn=6$ is required for stability (3 equations of motion and $cn/2$ contact force balances per particle), and the maximum attainable cn is 12 (Kepler; FCC, HCP). For assemblies with more realistic ratios of $d_{\text{small}}/d_{\text{large}}$, the maximum cn observed in an assembly can be >100 . The point, of course, is the topological complexity of the pore space grows rapidly with the polydispersity of the assembly. How robust is the authors' algorithm in the face of this growing informational entropy? (Note that I am not referring to the resolution of the calculations and the images -- I grant that will be a challenge, but one that can be overcome with sufficient computational might. Rather, do the authors' algorithms continue to perform at the level demonstrated in the manuscript when polydispersity increases?

Lajos, F. 1937. "Egyenlő gömbökből álló halmazok." *Vízügyi közlemények*, 19 (1): 128–144.

T. Matthew Evans
Professor, Geotechnical Engineering
Oregon State University

Reviewer #2 (Remarks on code availability):

The code is well-documented with adequate instructions for installation. Given its length and interdependency, verification and validation are not possible a priori, but only through a large suite of demonstration calculations compared to closed-form solutions and expected asymptotic behavior. The authors have mostly provided this in the manuscript, though there are some edge cases that seem to be potentially problematic or otherwise unexplored (see comments above).

Reviewer #3 (Remarks to the Author):

The authors have addressed my concerns, and I am happy to recommend the manuscript for publication. I appreciate the great efforts that the authors put in their response.

Reviewer #3 (Remarks on code availability):

The code appears to be usable.

Reviewer #4 (Remarks to the Author):

The authors provided a strong effort to answer all reviewers comments. I won't be judging the novelty claims on this manuscript as other reviewers involved are clearly a lot more knowledgeable than me on that aspect. I simply want to commend the authors for highlighting more clearly the assets and limitations of the current algorithm. The description of the pore space is of prime importance for number of application and this approach seems like a step forward, with the limitation that applying to experimental data sets will be difficult due to the requirement of particles definition, but not impossible.

Reviewer #4 (Remarks on code availability):

The main code is available on codeOcean and could be accessed. The readme is there and provide a quick run through of the installation and packages needed. Given the format provided I couldn't check the installation as everything is packaged already with CodeOcean but I couldn't find any obvious limitations. Without being able to install and run it on a different computer it is hard to say more than that.

Author Rebuttal, first revision:

Reviewer #2 (Remarks to the Author):

The authors have done a nice job replying to reviewer comments in general, and to my own, specifically. I find the manuscript to be much improved as a result of the authors' continued efforts. I have two remaining comments that I believe the authors should consider, but I view the manuscript as very much moving in the right direction.

We genuinely appreciate Reviewer #2's comments and interest in the work, which yet again, have led to an improved version of the manuscript.

EDF4: Why is R^2 for the 90/100 mixture significantly lower than for the other mixtures? Random chance? A boundary case where the model begins to unravel? What would happen at 95/100? 100/105? What does the transition from 80/100 ($R^2=0.95$) to 90/100 ($R^2=0.71$) look like? Does R^2 decrease gradually? Or, is there a sharp drop off at some point?

As we increase the diameter of the second particle population, the slope of the linear regression approaches 0 asymptotically. This means the # of 3D-pores no longer depends on the x-values (% mixtures) – a finding that we would expect - and therefore R^2 drops rapidly. Mathematically, the SSR approaches the SST when the slope of the regression is nearly zero, and therefore R^2 decreases. It is certainly an interesting question to study the

$$R^2 = 1 - \frac{\text{sum squared regression (SSR)}}{\text{total sum of squares (SST)}},$$

$$= 1 - \frac{\sum (y_i - \hat{y}_i)^2}{\sum (y_i - \bar{y})^2}.$$

curve of the R^2 values, which we imagine would interpolate smoothly between the provided R^2 values (Supp. Fig. 3b). Since our x-axis represents the proportion of the smaller particle population, a mixture of 100/105 would be nearly equivalent to 95/100 except in a smaller container size, making it less suitable for comparison against our other values. While we are now itching to report data for 85/100 and 95/100, doing so would require simulating 220 additional particle domains because we include 10 replicates for each category. Given $R^2 = 0.9995$ when fitting a curve to the slopes (Supp. Fig. 3c), inclusion of 85/100 and 95/100 will likely not affect the final interpretation.

Particle gradation: The authors have elected to focus on materials that are quite poorly graded (i.e., uniform). The minimum ratio of $d_{\text{small}}/d_{\text{large}}$ appears to be 0.4. In addition, only monodisperse and bidisperse assemblies of particles have been considered. Referring now specifically to spheres, it is well-known that packing geometry is quite simplistic for monodisperse assemblies. Indeed, for monodisperse assemblies it has long been known (Filep 1937) that there are a finite (and countable) number of microstates accessible in the system. (It has been shown numerically that the same is likely true for bidisperse assemblies, but no closed-form solution exists.) In a monodisperse assembly of frictionless particles a minimum of $cn=6$ is required for stability (3 equations of motion and $cn/2$ contact force balances per particle), and the maximum attainable cn is 12 (Kepler; FCC, HCP). For assemblies with more realistic ratios of $d_{\text{small}}/d_{\text{large}}$, the maximum cn observed in an assembly can be >100 . The point, of course, is the topological complexity of the pore space grows rapidly with the polydispersity of the assembly. How robust is the authors' algorithm in the face of this growing informational entropy? (Note that I am not referring to the resolution of the calculations and the images -- I grant that will be a challenge, but one that can be overcome with sufficient computational might. Rather, do the authors' algorithms continue to perform at the level demonstrated in the manuscript when polydispersity increases?

We had intentionally chosen to limit the scope of the manuscript to monodisperse particle domains for the sake of focusing the breadth – so as not to overwhelm the reader (a criticism in past versions of the manuscript). However, to address the concern from Reviewer #2 and to provide examples of the code running on polydisperse domains, we have extended Figure 3 (previously Fig. 2) to include polydisperse results, as well as run the additional domains in Code Ocean to report the full datasets. By including a ‘standard deviation’ term to our x-axis transformations in Equation 1 and 2, our equations adapt to predict # 3D-Pores (per pL) and median pore volume for polydisperse spherical packing. While not included in this manuscript, we have run LOVAMAP on incredibly polydisperse domains ($d_{\text{small}}/d_{\text{large}} \approx 0.02$) that were simulated to replicate the polydispersity seen in

microscope images of packed microparticles fabricated in lab using an emulsion technique, and we had no issues with the algorithm. We have also run the code directly on microscope image data of granular biomaterials that are inherently polydisperse, but these results are unfortunately outside of the scope of the current manuscript.

Lajos, F. 1937. “Egyenlő gömbökből álló halmazok.” *Vízügyi közlemények*, 19 (1): 128–144.

T. Matthew Evans

Professor, Geotechnical Engineering

Oregon State University

Reviewer #2 (Remarks on code availability):

The code is well-documented with adequate instructions for installation. Given its length and interdependency, verification and validation are not possible a priori, but only through a large suite of demonstration calculations compared to closed-form solutions and expected asymptotic behavior. The authors have mostly provided this in the manuscript, though there are some edge cases that seem to be potentially problematic or otherwise unexplored (see comments above).

We have now included a wider range of test domains in Fig. 3, which span variations in particle size, stiffness, shape, friction, configuration, polydispersity, and container shape. While test options are countless, we hope our current suite is considered sufficiently large to support our conclusions.

Reviewer #3 (Remarks to the Author):

The authors have addressed my concerns, and I am happy to recommend the manuscript for publication. I appreciate the great efforts that the authors put in their response.

Thank you Reviewer #3.

Reviewer #3 (Remarks on code availability):

The code appears to be usable.

Reviewer #4 (Remarks to the Author):

The authors provided a strong effort to answer all reviewers comments. I won't be judging the novelty claims on this manuscript as other reviewers involved are clearly a lot more knowledgeable than me on that aspect. I simply want to commend the authors for highlighting more clearly the assets and limitations of the current algorithm. The description of the pore space is of prime importance for number of application and this approach seems like a step forward, with the limitation that applying to experimental data sets will be difficult due to the requirement of particles definition, but not impossible.

Reviewer #4, we genuinely appreciated your originally comments as they added transparency, which strengthened the manuscript.

Reviewer #4 (Remarks on code availability):

The main code is available on codeOcean and could be accessed. The readme is there and provide a quick run through of the installation and packages needed. Given the format provided I couldn't check the installation as everything is packaged already with CodeOcean but I couldn't find any obvious limitations. Without being able to install and run it on a different computer it is hard to say more than that.

Final Decision Letter:

Date: 9th October 23 11:12:33

Last Sent: 9th October 23 11:12:33

Triggered By: Jie Pan

From: jie.pan@us.nature.com

To: tatiana.segura@duke.edu

BCC: rjsart@springernature.com,fernando.chirigati@us.nature.com,rjsproduction@springernature.com,jie.pan@us.nature.com,computationalscience@nature.com

Subject: Decision on Nature Computational Science manuscript NATCOMPUTSCI-23-0461B

Message: Dear Professor Segura,

We are pleased to inform you that your Article "Identification and analysis of 3D-pores in packed particulate materials" has now been accepted for publication in Nature Computational Science.

Once your manuscript is typeset, you will receive an email with a link to choose the appropriate publishing options for your paper and our Author Services team will be in touch regarding any additional information that may be required.

Please note that <i>Nature Computational Science</i> is a Transformative Journal (TJ). Authors may publish their research with us through the traditional subscription access route or make their paper immediately open access through payment of an article-processing charge (APC). Authors will not be required to make a final decision about access to their article until it has been accepted. Find out more about Transformative Journals

Authors may need to take specific actions to achieve compliance with funder and institutional open access mandates. If your research is supported by a funder that requires immediate open access (e.g. according to Plan S principles) then you should select the gold OA route, and we will direct you to the compliant route where possible. For authors selecting the subscription publication route, the journal's standard licensing terms will need to be accepted, including self-archiving policies. Those licensing terms will supersede any other terms that the author or any third party may assert apply to any version of the manuscript.

If you have any questions about our publishing options, costs, Open Access requirements, or our legal forms, please contact ASJournals@springernature.com

Acceptance of your manuscript is conditional on all authors' agreement with our publication policies (see <https://www.nature.com/natcomputsci/for-authors>). In particular your manuscript must not be published elsewhere and there must be no announcement of the

work to any media outlet until the publication date (the day on which it is uploaded onto our web site).

Before your manuscript is typeset, we will edit the text to ensure it is intelligible to our wide readership and conforms to house style. We look particularly carefully at the titles of all papers to ensure that they are relatively brief and understandable.

Once your manuscript is typeset, you will receive a link to your electronic proof via email with a request to make any corrections within 48 hours. If, when you receive your proof, you cannot meet this deadline, please inform us at rjsproduction@springernature.com immediately.

If you have queries at any point during the production process then please contact the production team at rjsproduction@springernature.com. Once your paper has been scheduled for online publication, the Nature press office will be in touch to confirm the details.

Content is published online weekly on Mondays and Thursdays, and the embargo is set at 16:00 London time (GMT)/11:00 am US Eastern time (EST) on the day of publication. If you need to know the exact publication date or when the news embargo will be lifted, please contact our press office after you have submitted your proof corrections. Now is the time to inform your Public Relations or Press Office about your paper, as they might be interested in promoting its publication. This will allow them time to prepare an accurate and satisfactory press release. Include your manuscript tracking number NATCOMPUTSCI-23-0461B and the name of the journal, which they will need when they contact our office.

About one week before your paper is published online, we shall be distributing a press release to news organizations worldwide, which may include details of your work. We are happy for your institution or funding agency to prepare its own press release, but it must mention the embargo date and Nature Computational Science. Our Press Office will contact you closer to the time of publication, but if you or your Press Office have any inquiries in the meantime, please contact press@nature.com.

An online order form for reprints of your paper is available at <https://www.nature.com/reprints/author-reprints.html>. All co-authors, authors' institutions and authors' funding agencies can order reprints using the form appropriate to their geographical region.

We welcome the submission of potential cover material (including a short caption of around 40 words) related to your manuscript; suggestions should be sent to Nature Computational Science as electronic files (the image should be 300 dpi at 210 x 297 mm in either TIFF or JPEG format). We also welcome suggestions for the Hero Image, which appears at the top of our [home page](http://www.nature.com/natcomputsci); these should be 72 dpi at 1400 x 400 pixels in JPEG format. Please note that such pictures should be selected more for their aesthetic appeal than for their scientific content, and that colour images work better than black and white or grayscale images. Please do not try to design a cover with the Nature Computational Science logo etc., and please do not submit composites of images related to your work. I am sure you will understand that we cannot make any promise as to whether any of your suggestions might be selected for the cover of the journal.

You can now use a single sign-on for all your accounts, view the status of all your manuscript submissions and reviews, access usage statistics for your published articles and download a record of your refereeing activity for the Nature journals.

To assist our authors in disseminating their research to the broader community, our SharedIt initiative provides you with a unique shareable link that will allow anyone (with or without a subscription) to read the published article. Recipients of the link with a subscription will also be able to download and print the PDF.

As soon as your article is published, you will receive an automated email with your shareable link.

We look forward to publishing your paper.

Best regards,

Jie Pan, Ph.D.
Senior Editor
Nature Computational Science

P.S. Click on the following link if you would like to recommend Nature Computational Science to your librarian: https://www.springernature.com/gp/librarians/recommend-to-your-library

** Visit the Springer Nature Editorial and Publishing website at www.springernature.com/editorial-and-publishing-jobs for more information about our career opportunities. If you have any questions please click here.