Replies to reviewer 2

For what concerns Rev.2 we proceed as follows: first we reply to the "15 major comments", then we reply to the "generic" ones, which we divided in 9 additional comments. Then to the "Implementation details" and finally to the "minor points". This is because some of the answers to the generic comments are present in the major ones.

Major points

1. There are some occasions where the authors mention the necessity of reaching a maximum packing. Why is this the case? I cannot find a reference for that. This is confusing because I first though that the code was designed to reach the maximum packing, but it failed. However what is shown later is that even without enforced packing the code produces aggregates that are too packed with respect to the physical evidence. This is because of a switch in the focus from packing to porosity. I would suggest to provide evidence for the maximum packing theory (or minimum porosity) and shape the manuscript in the sense that it only looks for the packing tau and not porosity (or viceversa).

Reply:

We were probably not clear enough on this point. As you said, maximum packing is not a reality in the real field of application of SCARLET (mostly volcanology). This is because field observations show that natural aggregates are rarely structured in the most packed configuration. However, the maximum packing problem has a wide historical background in literature and several benchmarks that are interesting to use, both for testing the algorithm and also to show how real aggregates are actually far from this scenario. Packing and porosity are closely related concepts (since $\phi_{agg} = 1 - \tau$), and in our field of application (volcanology) porosity is usually more used than packing. However, τ have been more investigated than ϕ_{agg} in literature, especially from a theoretical point of view. The need of analytical expressions, not just as a benchmark, but also as an upper limit to better understand how the different initial configurations translate in the "maximum packing plot"(Fig.10 of the new version) convinced us to keep sec. 3.1.2 and to improve its initial part, to clarify that the aim of SCARLET is not dedicated to the maximum packing. But it is still an interesting test bench.

After having clarified the meaning of packing and porosity in sec. 2.2, we added at the beginning of sec.3.1.2 the following introductory sentence:

"The determination of the aggregate porosity is based on the assumption that the external surface that circumscribes the inner components is convex. This choice is motivated from the aggregate morphology associated to PC type aggregates in volcanology. Also in other applications this can be the case but the user must be a-priori aware of this. In fact, this approach may lead to an overestimation of the porosity in case, for example, of fractal-like aggregates for which the overall fractal dimension is less from three. In those cases the convex hull approximation can be seen as an upper limit for the maximum porosity, which converges to the actual porosity for aggregates that are well described by a convex geometry."

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2. Lines 10, 11, 54, 55, and other parts. I do not see a clear justification for the design choice of a "central oriented collision". This is clearly not the case for snowflakes [14]. The fact that samples of volcanic ash present one block much larger than the others can be explained by a non continuous size distribution of monomers and again the process of differential sedimentation.

Reply:

This is an interesting observation that deserves a few lines of clarification. The choice of a central oriented collisional process is a consequence of field observations of volcanic ash aggregates that clearly show a large inner core at their center (Taddeucci et al., 2011; Bagheri et al., 2016; Gabellini et al., 2020). It is worth stressing that these observations are taken during a real volcanic fallout (not from preserved samples on the ground). The explanation of why this happens is based on the different coupling of different particle sizes within a highly turbulent flow. In fact, most of the collisions that lead to volcanic aggregates actually happen within the plume, more than in the sedimentation process in the atmosphere. The central oriented collisional process is what describes the fate of particles with a certain degree of decoupling from some sizes of turbulent eddies within the plume. These particles are described by the so-called Abrahamson kernel (Abrahamson, 1975; Textor and Ernst, 2004). If you set your system of reference centered on the particle you will statistically observe particles coming from random directions, for which the central oriented collisional process is totally justified. Moreover, this is not true just in volcanology, but in all the fields where collisions happen in a highly turbulent flow with different sizes involved. For example, also in planetary formation, this is the case (see Volk et al., 1980; Ormel and Cuzzi, 2007).

And what you actually observe in the field is really a big particle coated – almost symmetrically – of other particle sizes (usually smaller). It is the observation of this kind of object – called in literature "particle clusters" that have motivated the code. Not viceversa.

In general, the code is suitable to describe the formation of aggregates in which the decoupling of the particle from the surrounding flow assures collisions coming from random directions.

However, we agree that this point must be more clear. We added in the introduction the following sentence:

"*The physical explanation of the formation of PC type aggregates involves the theoretical description of a polydisperse particle population within a highly turbulent flow, such as it is the case for a volcanic plume or cloud (Kieffer and Sturtevant, 1984). In presence of a cascade of turbulent eddies and grains of different sizes, particles will show different degrees of coupling with the turbulent flow (Volk et al., 1980). This will produce a wide spectrum of relative velocities (Ormel and Cuzzi, 2007). In the limiting case of particles with a negligible velocity correlation with respect to the others (i.e. the kinetic theory limit), the object will aggregate following an inertial aggregation mechanism (Textor and Ernst, 2004). This is typical of large particles that will jump from one eddy to another with a poor correlation with the dynamics of the eddies. In their path across the turbulent flow these objects will encounter smaller particles that, on the contrary, do show a high correlations with the eddies. This process can be modelled as a central collisional process in which the big particle is the target of random collisions coming from random directions.*"

In addition, following your observation (but keeping the hypothesis of central oriented collisions, for which the code has been purposely designed) we added a Boolean variable that can be used by the user to fix the direction from which random particle come. This option can

be activated in all those cases where there is an a-priori assumption of a preference in the direction of the collisions. The aperture of the cone is controlled in the same way as usual (closet.cone_aperture_degree). In the text we do not make the assumption of knowing a preferred angle for the particles, but we think – after your comment – that it is important to add this option to the code. Following this, we aldo added the following section in the methodology (sec 2.3):

"*The user can also fix a preferred direction for the cone for all the cases in which collisions occur along a preferential direction. We will not make this assumption in the rest of the paper, assuming that the collisions described comes from a fully isotropic turbulence with random angles of collisions.*".

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3. Line 15. I do not see why the distance of the CM should be minimized. What is the physical force that acts in this process?

Reply:

The minimization of the distance between the CM of the N possible particles placed at iteration i-th and the CM of the already placed structure is an algorithmic choice based on the request of having a simple but effective parameter to determine higher or lower degrees of packing, according to the user's request. In fact, it is important to stress that the user can play with the initial setup of the simulation to give more or less importance to the minimization process. SCARLET-1.0 has been conceived to study the geometrical packing of central collisional processes, more than to reproduce the action of well parameterized forces on complex-shaped bodies (that are really dependent on the specific application and for which we should also know all the torques to be accurate).

The forces acting in the process are given by the differential coupling with respect to the turbulent flow, due to nonlinear combination of the Stokes numbers and the eddie scales involved in the process. However, the final result of this non-trivial process is simply a relative velocity with random angle of impact.

Our approach has been: regardless of the nature of the force involved, we assume that it is a vector point inward to the core and we leave to the Montecarlo approach the effort to investigate how the packing will change as a function of the Euler's angles and rays investigated. I found very interesting your comment 4 (the next one) because I have noticed that SCARLET-1.0 slightly resembles some of the steps that you implemented within the SAM algorithm (pag. 9934 of your really interesting paper!). The difference in our case is the use of STL files and the assumption that the final result of the inward pushing of the external force is to take the final configuration whose CM is closest to the core. This, with a high degree of control for the user, that can manipulate the initial settings to produce different packings and a high degree of freedom in using STL files (very flexible format) and the possibility to 3D print the result (so she/he can also test in the 3D world of the lab the outcome of the simulation). To sum up, the underlying idea is that in SCARLET one is free to explore different possible geometries in a Montecarlo approach, under the assumption that there is an external force that pushes radially to the inward and that modifying the Euler's angle one can test how well the arbitrary shape can be fitted in the structure.

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4. The cited bibliography in the introduction needs some work. In particular for snowflake aggregation the only paper among those cited that covers aggregation is [4]. Some useful resources might be [14, 6, 5]. Concerning available models there are some resources [7, 10, 11] which are usually referenced using the following papers[6, 9, 2]. All these models are capable of dealing with arbitrary shapes and it does not seem to be difficult to deactivate the aggregation kernel in order to simulate a central aggregation process, this is evident especially for [9] where this effect is fully explored. A comprehensive review of snowflake aggregation models can be found in [13].

Reply:

Really thanks a lot these suggestions on specific literature about snowflake aggregation. We apologize for being too vague. Thanks especially for reference [14] which is really nice to be inserted in this context (see previous comment). I removed one old reference, but I would like to keep also others from the previous literature because they offer some fruitful ideas on possible algorithms to simulate snowflakes formation or simply ice growth. For example, we explicitly added the reference of Ning and Reiter 2007 because of the use of cellular automata. Moreover, since our paper is mostly oriented to volcanology, we would like also to keep some of the old references because we believe is important in our community to show different algorithms in different fields, more than providing a focused analysis on aggregation of snowflakes. So we propose to reformulate the sentence and adding some of your suggestions: *"ice and snowflakes formation and aggregation (Kessler et al., 1984; Westbrook et al., 2004; Maruyama and Fujiyoshi, 2005; Reiter, 2005; Ning and Reiter, 2007; Gravner and Griffeath., 2009; Tyynela and von Lerber, 2019; Ori et a Karrer et al., 2020)"*

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5. Line 55. I do not see how the evidence of finding an aggregate composed of a piece much larger than the other constituents is supporting the idea of a central aggregation effect. The same result can be achieved from the differential sedimentation of discontinuous distributions of monomers. This seems physically sound to me if I imagine the initial distribution of monomers as resulting from the explosion of rocks where the internal inhomogeneous distribution of mass and hardness might play a substantial role. Also what would be the physical force that generates this central pole of attraction? I think that these argumentation needs more discussion or the citation of previous work.

Reply:

The answer to this comment has already been given in the reply to your comment 2. Here we just add a few line of clarifications (we hope that the new lines added at line 55 are more clear now). The central collisional process are given when particles with different Stokes numbers show different degrees of coupling with the surrounding eddies. Some of them will be sufficiently decoupled in order to cross the eddies and interact with particles fully coupled with the turbulence. This is described by Abrahamson (1975) and Ormel and Cuzzi (2007). A more specific reference for volcanology can be found in Textor and Ernst (2004). In volcanology we found that the concentration of ash present in a volcanic plume is order of magnitude larger than the one sedimenting in the atmosphere (or in the lower part of the cloud). Therefore, our

attention is mostly focused on the plume, where most of the aggregation is meant to happen according to the time-scales measured during field observations (Bagheri et al., 2016). But most interestingly, field observation and aggregate collection with UV resins show almost symmetrical objects with a large core inside, which suggest a central collisional process of a decoupled particle passing to turbulent eddies and having random collisions without preferred angle of impact. That's why we are interested in central collisional processes.

However, thanks to your comment 2, we added to the code the possibility of having one fixed axis of the cone, which would mimic a collision that happens on a preferred direction (such as a larger particle falling in the atmosphere surrounded by smaller particles).

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6. Line 200-207 (also figures 6 and 7) the approach of having trajectory cones centered around the CM is not convincing to me. It seems to implicitly assume that the CM is always inside the core particle and thus the cone always intersects the particle surface. What happens if the core particle is shaped like a C or a parabola? The cone might not intersect the shape. Alternatively, consider a flat surface. In this case the contact pointwill always be the same regardless of the angle omega. I know that these are rather exotic cases (having theCM outside the shape is actually rather common for snowflakes), but I still do not know if I should considerthis code as rather generic or applicable to only specific geophysical situations.

Reply:
We really thank the reviewer for this comment, because it helped improve the code. In fact, this point was not sufficiently clear in the text (in general was not sufficiently explained the option of *closet.origin_in_the_CM*). The user can select if the center of the cone is coincident with the CM of the core (option 1) or with one of the center of its sphere-composite representation (option 2) (randomly selected by the code). When the topology of the core is in such a way that its CM is outside the STL file, the code switches to option 2. Here is an example using a "donut" as core and spheres as coating.

In sec. 2.3:
"*The center of the cone is placed at the origin of the axes, i.e. the CM of the core, or at the center of one of the spheres that form the sphere-composite representation of the aggregate, according to the user defined parameter closet.origin_in_the_CM. In case of a central core whose CM is outside the STL file, the second option is activated by default*".

In the caption of Fig.6:

"Cone, rays and rotations associated with the placing of one single coating particle. In the figure, the cone is coincident with the center of mass of the core. However, the user can modify the Boolean variable closet.origin_in_the_CM in order to make the center of the cone coincident with one of the spheres of the sphere-composite representation of the STL file."

In the caveats:
"1. When the center of mass of the core is placed outside the STL file, SCARLET-1.0 automatically places the center of the investigation cone at the center of one of the spheres that form its sphere composite representation."

<div align="center">***</div>

7. Sections 2.2 and 2.3 The code uses the sphere equivalent representation and avoid the overlapping of the spheres. Somehow it must be possible to get back the STL representation of the aggregates. How is this done? What is the degree of overlapping of the final STL polyhedrons? This might have an impact on the aggregate porosity?

Reply:
One of the interesting feature of the code is the capability to generate the STL file of the final aggregate. This is done by means of the Matlab function "stlwrite.m", which considers the triangulated patch of the single components of the aggregate as a single object. The degree of overlapping is controlled by the variables *closet.delta* and *closet.delta_2* at the top of the program, where the step-size is defined as a fraction of the particle size. And it is dependent on the shape of the objects involved and the number of spheres used. Yes, it can have an impact on the porosity, especially if the number of sphere is underdetermined respect to the shapes involved. This is somehow the price implicit in the sphere-composite models (the advantage is the possibility of using arbitrary shapes). However, if the sphere composite representation of the object is reasonably accurate, the error due to the overlapping is negligible respect to other aspects, such as the definition of the external volume of the aggregate which is used to evaluate the porosity (see reply to comment 3 of reviewer1). The important limit here is that if two STL surfaces are not "reasonably" in contact the final STL file cannot 3D printed. By "reasonably" we mean that, depending on the resolution of the printer and its technical details, it can be that the overlapping required to the structure should be more remarked than the one which is considered acceptable for a theoretical description of an aggregate. In our tests with a real 3D printer we found that the use of the global STL file for actual 3D printing is really dependent on the printer itself. For high resolution printer (orders of few microns) the default tolerances in overlapping worked reasonably well without spending time tuning *closet.delta* and *closet.delta_2*. However we believe that is really dependent on the setup under analysis and the complexity of the shape.

To better address this important point, especially for those who will try to use SCARLET to 3D print objects, we added the following warning in the caveats:
"3. SCARLET-1.0 generates the STL file of the virtual aggregate. This format can potentially be used for 3D printing. Here we refer to "potentially" because the success of the procedure depends on many conditions: the shape of the aggregate, the degree of overlapping of its components, the stability of the structure and the characteristics of the 3D printer. In some cases the user should decrease the tolerances used in the calculation of the intersection between spheres in order to have a more solid structure suitable for 3D printing. Further tests

will be conducted on this and in the GitHub page of the software new releases and notes will be posted on this in the future.".

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8. Line 245-265 This part is quite difficult to follow. The ellipsoids are defined only for by flatness and elongation if one assumes that the generic size is always the same, or scale invariant. Is the size always the same? also the parameter gamma is the ratio between a (a property of the monomers) and R (a property of the aggregates)this means that a is uniquely defined only if the same ellipsoids are used within an aggregate and R will depend on the number of monomers. Am I right?

Reply: In this test we follow the paper of Man et al. 2005, in which they tested the packing of ellipsoids of a predetermined shape (e.g. the principal axes of the ellipsoids are in the ratio 0.8:1:1.25). In this text is not so important the actual mean radius of the ellipsoid (defined as $\bar{a} = (a_1 \cdot a_2 \cdot a_3)^{1/3}$). What matters is that all the ellipsoids have the same shape, all the average radius (i.e. they must be identical) and that the final radius of the sphere (R) that contains them (the so called "spherical container" in their paper) is much larger than $\bar{a}$.

From your comment we found that we were not clear enough in explaining the setup used in the simulations, our fault. We propose to modify the paragraph as follows, hoping that it results more clear:

"The research of the maximum packing has always played an interest in mathematics and other practical applications (Hales, 2005; Man et al., 2005; Farr and Groot, 2009). As it is evident from Eq.1, porosity and packing are oppositely related: a larger packing will reduce the porosity of the aggregate, and viceversa. Even if SCARLET-1.0 has not been specifically designed for the research of maximum packing, it is somehow interesting to has been test the algorithmed with respect to this branch of investigation, for which analytical limits are provided according to the shape of the single components under analysis. In this paragraph we will evaluate the packing τ of spheres and ellipsoids with principal axes ratios of (a_1= 1.25: a_2= 1: a_3= 0.8), for which the theoretical values of the maximum packing τ are available in literature (see Fig.2 of Man et al., 2005). This particular choice of the principal axes constrains the value of flatness f=a_3/a_2 =0.8 and elongation e=a_2/a_1 =0.8. In our simulations we fix a_1=10 a.u. (arbitrary units) and we derive a_2 and a_3 from f and e (e.g. a_2=8 a.u. and a_3=6.4 a.u.). The average radius of the ellipsoid is defined as a̅= ⟦(a_1·a_2·a_3)⟧ ^(1/3) and with R we indicate the radius of a sphere with same external volume of the overall aggregate. evaluating the packing τ of spheres and ellipsoids with axes ratios (L=1.25):(I=1):(S=0.8), for which theoretical values of maximum packing are available in literature (see Fig.2 of Man et al., 2005). This particular choice of the largest axis (L), the intermediate (I) and the smallest one (S) guarantees an equal value for flatness (f=S/I=0.8) and elongation (e=I/L=0.8). The goal of the test is to relate the computed values of τ with the initial setup adopted in each simulation for solid angles, number of rays and number of rotations respectively, i.e. (Ω, N_r, N_o). Virtual aggregates created in this test are made of identical particles with the same size. Packing is displayed in Fig.9 as a function of the dimensionless parameter γ=a̅/R. ,where a̅ is the characteristic size of the ellipsoid or the sphere,and R is the radius of a sphere with same external volume of the aggregate.More in detail,a̅ is equal to the radius when spheres are adopted as particles; a̅= ⟦(a_1·a_2·a_3)⟧ ^(1/3) for an ellipsoid with principal axes 2a_1,2a_2,2a_3.The use of ellipsoids with equal values for flatness and elongation results in a simplified value for a̅=(L·e)/2. A variable number

of particles, comprised between a minimum of 10 and a maximum of 1000, has been used in all the simulations in order to achieve different values of γ.The results presented in Fig.9 show that the condition of maximum packing for ellipsoids can be easily reached only for values of γ>0.3. For values of γ<0.3 a larger number of rays must be adopted to reach the same degree of packing. For spheres and for the conditions analyzed in this example, the maximum packing is reached for γ>0.1. As expected and it is clearly shown for spheres, the increase in number of rays is not linearly related neither to the decrease of γ no the increase of τ. In the search of the maximum packing, it seems that a large number of rays can have larger benefits respect to the increase of the number of rotations, at least for the shape analyzed in this example. In addition, we notice that for spheres the minimum value for packing τ is about 20%; a value that correspond to a maximum porosity of 80% (see Eq.1). This threshold can be considered as a good estimation of the maximum value for porosity reachable by means of the SCARLET-1.0 algorithm."

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9. Line 308 I suggest to not refer to this as a time evolution. As a matter of fact the aggregation code does not involve process rates. Time is never a variable here. What the authors can analyze is the evolution with respect to the number of collisions. An "aggregation stage" perhaps. I would also be curious of knowing why this is not observable in field and lab studies. One can envision the possibility of collecting many samples and derive the properties of them as a function of the number of monomers, which is precisely what the code can give.

Reply:
We found this comment really important because it helped us a lot improve the message that we wanted to outline in this text. "Change in time" is too confusing here, because time is only implicitly present here and under some assumptions that may not be immediate to the reader. We propose to modify it as follows: "Porosity of volcanic ash aggregates as a function of the aggregation stage".
To answer your curiosity: in field observations, during a real volcanic fallout, you usually collect the final product of aggregation (the aggregate). And also this is actually not that easy, because you have to be in the right place at the right moment, using the right sampling strategy and to be below a volcanic eruption and its fallout. The tricky part is that volcanic aggregates are very fragile and they break at the impact with the soil or the sampling surface. Moreover, since most of the aggregation does not happen in the sedimentation process, but in the volcanic plume and in the volcanic cloud, it is almost impossible to conduct a sampling campaign during the stages when aggregation occurs. There will be in the next months some challenging field campaigns to sample directly in the volcanic cloud, but for the moment this is the frontier of the investigation.
In the lab, of course, things are easier, but still not easy. Because volcanic particles are not easy to suspend in a vertical wind tunnel due to their irregular shape. And in order to reach enough collisions, particle concentration should be quite high and the time of suspension of order of minutes (which is not that trivial when different sizes are involved and the shape is totally irregular). Moreover: volcanic plumes have Reynolds number not far from 10^{10}-10^{11}. A wind tunnel is orders of magnitude away from a realistic environment.
So...yes, you can collect many samples during an eruption. This is easy (if you are in the right location at the right time). However, from a practical point of view, you will collect "broken"

aggregates. In this case you only know the population inside them, but not the different stages that led to the creation of the entire aggregate. You can use something more advanced for the collection, such as UV resins. But then the only way to access the details of the internal structure of the aggregate is by means of a micro X-ray tomography (the size of the aggregates is between 100-1000 microns). This is still feasible, but quite advanced, since to our knowledge we will be the first in our community to do this kind of study (we already have the samples, but due to the pandemic the analysis went slower than planned). Once the micro X-ray tomo will be done, it will be interesting to use SCARLET with this dataset.

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10. Line 357 The expected mass loss is a new concept for me. Also what is this Na replication actually doing?

Reply:
Yes this is an interesting point and it outlines the differences between each different field of study. In practice the sampling technique used for the campaign involved in the example are based on the use of sticky papers and High-Speed Cameras (HSC). With the HSC you can precisely record the moment of the impact, derive some fruitful information from the falling phase and check what happens during the impact with the sticky paper (a particular paper that can be used later in the Scanning Electron Microscope (SEM)). This is described in Bagheri et al. 2016 (if you are more interested and curious take a look to Fig.5 of that paper). At the moment of the impact a lot of fine ash of the coating (and also the big core!) escapes from the sticky paper. What remains on the sticky paper is just part of the total coating. Moreover, due to the small sizes involved (tens of microns) is also difficult to completely conserve all the material that has remained on the sticky paper itself. Sometimes aggregates are then analyzed with special machines that use laser diffraction to automatically measure the sizes (but you lose sometimes mass also in this process) or by using the SEM. In the SEM you can obtain information on the sizes of the particles of the coating, but unfortunately there is some overlapping between them. In fact, the coating on the sticky paper is three-dimensional more than two-dimensional, and when you coat them with conductive metal (usually gold) you freeze the 3D structure. So you hide some particles below others. All these variables in the collection and analysis lead to a not negligible mass of the original coating of ash in the aggregate shell. In this example, which is just an application because of the huge uncertainties behind the collection process, we postulate that the final grain-size analyzed at the SEM is still representative of the entire initial population of the coating. But with a loss of particles. So we replicate N_a times the number of particles observed in each bin at the SEM. We hope that now is more clear.
We also added the following sentence:
"This means to multiplicate N_a times the number of particles reported in each bin in the work of Bagheri et al. (2016)."

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11. I disagree with the statement at line 359. The small particles might be individually a second order contribution to the final mass. But collectively they can contribute more than the largest. This is simply because the contribution to the final mass depends also on the number

concentration and according to fig 14 there are a lot of small pieces. More importantly, the contribution of small particles to packing is even more important. This is because small pieces are the only one able to fill the gaps in between larger pieces and achieve higher packingvalues. as packing approaches 1 the importance of having small pieces accounted for is larger and larger.

Reply:

We understand the criticism of this comment. However, we still believe that neglecting particles of 15 and 23 microns, respect to the others, is totally within the accuracy implicitly assumed in the example, for which we miss about 80% of the number of particles actually present in the original coating! The error on the porosity can be estimated to be about 7% in the porosity if we neglect the sizes of 15 and 23 microns, which is totally in agreement with the approximations considered in the example. This number can be estimated considering particles as spheres, that the final size of the aggregate is about 670 micron and that we know the sizes of the core and the coating

$$\phi_{tot} = 1 - \frac{V_{solid}}{V_{tot}} = 1 - \frac{V_{core} + V_{coat\_simpl} + V_{miss}}{V_{tot}}$$

$$\phi_{appr} = 1 - \frac{V_{core} + V_{coat\_simpl}}{V_{tot}}$$

$$\frac{\phi_{tot}}{\phi_{appr}} = \frac{\frac{V_{tot} - (V_{core} + V_{coat_{simpl}} + V_{miss})}{V_{tot}}}{\frac{V_{tot} - V_{core} - V_{coat\_simpl}}{V_{tot}}} = \frac{V_{tot} - V_{core} - V_{coat_{simpl}} - V_{miss}}{V_{tot} - V_{core} - V_{coat\_simpl}}$$

$$= 1 - \frac{V_{miss}}{V_{tot} - V_{core} - V_{coat\_simpl}}$$

But:

$$\frac{V_{miss}}{V_{tot} - V_{core} - V_{coat\_simpl}}$$

$$= \frac{k \cdot (258 \cdot 15^3 + 138 \cdot 23^3)}{670^3 - 270^3 - k \cdot (62 \cdot 32^3 + 24 \cdot 40^3 + 22 \cdot 48^3 + 8 \cdot 56^3 + 6 \cdot 65^3 + 73^3 + 2 \cdot 80^3)}$$

$$= 0.07$$

Where k=6 is the correction term for the missing particles. Therefore $\phi_{tot} = 0.93 \, \phi_{appr}$.

However, we agree that this sentence should be more clear. We propose to add the following sentence to the text:

"This assumption leads to an overestimation of the porosity of the order of 10% respect to the case where the entire grain-size distribution is considered. This compromise is then totally acceptable if we consider the uncertainty related to the poor constraint on the grain-size distribution actually present in the original sample before its impact with the sticky paper."

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12. Line 378 this measured values between 80-97%, why are them not reported in the figure 16 and 17 like it was the case for figure 15? I have the feeling that in contrast to what is affirmed here even the loose packing is not capable of reproducing the observed porosities.

Reply:

No, the reason is simply related to the fact that for Fig.15 we used a much better field technique that allowed us to measure the density of the falling aggregate prior to its impact with the sticky paper. In addition, the use of the High-Speed Camera allowed us to film the core before bouncing away from the sticky paper. Therefore we had a much better constraint both on the grain-size involved and on the actual density of the specific sample.

In fig.16 the sample is related to the eruption of 2010, where simply the sticky papers were involved. No direct measurement on the aggregate density of the specific sample analyzed. The values of estimated porosity reported in the paper are taken from literature and they are all derived from indirect observations (for PC1 type). Therefore we preferred to not overlap in the plot the value of the measured porosity (as in Fig.15), simply because even in the case of a not perfect matching, is difficult to say that this is due to the virtual reconstruction or to large errors in the estimation of the density or the grain-size (which in this case were not directly measured, differently from Fig.15).

We think that the sentence of line 370 is clear in specifying this and the limits of this particular example.

13. An effect that was not commented emerges from fig 16. The sequential densely packed do not reach a plateau, but their porosities start decreasing after some number of collisions.

Reply:

We really thank the reviewer because this is an interesting observation that is worth adding to the text. We propose to ass in the discussion (sec. 4.2, at the very end), the following sentence:
"A final aspect that is worth to be discussed is that in case of a sequential deposition of the coating, after reaching the plateau, the porosity of the aggregate starts decreasing. This can be explained by the fact that adding fine ash after coarse ash will not significantly alter the total volume of the aggregate, but this process increases the mass of the aggregate and reduces the voids. This leads to the observed decrease in porosity."

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14. Line 425-427. I do not really understand the argumentation here. The authors suggest that there is a violation of the contact condition. Is this condition the idea that aggregates stick immediately on contact? Also they speculate that liquid bonds might be responsible for this. Is there any evidence of this process? Also, it is not immediate to me understanding how this liquid bond works. Perhaps the particle collide forming a liquid bond, but they bounce. While bouncing the liquid solidifies generating the permanent bond? Is this the process suggested? Shouldn't there be evidence of this in the samples? I know I might sound repetitive, but also the implementation of a collection kernel (differential sedimentation, brownian or else) would increase the final aggregate porosity.

Reply:

A few clarifications are needed. From an historical point of view (early '80s), in volcanology aggregation has usually been classified within two categories: *wet* and *dry* aggregation. *Wet* means a macroscopic layer of liquid water or ice is involved (Gilbert and Lane, 1994). *Dry* means that viscoelastic dissipation, electrostatic charges, dipole induced and other sticking mechanisms where water is not involved are present.

SCARLET works assuming that particles are physically in contact. This means that there is no other medium present within particles. This is the case, in general, for what we refer to as "Particle Clusters" (PC1, PC2, PC3), which usually belong to the "dry" family. This is not the case for other types of aggregates (e.g. Accretionary Pellets (AP) and mud aggregates, for example), where a macroscopic liquid water can be present. However, this classification of dry and wet has been "criticized" in the last decade (Brown et al., 2012), because also for PC type aggregates cannot be completely excluded the presence of liquid water in some cases. The difference is that in PC type the amount of water is probably less evident than in the other types.

More than how immediate is the sticking, here is important what produces the sticking. If the sticking is produced by the presence of water, there is a secondary medium between solid particles that cannot be fully described by SCARLET, this was the point of the sentence in the paper.

The liquid bonds can act in three ways: ice bridges, if the temperature is low enough for the process to happen; salt bridges, in case of eruptions where seawater is involved; viscoelastic dissipation in case of liquid water (see for example Ennis et al., 1991).

Revealing the presence of liquid bonds in the sample is much more tricky than you can expect. It is relatively "easy" when the water with salts is involved, because then you can observe at the SEM the remaining salt bridges (as for example in some AP samples). It is almost impossible when water acts by means of its hydrogen bonds and by means of its viscoelastic dissipation, because after the moment of the collection the water can evaporate or the amount involved be relatively small if compared to the size of the aggregate.

I didn't fully get the meaning of your sentence in this context ("*the implementation of a collection kernel (differential sedimentation, Brownian or else) would increase the final aggregate porosity*").

What a particle collects in its path from the eruption down to its collection results in population of particles that you will catch in the final grain-size distribution. This regardless from the mechanisms of the collision (mostly due to collisions in the highly turbulent plume and sedimentation in the cloud. Brownian motion in a volcanic eruption is several order of magnitude less important than any other mechanism, see Costa et al. (2010)). Here the problem is: once you know the particles that different collision mechanisms led to interact with the central core, is there any other phase involved that can alter the assumption that particles are "touching" each other? In some cases no (see Gilbert and Lane, 1994).

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15. Line 440. Again, if you let the colliding particle aggregate among them before sticking to the larger one (implement an aggregation kernel) the history would be much different and also the final properties.

Reply:

The reconstruction of the aggregate proposed in this example is based on the assumption of a core that performs central oriented collisional process as discussed in previous comments. This assumption is motivated by field observation on the symmetry of aggregates (Gilbert and Lane, 1994; Brown et al., 2012; Bagheri et al., 2016) and by the theoretical description of what happens in a highly turbulent flow under the assumption of the validity of the Kolmogorov cascade eddies (Ormel and Cuzzi, 2007). So that sentence at line 440 is within this context.

However, it is true that the history would have been different in other scenarios. But then we have to understand how realistic is the alternative scenario.

Because in order to have particles of the same sizes to collide together and produce aggregation before encountering the large particle is not as simple as it may seem. Particles of nearly the same size will be show the same degree of coupling in a turbulent flow (statistically speaking). So the collision rate is not that high and this is in contrast with the assumption that they should first form aggregates and then later encounter a different population of sizes (the core). For sure it can be that in the Abrahamson mechanism, the larger Stokes number associated with the core will produce a preferential direction in the collision (if the particle rotation is neglected). But still I do not see clearly why the core should encounter aggregates of smaller sizes already formed, if the timescales of the processes are so different. This, of course, does not exclude alternative scenarios. But the one described in the example is supported both by observations and a theoretical explanations.

However, with SCARLET you can also simulate the scenario of a core interacting with already formed aggregates. But we think that is not worth investigating it because we lack of evidences from field observations and the scenarios to be investigated are countless and arbitrary.

<div style="text-align:center">***</div>

Reply to generic comments of reviewer 2

The paper writing can be improved. Section 2 reads vaguely as a user manual with some explanation of the details of the model. The code already comes with a User Guide and I find the explanations written in the paper on how to call the functions rather repetitive and distracting. The fact that the paper uses symbols and variable names that are different from those used in the code creates unnecessary confusion. For example at page 7, the variable Nr is also closet.NraysXSA and so on. My suggestion is to dedicate a separate section for the model description (which should be the focus of a GMD paper) where only mathematical symbols and clearly defined variables are used.

Reply: We agree with the reviewer that section 2 was too repetitive and not so clear in explaining the aggregation process that we want to describe. Moreover there was too much confusion in mixing the names used in the paper as a mathematical description and those used by the code and that are also explained in the user guide.

So, in order to follow the advice of the reviewer we completely reformulate sec.2, that we hope is now more simplified and clear. The variable names used within the code are no longer in the paper, but just in the user guide. We also added the new table 3 in order to facilitate the reader in not being confused with the different parameters used in the text.

We also agree that in the spirit of a GMD paper a separate model description should be present. We introduced it at the beginning of sec.2 where the physics behind the collision is reported.

However, we kept a more detailed explanation of the two main subroutines of SCARLET in the second part of sec.2 because we think they are important to help the reader understanding the philosophy of the algorithm and its peculiarity (and limits of course), which is also in the spirit of GMD.

<div align="center">***</div>

After, the authors can give some details of the implementation and provide code examples on how it is possible to interact with the software. The description of the methods can be generically improved. Sometimes, variables are used without being introduced before. For example "deq" is used at page 6 and called "characteristic size", than it is defined for ellipsoids only at page 10 and it seems to be a spherical equivalent diameter, but at page 7 (line 194) it is the maximum dimension and at page 8 the spherical-equivalent diameter is used to define the size of the aggregates and it is called Dagg.

Reply: Yes you are right. There was some confusion in the labelling of some variables within the text. We hope now they are sufficiently clear and coherent throughout the text. In addition we added a final table where all the variables are summarized, we hope this will help the reader.

As mentioned in the previous comment, we dedicated the second part of section 2 to describe more in detail the structure of the code and its implementation. So, basically, we introduced a new initial part in sec.2 to describe the model. And the second part is a rearrangement of the previous version where we cut the repetitions and unnecessary concepts.

<div align="center">***</div>

The units of the quantities are also not specified, I believe this is because the shapes can be scaled and do not require tobe defined into specific units (i.e. if the STL files is defined in meters the resulting Dagg will be meters, V will bem**3 and so on, same for density). I think it will be useful to make this explicit: the authors should say that theunits of the output will depend on the units used for the input. On the other hand, there is, at least, one quantity that must hold units and this is the solid angle omega which also appears with numbers in various places of the text and figures, but without units.

Reply: yes you are right, the reason why we didn't put units is because we deal with objects that can be scaled. For our convenience we considered all the sizes in micrometers, but in reality they have arbitrary units.

But we agree with you that this should have been explicitly stated in the text (we hope that in the new version of the paper it is now clear). For what concerns the solid angle, you are right too! And – my fault – I was not clear at all in defining what actually the code uses to calculate the solid angle (the solid angle is given by the angle respect to the unit vector $\hat{v}$ which represents the center direction of the cone). And it is also much clear for the user to keep all the angles between $0 \leq \Omega \leq 180$ (there is a periodicity, but it is not gain in using it). The solid

angle is then computed by the function "solidAngleRndPoints" (see previous comments). This was not clear in the text, my fault, and I also made it clear in the user guide.

$$***$$

I also have the feeling that the authors make implicit assumptions sometimes and this does not help understanding the argumentation, I will try to point where this occur in my detailed review. The model aims at being generically applicable to various collision phenomena in geophysics. However, at least for what concerns snowflake aggregation (my field of expertise), it fails to cite adequate literature and the conspicuous production of models that happened in the past 20 years. Sometimes, the cited literature is also inappropriate as It does not sustain the argumentation in the text.

Reply:

We agree with the reviewer that the previous version of the manuscript was too general and it lacked in explaining properly the field of application. We modified the abstract and the appendix to make it more clear. For what concerns the literature: this is not a paper focused on snowflake formation, which was mostly cited as an example for the interlocking where the shape of the monomers cannot be fruitufully approximated as spheres. The cited literature was mostly reported to provide a wide range of different approaches to the problem of aggregation and also different algorithms respect to those similar to SCARLET, such as cellular model of Ning . and Reiter (2007) for example.

However, according to previous comments we implemented the cited literature, but without giving to the paper a too much oriented attention to snowflake formation which is not the aim of the manuscript.

For what concern a better specification of the field of application of the code we modified the introduction as follows:

"In volcanology, despite the importance attributed to ash aggregation, no specific code has been designed so far for the study of particle packing geometries or, in general, the dependency of aggregate densities respect to their structural configuration. Several types of different aggregate geometries have been described in literature, from Particle Clusters (PC) to Accretionary Pellets (AP) (see Brown et al., (2012) for a complete review). . However, this is not always the case, as it has been proven for volcanic ash, where recent field observations In particular, recent field observations have revealed how PC3 objects have a key role in ash sedimentation (Bagheri et al., 2016; Gabellini et al., 2020). PC3 are have revealed that in many cases volcanic ash aggregates are roughly spherically symmetrical in shape and they are composed of a big particle of about 200-1000 m (named the core) at the center of the structure, with and a large variety of smaller sizes typically less than 100 m around it (the coating). (Bagheri et al., 2016). These two elements poorly fit with a typical monomer-like description at the base of fractal theory, in which the characteristic length of the aggregate, R, is related to the number of monomers involved, N, by means of the power law $N \propto R^{(D\_f)}$ (Jacobson, 2005).

The physical explanation of the formation of PC type aggregates involves the theoretical description of a polydisperse particle population within a highly turbulent flow, such as it is the case for a volcanic plume or cloud (Kieffer and Sturtevant, 1984). In presence of a cascade of turbulent eddies and grains of different sizes, particles will show different degrees of coupling

with the turbulent flow (Volk et al., 1980). This will produce a wide spectrum of relative velocities (Ormel and Cuzzi, 2007). In the limiting case of particles with a negligible velocity correlation with respect to the others (i.e. the kinetic theory limit), the object will aggregate following an inertial aggregation mechanism (Textor and Ernst, 2004). This is typical of large particles that will jump from one eddy to another with a poor correlation with the dynamics of the eddies. In their path across the turbulent flow these objects will encounter smaller particles that, on the contrary, do show a high correlations with the eddies. This process can be modelled as a central collisional process in which the big particle is the target of random collisions coming from random directions.

For those cases where the presence of an inner large particle is present, volcanic ash aggregates are better simulated with a central collisional process, in which a sequence of particles collides towards the inner core, considered as a pole of attraction.

Another important aspect concerning aggregation algorithms is the capability of accurately describe the shapes of the particles involved in the process. In some circumstances the morphology of the colliding objects is sufficiently well described in terms of equivalent spheres. In other cases, a more accurate description is needed, as for example in the study of particle packing or in all those cases where particle interlocking is the leading mechanism for aggregation, such as in snowflakes formation (Gravner and Griffeath., 2009). In these problems the user would benefit from an algorithm that offers the possibility of dealing with arbitrary shapes in the simplest way possible.

All these aspects motivated us to create SCARLET-1.0 (SpheriCal Approximation for viRtuaL aggrEgaTes), a MATLABatlab package designed for the study of volcanic ash aggregation virtual aggregates generated by central collisional processes of particles with arbitrary 3D shapes."

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I cannot speak about interstellar dust grains, but the model is not applicable to the problem of snow aggregation. Judging by the results presented in this paper it also seems to not fully capture the properties of volcanic ash aggregates. For these reasons I would suggest the authors to focus on the application they designed the model for and clearly demonstrate the suitability of the model assumptions for that specific problem. If this is not done, the code remains a nice piece of software, but with a limited applicability to geophysical problems and the authors might consider publishing it on a different journal.

The authors stress a lot about the generality of the model. However, I have the feeling that SCARLET might be too generic for having an application in geophysics.

Reply:
We agree with the reviewer that a better explanation of the hypothesis behind the model are needed. And also why we chose this specific design and the field of application.

The application of the algorithm is mostly related to volcanic eruption. But we believe it can be fruitful also in planetary sciences or whenever you have particles with different Stokes number in a turbulent flow. Fixing the direction of collision (a new possibility that we introduced one reading one of your previous comments) it can also be used to mimic the sedimentation of a particle, but in this case probably neglecting the secondary motion due to the drag that can later rearrange the particles in the wake of the main object is missing. Moreover, and quite

important, the code can be used in all those applications where the user needs to 3D print the object or work with the STL file of the aggregate (for example in CAD applications). This can potentially have uses in geophysical applications, such as the study of the terminal velocity of irregular shapes.

We propose to modify the first part of the model description as follows, accordingly to what has been suggested by the reviewer:

"*SCARLET-1.0 is written in MATLAB (tested for MATLAB R2015a) and it has been motivated by the need of a better understanding of the geometrical packing of volcanic ash aggregates observed during volcanic eruptions (Taddeucci et al., 2011; Brown et al., 2012; Bagheri et al., 2016; Gabellini et al., 2020). Iit simulates the random collisions encountered by a single grain (the core) in its path across an environment where N_p particles of arbitrary sizes and shapes are present. This happens in nature when particles with different dynamical properties are released in a turbulent flow. In this case the complex interaction of a cascade of turbulent eddies and the presence of particles with different sizes and masses produce a wide spectrum of particle Stokes numbers and thus a complete set of different couplings with the flow (Ormel and Cuzzi, 2007). The limiting cases for the situation under analysis are described in literature as the Saffman-Turner limit (Saffman and Turner, 1956), for particles fully coupled with the fluid and with a size smaller than the Kolmogorov scale, and the kinetic theory limit (Abrahamson, 1975; Textor and Ernst, 2004), in which large particles are poorly coupled with the eddies. The final result of the process is a population of particles with relative velocities that are higher as the decoupling with the flow is more pronounced (Volk et al., 1980). This is for example what happens in the case of coarse ash within a turbulent volcanic plume or cloud (Textor and Ernst, 2004; Bagheri et al., 2016; Gabellini et al., 2020), and also for dust grains in protoplanetary disks (Ormel et al., 2007; Okuzumi et al., 2009). The presence of a relative velocity between the core (or the aggregate) and the colliding particle creates a relative kinetic energy that must be dissipated in order to have a successful sticking. the aggregation process of N_p arbitrary objects, here named "the coating particles", around a central one, here referred to as "the core". In SCARLET we do not focus on the dissipation mechanisms, which require a full understanding and constraint on non-trivial quantities such as the presence of viscoelastic forces, water layers, electrostatic charges, plastic deformations, etc. We simply assume that after the collision the i-th particle will dissipate the relative kinetic energy available and it will stick to the central structure. This is an acceptable compromise since we are mostly interested in the final geometrical packing of volcanic ash aggregates, more than on the precise description of the dynamic of the collision (for which many key quantities are missing).*

In summary, within the limit of the kinetic theory the large core shows a negligible correlation with the velocity of the surrounding particles and it acts a central pole of accretion for the colliding particles in its path across the turbulent eddies. Considering that the core can rotate and that the turbulence is assumed isotropic, the angles of collisions respect to the central body are assumed to occur at random orientations.

In SCARLET we assume a Lagrangian perspective and we fix the System Of Reference (SOR) at the Center of Mass (CM) of the core. The core, of arbitrary shape, is the target for the collisions and the population of particles encountered as the aggregation process evolves is defined as a vector of particles with different sizes and arbitrary shapes. According to the physical process that we want to describe, collisions happen at random angles around the central structure and the collision cone is centered in the CM of the core. This is the main mode of operation of the code and the motivation that lead us to create it according to what has been recently observed in volcanic eruptions (Bagheri et., 2016; Gabellini et al., 2020). However it is worth anticipating

here that SCARLET-1.0 allows relaxing some of these constraints, if needed by the user. For example, collisions can occur following a fixed direction of collision and the collision cone can be centered in alternative locations respect to the CM of the core. This will be explained in the following sections.”

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 To me the aggregation process can be divided into 4 main events: a) the generation of monomers distributions, b) the selection of which particles have to collide, c) the collisiontrajectory and d) the contact dynamics.

a) Is not treated in SCARLET, while it is a fundamental aspect at least for snowflake aggregation [5, 6].

b) In SCARLET the order of aggregation is again arbitrary. The user decides the sequence of collisions and the design choices prevent from making an aggregate of previously aggregated particles (without considerable effort). Again this is a process that is essential for snow aggregation because it follows the process of collision through differential sedimentation [14]. Considering that volcanic ash fall in the Earth atmosphere I do not see why it should follow a different process, although it is true that differential sedimentation can only act after the initial inertia due to the volcanic explosion is dissipated and probably electric or chemical process also have a contribution in the dynamics. The authors should clarify which physical processes play a role and how SCARLET models them.

c) SCARLET makes a very precise assumption about the collision trajectory. Each colliding particle is attracted by the center of mass (CM) of an evolving core object. This is not the case for snowflakes (the authors cite [4]where this is explicit), I guess this might be true for planet formation due to gravitational attraction, but again I donot see an immediate justification for volcanic ashes (perhaps electrostatic forces?).

d) SCARLET explores various orientations of the colliding particle and select the one that minimizes the distance between the CMs of core and colliding particle. Again this is not seen in snowflake formation where the crystal attitude is generally determined by aerodynamic forces, and I do not see a justification for volcanic ashes. It might be true for planet formation, but I suppose that the most important effect of collision would be the break-up of the colliding objects. Considering my list of aggregation events. SCARLET implements only c) and d). This of course increases the applicability of SCARLET to different problems, but reduces the scope of the model from aggregation of particles to simple collision-collection.

Reply:
Aggregation of volcanic ash is quite different from snowflakes, we agree. The grain-size distribution is usually constrained just at the end of the process and measured on collected samples as a bulk quantity. So the order of the collision processes is completely lost. However in SCARLET you can perform random or sequential collisions, with an easy manipulations of the vectors.

For what concerns the aggregation of already aggregated structure. Here we must be clear: the fact that we only simulated the interaction of single particles does not mean that in general the code cannot deal with collision of aggregates. It just requires a bit of iterative calls of the code. The reason why we performed this scenario is because we want to simulate what

happens when a big core encounters a population of particles. Something that really happens in a turbulent flow where smaller particles have a low relative velocity that brings few collisions (i.e. the probability of forming aggregates for particles coupled within the same eddy is relatively low (Ormel and Cuzzi, 2007)). We also believe that is not only the case for a volcanic eruption but also in other contexts such as in planetary formation (see the above reference), or in any other turbulent flow where there are different degrees of coupling between particles in presence of a wide range of particle Stokes numbers.

The use of the CM is here used to mimic a central collisional process as the one above described (and in previous comments). How much the minimization will influence the final result is a user's choice. We do not actually think that this a weak point. Instead we think it is a point of strength because the user can investigate different degrees of packing and provide maximum and minimum bound of a process for which, in any case, a quantitative information is missing. The idea is that the relative velocity produced by all the mechanisms explained in previous comments leads the impact of the two objects. However, this relative velocity can span different values, due to the complexity of the coupling of particles and fluid. Therefore the dissipation of the relative kinetic energy can be "fast" (so they stick just touching) or let the particle penetrate much deeper in the structure. This is mimic with having the possibility of performing simulations in which different degrees of packing can be tested. Also in other models from different scientific fields, such as the interesting one that you highlighted in a previous comment (Fig.1 in Ori et al. 2014), we see that for example complex mechanisms of collisions between irregular shapes can be modelled as a contact of randomly oriented bodies in which a punctual description of the forces involved is summarized by a sampling of possible orientations. In addition, the code provides the possibility of relaxing the condition of having the collision cone coincident with the CM of the core. This is not the case where we are interested, but we think that it is up to the user to decide which option to choose. For what we observed with the HSC during a volcanic eruption (Bagheri et al., 2016; Gabellini et al., 2020), the almost symmetrical shape of ash aggregates around the inner core is really well represented by a center oriented collision.

However, we agree with the reviewer that the model description part would actually benefit of a section with a list of points as suggested by the reviewer in his comment.

However, due to specific characteristic of SCARLET of working with STL files and sphere-composite representations, we will add two more bullet points as indicate in the following.

"Here the main steps in which the aggregation process is modelled in SCARLET-1.0 are summarized:

1. *Definition of the shapes involved in the aggregation process: every different shape used in the simulation must be already present in the folder as an STL file. STL files can be generated by means of specific CAD softwares, they can be downloaded from the internet or simply obtained by 3D scanners. The use of binary STL files is preferred respect to the ASCII format.*

2. *The sphere-composite representation of each STL file: SCARLET-1.0 uses a dedicated script named "fromStlToSpheres" to construct the sphere composite representation of the original shape (see section 2.1). STL files are defined by a set of points and facets. The script takes advantage of the MATLAB function "inpolyhedron" to generate random points within the surface. Once that the point created is within the STL shape and outside an already existing sphere, it generates a new sphere (see sec 2.2). Each single shape that the user wants to involve in the simulation must be pre-processed by "fromStlToSpheres".*

3. _Generation of the monomers distribution_: the grain-size distribution of the coating is defined a priori by the user who defines a vector of sizes with a length equal to the number of particles involved. Each element of the vector indicates the maximum length of the i-th particle (in arbitrary units). The shape attributed to the i-th particle is randomly selected within those available for the coating (see sec.2.1). The core size is defined by the user and it corresponds to the maximum its maximum length (in arbitrary units). The shape attributed to the core is by definition the first element of the structure generated by the pre-processing function "fromStlToSpheres".

4. _Selection of which particles collide_: particles are selected sequentially from the previously defined vector of sizes. Therefore the contemporary deposition of two particles is forbidden (i.e. only binary collisions are treated).

5. _Collision trajectory_: in order to place the i-th particle, a cone is generated in the CM of the core (or, alternatively, in one of the spheres that form the sphere-composite representation of the aggregate). A cone with random orientation and aperture Ω is centered in CM. Within the code N_r random rays are uniformly generated and along each ray the shape associated with the i-th particle – scaled to its actual size – is randomly rotated N_o times following the ZXZ convention on the Euler's angles. Each rotated object, along each ray, is by default placed outside the aggregate. Then a coarse inward movement along the ray is performed until the overlapping of the sphere-composite representations of the aggregate and the i-th particle happens. At this stage a finer outward movement along the ray is done until there is overlapping between the spheres. The two steps process guarantees faster movements when the objects are far away and a fine tuning when they overlap.

6. _Contact dynamics_: the contact dynamic assumes an instantaneous sticking when the intersection of the spheres is no longer present (see step 5). This means to assume that the relative kinetic energy is completely dissipated at the moment of the impact, with no rearrangement of the already existing structure. For each i-th particle a total of $N\_r \times N\_o$ configurations are tested but we only take the one that minimizes the distances between the CM of the aggregate and the particle. This is equivalent to assume that among the $N\_r \times N\_o$ configurations the inward force and the torque oriented the particle in such a way to minimize this distance."

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What the paper lacks the most is a clear justification of the design choices made for the model. Since I am not an expert of volcanic ash aggregation I might lack some knowledge that is trivial to volcanologists. Because of that, please correct me if I am wrong.

Reply: We agree with the reviewer that a clear justification of the design was lacking in the previous version of the paper. And no problems at all for some lack of knowledge about some details about aggregation of particles in volcanology. Actually, it helped a lot to rewrite better some parts without taking from granted some aspects that for us were "natural", but that were not to people from a different scientific community. We totally followed your advice to be more clear in the justifications behind the design of the code and we hope that now they are clear in the new "model description" part (see previous comments)

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I suggest the authors to do a major revision of the study and include a model description in their revised manuscript. I honestly do not understand what is the rationale behind the choice of making the aggregation trajectories converging towards the CM of the core particle.
What is the dynamical model of ash aggregation? Why should the CM of the aggregating and the core particle be minimized? Failing of explaining this breaks the link between the model and the physics. The model will indeed produce aggregated particles, but not particles that are resulting from a physical process. If the physics behind ash aggregation is unknown it will be only logical to make simplifications and assumptions, but again this must be clearly stated in the description of the model.

Reply: The answer to this observation has been largely discussed in some of the previous comments. We hope to have accomplished the major revisions that the reviewer suggested. Most of the observations inspired by the reviewer have been carefully analyzed and we think that the manuscript really improved a lot thanks to his help.

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While reviewing this paper I tried to gain some bibliographic information about volcanic ash aggregation. This recent paper [1] lists three processes that are acting in the determination of an aggregation kernel, namely Browninan motion, fluid shear and differential deposition. My experience in snowflake aggregation makes me believe that the discrepancies found in the paper between observed and modeled aggregates are likely to be a result of the lac kof a physical model for ash aggregation. This is simply because if one allows the larger object to collect already aggregated particles the packing efficiency of the end process will be much lower. I suggest the authors to take this into account because: 1) The existence of an aggregation kernel is not incompatible with the evidence of having a large object collecting smaller ones; 2) Leveraging on the knowledge taken from snowflake aggregation, it is very likely that the lack of porosity of the modeled aggregates is due to an incorrect aggregation dynamics; 3) The inclusion of an aggregation kernel binds the model to the physics.

Reply: We largely discussed in the previous comments all these aspects. We only want to add here one additional comment.
Brownian motion in a volcanic plume or cloud should be considered negligible (see Costa et al., 2010). Brownian collisions mostly happen in a still fluid conditions as consequence of the termal motion of atoms and, as a secondary consequence, between particles. However this is not the case in volcanic plumes, where the Reynolds number can reach the tremendous number of 10^{11} (Kieffer and Sturtevant, 1984). On the other hand, differential deposition can actually happen, especially in the free atmosphere below a volcani cloud or in the volcanic cloud itself where the turbulence is reduces. This process can be partially simulated setting a fixed direction of provence for the collision cones (but the wake accumulation effect due to the drag is of course not captured). However, if the particle is much larger than the particles it will encountered probably the final scenario will not that different from a realistic one (if we

neglect small particles that avoid the collisions following the streamlines of the flow. But, still, aggregation in a settling process is much more important for rain droplets (coagulation) or snowflakes. Much less in a volcanic eruption, where the leading collision mechanism happen due to turbulence. Fluid shear is a special case of collisions in a turbulent flow. Unfortunately, in most of the cases, the collision kernels used in volcanology derive from other fields, where the degree of turbulence is much smaller than in a volcanic eruption. Concepts like fluid shear or the Saffman-Turner regime just give a partial representation of the complexity of what can happen in a highly turbulent environment. A good paper about it is Textor and Ernst (2004) which provides an overall view of what should be considered as a collision mechanism in a volcanic plume. What the theory suggests, and it is actually quite reasonable, is that the central collisional process that produce the roughly spherical aggregates with an inner core observed in the field, are the result of the inertial aggregation mechanism. Which appears when poorly coupled large grains meet a population of highly coupled smaller particle in a "Kolmogorov-like" turbulent eddy cascade.

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Reply to "Implementation details" of Rev.2

From what I can understand from the code there are two passages that seems to not fit the what it is stated in the manuscript. The Euler matrix used at line 209 should explicit the convention used. There are many of them, from what I see from the code implementation it seems the authors used the ZXZ convention, please clarify this.

Reply: Yes we use the ZXZ convention (also named the "x-convention"). Thanks for noticing this because we agree with you that this part of the paper was too vague.
We propose to add the following lines:
"[…] *according to the Euler rotation matrix (we adopt the Z-X-Z convention). In this convention the i-th particle is first rotated along the $\hat{z}$ axis of an angle φ ($\varphi \in [0,360°]$), then along the $\hat{x}$ axis of an angle ϑ ($\vartheta \in [0,180°]$) and finally around the new $\hat{z}$ axis of an angle ψ ($\psi \in [0,360°]$)."*

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The Euler angles at line 209 are picked from the wrong domain. At line 284-287 of the code SCARLETv1.m the phi, theta and psi angles are sampled uniformly from the (0, 180), (0, 180) and (0, 360) domains respectively. Actually, for random orientations in space the phi angle should be sampled in the (0, 360) domain and the theta angle should be sampled uniformly in the (0, 1) domain of cos(theta). The current implementation generates orientations that are more dense around the "poles". This is not a big problem for the results since the angles are only useful for the Monte Carlo minimization. Sampling a non-uniform distribution means that on average more orientations have to be sampled in order to reach the same convergence. However, the Monte Carlo minimization could benefit from a performance boost, and the effects (if any) should be visible in Fig. 9. In general it is better to not make implementation mistakes. Also the versor v (line 201) components are derived from a normal distribution. This is something different from having the versor in a random direction on the unit sphere which is achieved with the procedure detailed before. In any case this is different from what is written in the paper at line 201. Both what is implemented and what is written

in the paper does not provide uniformly distributed colliding trajectories. This might have an impact on the final aggregate properties because if the collision trajectories are not uniformly distributed the resulting aggregate is more likely to have "blobs" of colliding particles.

Reply:

We really thank the reviewer for having spotted the little bug on the phi angle domain. Sorry for this mistake. Having used highly symmetrical objects, such as spheres and ellipsoids, should have prevented large consequences on the missing angle in phi. We correct this number. For what concerns the not uniform random orientation of the Euler angles we agree with you that is not totally correct, since we have an oversampling at the z-poles. However, as you also pointed out, the Monte Carlo minimization process should not be affected by this, since we only take the closest oriented shape from the ones generated at this stage. It is true that the efficiency could be slightly reduced by this oversampling at the poles, but since the parallelization is done at this stage, we do not think it is a major drawback and we would prefer to keep this part of the code as it is.
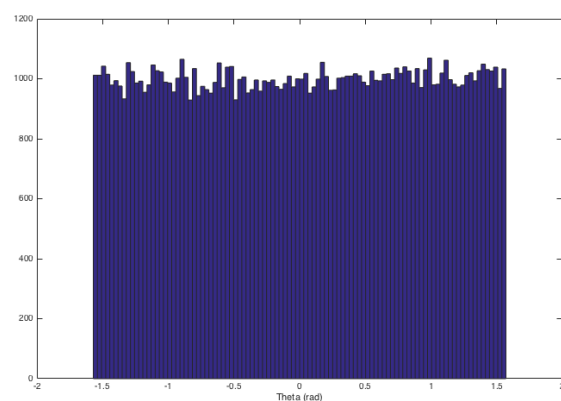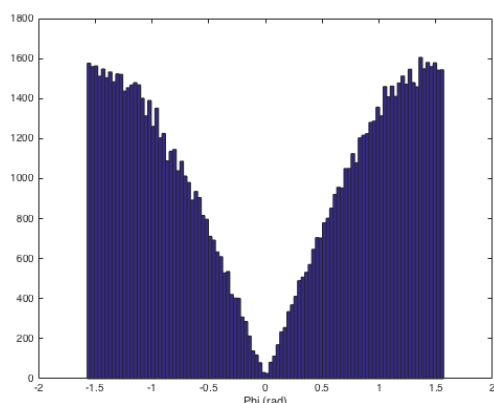
On the other hand, we believe that it is more important discussing here the problem of the uniformity of the random directions of provenance of the colliding particles. We would like to thank the reviewer for having focused our attention on this aspect and the *potential bug* associated. At the end, as we will show you, there was not a mistake or a bug on this, but it has been important to verify this step. Because it could have created preferred directions of collisions instead of a random ones.
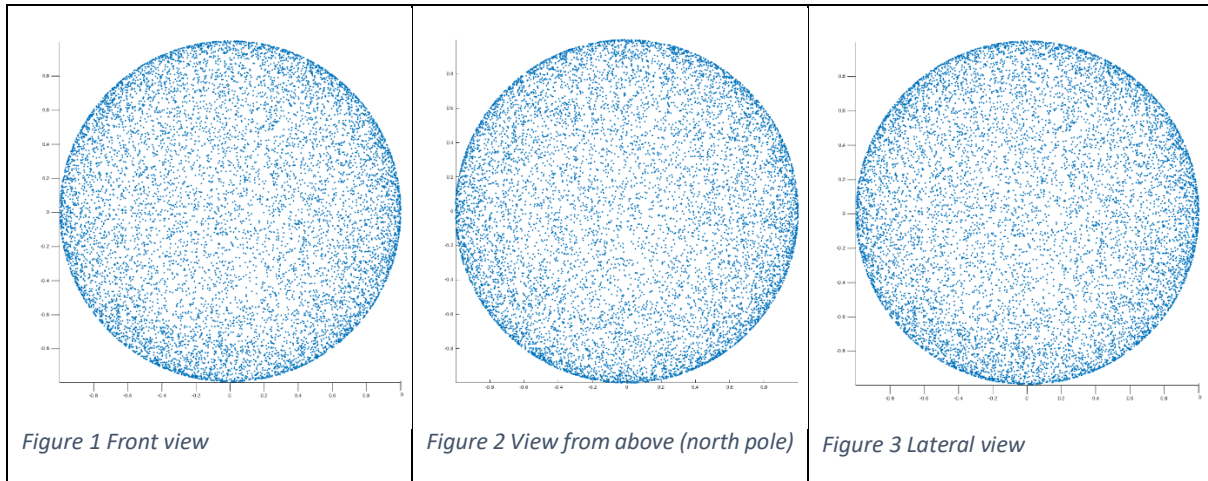
The problem of uniformly distributed points on the surface of a sphere is a well-defined problem with different discussions on the web and algorithms available. This aspect concerns SCARLET in selecting a random orientation for the cone and in generating random directions within the cone. There are two specific functions that perform these tasks, namely "generateUnitVector" and "solidAngleRndPoints" (the second one performs also additional operations and it has a different implementation respect to the first one).

Investigating these two functions we actually found out that both the implementations are correct and they provide a uniform concentration of points on the sphere, i.e. uniformly distributed collision trajectories.

For what concerns "generateUnitVector". It generates random points equally distributed on the surface of a sphere, but both provide the right result.
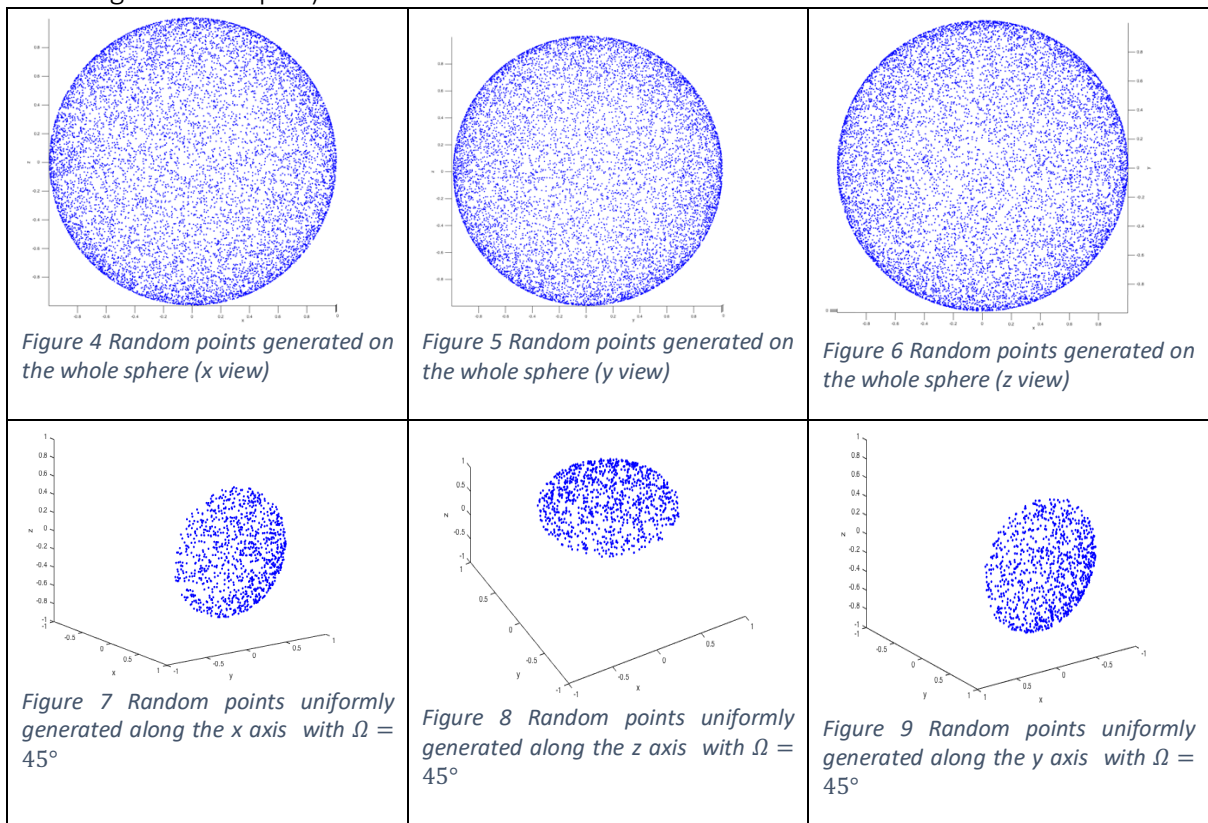
The fact that a Gaussian distribution is involved is actually <u>necessary</u> for the specific algorithm to work. In fact, this is the key aspect of the papers of Muller 1959 and Marsaglia 1972 (see https://mathworld.wolfram.com/SpherePointPicking.html).

| Figure 1 Front view | Figure 2 View from above (north pole) | Figure 3 Lateral view |

From the histogram is clear that in order for the solid angle to contain the same number of random points independently of the orientation, the distribution along the phi angle needs to show much more points along the equator respect the pole due to the greater number of same solid angles, with the same phi, but a different theta. And the uniform distribution on theta is what we expect due to the symmetry. The final proof is the complete invariance for rotations of the number of points present on the sphere. This is clearly shown in Fig.1,2,3: no more dense distribution around the poles. So we think that the specific part of the code that has been criticized is actually correct and no blobs are expected (i.e. no preferential directions for the collisions)

We also tested the uniformity of the points generated by "solidAngleRndPoints" (here in the following two examples)

| Figure 4 Random points generated on the whole sphere (x view) | Figure 5 Random points generated on the whole sphere (y view) | Figure 6 Random points generated on the whole sphere (z view) |

| Figure 7 Random points uniformly generated along the x axis with $\Omega = 45°$ | Figure 8 Random points uniformly generated along the z axis with $\Omega = 45°$ | Figure 9 Random points uniformly generated along the y axis with $\Omega = 45°$ |

***

What I might suggest is to consider the reciprocity relation: having random colliding trajectories is equivalent of having always the same trajectory (let's say the vertical z axis [0, 0, 1]) and the central particle rotated randomly. This simplifies the implementation quite a bit because we already have a function that rotates the particles (with the corrections suggested before) and this avoids unnecessary coding. Also sampling the rays in a cone always centered around the z axis is simpler in my opinion.

Reply: on the base of the previous considerations we would prefer to keep SCARLET with its own subroutines and its "algorithmic philosophy". This because we think that for the volcanological community – to whom it is mainly oriented – it can be already enormously fruitful with this structure and functions. Probably, in the future, better implementation and efficiency-oriented versions of SCARLET will be released. But at this stage we think that the coherence of the package and the methodology used is already of large interest for practical application in our community. However, we would sincerely thank the reviewer for the advices provided at this stage of the review. Because his experience helped us a lot in improving and testing the algorithm. Really thanks.

***

Reply to minor points

1. Line 13 (and others). The benefit of 3D printing is not clear. Perhaps this resource demonstrate a scientific application [8]

Reply: 3D printing of virtual aggregates is one of the most interesting benefits of the algorithm, considering that it allows proving theories related to packing problems in lab experiments. For example the drag exerted on air on an aggregate for which you know exactly all the caracteristics. We thank the reviewer for this reference, that we added in the introduction (not in the abstract)

***

2. Line 14. The concept of a vector of sizes and shapes is not clear at this stage. I would rather say "a list ofcolliding shape"

Reply: Yes you are right, we modified as you suggested.

***

3. Line 14 (and many other places). A spherical equivalent representation of a irregular shape is not necessary foran analytic solution of the collision problem. The solution for a generic polyhedron (such as those representedin STL shapes) is formulated analytically by considering all vertexes and faces instead of simply

center andradius of the spheres. This is implemented in [10, 11] for the aggregation of snowflakes.

Reply: For sure many other analytical methods to compute the intersection of generic polyhedrons exist. Nonetheless, also the sphere-composite representation allows doing it, and in quite efficient and general way. In the text we do not mean that the sphere-composite method is the only one that allows for analytical solution of the problem. We think we have been quite clear on this.

***

4. Line 16 (and many others). I personally do not like thinking about functions and parameters as entities thatcan take responsibilities (at least not in a scientific paper). Instead of saying that those objects are "in charge"of something I think it is more clear to just explicitly say what is their meaning or what happens when changed,invoked, called ...

Reply: We agree with you. We reformulated the sentences where this occurred. Thanks for highlighting this.

***

5. Lines 16, 102, 146, 250, 360, 400, 402, 702, and in connection my generic comments. The angle omega is calledsolid angle, however in the code it is called "aperture angle". The values of omega does not have units but therange of values are compatible with linear angles in degrees. This means that omega is not a solid angle, butthe aperture angle. Please clarify and correct.

Reply: My fault, because the text contained some old version that worked with a different user-defined control on this angle. We clarified this in the text and in Fig.6.

***

6. Line 19. The fact that SCARLET uses STL shapes is not unique (similar approaches in [10, 11, 7]) and doesnot extend the range of applications. It only means that it can deal with particles of arbitrary shapes. The implemented physics is the main limitation for its applicability.

Reply: In the text we do not mean that SCARLET is the only code doing this. But what we state is the versatility of the process that you get in using STL files (and probably this class of softwares will have an increasing role in the community thanks to the direct connection with the world of 3D printing). Therefore, we don't agree on fact that it does not extend the range of applications. One thing are the limits of the implemented physics – that can be always improved in further version of the code where you can add more and more processes. Another thing is the

easy-way how you can deal with arbitrary shapes. There is a countless amount of ways in which you can work with arbitray shapes. But dealing with STL format is probably one of the most versatile ones: you can create mathematical shapes using CAD; you can get real complex shapes using 3D scanning; etc. And you can translate this in the real world by means of 3D printing. In my opinion the range of applicatins is really wide and promising.

***

7. Line 27 The term "minimization" is rather generic and not clear at this stage. I think the authors should referto the forced compaction of the aggregating particles.

Reply: we agree with you. At this stage of the paper is confusing the term "minimization". We replaced it with "compaction".

***

8. Line 39 I think that [5] might be a perfect example of how virtual reality can compensate the lack of directobservations.

Reply: Thanks for this suggestion. We added it to the paper.

***

9. Line 51-53. The arguments presented is only valid for monodispersed distributions of monomer sizes, but thetheory of fractal aggregation is not limited to those cases [14, 12]. Moreover, fractal aggregates are in generalthe result of the modeling of physical processes and not a model by itself (although there are cases wherefractals are used as models of particles). The fractal properties of aggregates emerge in general when the sizeof an aggregate is much larger than the size of any of its constituents.

Reply: yes we modified the whole sentence in the introduction

***

10. Line 75. The term opensource does not oppose under-license. The vast majority of opensource software arereleased under license (including SCARLET).

Reply: At the end we removed the sentence because was not fitting well in the main body of the test.

***

11. Line 86 I think that the MATLAB software and programming language is written capitalized. The companyreleases the IDE two times a year and the releases are

named for example as R2015a, while the version numberfollows a different scheme. It seems that R2015 implements either 8.5 or 8.6 version of the language. Also,it is not clear if 2015 means that the package has been only tested with the release R2015 (a or b?) or if itcompatible only for R2015 and newer versions.

Reply: For the capital letters of MATLAB you are right, we corrected this. For the rest we modified the text saying that we tested it in the 2015a release

***

12. Line 80. I do not see the scientific value of including aggregates of t-rex and LEGO characters. However Ifind it not much distracting, so they cause no harm (and I personally find it funny). However I am not sure if LEGO must be written making explicit that it is a trademark and if the company is ok with their name beingused in this publication.

Reply: About the name LEGO we agree with you. It is better to not show it. About the rest…We introduced these "funny shapes" to veicolate the following two basic messages: 1. "you can really use the code with arbitrary and unexpected shapes" and 2. "have fun with it!". In the second sentence we condensate our hope to make this code (but also others analogue that deal with aggregation problems) more accessible also to a non-expert community. We think is not a negative point, but a strong and positive one. Also because some codes can also have "a second (and unexpected) life" in different fields respect to the one for which they have been conceived.
But it is true that is somehow weird to see something like that in a scientific context (but that's why we only put it in the appendix).

***

13. Line 98. "User-defined number" should have a hyphen. At line 102 the hyphen is used correctly. Conversely"User Guide" at Line 110 should not be hyphenated. It might worth checking.4

Reply: Thanks! We corrected this typo

***

14. Line 112. I think GMD require a fixed DOI for the code. So the zenodo link should always refer to the fixedversion 1.0 of the code and not being updated.

Reply: Yes, this is true (that's why we added the zenodo link). However we would also like to highlight to the interested readers that the new versions of the code will be available on the GitHub page. I will see with the editor if it is possible to keep both in the text, or at least to say something at the end of the paper.

***

15. Line 155. Cancel "as a matter of fact"

Reply: We reformulated all the sentence.

<div align="center">***</div>

16. Line 156 The subject here is the triangles which are uniquely defined by the coordinates of their vertices. Thefaces are themselve the triangles. The whole first paragraph (line 154-159) is superfluous. You only need tosay that STL represents any surface as a set of triangular facets in the 3D space.

Reply: I need to introduce Nv for later in the text. I simplified a bit the text, but I kept some of the all sentences.

<div align="center">***</div>

17. Line 160. I believe that theinpolyhedronfunction is not built-in but rather a contribution [3] which has beenported in the code

Reply: inpolyhedron born as a contribution but it is now a built-in function of MATLAB, at least in the R2015a.

<div align="center">***</div>

18. Line 160-164 This paragraph does not read very algorithmically. We first place one point, than we checkagainst other points which we did not place yet. We check also if points are inside spheres that are not definedanywhere. Since this looks like an iterative process you can clearly explain it iteratively: Say how to place thefirst sphere, than say how given N spheres you can find the sphere N+1.

Reply: We simplified a lot sec. 2.2 according to another comment you made in another part of the reviews.

<div align="center">***</div>

19. Line 167. I think it is better to explain from the beginning what is the effect of using Oext=1, Otherwise, onemight think that this takes out some mass. Also, from what I understand of the documentation the functionboundarythis is a convex hull finding algorithm. I think it is clear and simple if you call it convex hull. Theidea of "discarding internal spheres" or "those fully covered" is not straightforward.

Reply: we removed this piece because it was too misleading here. We just left it in the user guide.

<div align="center">***</div>

20. Line 170. I think the word trial has very specific meanings in english (in law, medicine and so on). A betterword might be tries, or attempts.

Reply: we reformulated this sentence

***

21. Line 197. I though that at this stage the shapes are represented with their sphere-analogues. Why is themaximum dimension of the STL cited here? How it relates to the maximum dimension of the sphereicalrepresentation?

Reply: We were not clear in saying that we refer to the core at this stage. Its shape is indeed represented by its sphere-analogue, but it still need to be properly scaled before starting the loops.

***

22. Line 199. Might be worth making explicit what is the nesting relation among the loops. The fact that Loop 2is nested inside loop 1 becomes clear while reading, but I do not see the point of having Loop 3 nested in loop2. Loop 3 could be one of the routines performed inside loop 1 and the same set of rotated colliding particlescan be reused for each ray. Potentially it makes possible to parallelize loop 2 and loop 3 independently andincrease performances.

Reply: Yes and actually this is something that we are interested to add in a future release of the code. At this stage we designed the loops in this ways because we want to investigate all the collisions as completely independent. The reuse of rotated shapes is an interesting alternative which speeds up the code (since it allows a sequtential double parallelization) but reduced the independency if the collisions. In future release we will probably add also this configuration to the package.

***

23. Line 201 Components cannot be randomly oriented, they are scaler values. Look also at my comments on the implementation.

Reply: Yes, what is randomly oriented is the unit vector (not its single components). For what concerns the uniformity of the random points refer to my comment in the "implementation" section.

***

24. I assume, but it is not written anywhere, that the colliding particles are placed with their CM on the rays. Am I correct?

Reply: yes you are correct

25. Line 235 Can you please provide a better reference for the algorithm that computes volumen from STL? Likean URL?

Reply: We agree with your comment, the actual URL to the reference was missing. We added in the reference list.

26. Line 243 Eq.3 Dagg does not provide additional information with respect to Vext. Would it be better tocalculate the final Dmax?

Reply: We prefer to keep it as it is in the present release, which is more oriented towards the reconstruction of PC type aggregate (almost spherical in their global shape). But for sure in a future release, different characteristic sizes for the aggregate will be introduced (especially when alternative ways of measuring the porosity will be added).

27. Line 313 I think that ash concentration would be overestimated by models if aggregation rates are underesti-mated not generically an uncorrect parameterization.

Reply: Yes, you are right

28. Line 325 How can the high-speed camera derive the density of the object before they are captured? This is an ever-green problem in snow microphysics.

Reply: First of all you have to verify that the object moves at its terminal velocity (if it is not, things gets more complicated, even if still possible). Then you have to constrain the size and shape of the object (but we are luckier than in your field because all these aggregates are not that far from being a sphere). Then knowing the size, the projected area, the terminal velocity, you can invert for the density. Due to the errors that affect some of the key features of the aggregate, such as the missing third dimension on the HS video, it is always better to calculate the density-distribution using a Monte Carlo method.

29. Figure 9 The other of the parameters changes from the figure to the caption. And I believe that also thenumber of monomers should be a parameter here.

Reply: Yes, thanks for highlighting this. The correct order was the one in the plot. We prefer to simply add the range of number of monomers used in the all test

because here we are more interested in gamma and tau, more than the specific components of each point.

***

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