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Key Points:
- Antarctic cosmic spherules with cumulate textures are reported
- Numerical simulation shows that settling of relict olivine is possible at entry velocities of 16 km s^{-1} relating to dust eccentricities of 0.35
- The relative abundance of cumulate olivine spherules provides a proxy for variations in the orbital properties of dust over time

Supporting Information:
- Supporting information S1

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Olivine settling in cosmic spherules during atmospheric deceleration: An indicator of the orbital eccentricity of interplanetary dust

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Abstract A new type of cosmic spherule is reported with textures suggesting settling of olivine during atmospheric deceleration. Numerical simulations of entry heating reveal that relict forsterite, which survives melting, can settle over the 1–2 s of flight at high entry angles and entry velocities up to 16 km s^{-1}. Enhanced crystallization of phenocrysts by heterogeneous nucleation on accumulated relict forsterites is the most likely origin of the observed cumulate textures in cosmic spherules. Such textures in cosmic spherules reveal interplanetary dust with higher encounter velocity with the Earth that correspond to orbital eccentricities >0.3. The relative abundance of cumulate spherules suggests that 14% of ordinary chondrite-related, S(IV)-type asteroid dust over the last 800 kyr had relatively high orbital eccentricity owing to secular and planetary perturbations. The textures of cosmic spherules collected from sediments can therefore be used to trace dust orbital variations with time, which may influence terrestrial climate.

1. Introduction

Micrometeorites (MMs) are extraterrestrial dust particles <2 mm in size that are recovered from the Earth's surface [*Genge et al.*, 2008] and have been collected from Antarctic blue ice and snow, Antarctic traps, and aeolian deposits [*Maurette et al.*, 1991; *Taylor et al.*, 2000; *Duprat et al.*, 2007; *Rochette et al.*, 2008]. The flux of MMs captured by Earth is currently 40,000 t a^{-1} [*Love and Brownlee*, 1993]; however, changes over geological time occur due to secular and planetary perturbation of dust orbits [*Kortenkamp and Dermott*, 2001] and major dust production events in the solar system [*Alwmark et al.*, 2012]. Changes in the flux of extraterrestrial dust have been detected in the ^{3}He content of ocean sediments and have been suggested to affect terrestrial climate [*Farley and Patterson*, 1995]. No reliable method currently exists to constrain the orbital properties of MMs by which the origin of flux variations could be constrained.

The majority of MMs are thought to represent samples of primitive asteroids similar to the parent bodies of the carbonaceous and ordinary chondrite meteorites [*Kurat et al.*, 1994; *Genge et al.*, 1997; *Genge et al.*, 2008; *Genge*, 2008; *Cordier et al.*, 2011], although some particles are also thought to be derived from comets but their abundance is uncertain [*Noguchi et al.*, 2015]. Most MMs experience significant heating during atmospheric entry to form partially melted scoriaceous particles and extensively melted cosmic spherules [*Kurat et al.*, 1994; *Genge et al.*, 2008]. Entry heating effects are strongly dependent on atmospheric entry parameters, such as entry velocity and angle [*Love and Brownlee*, 1991]. Mineralogical and textural features arising during atmospheric entry could, therefore, be used to identify the dynamic behavior of dust particles.

In this paper we report the discovery of a new subtype of silicate-dominated (S-type) cosmic spherule that exhibits systematic variation in olivine crystal size that is compatible with settling during atmospheric flight. We present numerical simulations of olivine settling during entry heating to demonstrate that such particles had relatively high entry velocities. Cumulate olivine spherules, therefore, provide a unique means of identifying dust particles with discrete entry parameters.

2. Cumulate Olivine Cosmic Spherules

Cumulative olivine (CumPo) cosmic spherules are a type of silicate-dominated porphyritic spherule in which olivine phenocryst size increases from one side of the spherule to the other (Figures 1d–1f). Nineteen CumPo

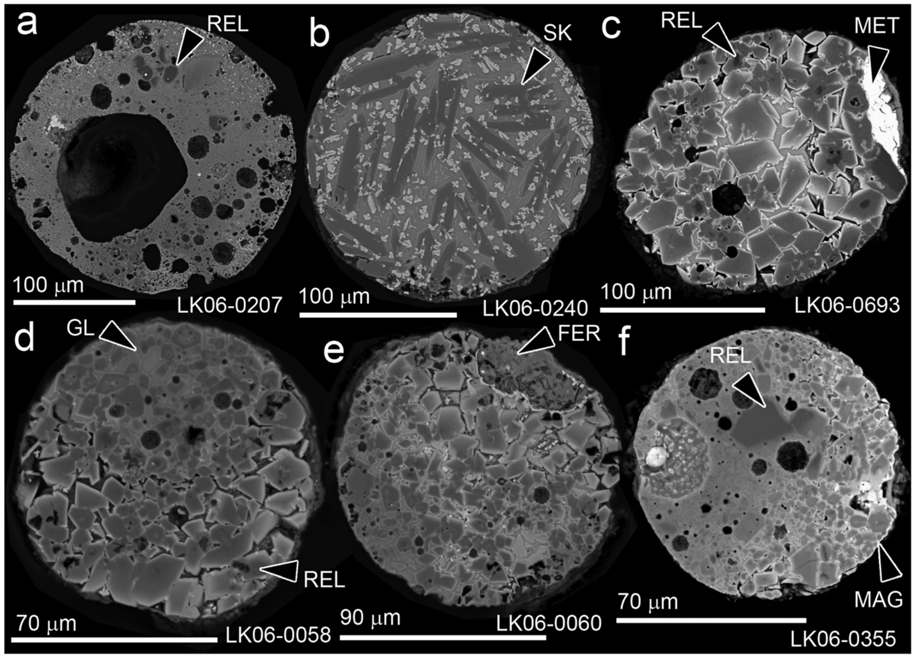

Figure 1. Backscattered electron images of porphyritic cosmic spherules. Showing the features of (a–c) noncumulate varieties and (d–f) cumulate porphyritic spherules. Examples are shown of the most abundant porphyritic, noncumulate spherules with (Figure 1a) micro-PO spherules containing relict forsterite (REL), (Figure 1b) skeletal-PO spherule with re-entrant skeletal olivine phenocrysts (SK), and (Figure 1c) coarse-PO spherule with relict forsterite (REL) and FeNi metal (MET). Figures 1d–1f show the cumulate porphyritic spherules showing variations in olivine phenocryst grain size to one side of the particle showing (Figure 1d) relict forsterite (REL) within phenocrysts set in glass (GL), (Figure 1e) an increase in olivine phenocryst size toward an FeNi metal bead altered to ferrihydrite (FER), and (Figure 1f) an increase in olivine phenocryst toward the magnetite rim (MAG).

spherules were recognized among 346 porphyritic cosmic spherules from moraine at Larkman Nunatak in Antarctic (supporting information S1) and apart from their systematic variations in crystal size are identical to coarse-porphyritic spherules (Figure 1c; 132 particles). Cumulative olivine spherules are distinct from (1) microporphyritic spherules (Figure 1a; 71 particles), which have crystal sizes less than 5% of particle size; (2) skeletal-porphyritic spherules (Figure 1b; 102 particles), which have re-entrant, usually coarse, olivine crystal shapes; and (3) dendritic-porphyritic (19 particles), which often have relict iron-rich olivines within a mesostasis-containing olivine dendrites.

Coarse-porphyritic and CumPo spherules are all dominated by equant olivine phenocrysts, which usually exhibit normal zoning with Mg-rich cores (Fa_{13}) and Fe-bearing rims (Fa_{35}), within a glassy mesostasis that often contains magnetite. Relict forsterite (Fa_1–Fa_7) is also commonly observed within these spherules forming the cores of olivine phenocrysts. Metal beads, or magnetite rims, which probably indicate the previous presence of metal in-flight, are also present within 35 particles (Figures 1c, 1e, and 1f).

Cumulate olivine particles show an increase in olivine crystal size toward one side of the particle with maximum dimensions increasing by a factor of 2–5 up to 25 μm (Figures 1d–1f). In eight particles olivine crystal size increases toward a metal bead or the ferrihydrite alteration products of a metal bead. Forsterite relicts within these particles are most abundant within the cores of the larger olivine phenocrysts close to the metal bead. The spherules range in diameter from 91 to 211 μm with an average of 127 μm.

The locations of metal beads in cosmic spherules indicate the flight direction of the particle during atmospheric entry due to migration of metal during deceleration to the leading face [*Genge and Grady*, 1998]. The occurrence of CumPo spherules that have an increase in grain size toward the leading face of the particle thus suggests settling of olivine under deceleration due to its higher density than the surrounding silicate melt.

3. Simulation of Settling During Entry Heating

A numerical simulation based on the model of *Love and Brownlee* [1991] is used here to simulate atmospheric deceleration (supporting information S2). To model the settling of olivine crystals a more realistic treatment of the nature of the melting process was added to the model in which the temperature-dependent fraction of melt is incorporated from thermodynamic calculations to predict the compositions and abundances of the stable equilibrium phase assemblage. Phase calculations were performed using the MELTS code [*Ghiorso and Sack*, 1995; *Asimow and Ghiorso*, 1998] at a pressure of 1 atm and fO_2 fixed at the iron-wustite buffer. The phase abundances and compositions were derived over a range of temperatures from the eutectic to the liquidus. The densities of silicate partial melt, olivine, clinopyroxene, and magnetite over the temperature range of melting were predicted following *Niu and Batiza* [1991]. Melt viscosity was calculated from liquid compositions using the formulation of *Giordano et al*. [2008] for silicate magmas. The bulk composition used in this study corresponds to an ordinary chondrite spherule and is given in supporting information together with the temperature-dependent phase abundance, density, and viscosity.

The motion of olivine crystals of different sizes during atmospheric entry were followed by numerical integration of their equation of motion (equation (1)), where a is the deceleration derived from the entry heating model, η is the viscosity, R is the radius of the vesicle, ρ_m is the melt density, and ρ_c is the olivine density. Stokes law was used to calculate crystal velocity, and the calculation thus assumes the crystal travels at terminal velocity.

$$\frac{\partial x}{\partial t} = \frac{2(\rho_c - \rho_m)aR^2}{9\eta} \tag{1}$$

The viscosity of crystal-melt mixtures at subliquidus temperatures was evaluated using the Einstein-Roscoe relation (equation (2) [*Einstein*, 1906; *Roscoe*, 1952]. The effective viscosity of the mixture is dependent on the volume fraction of crystals ϕ and a threshold parameter $\omega = 0.6$. Crystal content causes significant increases in the effective viscosity. The density of olivine is assumed to be constant at 3400 kg m^{-3} [*Niu and Batiza*, 1991].

$$\eta_{\text{eff}} = \eta\left(1 - \frac{\phi}{\omega}\right)^{-2.5} \tag{2}$$

Two different models of crystal settling were undertaken. Relict crystal migration was calculated as the distance travelled over the suprasolidus proportion of flight since these crystals survive melting. In contrast, phenocryst migration was only calculated during cooling at subliquidus temperatures and crystal size was assumed to increase proportional to the crystal to melt ratio. A more detailed discussion of the methodology can be found in the supporting information [*Brearley and Jones*, 1998; *Genge et al*., 2005; *Genge*, 2006; *Vondrak et al*., 2008].

4. Settling of Crystals in Cosmic Spherules

The results of simulations of settling of 5 μm radius olivine crystals in particles with different entry angles and initial sizes for entry velocities of 12 and 16 km s^{-1} are shown in Figure 2. Crystal migration distance increases with deceleration and thus is largest for particles entering the atmosphere at the highest angles with large initial size. At entry velocities of 12 km s^{-1} settling distance reach 0.25× particle radius (pr) only at vertical entry angles for particles heated to peak temperatures up to the liquidus and only particles with an initial radii of 40 μm will experience such conditions. Significantly larger settling distances up to 2.5 pr, however, occur at higher entry velocity (16 km s^{-1}) at subliquidus temperatures with settling exceeding 0.25 pr over a wide range of entry angles (90–20°) and initial particle sizes of 25–75 μm.

Crystal size strongly influences settling distance over the suprasolidus portion of flight with less settling occurring for small crystals. The settling distances for a 2.5 μm radius olivine crystal at an entry velocity of 12 km s^{-1} are shown in Figure 3 and are factor of 4 smaller than those for a 5 μm crystal, consistent with the dependence of settling velocity on R^2. Values of migration distance at an entry velocity 12 km s^{-1}, even at maximum entry angle, at subliquidus temperatures, are <0.1 pr. At 16 km s^{-1} particles with entry angles

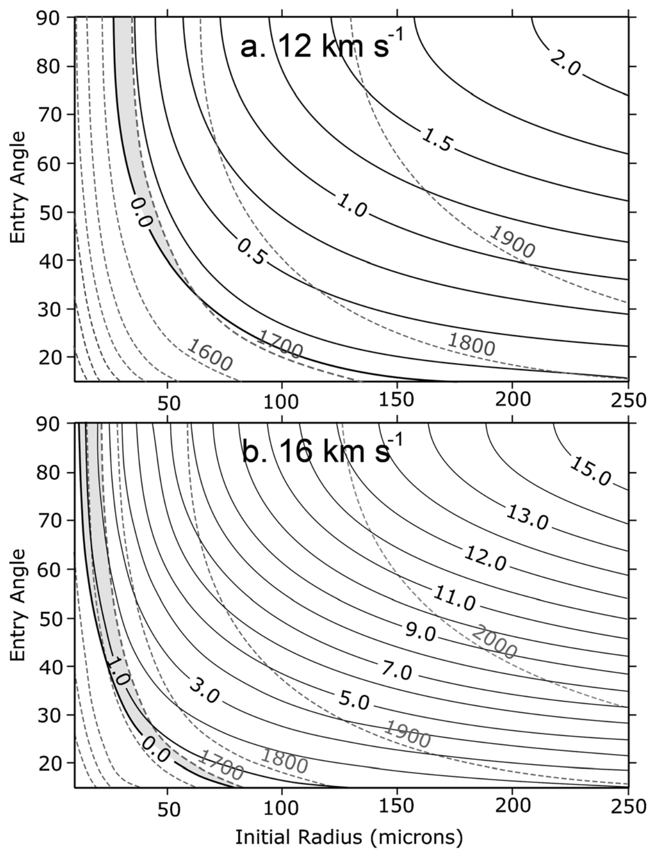

Figure 2. (a and b) The migration distance of 5 μm radius olivine crystals within cosmic spherules of different initial sizes, entry angles, and entry velocities. The dashed grey lines show the peak temperatures attained by particles during atmospheric entry, where 1700 K is the liquidus temperature. The opportunity for settling at subliquidus temperatures is shown by the shaded area.

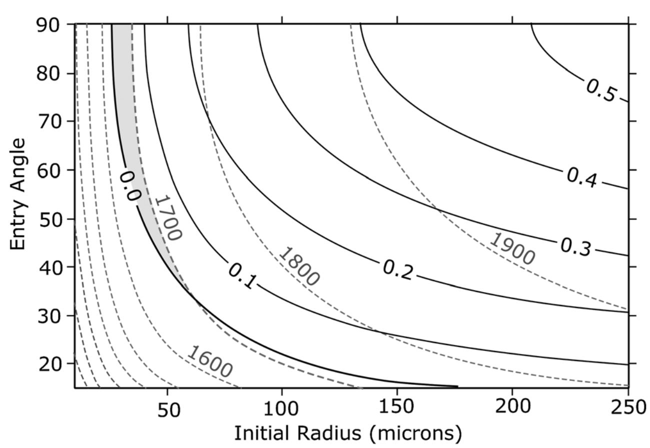

Figure 3. The migration distance of 2.5 μm radius olivine crystals within cosmic spherules of different initial sizes and entry angles at an entry velocity of 12 km s^{-1}. The dashed grey lines show the peak temperatures attained by particles during atmospheric entry, where 1700 K is the liquidus temperature. The opportunity for settling at subliquidus temperatures is shown by the shaded area.

>30° and with radii of <75 μm have settling distance of >0.25 pr, where they attain liquidus peak temperatures.

Settling distances of phenocrysts formed during cooling from peak temperature are much smaller than those of relict crystals (Figure 4). The combination of small initial size and decreasing deceleration after peak temperature leads to values of <0.004 pr at 12 km s^{-1} even at the largest entry angles.

5. The Origins of Cumulate Olivine Spherules

Concentration of olivine crystals to the leading face of CumPo spherules is likely to require migration distances of at least 0.25 pr. Simulated migration distances for phenocrysts, however, are significantly smaller at <0.004 pr even given optimal high entry angle, where deceleration is maximized. The settling and accumulation of phenocrysts due to deceleration, therefore, cannot explain the textures of cumulate spherules.

Other possible explanations for the observed grain-size variation could be thermal or compositional gradients that could influence olivine growth rates. Thermal gradients are likely if particles are oriented during flight since heating occurs by the collision of atmospheric molecules with the leading face. A preferred orientation for cosmic spherules during flight is suggested by the presence of single metal beads which have migrated by deceleration to the leading face of particles [*Genge and Grady*, 1998]. Preferred orientation is likely to occur by spin-up of particles owing to their asymmetric shapes and would also be required for olivine migration. Compositional differences might also explain variations in crystal growth rate. The compositions of cosmic spherules tend to be more iron rich close to the metal bead. In cumulate olivine spherules the iron-rich areas are associated with larger crystal sizes. The best evidence that neither compositional differences nor thermal gradients are responsible for the textures of cumulate

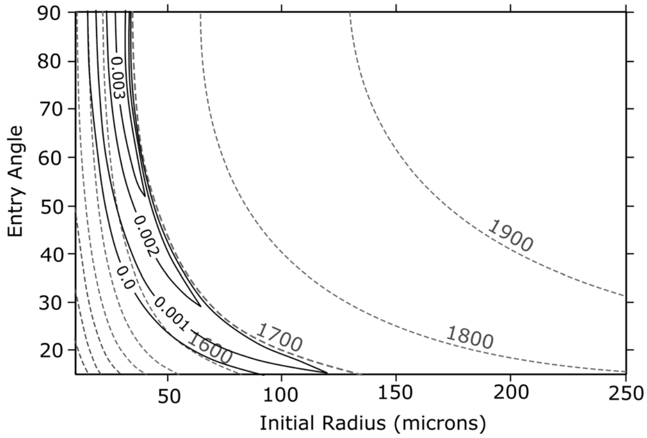

Figure 4. The migration distance of olivine phenocrysts growing to a maximum radii 5 μm in cosmic spherules with different initial sizes and entry angles at an entry velocity of 12 km s^{-1}. The dashed grey lines show the peak temperatures attained by particles during atmospheric entry, where 1700 K is the liquidus temperature. The opportunity for settling at subliquidus temperatures is shown by the shaded area.

olivine spherules is that noncumulate coarse-porphyritic spherules with metal beads (Figure 1c), which were also oriented, lack grain-size variations and yet must have been subject to the same gradients.

Although the migration distances of olivine phenocrysts are too small to facilitate accumulation during atmospheric deceleration, relict grains have significantly higher migration distances since they are present throughout suprasolidus flight. Although the textures observed in cumulate olivine spherules are produced by olivine phenocrysts, relict forsterite is observed in the cores of many of the larger crystals. Relict crystals play an important role in crystallization since they act as nucleation sites for phenocrysts. Crystallization during rapid cooling is often impeded by a lack of crystal nuclei on which crystals can grow [*Berkebile and Dowty*, 1982]. Nuclei can form spontaneously in a melt, by homogeneous nucleation; however, the maximum nucleation rate usually lies at lower temperature than the maximum in crystal growth. Existing crystals act as heterogeneous nuclei and are effective at promoting crystal growth since they are already present at temperatures at which crystallization rate is a maximum [*Berkebile and Dowty*, 1982]. The survival of relict Mg-rich olivine in porphyritic spherules is thus probably responsible for the rapid growth of iron-bearing olivine phenocrysts and explains how these cosmic spherules can contain phenocrysts up to 25 μm in length, grown in 1–2 s, when experimentally derived equant olivine growth rates are <0.1 μm s^{-1} [*Jambon et al.*, 1992]. Migration of relict olivine toward the leading face of particles is therefore likely to produce the cumulate textures.

Cumulate olivine porphyritic spherules could potentially form at entry velocities of 12 km s^{-1} but only if they have near-vertical entry angles and a limited range of sizes of close to 80 μm diameter. At higher entry velocities of 16 km s^{-1} relict olivine settling is more significant with large olivines having significant migration distances at subliquidus temperatures over a wide range of entry angles. At these higher velocities cumulate olivine spherules could form over a diameter range of ~50–150 μm. Although this encompasses the majority of cumulate olivine spherules it does not explain the occurrence of the largest observed examples at 211 μm.

A possible explanation for the occurrence of rare large CumPo spherules is the survival of relicts to supraliquidus compositions. Under equilibrium melting the liquidus temperature is a function only of the system composition and is dependent on diffusive transport of components, an inherently slow process. Nonequilibrium melting behavior is more likely within micrometeoroids. Given perfect disequilibria, where no chemical exchange between phases is possible, the melting temperature of forsterite relicts is their single-phase melting temperature of 2063 K significantly higher than the equilibrium liquidus temperature. In reality some chemical exchange will always occur, and relicts will progressively melt and decrease in size above the equilibrium liquidus. The duration of melting is thus important in the survival of relict olivine.

The duration of suprasolidus flight at different entry angles and initial particle sizes is shown in Figure 5 for entry velocities of 12 and 16 km s^{-1}. Duration of melting decreases with entry angle and with increasing entry velocity. Relicts are most likely to survive therefore at higher entry angles and higher entry velocities; however, increasing entry velocity also increases peak temperature resulting increased melting of larger particles. It seems likely therefore that large CumPo spherules form by survival of relicts to supraliquidus temperatures and are most likely to form at entry velocities >16 km s^{-1}.

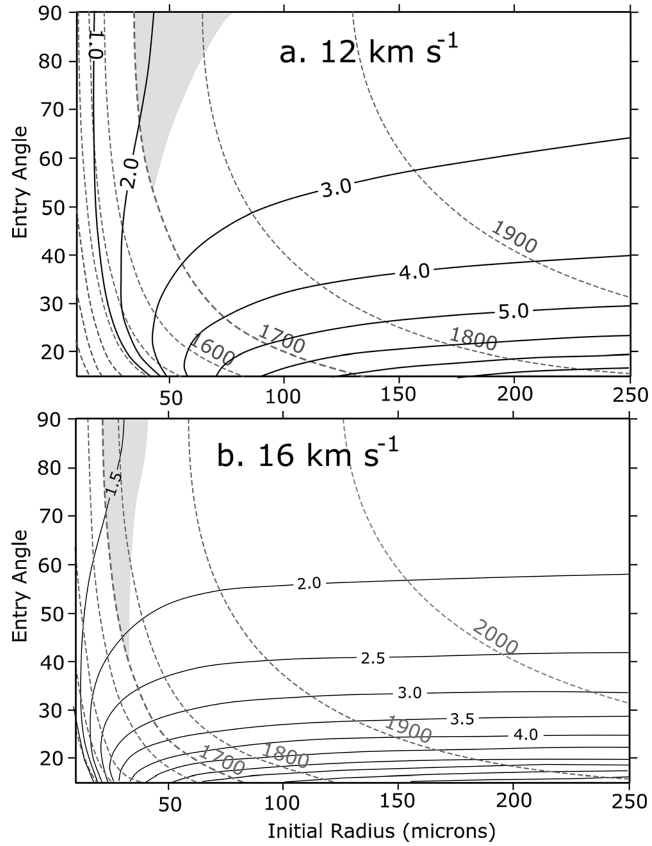

Figure 5. (a and b) The duration in seconds above the solidus for particles with different initial radii, entry angles, and velocities. The dashed grey lines show the peak temperatures attained by particles during atmospheric entry, where 1700 K is the liquidus temperature. The most likely conditions under which relict Mg-rich olivines will survive above the liquidus are shown shaded.

Although the simulation results give confidence that olivine settling during entry velocity is possible, particularly at entry velocities higher than 12 km s^{-1}, real behavior will differ from the models. First, relict forsterites will decrease in size during melting, resulting in decreases in their settling rates; however, given large rates possible at 16 km s^{-1} at high entry angles even for small relicts settling is highly likely to still occur. The growth of phenocrysts around relict olivines at subliquidus temperature will also increase settling rate during cooling, although this is likely to be relatively minor as effective viscosity increases significantly with crystal abundance.

6. Implications for the Terrestrial Dust Flux

The observation that CumPo spherules require higher entry velocities to form, while other coarse-porphyritic olivine spherules must form at low entry velocities of 12 km s^{-1} or less provides a constraint on the entry velocity distribution of the micrometeoroid flux to Earth. Cumulate olivine porphyritic spherules represent 14% of the coarse-porphyritic spherule population in this study, thus suggesting that the majority of their precursor micrometeoroids have low entry velocities of $<$12 km s^{-1} but with dispersion of 14% of this dust up to at least 16 km s^{-1}. Given that entry velocity is largely dependent on orbital eccentricity, the precursor micrometeoroids must have had eccentricities up to 0.35 when they were captured by the Earth.

Observations of sporadic meteors indicate that low-velocity asteroidal dust dominates at the Earth's orbit and becomes less abundant with increasing entry velocity [*Carrillo-Sanchez et al.*, 2015]. Coarse-porphyritic olivine and CumPo spherules add a further constraint since their oxygen isotope compositions indicate precursors related to ordinary chondrites [*Suavet et al.*, 2010; *Van Ginneken et al.*, 2015] and thus their parent bodies are probably S(IV) asteroids [*Binzel et al.*, 1996]. Previous studies have suggested that ordinary chondrite micrometeorites are related to the 5.8 Myr breakup of the Karin group, part of the Koronis family asteroids [*Genge*, 2008].

The evolution of asteroidal dust band particles by Poynting-Robertson light drag results in circularization of orbits, leading to low eccentricities by the time they are captured by the Earth [*Kortenkamp and Dermott*, 2001]. The observation that ordinary chondrite-like S(IV) dust, represented by coarse-porphyritic olivine spherules, predominately enters the atmosphere at low velocities $<$12 km s^{-1} is thus consistent with particles within asteroidal dust bands. The occurrence of CumPo spherules, however, indicates that 14% of these particles have higher eccentricities. Increases in eccentricity can result from passage through secular resonances, in particular for larger ($>$50 μm) particles that pass through such regions relatively slowly or planetary perturbations, in particular resonant trapping [*Nesvorny et al.*, 2006]. Orbital models suggest that eccentricities of 0.35 are possible for Koronis asteroidal dust [*Nesvorny et al.*, 2006]. Given that the Larkman

Nunatak collection has sampled micrometeorites over the last 800 kyr [*Van Ginneken et al.*, 2016], it represents a time-averaged record of orbital variations in ordinary chondrite dust over most of the Quaternary. Variations in the extraterrestrial dust flux are, however, indicated by a 100 kyr periodicity in ^{3}He within deep-sea sediments [*Farley and Patterson*, 1995], suggested to be the result of changes in the Earth's eccentricity and secular perturbations, but are 50 kyr out of phase with the ^{3}He record [*Kortenkamp and Dermott*, 2001]. A 41 kyr periodicity in ^{3}He flux is also reported for the early Quaternary [*Winckler et al.*, 2004]. Such fluctuations in the extraterrestrial dust flux may have climatic effects due to production of cloud nuclei [*Plane*, 2012] and have been suggested as responsible for the Quaternary glacial cycle [*Winckler et al.*, 2004]. The results of this study suggest that the mineralogy and textures of time-correlated collections of MMs recovered from sediments can place important constraints on the orbital properties of interplanetary dust accreting to Earth and the underlying causes of fluctuations in the extraterrestrial dust flux and its effect on our planet.

7. Conclusions

Coarse-porphyritic olivine spherules are reported with spatial variations in grain size that suggest that olivine settling occurs during atmospheric deceleration within these melted particles. These particles are suggested to be derived from S(IV)-type asteroids. Numerical simulations of atmospheric entry heating combined with a treatment of partial melting and crystal settling reveal that phenocrysts of olivine cannot settle during the few seconds of suprasolidus flight. Relict Mg-rich olivine grains, which are observed within the cores of large phenocrysts, however, were present throughout suprasolidus flight and experience settling over distances significant compared to particle radii. Relict olivines are suggested to promote crystal growth resulting in the observed variations in phenocryst grain size in cumulate particles. The settling of relict Mg-rich olivines is most effective at entry velocities of ~16 km s^{-1}, suggesting that cumulate particles had orbital eccentricities of ~0.35, higher than most asteroidal dust band particles. Planetary or secular perturbation of dust orbits over the last 800 kyr is suggested to be responsible for increasing the orbital eccentricity of these extraterrestrial dust particles.

Acknowledgments

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