

# An urban collection of modern-day large micrometeorites: Evidence for variations in the extraterrestrial dust flux through the Quaternary

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## ABSTRACT

We report the discovery of significant numbers (500) of large micrometeorites (>100 µm) from rooftops in urban areas. The identification of particles as micrometeorites is achieved on the basis of their compositions, mineralogies, and textures. All particles are silicate-dominated (S type) cosmic spherules with subspherical shapes that form by melting during atmospheric entry and consist of quench crystals of magnesian olivine, relict crystals of forsterite, and iron-bearing olivine within glass. Four particles also contain Ni-rich metal-sulfide beads. Bulk compositions are chondritic apart from depletions in the volatile, moderately volatile, and siderophile elements, as observed in micrometeorites from other sources. The reported particles are likely to have fallen on Earth in the past 6 yr and thus represent the youngest large micrometeorites collected to date. The relative abundance ratio of barred olivine to cryptocrystalline spherule types in the urban particles of 1.45 is shown to be higher than a Quaternary average of ~0.9, suggesting variations in the extraterrestrial dust flux over the past 800 k.y. Changes in the entry velocities of dust caused by quasi-periodic gravitational perturbation during transport to Earth are suggested to be responsible. Variations in cosmic spherule abundance within the geologic column are thus unavoidable and can be a consequence of dust transport as well as major dust production events.

## INTRODUCTION

Micrometeorites (MMs) are extraterrestrial dust particles that survive atmospheric entry and reach Earth's surface (Genge et al., 2008). A popular belief among amateur astronomers is that modern-day extraterrestrial dust can be collected on roofs in urban environments. Studies by Nininger (1941) reported large numbers of magnetic spherules collected in urban areas; however, later studies showed that the abundance of magnetic particles decreases away from urban areas, and that urban spherules are largely artificial in origin (Buddhue, 1950; Handy and Zimmerman, 1953). Despite these studies, amateur collection projects in built-up areas have been common, even though most researchers in MMs consider this occurrence an urban myth. Collection of MMs for research has focused on environments where terrestrial sedimentation rates and input of artificial particles is minimal, including deep-sea sediments and Antarctic ice and snow (Brownlee and Bates, 1983; Maurette et al., 1991; Taylor et al., 2000; Duprat et al., 2007; Rochette et al., 2008). These samples of the extraterrestrial dust flux allow study of the dust population within the early solar system and the nature and evolution of

their parent bodies. The preservation of such particles in sediments also provides a record of events occurring beyond our planet over geological time (Dredge et al., 2010; Tomkins et al., 2016).

Micrometeorites are thought to include materials derived from both asteroids and comets (Genge et al., 1997; Genge, 2008; Noguchi et al., 2015). Although a proportion of smaller dust particles survive atmospheric entry without significant heating, the majority of particles undergo melting during their passage of the atmosphere (Love and Brownlee, 1991). Most abundant, particularly at large sizes, are cosmic spherules, i.e., completely melted droplets dominated by quench textures. These spherical particles provide a useful proxy for the total flux of dust because they are relatively easy to identify (Maurette et al., 1991).

We report the results of a study of 500 MMs collected among particles recovered by Project Stardust (Oslo, Norway; <http://project-stardust.com>) in urban areas. A subset of 48 of these particles are shown here to be cosmic spherules on the basis of their compositions, mineralogies, and textures, and represent the youngest large MMs yet recovered.

## SAMPLES AND METHODS

Particles were collected from accumulated sediments in the gutters of roofs mostly in Oslo, Norway, although one is from Paris, France. The total mass of gutter sediment collected was 300 kg from a total roof catchment area of ~30,000 m<sup>2</sup>; the sediment samples were processed by magnetic separation, washing with water, and size fraction separation. Particles were selected under a binocular microscope on the basis of several criteria: (1) spherical or subspherical shape; (2) color and luster (black vitreous, black to gray metallic, and translucent vitreous particles were selected); and (3) the presence of surface dendrites or metallic surface protrusions. Among 500 particles, 48 were embedded in resin and polished for further mineralogical and compositional characterization by scanning electron microscopy (Zeiss EVO 15LS) and electron microprobe analysis (Cameca SX100) at the Imaging and Analysis Centre of the Natural History Museum (London, UK); standard matrix corrections were applied. Bulk compositions were determined by averaging 5–20 wide beam (10 µm) electron microprobe analyses.

## RESULTS

The studied particles are all spherules ranging from 300 to 400 µm in diameter and are identified as MMs on the basis of mineralogy, texture, and bulk composition. The identity and abundance of spherules are shown in Table 1.

### Mineralogy and Texture of Silicate-Dominated Particles

Three textural and mineralogical groups of silicate-dominated spherule are recognized: (1) porphyritic olivine spherules dominated by phenocrysts of olivine within glassy mesostasis (Figs. 1A–1C), (2) barred olivine spherules dominated by parallel growth dendrites of olivine with interstitial glass (Figs. 1D–1F), and (3) cryptocrystalline spherules dominated by radiating clusters of fine olivine dendrites within glass (Figs. 1G and 1H).

TABLE 1. RELATIVE ABUNDANCE OF PARTICLE TYPES IN PERCENT FOR SEVERAL DIFFERENT COLLECTIONS

| Subtype  | Urban | Cap Prudhomme* | South Pole water well† | Larkman Nunatak‡ |
|----------|-------|----------------|------------------------|------------------|
| BO       | 48    | 23             | 54                     | 27               |
| C        | 33    | 27             | 16                     | 31               |
| PO       | 19    | 50             | 30                     | 42               |
| BO/C     | 1.45  | 0.9            | 3.4                    | 0.9              |
| relicts  | 10    | —              | —                      | —                |
| metal    | 10    | —              | —                      | —                |
| vesicles | 15    | —              | —                      | —                |

Note: Cap Prudhomme, South Pole Water Well, and Larkman Nunatak are all Antarctic collections. The abundance of particles with relicts, metal, and vesicles is also shown for the urban spherules. BO—barred olivine; C—cryptocrystalline; PO—porphyritic olivine. Dashes represent no data.

\*Genge et al. (1997).

†Taylor et al. (2000).

‡Suttle et al. (2015).

Nine spherules have porphyritic textures and are dominated by phenocrysts of olivine within an Na-poor aluminosilicate glassy mesostasis that also often contains magnetite (Figs. 1A–1C). The abundance of olivine within these particles varies from ~45 to 70 vol%. Olivine crystals vary from equant (Fig. 1A) to skeletal (Fig. 1B), including those arranged in domains of parallel crystals. Olivine phenocrysts exhibit normal zoning with magnesian-rich cores (fayalite,  $Fa_{16}$ ) and iron-rich rims ( $Fa_{47}$ ). Phenocrysts can also contain relict magnesium-rich inclusions ( $Fa_{1.5-3}$ ) with discontinuous zoning with the enveloping crystals (Figs. 1A and 1B). Relict cores range in size from 24.0 to 1.5  $\mu\text{m}$  and are present in apparent abundances of <4 vol%. Rounded iron-bearing olivines ( $Fa_{25}$ ) are observed in two spherules (P506 and P305) containing dendritic olivine phenocrysts and have external rims of magnesian-rich olivine (~ $Fa_{11}$ ) (Fig. 1C). Iron-rich olivines have Ca and Cr contents below wavelength-dispersive X-ray spectroscopy detection limits in contrast to equant, skeletal, or dendritic olivine phenocrysts (to 0.4 and 0.5 wt%, respectively) and forsterite cores (0.13–0.29 and 0.07–0.41 wt%, respectively).

Magnetite occurs in the mesostasis of the five porphyritic spherules with equant and skeletal phenocrysts and is present largely as cruciform dendritic crystals (Fig. 1B). It contains as much as 4.8 wt% Cr and has Ni below energy-dispersive X-ray spectroscopy detection limits. Magnetite also occurs as an incomplete discrete rim around the exterior of particle P517 (Fig. 1A) and contains 0.5 wt% Ni and no detectable Cr in contrast to magnetite within the mesostasis.

There are 23 spherules that have barred olivine textures and are dominated by parallel growth olivine dendrites with interstitial glass and equant to dendritic magnetite (Figs. 1D–1F). Barred olivine spherules can be subspherical (equidimensional), elliptical, and ovoid in shape.

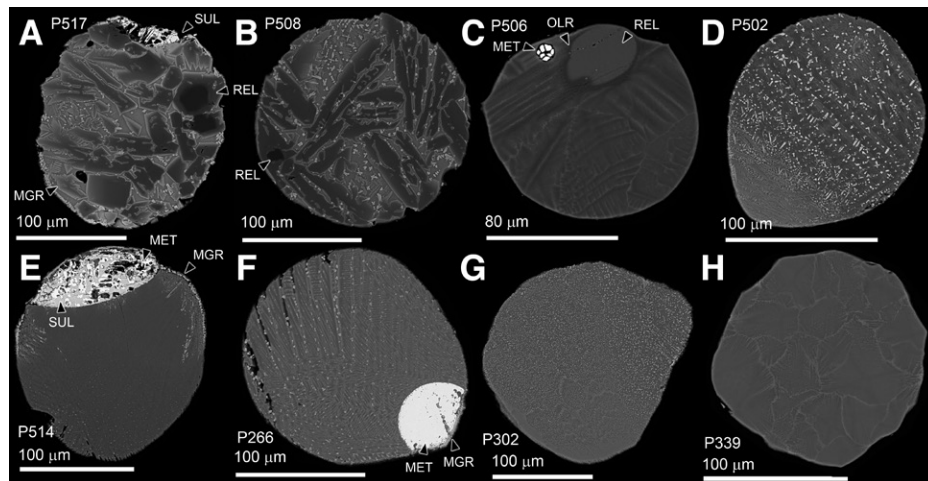


Figure 1. Backscattered electron images of urban cosmic spherules. REL—relict grain, SUL—sulfide, MET—metal, MGR—magnetite rim, OLR—olivine rim. A–C: Porphyritic spherules. D–F: Barred olivine spherules. G, H: Cryptocrystalline spherules.

In ovoid spherules the width of olivine dendrites is often smallest at the high curvature end of the particle, with the largest either at the median of the particle or at the low-curvature face (Fig. 1D). The widest olivine bars, in P502, give compositions of  $Fa_{22}$ .

We identified 15 cryptocrystalline spherules; they are dominated by radiating clusters of olivine dendrites with interstitial glass (Figs. 1G and 1H). Most particles also have interstitial submicron equant magnetite crystals, although these are absent in three particles. Olivine dendrites mostly have widths <1  $\mu\text{m}$ ; however, several particles exhibit coarser crystals, often increasing to 5  $\mu\text{m}$  across a particle and forming areas with textures similar to barred olivine spherules. Three cryptocrystalline spherules have numerous internal domains of radiating olivine dendrites producing a cellular-like structure (Fig. 1H).

Metal and sulfide beads are observed in six spherules, four barred olivine spherules (P514 and P266; Figs. 1E and 1F), and two porphyritic spherules (P506 and P517). All beads are located at one end of particles along the long axis. Metal Ni content varies from 75.0 to 21.8 wt%; sulfides consist of troilite containing 27.0–6.7 wt% Ni. Metal beads often have an external rim of Ni-bearing magnetite. Metal is partially replaced in some particles by ferrihydrite.

### Bulk Composition of Silicate-Dominated Particles

The bulk compositions of spherules were determined by averaging wide beam electron microprobe analyses and are shown in Figure 2. The compositions of spherules are mostly within a factor of 3 of CI chondrite for Mg, Al, Si, Ca, Mn, Fe, and Ti; however, all are depleted in Na (<0.1), K (<0.01), and S (<0.01) relative to CI chondrite and most are depleted in Co and Ni. The compositions of Fe–Mg–Si are also shown (Fig. 2) and illustrate a wide range plotting on

an approximate trend line between a forsterite composition and pure Fe.

## DISCUSSION

### Identification of Micrometeorites

All 48 spherules have mineralogies, textures, and compositions consistent with those of S-type cosmic spherules recovered from Antarctica and deep-sea sediments. All spherules are dominated by olivine phenocrysts or dendrites within a glassy mesostasis, usually with accompanying magnetite. Previous studies of cosmic spherules noted the dominance of olivine as a quench phase due to crystallization during rapid cooling after entry heating in the atmosphere. Olivine preferentially forms because pyroxene growth is kinetically impeded (Taylor and Brownlee, 1991). The range of olivine compositions is likewise similar to those reported from phenocrysts in cosmic spherules (Kurat et al., 1994; Genge et al., 1997). Rounded forsterite grains were

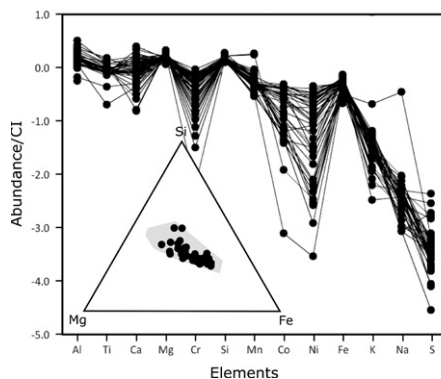


Figure 2. Bulk compositions of urban cosmic spherules showing abundances of elements compared to CI chondrite, and major element compositions (in wt%) (inset) compared to Antarctic micrometeorites (gray; Genge et al., 1997).

previously noted in spherules and identified as relict grains that survived melting during entry heating (Kurat et al., 1994; Genge et al., 1997). As noted in previous studies, a proportion of olivine contains detectable Ni (Cordier et al., 2011).

Glass compositions within porphyritic spherules are also similar to cosmic spherules, being iron-rich calcium-bearing aluminosilicate glass containing little Na (Genge et al., 1997). The Na-poor nature of cosmic spherules is thought to be characteristic of significant evaporation undergone by dust-sized particles during their entry heating (Genge et al., 1997). Meteoroid ablation spherules, formed by separation from larger meteoroids during entry heating, are thought to have higher Na contents than cosmic spherules (Genge and Grady, 1999).

The occurrence of Fe-Ni metal and sulfides within seven urban particles is strong evidence of their extraterrestrial origin because, apart from rare reduced basalts (Bird et al., 1981) and komatiites (Barnes et al., 2011), these phases are very rare among terrestrial rocks. Metal-sulfide beads are relatively common among S-type cosmic spherules and are present within >50% of particles recovered from the South Pole Water Well, but are less commonly encountered in polished section (Taylor et al., 2011).

The textures of the urban particles show close similarities to those of cosmic spherules and exhibit three of the four textural types observed within micrometeorite collections, i.e., porphyritic olivine, barred olivine, cryptocrystalline, and glassy (V type) (Genge et al., 2008). The existence of this range of textures relates to the variation in peak temperatures, cooling rates, and the nature of precursor materials, and is thus a fundamental feature of the micrometeorite flux. The lower abundance of porphyritic olivine spherules and the absence of V types among urban spherules are probably due to preferential selection of particles that have dendrites on their outer surfaces. Glassy V-type spherules are furthermore difficult to recognize in urban environments due to the large abundance of translucent, vitreous spherules of artificial origin (Larsen, 2016).

In detail the textures of urban spherules show a variation similar to that of cosmic spherules (Taylor and Brownlee, 1991; Genge et al., 1997, 2008; Taylor et al., 2007; Rochette et al., 2008). The common location of metal beads at one end of elongate particles was noted in several studies (e.g., Genge et al., 1997; Genge and Grady, 1998) and is suggested to indicate the flight direction owing to deceleration in the direction of motion. The occurrence of metal beads in such locations in six spherules in the current study is therefore excellent evidence that these particles underwent deceleration during atmospheric entry.

Iron-type (I type) cosmic spherules are found in small abundances (<5%) in micrometeorite collections (Genge et al., 1997; Taylor et al.,

2007; Rochette et al., 2008), but are difficult to recognize among urban particles due to the large number of metallic artificial spherules, including spray from welding and ablation from grinding wheels and high-speed drills used in construction (Larsen, 2016).

The bulk compositions of urban spherules provide the most conclusive evidence for their identification because they have chondritic abundances of Al, Ca, Ti, Mg, Si, Mn, and Fe, thus distinguishing them from terrestrial particles and impact spherules. Furthermore, the depletion in moderately volatile and volatile elements is consistent with partial evaporation as MMs rather than as the ablation debris of larger meteoroids (Genge and Grady, 1999). Depletions in Ni and Co are also observed in cosmic spherules and are thought to be due to separation of metal (Genge and Grady, 1998).

### Parent Bodies of Urban Micrometeorites

The sources of the recovered particles can be evaluated through their mineral and bulk compositions. All particles have chondritic bulk compositions and thus indicate that none has an achondritic source. Van Ginneken et al. (2015) showed, on the basis of oxygen isotope compositions, that 84% of coarse-grained porphyritic spherules are derived from ordinary chondrite parent asteroids, although the presence of forsterite relicts necessitates an unequilibrated source. Iron-bearing olivine relicts, in contrast, have low Ca abundances within the range of equilibrated ordinary chondrites, similar to MMs described in Genge (2008) and Rudraswami et al. (2011).

Oxygen isotope compositions suggest that the majority of barred olivine spherules are formed from fine-grained CM2 and CR2 carbonaceous chondrite precursors (Van Ginneken et al., 2015). Furthermore, several particles with high Ni olivine contents were suggested by Cordier et al. (2011) to be derived from C-poor, metal-rich precursors consistent with H or L ordinary chondrites, or type 3 carbonaceous chondrites. The range of sources of the observed urban particles is therefore similar to other micrometeorite collections.

## IMPLICATIONS

### Micrometeorite Flux

The urban particles reported herein represent the youngest large MMs collected to date; most are likely to have residence ages <6 yr (on the basis of regular cleaning of gutters on commercial buildings) with a maximum age of 50 yr (the maximum age of a building sampled). The minimal alteration of particles, with only minor rusting of metal to ferrihydrite, testifies to their short terrestrial residence.

The number of MMs expected to fall in a catchment area of 30,000 m<sup>2</sup> can be calculated from estimates of the flux of spherules at Earth's

surface of 6 t day<sup>-1</sup> for the size range 50–300 μm (Carrillo-Sánchez et al., 2015); together with size distributions from the South Pole Water Well (Taylor et al., 2007), this suggests ~2 spherules m<sup>-2</sup> yr<sup>-1</sup> in the size range of the urban particles. The recovered number of 500 particles is therefore consistent with the best estimates for the present-day flux and suggests a recovery efficiency of ~0.1%.

More significant is the observation that the relative abundance of micrometeorite types varies (Table 1) between micrometeorite collections with the barred olivine/cryptocrystalline (BO/C) ratio of 1.45 for urban spherules, higher than that of Larkman Nunatak (0.9) and Cap Prudhomme (0.9; Genge et al., 1997), but lower than that of the South Pole Water Well (3.4; Taylor et al., 2007). Larkman Nunatak moraine has been accumulating for at least 800 k.y. (Van Ginneken et al., 2016), while the blue ice of Cap Prudhomme is of unknown age but is likely to be <200 k.y., and accumulated over a shorter time period. The South Pole Water Well, in contrast, samples MMs that fell between A.D. 800 and 1500 (Taylor et al., 2007). The higher BO/C ratio in both young collections, if not a sampling bias, suggests a change in the extraterrestrial dust flux during the Quaternary.

Barred olivine spherules are thought to form at lower peak temperatures than cryptocrystalline particles (Taylor and Brownlee, 1991). A decrease in the average entry velocity of extraterrestrial dust would thus increase the BO/C ratio and could be explained by the influx of particles from a low-velocity asteroidal source. The time scale of decay of BO/C ratio from A.D. 800 to the present, however, is not consistent with orbital evolution of asteroidal collisional debris that occurs over several million years (Alwmark et al., 2012). A more likely explanation may be that there are variations in the entry velocities of dust caused by gravitational perturbations during transport to Earth, which could occur over shorter time periods and be quasi-periodic in nature (Kortenkamp et al., 2001). The observed change in BO/C ratio illustrates that data on particle type abundances from time-constrained micrometeorite collections have the potential to provide a sensitive means of investigating changes in the flux of extraterrestrial dust arriving on Earth. Differences in average entry velocity caused by gravitational perturbations will also influence the flux of dust that survives evaporation to accumulate within sediments. Variations in Earth's past extraterrestrial dust flux, determined by recovery of fossil MMs (Tomkins et al., 2016; Dredge et al., 2010), need not imply changes in the flux at the top of the atmosphere.

## CONCLUSIONS

The discovery of 48 cosmic spherules, collected primarily from roof gutters in Norway,

is reported from a collection of 500 spherules identified as micrometeorites. All particles are S-type cosmic spherules and have mineralogies, textures, and compositions identical to those of cosmic spherules collected from Antarctica and deep-sea sediments, including porphyritic, barred, and cryptocrystalline types and particles that retain Fe-Ni metal and sulfide beads. The discovery demonstrates that, contrary to current belief, micrometeorites can be collected from urban environments. The abundance of recovered spherules is consistent with estimates of the global spherule flux to Earth of  $\sim 6 \text{ t d}^{-1}$ . The relative abundance of spherule types within the modern urban spherules and ca. A.D. 800–1500 spherules from the South Pole Water Well compared with collections of spherules that accumulated over longer periods provides the first evidence for short-term variations in the extraterrestrial dust flux through the Quaternary.

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