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Sampling the constant drizzle of meteoric dust in the upper stratosphere

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Abstract. In our effort to sample the constant drizzle of meteoric dust DUSTER (Dust from the Upper Stratosphere Tracking Experiment and Retrieval) collected a surprisingly mineral-rich population of mostly nanometer, and lesser amounts of micrometer, particles. Our analysis shows that bolide disintegration could be a possible source for this dust in the upper stratosphere.

Keywords: meteoric dust, dust collection, upper stratosphere, bolides

1. Introduction

Meteoroid ablation and evaporation release metal species and molecules into the upper atmosphere where they contribute to the mesospheric metal complex between 100 and 80 km altitude. What happens next is a bit uncertain. It was suggested that vapors of small extraterrestrial particles would condense in the mesosphere followed by settling to lower altitudes (Megie and Blamont 1977). In the sulfate aerosol layer of the stratosphere the smallest ($\leqslant$500 nm) and the largest ($\geqslant$10 μm) sulfate particles had presumably nucleated on this condensed meteoritic dust (Hunten et al. 1980). There is no evidence to connect this putative, condensed meteoritic dust from mesospheric to these sulfate particle nucleation centers. Today we know there is a continuous supply of meteoric smoke nanoparticles, 4 to 20 nm in diameter, between $\sim$85 km to $\sim$35 km altitudes, incl. fayalite (Fe_2SiO_4) and pyroxene ($Mg_{0.4}Fe_{0.6}SiO_3$ Hervig et al. (2009)) that act as condensation nuclei for Polar Mesospheric Clouds (a.k.a., noctilucent clouds). Other meteoric nanograin compositions are listed as carbon (C), wüstite (FeO), or magnesiowüstite, $Mg_xFe_{1-x}O$; $x = 0.1$–0.6 (Hervig et al. 2012). These meteoric dust compositions are inferred from remote sensing data that cannot make unique identifications. If we want to know their chemical composition, size, shape, morphology and structural state (crystalline or amorphous) we need to collect these meteoric particles.

2. Particle Collections and Source Connections

The top of the stratospheric aerosol layer at 30 km altitude (Renard et al. 2008) is a natural lower boundary for any searches of interplanetary materials. The upper stratosphere is quite accessible by high altitude balloons that can carry dust

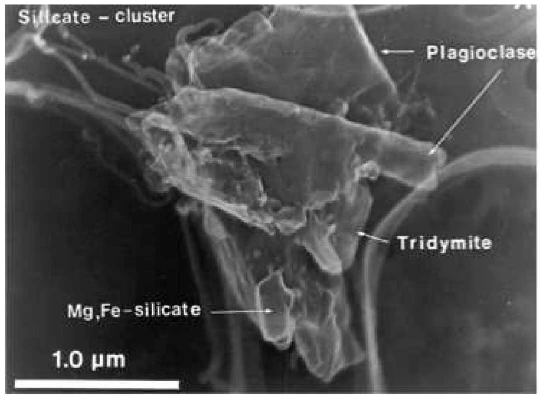

Figure 1. This cluster of Ca,Al- and Mg,Fe-silicate minerals and SiO_2 (tridymite) collected in the stratosphere between 34-36 km during May, 1985 was present in a sample of sub-micron grains that were clearly volcanic ash fines. Occam's razor then dictated that this particle too is volcanic ash. At that time claiming it was extraterrestrial dust from the Zodiacal cloud could not be supported based on the state of knowledge in 1985. Today, the same claim is not preposterous. Reproduced from Rietmeijer (1993); Journal of Volcanology and Geothermal Research.

collectors. Obviously interplanetary dust collections should avoid periods directly following volcanic eruptions of major magnitude, e.g. Mt. Pinatubo, El Chichón and Mt St. Helens, as their fine dust entrained in the rising plume could reach above 30 km (Rietmeijer 1993). The upper part of the stratosphere presents a potentially mixed environment, a crossroads, of terrestrial and extraterrestrial dust. The terrestrial component would be overwhelmingly dominated by the finest volcanic ash particles (Fig. 1). But we learn as we go on. It turns out that 85%–95% of the observed mid-infrared emission of the Zodiacal cloud is produced by particles from Jupiter-Family (J-F) comets and that ~85% of the total mass influx at Earth is J-F comet dust (Nesvorný et al. 2010). Their atmospheric velocities are typically low; and as low as $\sim$12 km s^{-1}, which means that many J-F comet particles might survive flash heating. It is then not too farfetched to postulate that this compact aggregate particle of silicate minerals (Fig. 1) could have survived but of course comets do not contain chemically differentiated materials. This notion must be revisited in the light of the mineralogical results from the Stardust mission. The big surprise was that the dust from 81P/comet Wild 2 closely resembled asteroid-like minerals and mineral-grain clusters (Brownlee et al. 2006; Zolensky et al. 2008; Joswiak et al. 2012; Dobrică et al. 2009). Thus, it is not unthinkable that this silicate-cluster has its analogs among the dust in comet Wild 2 that began life as a Kuiper Belt Object that became part of a constant supply of Interplanetary Dust Particles (IDPs) and micrometeorites (MMs) to the lower stratosphere and the Earth surface of J-F comet dust in addition to meteoric dust. Meteoric (smoke) nanoparticles have yet to be collected but for a few possible exceptions. Crystalline metallic noctilucent cloud particles $\leqslant$500 nm in diameter, and clusters

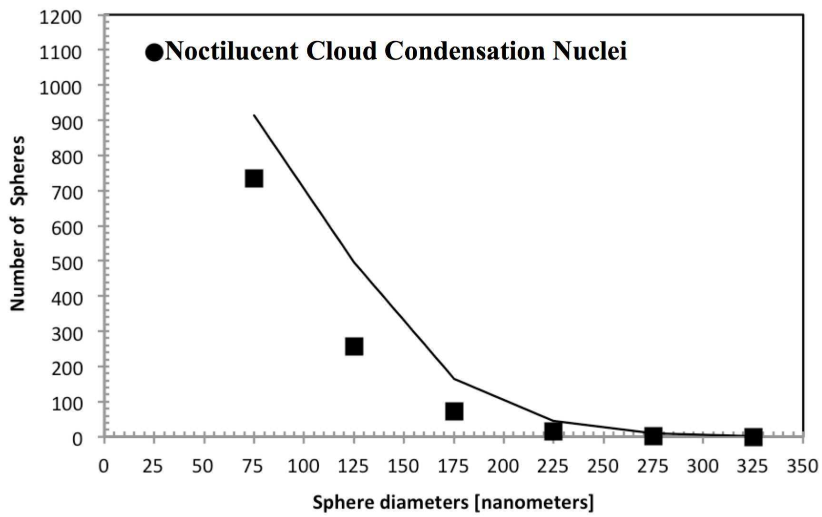

Figure 2. The size distribution of meteoric NiO and taenite smoke nanospheres from 5 to 30 nm in diameter (Hemenway et al. 1961) (solid square) and the smallest meteoric smoke particle, or noctilucent cloud condensation nuclei, size (dot). The solid squares are the midpoints of each of the six size bins listed in Hemenway et al. (1961). The trend shows that these nanospheres could be evolved meteoric smoke particles by a process of grain growth.

thereof ∼500 nm in diameter, had pure iron and FeNi- compositions, while other submicron noctilucent particles had Si and Fe, Si and Ca and pure Si compositions (Witt et al. 1964). The size distribution from 20 to 800 nm supports they could be chemically-evolved meteoric dust (Hemenway et al. 1964). Also, NiO and taenite (high-Ni Fe,Ni-metal) spherical nanometeorites intercepted settling in the lower stratosphere at 20 km altitude in the Arctic November 1960 (Hemenway et al. 1961) could be evolved meteoric smoke (Fig. 2). The NiO and taenite compositions are quite acceptable for extraterrestrial dust but unlikely for natural terrestrial dust in this size range.

Meteoric particles 0.2 to 3 microns in diameter were also collected in the lower stratosphere at ∼20 km altitude. It was concluded that this extraterrestrial component residing in the mesosphere and stratosphere did not have a chondritic composition for Fe, Ni, Mg, Mn, Ca, Na and K (Cziczo et al. 2001). Apparently the tacit assumption was that meteoric dust once it had settled into the lower stratosphere should have the chondritic composition of the annual influx of interplanetary materials to the Earth's atmosphere. The observed Ca abundance was well below its chondritic abundance (Cziczo et al. 2001), while no Al and Ti abundances were reported. It could be an indication that differential ablation (Janches et al. 2009) is on average for all incoming extraterrestrial materials more efficient than anticipated.

3. Do Meteoric Dust Aggregates Exist?

Laboratory simulations of photo-chemical oxidation of mesospheric Mg, Fe and Si metals by O_3 when settling through the upper atmosphere showed potentially diverse meteoric nanoparticle compositions. The ∼10 nm in diameter meteoric dust analogs were open aggregates of SiO_2 (silica), Fe_2O_3 (hematite) and FeOOH (goethite), fayalite, forsterite (Mg_2SiO_4), ferrosilite ($FeSiO_3$), enstatite ($MgSiO_3$), amorphous olivine [$(Mg,Fe)_2SiO_4$] and amorphous pyroxene [$(Mg,Fe)SiO_3$] (Saunders and Plane 2006, 2011). The largest meteoric analog grains in these aggregates were ∼200 nm in diameter showing that initially meteoric nanoparticle sizes could be increased via simple grain growth. The formation of open nanograin aggregates is probably an artifact of high particle densities in the experiments as it is in almost all experiments of this nature (Rietmeijer and Nuth III 2012; Rotundi et al. 1998). It is unlikely that similarly-high particle densities exist in the meso- and stratospheres except perhaps during strong winds. The finding of branched chains of nanograin aggregates with typical lengths of 1–2 microns above 35 km altitude (Bigg 2012) could be interesting evidence that meteoric dust aggregation is possible in the upper atmosphere. Still, lacking compositional data of these branched chains we cannot exclude the possibility that they can be chondritic porous (nano)IDPs. Clusters and short strings of electron-dense spheres (∼10 to ∼100 nm in diameter) collected $\geqslant$35 km altitude were interpreted as melted metallic particles from ablating meteoroids (Bigg 2012) but lacking chemical analyses this interpretation cannot be confirmed albeit also not be denied.

4. DUSTER collecting meteoric dust

DUSTER (Dust from the Upper Stratosphere Tracking Experiment and Retrieval) is an autonomous instrument designed for the non-destructive collection of dust particles, 200 nm to 40 microns in size, between 30 and 40 km altitude (∼12 to 3 mbar) in the upper stratosphere. This balloon-borne instrument has an active sampling system that was specifically developed to minimize and control contamination during instrument assembling and autonomous flight performance (Della Corte et al. 2012, 2013). The instrument contains an active collector operational at altitude and a "blank collector" that functions as a monitor of particulate contamination during all pre- and post-flight operations, and all operations in the laboratory where a class-100 clean room is used. Another unique DUSTER feature is the rigorous protocol to accept an individual particle on the active collector as "collected" during stratospheric sampling. That is, all collection surfaces (standard transmission electron microscope holey-carbon thin films on Cu-mesh grids) are automatically scanned using a Field-Emission Scanning Electron Microscope (FESEM) and any particles present are documented. Upon return after collection these same surfaces are re-scanned and "new" particles are recognized. When the blank's integrity was compromised during the actual period of stratospheric collection, or at any time after closing the actual collector during descent, recovery or transportation to the laboratory in Naples, these added particles (relative to the pre-flight analyses) are proof of contamination. When the blanks integrity was preserved, the difference

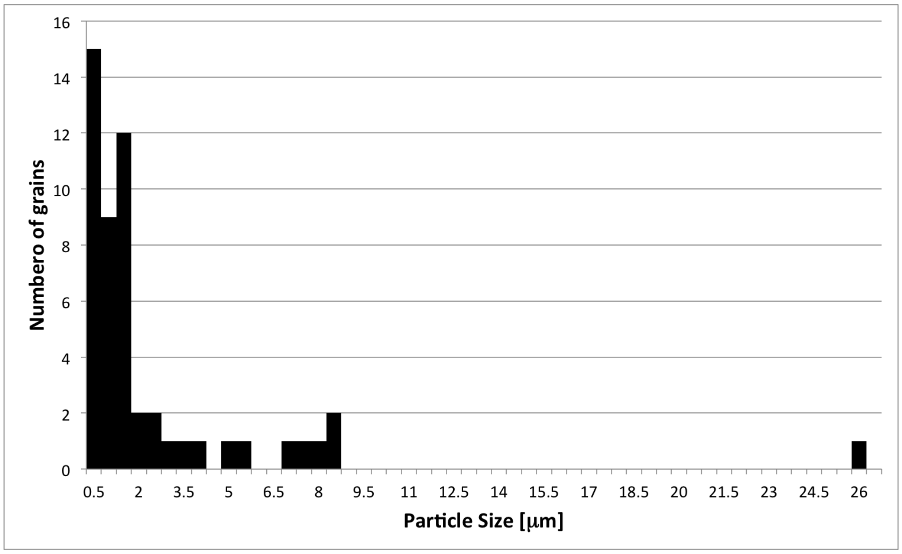

Figure 3. Histogram showing the number of particles collected in the upper stratosphere during 2008 and 2011 as a function of geometric mean particle diameter (microns).

between pre- and post-flight dust loadings on the actual collector are particles that were collected in the upper stratosphere (for more details see: Della Corte et al. (2012) and Della Corte et al. (2013)). It is possible that individual particles can be removed from the collector for further analyses by Fourier Transform Infrared Spectroscopy (FTIR) and micro-Raman spectroscopy (cf. Ciucci 2011). Fifty-one particles 0.2 μm to 26 μm in size were collected at altitudes between 32 and 38.5 km (Fig. 3).

The particles are mostly (1) Ca-bearing calcite and/or aragonite (De Angelis et al. 2011) with evidence of thermal erosion and incipient melting (Fig 4a), (2) irregularly-shaped carbon particles (Fig. 4b), and (3) carbon and low-Si C-O-Si spheres (Fig. 4c). Aluminosilica and aluminum-oxide grains are also present. Rare aggregate particles include (1) massive (Fig. 4d) and smoke-like (Della Corte et al. 2013) carbon aggregates, (2) quenched 'bunch-of-grapes' CaO aggregates (Fig. 4e), and (3) fine-grained CaF_2 aggregates (Fig. 4f).

5. Bolide Disintegration: A possible new source of meteoric dust

Such particles, but more critically, such assemblages of particles were never before identified among dust collected in the upper stratosphere. The grain compositions and morphologies point to an environment wherein each particle experienced flash heating up to $\sim$4,000K followed by ultra-rapid cooling that caused grain melting and quenching into spheres and formation of liquid spays that quenched into smoke-like aggregates (Figs. 4d, -e, -f). That is, the environment had to provide containment of these grains as a dense cloud. We propose that conditions during

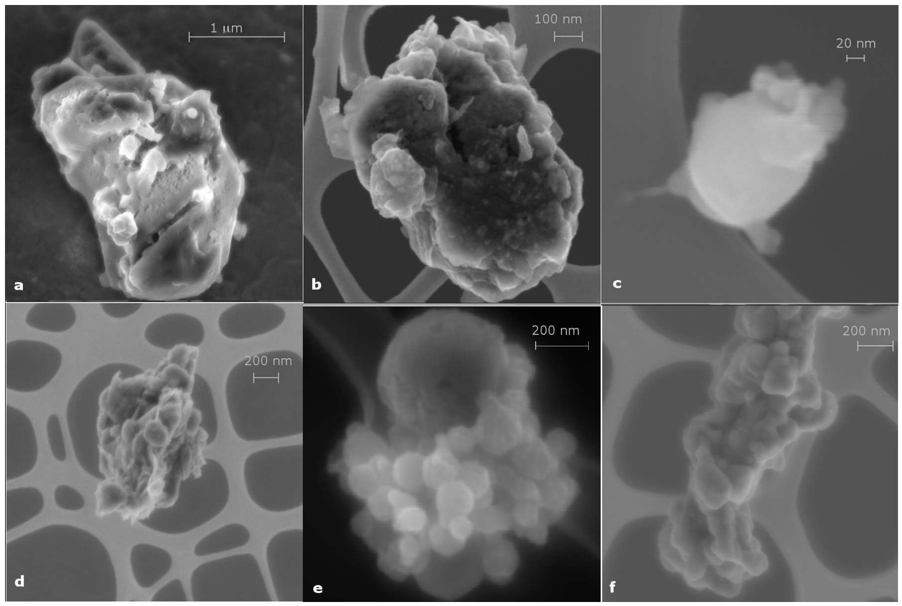

Figure 4. *(a)* thermal erosion of Ca-bearing calcite or aragonite, *(b)* an irregularly-shaped carbon particle, *(c)* a C-O-Si sphere, *(d)* an almond-shaped massive carbon aggregate of (sub)spherical grains (it lacks the typical smoke morphology of soot particles collected in the lower stratosphere, *(e)* an agglomerate of CaO nanospheres $\sim$10 to 120 nm in diameter attached to a 250 nm CaO sphere (Della Corte et al. 2013), and (f) a compact cluster of fine-grained CaF_2 grains.

disintegration of a low-tensile-strength bolide meet these environmental requirements. We suggest that similar dust particle associations will be found in the dust clouds that are associated with large bolide events, a.o. the Tagish Lake bolide.

6. Conclusions

There is a constant drizzle of nano- to micrometer size dust settling from the mesosphere to the lower stratosphere (and all the way down to the Earth surface) that consists mostly of J-F family debris in the form of most of the collected IDPs and meteoric dust due to meteor ablation in the mesosphere. The collected CaO, spheres were very much part of the dust associations collected by DUSTER but on the basis of size alone they resemble evolved meteoric smoke particles from noctilucent clouds (Hemenway et al. 1961, 1964). They are in fact oxidized mesospheric metal nanograins. Other particles collected by DUSTER are not meteoric smoke particles as they formed during bolide disintegration in the upper stratosphere. They represent a possible new source of meteoric dust that was not previously sampled.

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