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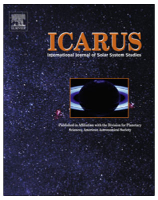

Laboratory analyses of meteoric debris in the upper stratosphere from settling bolide dust clouds

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ABSTRACT

Bolide and fireball fragmentation produce vast amounts of dust that will slowly fall through the stratosphere. DUSTER (Dust in the Upper Stratosphere Tracking Experiment and Retrieval) was designed to intercept the nanometer to micrometer meteoric dust from these events for laboratory analyses while it is still in the upper stratosphere. This effort required extraordinary precautions to avoid particle contamination during collection and in the laboratory. Here we report dust from the upper stratosphere that was collected during two campaigns one in 2008 and another in 2011. We collected and characterized forty five uncontaminated meteoric dust particles. The collected particles are alumina, aluminosilica, plagioclase, fassaite, silica, $CaCO_3$, CaO, extreme F-rich C—O—Ca particles, and oxocarbon particles. These particles are found in friable CI and CM carbonaceous chondrite, and unequilibrated ordinary chondrite meteoroids that are the most common source of bolides and fireballs. The oxocarbons have no meteorite counterparts. Some F-bearing $CaCO_3$ particles changed shape when they interacted with the ambient laboratory atmosphere which might indicate their highly unequilibrated state as a result of fragmentation. Equilibrium considerations constrain the thermal regime experienced by the collected particles between ~2000 °C and ~1000 °C, as high as 3700 °C and as low as ~650 °C after 9 s, followed by rapid quenching (μs) to below 1600 °C, but equilibrium conditions during these events is most unlikely. So far the observed thermal conditions in these events put the temperatures between ~4300 °C and ~430 °C for 5 s and high cooling rates. Such conditions are present in the immediate wake of meteors and fireballs.

1. Introduction

Meteoric dust trains and persistent dust clouds can linger for hours until they are sheared apart by upper atmospheric winds during continued sedimentation in the mesosphere and stratosphere. This dust is due to fragmentation of meteorite-dropping bolides and fireballs when decelerating in the atmosphere, and interacting with the ambient atmosphere while still entrained inside hot thermally-evolving dust trains (Fig. 1). Lingering persistent dust clouds can be traced settling in the atmosphere until they are dispersed by the upper atmospheric winds. This meteoric dust is fundamentally different from meteoric smoke particles that are the result of photolysis-driven oxidation of mesospheric metals (Plane, 2003), *viz.* Na, K, Ca, Fe and Mg, deposited between ~85 and 110 km altitudes (McNeil et al., 1998; Murad and Williams, 2002; Janches et al., 2009). The resulting meteoric metal-oxide molecules become the nuclei of noctilucent cloud particles that can be traced falling in the stratosphere (Hervig et al., 2009, 2012; Neely et al., 2011) and well into the upper troposphere (Cziczo et al., 2001).

Bolides, *i.e.* meteors brighter than −17 magnitude, and fireballs, *i.e.* meteors brighter than −8 magnitudes, are unpredictable events (Ceplecha et al., 1999). We will use these terms interchangeably. The well documented meteoroid-meteor-meteorite sequence of the Asteroid 2008 TC_3 on its pre-entry trajectory, its fireball stage, and delivery and recovery of the Almahata Sitta meteorite was the "perfect" fireball (Jenniskens et al., 2009; Millera et al., 2013). It was possible to locate its source in the asteroid belt. With the increased use of cell-phones, dashboard and security cameras, and the Internet these events receive ever increasing world-wide coverage that triggers teams of experts to rush in and conduct eye-witness interviews, collect video footage and search for surviving meteorite fragments. There were four well-documented meteorite-dropping events between 2010 and 2015 (Jenniskens,

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Fig. 1. Smoke train of the Chelyabinsk bolide with the double plumes clearly visible. (Photo: Nikita Plekhanov). Source:http://en.wikipedia.org/wiki/Chelyabinsk_meteor#/media/File:Chelyabinsk_meteor_trace_15-02-2013.jpg.

2013), *viz.* the Križevci meteorite (Meteoritical Bulletin Database) and the Sutter's Mill (Jenniskens et al., 2012), Novato (Jenniskens et al., 2014) and Chelyabinsk (Borovička et al., 2013; Popova et al., 2013) meteorites but they are only a fraction of these very bright meteors.

Fireballs experience fragmentation events along their trajectory, sudden disintegration (Klekociuk et al., 2005), and the formation of incandescent dust trains and clouds, before reaching 'rest' velocity and the onset of ballistic fall of remaining fragments of recoverable meteorites, as well as the formation of persistent dust clouds, *e.g.* the Morávka (Borovička and Kalenda, 2003) and Tagish Lake (Brown et al., 2000) fireballs. Since ordinary chondrite meteorites make up 81% of all meteorite 'falls' and 93% of all meteorite 'finds' worldwide (*cf.* Rietmeijer, 2002) it stands to reason that most bolides and fireballs should have an ordinary chondrite bulk composition. This is the case, witness the Chelyabinsk LL5 ordinary chondrite (Popova et al., 2013), Novato L6 ordinary chondrite (Jenniskens et al., 2014) and Morávka H5-6 ordinary chondrite (Borovička et al., 2003) bolides. A small fraction of fireballs will have rare carbonaceous chondrite compositions such as the Tagish Lake CI-CM carbonaceous chondrite (Brown et al., 2000), reclassified as an ungrouped Type 2 carbonaceous chondrite (Zolensky et al., 2002), and Sutter's Mill CM carbonaceous regolith breccia (Jenniskens et al., 2012). The thermal conditions and timescales of the physiochemical processes inside these debris-laden trains are poorly known but recently data became available (Borovička and Charvát, 2009; Popova et al., 2013).

Qualitatively, the event starts with rapidly rising temperatures causing near-instantaneous evaporation and flash-melting of the smallest dust and the survival of the largest grains. Following peak heating, ultrafast vapor condensation and ultrafast melt quenching will probably produce newly formed compounds that will mingle with any surviving dust and dust fragments. Surviving dusts might show signs of thermal erosion (Rietmeijer et al., 2003). Ultrafast evaporation and melting followed by ultrafast quenching tends to favor dissipative behavior causing the formation of metastable eutectic compounds (Nuth et al., 2000; Rietmeijer and Nuth, 2012). It is uncertain to what extent the surviving and newly-formed non-equilibrium dust particles may interact with atmospheric nitrogen, oxygen (*incl.* ozone) and water entrained in the trains of these fireball events. These events are still very much targets-of-opportunity especially with regard to the physiochemical processes experienced by the copious amounts of dust inside their active trains but all of it will settle through the atmosphere while being dispersed. It should be possible to intercept some of this dust while it is still high in the atmosphere. Given the kinetically-controlled interactions inside dust-carrying bolide trains the settling dust particles may be an unpredictable mixture of original, modified and newly-formed dust.

The stratosphere above the stratospheric aerosol layer at 30 km altitude (Renard et al., 2008) is a prime environment for collecting the sub-micron fraction of meteoric dust from bolide disintegration events. Also, most dust from anthropogenic sources, *e.g.* soot from commercial air traffic jet fuel burning (Blake and Kato, 1995) and massive biomass burnings, and natural sources, such as major dust storms and most volcanic activities, are primarily contained below these altitudes. Some fraction of the very fine ash, 0.5 to ~50 μm in diameter, entrained in rising ejecta plumes from eruptions of major magnitudes, *e.g.* Mt. Pinatubo and El Chichón, may reach the upper stratosphere (Rose and Durant, 2009). It is only prudent to avoid the immediate aftermath of such events when planning extraterrestrial dust collections.

This paper reports the laboratory analyses of stratospheric dust collected in the upper stratosphere at the northern hemisphere in 2008 and 2011 between 30 and 40 km altitude (~12–3 mbar) by DUSTER (Dust in the Upper Stratosphere Tracking Experiment and Retrieval). DUSTER was developed with stringent contamination controls to insure a high level of confidence that the reported dust particles were in fact present in the stratosphere during the times of collection at the reported altitudes. Proof-of-concept of DUSTER's operational and laboratory protocols was provided by the successful collection and laboratory characterization of CaO and CaF_2 nanograins that were arranged in the delicate "bunch-of-grape" particles that is characteristic of rapidly quenched vapors or liquids (Della Corte et al., 2013).

2. Experimental procedures

2.1. Dust collector

This balloon-borne instrument for non-destructive dust collection in the upper stratosphere was designed to track and document all particles on the collector surfaces during pre- and post-flight laboratory procedures with the differences being the particles that were present along its stratospheric trajectory. DUSTER is a combination of collector hardware and laboratory protocols. The following is a summary of design and laboratory protocols to ensure cleanliness at all stages of collector handling by taking precautions to ensure high-quality contamination control with the ability to recognize contaminant particles when they occurred (Della Corte et al., 2012, 2014).

DUSTER relies on inertial-impact capture of dust particles between 200 nm and 40 μm at 7 m/s directly onto a clean substrate with no need of sticking material. The collection chamber houses two collectors, (1) the "actual collector" that is opened only when the intended sampling altitude is reached and (2) the "blank collector". The "blank collector" which is identical to the "actual