RECOVERY AND CLASSIFICATION OF SPHERULES FROM THE PACIFIC OCEAN SITE OF THE CNEOS 2014-01-08 (IM1) BOLIDE

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Abstract: We have conducted an extensive towed-magnetic-sled survey during the period 14-28 June, 2023, over the seafloor about 85 km north of Manus Island, Papua New Guinea, centered around the calculated path of the bolide CNEOS 2014-01-08 (IM1). We found about 850 spherules of diameter 0.05-1.3 millimeters in our samples. They were analyzed by micro-XRF, Electron Probe Microanalyzer and ICP Mass spectrometry. We identified 22% of the spherules as the product of planetary igneous differentiation and labeled them as D-type spherules. A small portion of the D-spherules show an excess of Be, La and U, by up to three orders of magnitude relative to the solar system standard of CI chondrites. These "BeLaU"-type spherules have a chemical composition unlike any known solar system material.

Introduction: On January 8, 2014, US government satellite sensors detected three atmospheric detonations in rapid succession about 84 km north of Manus Island (Loeb et al. 2023). Analysis of the trajectory suggested an interstellar origin of the causative object CNEOS 2014-01-08: an arrival velocity relative to Earth more than ~45 km s^{-1}, and a vector tracked back to outside the plane of the ecliptic (Siraj & Loeb 2022a). The bolide broke apart at an unusually low altitude of ~17 km. The object was substantially stronger than any of the other 272 objects in the CNEOS catalog, including the ~5%-fraction of iron meteorites (Siraj & Loeb 2022b). The fireball light energy suggest that about 500 kg of material was ablated by the fireball and converted into ablation spherules with a small efficiency (Siraj & Loeb 2023).

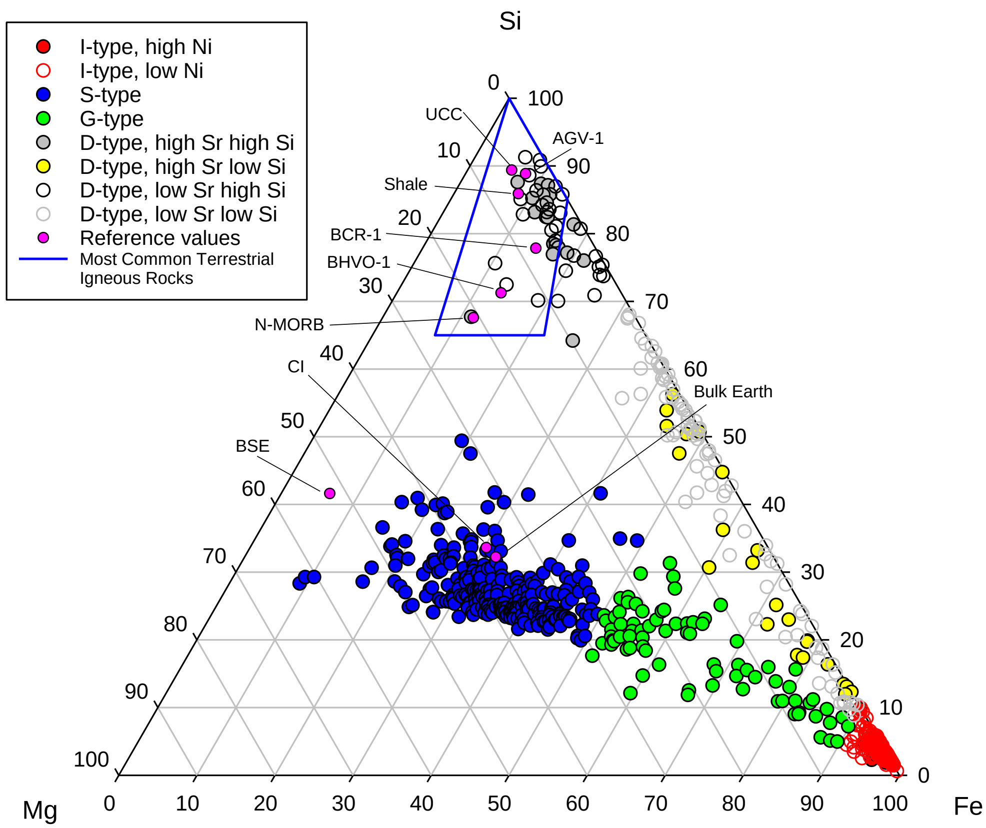
Sampling Expedition: The expedition searched for remnants of the bolide, labeled hereafter IM1. It utilized a 40-meter catamaran workboat, the M/V Silver Star. A 200-kg sled was used with 300 neodymium magnets mounted on both of its sides and video cameras mounted on the tow-bridle. Approximately 0.06 km^2 were sampled in the target area. The fine material collected on the neodymium magnets was extracted and brought in a wet slurry up to a laboratory. Subsequently, both magnetic and non-magnetic separations were processed through sieves and dried. Spherules were handpicked with tweezers using a binocular zoom microscope. They ranged in size from 100 microns to 2 mm. We obtained a total of 850 spherules by this method.

Analytical Methods: Most samples (780) were first analyzed by micro-XRF with a Bruker Tornado M4 for their bulk major element composition, followed by imaging (SEM and chemical mapping) and spot chemical analyses of about 100 samples with a JEOL Model JXA 8230 Electron Probe Microanalyzer (EPMA). Measurements of elemental abundances for about 60 major and trace elements were performed for 70 samples with an iCAP TQ triple quadrupole ICP-MS (ThermoFisher Scientific).

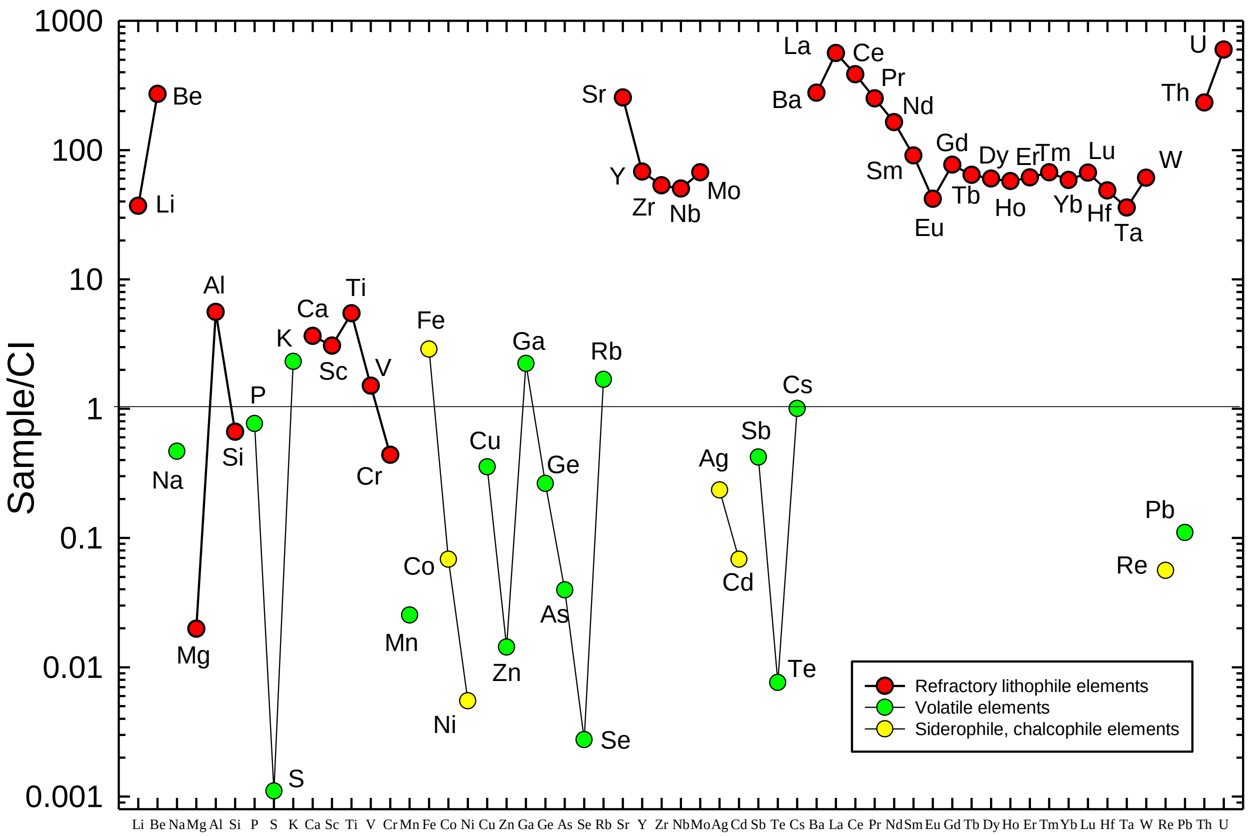
Classification: Cosmic spherules are sub-divided into three compositional types (Blanchard M.B. et al. 1980). These are the silicate-rich spherules or S-type, the Fe-rich spherules or I-type and glassy spherules or G-types. Relatively rare spherules have been labeled *differentiated* as they have similarities to achondrite meteorites and have been treated as a subgroup of S-type spherules (Folco & Cordier 2015).

The major element compositions of 745 spherules from the IM1 site, measured by micro-XRF, are plotted in a Mg-Si-Fe ternary diagram (**Figure 1a**). About 22% of the spherules have low Mg and plot close to the Si-Fe side of the diagram. The high-Si part of this group plots within the range of terrestrial igneous rocks that are shown for comparison. These spherules are thus called differentiated, meaning they are likely derived from crustal rocks of a differentiated planet; we label them as *D-type spherules*, characterized by Mg/Si < 1/3.

(1a)

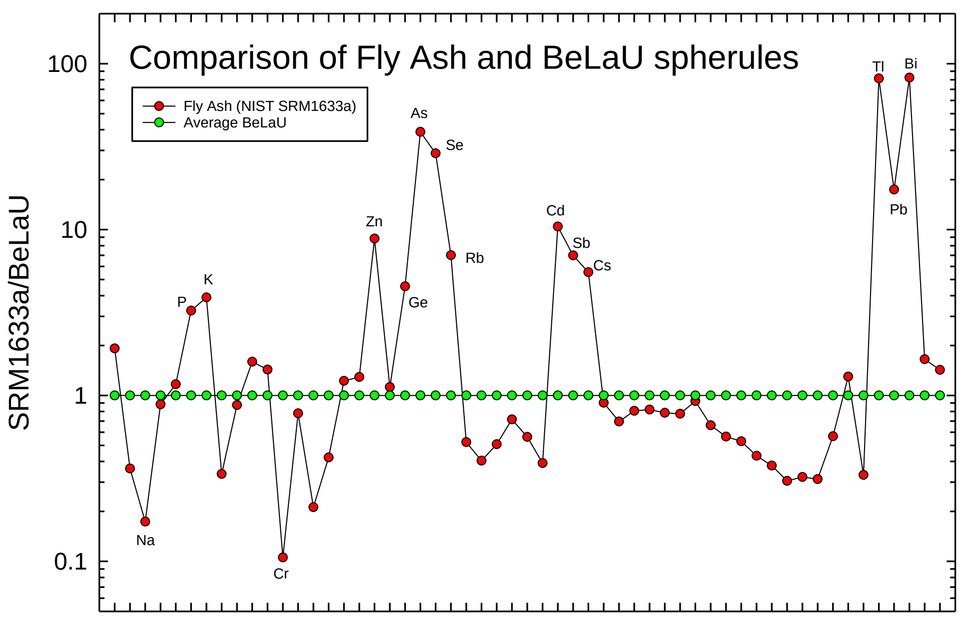


(1b)



Sample/CI
1000
100
10
1
0.1
0.01
0.001
Refractory lithophile elements
Volatile elements
Siderophile, chalcophile elements
Li Be Na Mg Al Si P S K Ca Sc Ti V Cr Mn Fe Co Ni Cu Zn Ga Ge As Se Rb Sr Y Zr Nb Mo Ag Cd Sb Te Cs Ba La Ce Pr Nd Sm Eu Gd Tb Dy Ho Er Tm Yb Lu Hf Ta W Re Pb Th U

(1c)



Comparison of Fly Ash and BeLaU spherules
Fly Ash (NIST SRM1633a)
Average BeLaU
SRM1633a/BeLaU
100
10
1
0.1
Li Be Na Mg Al P K Ca Sc Ti V Cr Mn Fe Co Ni Cu Zn Ga Ge As Se Rb Sr Y Zr Nb Mo Ag Cd Sb Cs Ba La Ce Pr Nd Sm Eu Gd Tb Dy Ho Er Tm Yb Lu Hf Ta W Tl Pb Bi Th U

Figure 1. ***Panel (a):*** Atomic Mg-Si-Fe plot of micro-XRF data for 745 IM1 site spherules. The spherule groups are compared to reference values of Earth materials (Bulk Earth, bulk silicate Earth (BSE), upper continental crust (UCC), shale, normal midocean ridge basalts (N-MORB), Hawaiian basalt (BHVO-1), Columbia River basalt (BCR-1), Guano Valley andesite (AGV-1) and CI meteorites. ***Panel (b):*** Typical BeLaU abundance pattern of a spherule in our sample (S21). ***Panel (c):*** Comparison BeLaU with the NIST coal fly ash standard SRM1633a for 55 elements**.**

For the primitive spherules we use 100Fe/(Fe+Si+Mg) > 90 to distinguish I-types from S- and G-types. The S- and G-type dividing line is 100Si/(Fe+Si+Mg) = 50, relatively consistent with previous literature (Folco & Cordier 2015; Brownlee, D. et al. 1997; Taylor S. et al. 2000). The primitive spherule groups are compared to reference values of Bulk Earth, bulk silicate Earth (BSE), and CI meteorites that are and should be on the primitive trend.

The differentiated spherules are divided into high Sr (>450 ppm) and low Sr groups (<450 ppm). They are further subdivided on the Si-content by using 100Si/(Fe+Si+Mg) = 60 as the dividing line for high Sr spherules. The D-type spherules have been divided into four distinct groups. This results in 8 distinct spherule groups that are all shown in **Figure (1a)**.

BeLaU Spherules: We use a different ternary plot to identify spherules with particularly high contents of refractory lithophile elements, based on the enrichments of Be, La and U relative to Mg and Fe. First the concentrations of these elements are divided by the concentrations of the same elements in CI chondrites. The CI-normalized values (Be_{CI}, La_{CI}, U_{CI}, Mg_{CI} and Fe_{CI}) are used to calculate the plotting parameters for the ternary diagram. The BeLaU parameter (Be_{CI}+La_{CI}+U_{CI})/(Mg_{CI}+Fe_{CI}+Be_{CI}+La_{CI}+U_{CI}) was divided by 100, while the M-parameter was multiplied by 10 to make use of the entire area inside the ternary diagram. We define a BeLaU spherule as having BeLaU > 80, and we define high and low Si varieties based on the Al/Fe ratio. This procedure identifies 10 of D-type spherules as BeLaU low Si spherules and 2 as BeLaU high Si spherules. Based on 60 elements from the periodic table, their chemical composition shown in Fig. (1b), is unlike any known solar system material. The BeLaU composition shows an excess of Be, La and U, and other elements by up to three orders of magnitude relative to the solar system standard of CI chondrites. Many of these enhanced elements are 1-2 orders of magnitude more abundant than shown in Figure 2 of Rudraswami et al. (2016) for S-type spherules.

It has been claimed that the compositions of BelaU spherules are consistent with coal ash (Gallardo 2023). The National Institute of Standards and Technology (NIST) has provided standards of coal fly ash. The best documented standard for many elements is SRM 1633a (Jochum et al. 2005). We compare the average composition of BeLaU spherules (Loeb et al. 2023) for 55 elements with the SRM1633a coal fly ash standard in **Figure (1c)**. Many volatile elements (Zn, As, Se, Cd, Tl, Pb and Bi) are enriched in the coal fly ash by factors of about 10 to 100 compared to the BeLaU spherules. Some refractory elements (Be, Ca, Cr, Fe, Y, Tm, Yb, Lu W) are depleted by factors of 3 to 10 in coal fly ash when compared BeLaU spherules. Thus, BeLaU spherules do not have the composition of coal fly ash making the claim of Gallardo (2023) invalid.

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