



# Ni-rich spinels and platinum group element nuggets condensed from a Late Archaean impact vapour cloud

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## ABSTRACT

Deciphering Earth's impact history before ~2 Ga relies heavily on the lunar record and terrestrial spherule layers, which are distal ejecta from large impacts. This study focuses on the Paraburdoo and Reivilo spherule layers in Western Australia and South Africa respectively, that were probably formed by one impact around 2.57 Ga. Both layers contain an aggregate thickness of ~2 cm of spherules, known as microkrystites. These spherules are up to ~0.6 mm in diameter and crystallized during flight, but were diagenetically replaced by K-feldspar and phlogopite with remarkable textural retention. Unlike any other Archaean layer, except for the 3.2 Ga S3 layer in the Barberton greenstone belt, the Paraburdoo and Reivilo spherules contain Ni-rich spinel crystals and high concentrations of meteoritic material (up to 357 ng g<sup>-1</sup> Ir for bulk samples of several gram). These exceptional characteristics shed new light on the distribution of the meteoritic component carrier phases (metallic alloys dispersed in the pristine glass) and the processes involved in impact spherule formation and secondary alteration.

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## 1. Introduction

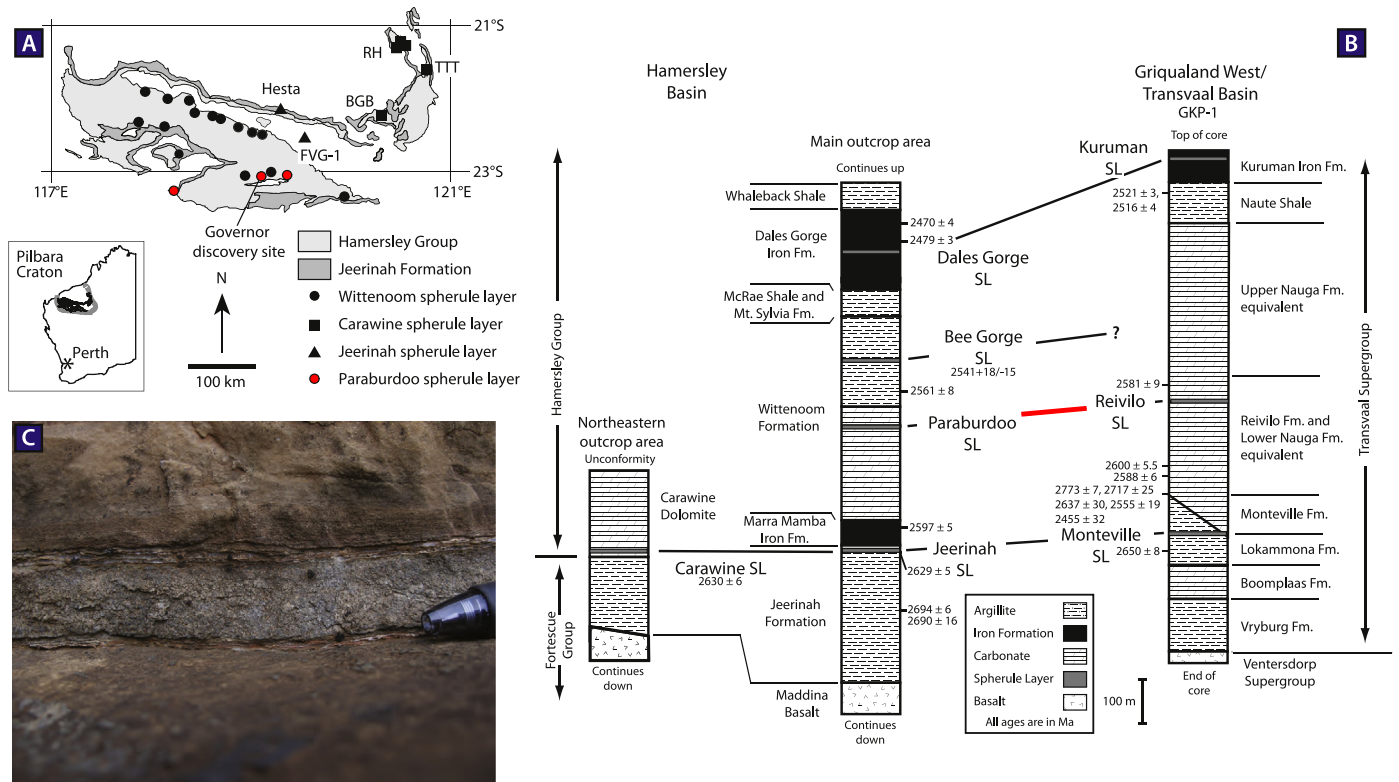
Extrapolation of the lunar cratering record implies that Earth was heavily bombarded during the Hadean and Archaean eras (Neukum and Ivanov, 1994; Bottke et al., 2012; Glass and Simonson, 2013; Johnson and Melosh, 2012). While claims for large Archaean impact craters remain controversial (Garde et al., 2012; Jourdan et al., 2012; Reimold et al., 2013; Garde et al., 2013), the most widely accepted evidence for this bombardment arises from spherule-rich layers in Archaean and Early Proterozoic sedimentary and volcanic strata on the Pilbara (Western Australia) and Kaapvaal (South Africa) cratons (Bottke et al., 2012; Glass and Simonson, 2013; Johnson and Melosh, 2012). At least four discrete impact events occurred close to the Archaean–Proterozoic Boundary (APB) over about 140 Ma, from approximately 2.63 to 2.49 Ga (Fig. 1; Hassler et al., 2011; Simonson et al., 2009a, 2009b); spherules from one of these events are the focus of this study.

Ejecta found more than ~10 crater diameters away from a large crater consists primarily of glassy spherules – droplets solidified from melt or condensed from vapour (Glass and Simonson, 2012, 2013). Most impact spherules are smaller than 1 mm in diameter and are either microtektites, consisting of pure glass, or microkrystites, containing primary crystallites in addition to glass. Microkrystites are generally more mafic in composition, can be highly contaminated by the impactor as indicated by elevated siderophile element contents (French and Koeberl, 2010), and may contain unusually Ni-rich spinels. As such, microkrystites probably represent droplets condensed from vaporized target rocks and impactor (Koeberl et al., 2012).

The Paraburdoo and Reivilo spherule layers have comparable stratigraphic positions in the sedimentary sequences of the Hamersley (Pilbara craton) and Griqualand West (Kaapvaal craton) basins, respectively (Hassler et al., 2011), and show similar interpolated U–Pb ages of around 2.57 Ga (Trendall et al., 1998; Sumner and Beukes, 2006; Fig. 1). These layers also share numerous characteristics not seen in any of the other APB spherule beds (Hassler et al., 2011). Both consist of highly crystallized microkrystites with abundant pseudomorphs of plagioclase and skeletal ferromagnesian (olivine and possibly clinopyroxene) crystals replaced mainly by K-feldspar and a phlogopite-like phyllosilicate,

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**Fig. 1.** Overview of the Hamersley and Griqualand West basins stratigraphy, the locations of the Paraburdoo spherule bed outcrops, and appearance of the spherule bed in the field. (A) Generalized geologic map of the Hamersley basin showing areas of occurrence of the Hamersley Group and Jeerinah Formation (uppermost Fortescue Group) and most locations where spherule layers have been studied (adapted from [Simonson et al., 2009b](#) and references therein). (B) Schematic stratigraphic columns of relevant parts of successions in Hamersley and Griqualand West basins showing spherule layer stratigraphy; black and red lines indicate proposed correlations (from [Hassler et al., 2011](#) and [Simonson et al., 2009b](#) and references therein). (C) Paraburdoo spherule layer at the Governor site outcrop (~1.6 cm thick), one of the most complete exposures. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

respectively. The skeletal texture is indicative of rapid cooling of a melt with mafic to ultramafic composition. Moreover, the bubble cavities and replaced cores of impact glass common in spherules in other APB layers rarely occur in these two layers. The Paraburdoo layer is ~2 cm thick, normally graded, and probably deposited as a direct fallout layer. The Reivilo spherule layer also contains an aggregate thickness of ~2 cm of spherules, but they were reworked into a thicker layer by waves, currents and/or slope processes at some sites in the Griqualand West basin ([Hassler et al., 2011](#); [Simonson et al., 2009a, 2009b](#)).

## 2. Materials and methods

Three bulk rock samples from the Paraburdoo layer discovery site Governor (S23°03'35.2", E118°49'9.6"; Fig. 1; Supplementary Fig. S1), one sample collected at Weeli Wolli (S23°02'42.9", E119°09'32.4"), and two spherule bed aliquots sampled from the Paraburdoo outcrop site (S23°14'51.3", E117°40'59.5") were removed from the exposures using hand tools (rock pick and chisels). Where possible, the samples were extracted by quarrying the surrounding rock to avoid contact with the spherule layer. None of the material used for further processing showed any evidence of contact with the used tools. The samples were broken into smaller pieces with an agate mortar and pestle, part of which was then ground to powder with an agate ball mill, and thoroughly homogenized. Original ground sample masses varied between 7.5 and 30.8 g. These powders were used in all subsequent destructive analyses. Powder aliquots of 2 to 5 g were dried at 110°C and loss on ignition (LOI) was determined by heating at 950°C. Samples were analyzed for major elements and several trace elements (Cu, Zn, Rb, Sr, Y, and Zr) by X-ray fluorescence spectrometry on

pressed powder pellets (Table 1), using the ARL 9400 instrument at the University of Liège (Belgium). Accuracy and reproducibility were evaluated on the basis of a collection of international standards ([Bologne and Duchesne, 1991](#)). The concentrations of the other trace elements were determined at Ghent University via inductively coupled plasma-mass spectrometry (ICP-MS) after acid digestion of ~100 mg of sample (Table 1; [Goderis et al., 2010](#)). Accuracy was assessed by analysis of certified reference materials (CRMs) BE-N, PM-S, DNC-1, and WPR-1. The concentrations of the platinum group elements (PGEs) and Au were measured at both Ghent and Cardiff Universities, via a combination of sample preparation relying on nickel-sulfide (NiS) fire assay techniques with ICP-MS, following the procedures described in [Goderis et al. \(2010\)](#), [Huber et al. \(2001\)](#) and [McDonald and Viljoen \(2006\)](#). Replicate PGE analyses were carried out on duplicate or triplicate powder aliquots, preferably using 15 g sample aliquots or more, however, due to sample size, this was not possible for samples IV-69IE and IV90 (Table 2). The accuracy of the PGE analyses was assessed by analysis of the CRMs TDB-1, WMG-1, and WPR-1, as reported in Table 2.

Simultaneously, the unground bulk rock fractions of the Governor site samples were prepared as thin and thick sections and studied using optical microscopy (detailed petrographic descriptions can also be found in [Hassler et al., 2011](#)), an M4 Tornado  $\mu$ XRF spectrometer at Bruker AXS Microanalysis GmbH in Berlin (Fig. 2), a JEOL model JSM-6400 Scanning Electron Microscope (SEM) equipped with a Si-Li detector (Pioneer) for energy-dispersive X-ray elemental analysis and a Dilor XY Raman spectroscope (HORIBA/Jobin Yvon) with an Olympus BH2 microscope, liquid nitrogen cooled CCD detector, and Coherent Innova 70C Ar/Kr mixed gas laser (512 and 647 nm) at Vrije Universiteit Brussel

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