

Geochemical evidence for an impact origin for a Late Archean spherule layer, Transvaal Supergroup, South Africa

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ABSTRACT

A Late Archean layer rich in sand-sized spherules of former silicate melt in the Monteville Formation (Transvaal Supergroup, South Africa) has Ir concentrations as high as 6.4 ppb and is clearly enriched in Ir relative to associated tuffs, carbonates, and shales. The Monteville spherule layer is also enriched in other siderophile elements, including the platinum group elements (PGEs). The PGEs in the spherule layer produce a flat (meteorite like) pattern when they are normalized to chondritic abundances. The abundances of Ir and other siderophile elements are similar to broadly contemporaneous spherule layers in the Hamersley basin of Western Australia. That the mineral compositions, textures, and sedimentary structures of the spherule layers in the Transvaal Supergroup and Hamersley basin are also very similar suggests that they were all formed by the same processes. We think that the best way to explain the high Ir concentrations and other characteristics of the Monteville spherule layer is that it represents distal impact ejecta. There are, however, significant differences between the Monteville spherule layer and Early Archean spherule layers in the Barberton greenstone belt, including much higher average and maximum concentrations of Ir in the latter. The data presented here clearly show that each Precambrian spherule layer is unique and needs to be characterized individually, as is true for the impact spherule layers of the Phanerozoic.

Keywords: impacts, spherules, Archean, iridium, platinum group elements.

INTRODUCTION

Discrete layers rich in distinctive sand-sized spherules of former silicate melt that have been interpreted as distal impact ejecta occur in four formations in the Hamersley basin of Western Australia (Simonson, 1992; Simonson et al., 2000). The impact interpretation is supported by elevated concentrations of Ir (to 1.69 ppb) and certain other siderophile elements in at least two of these layers (Simonson et al., 1998). Many stratigraphic and geochronologic similarities have been noted between the Hamersley basin and the Transvaal Supergroup of South Africa (e.g., Button, 1976; Cheney, 1996). A search for possible correlatives of the Hamersley spherule layers in roughly contemporaneous units of the Transvaal Supergroup yielded a single spherule-bearing layer in the Late Archean Monteville Formation (Simonson et al., 1999). The Monteville Formation is located in the Griqualand West basin, where it is near the base of a thick succession of marine strata deposited on the southwestern edge of the Kaapvaal craton and is known as the Campbell Group or Subgroup (Beukes, 1987; Altermann and Wotherspoon, 1995). Even though most of the units in the Campbell Group accumulated in shallow water, much of the Monteville Formation was deposited in deeper water, below wave base (Beukes, 1987; Altermann and Siegfried, 1997). The Monteville Formation and associated units are essentially flat lying and undeformed, except for narrow belts along the southern and western margins (Beukes, 1983).

Simonson et al. (1999) identified the spherule layer at a constant stratigraphic level within the Monteville Formation at several surface sites and in

10 cores distributed throughout an area of ~17 000 km². This encompasses all three of the areas reported in Simonson et al. (1999) plus a new site discovered in 1998 ~85 km southwest of Monteville farm. The nature of the layer varies considerably from one locality to another, but at all of these sites, the spherules range to ~1 mm in diameter and consist mainly of K-feldspar, except where they are replaced by carbonate. The internal textures of the spherules are very similar to those of the Hamersley layers and differ from other types of sand-sized spheroidal clasts, such as carbonate ooids and volcanic accretionary lapilli (Simonson et al., 1999). The Monteville spherule layer does not contain any conventional volcanoclasts, and the stratigraphically closest tuffaceous layer in the cores is located 46.5 m below the spherule layer.

The Monteville spherule layer also displays sedimentary structures that are comparable to those of the spherule layers of the Hamersley basin (Simonson et al., 1999; Hassler and Simonson, 2001). The Monteville spherule layer may actually be a product of the same event as one of the Hamersley spherule layers, but it is not clear at present which one because the age of the Monteville spherule layer is not well determined. SHRIMP dating of zircons from tuffs in the general stratigraphic vicinity of the Monteville spherule layer has generally yielded dates ca. 2.55 Ga, but a tuff collected stratigraphically ~50 m below the Monteville spherule layer produced a date of 2650 ± 8 Ma (see discussion in Simonson et al., 2000).

SAMPLES AND PETROGRAPHY

We analyzed 4 spherule-rich core samples from the Monteville spherule layer and 16 samples of associated strata free of spherules to determine if there was any siderophile element enrichment that could repre-

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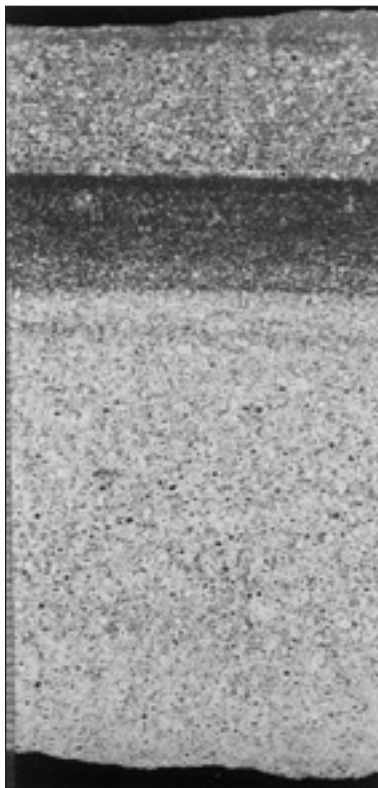


Figure 1. Cut face of core sample P11-1 showing three parts: thick, light gray basal layer rich in spherules, some of which are flattened by compaction; medial dark gray layer rich in quartzose sand; and upper light gray layer of spherules mixed with quartzose sand. Sample broke along contact with underlying shale at base and is 8 cm tall.

sent an extraterrestrial component in the spherules. The samples were all collected from two closely spaced cores, P9 and P11, drilled near a breccia-hosted zinc deposit. This deposit subsequently became the Pering mine, which is located at long 24°16'E and lat 27°26'S in the Griqualand West basin of South Africa (Wheatley et al., 1986). The spherule-rich layer in core P11 is 8 cm thick (Fig. 1), and a quarter core from this layer was split into three pieces that were analyzed separately. The fourth spherule-rich sample came from the layer in core P9. The remaining 16 samples are all from core P11 and were taken from various rock types in an interval from 54.8 m below to 31.8 m above the spherule layer. Of the samples, 14 are rich in carbonate and contain minor shale, which is uniformly black and frequently pyritic. The remaining two samples are interbedded tuff and shale (Table 1) and come from within a few meters of the contact between the Monteville Formation and the underlying Lokamonna Formation.

The shapes of the spherules in the Monteville layer range from well rounded to flattened (compacted) (Fig. 1), and they are texturally diverse internally. Most contain fibrous to lath-shaped crystals that typically radiate inward from the edges (Simonson et al., 1999, their Fig. 3). They consist primarily of K-feldspar with associated fine muscovite and minor amounts of FeS₂ and quartz. Most of the spherules contain micrometer-sized inclusions of rutile, zircon, and/or apatite, and many also contain round masses of finely felted muscovite that appear to be infilled gas bubbles. The spherule layer also contains a subordinate amount of quartzose, very fine to medium sand interpreted as continental detritus transported southward by bottom currents (Simonson et al., 1999). No evidence of shock deformation has been seen in any of these quartz grains. Small amounts of unshocked quartzose sand are also found in most of the Hamersley spherule layers (Simonson et al., 1998). The Monteville spherule layer contains both detrital grains (rip-up clasts) and tiny authigenic crystals of pyrite. In general, sulfide is scarce in the spherule samples, and no other opaque phases were observed petrographically. In contrast, concretionary pyrite is widespread in the associated shales.

Various amounts of finely crystalline matrix composed of the same minerals as the spherules are associated with the spherules. On the basis of the presence of cross-bedding and the locally well-sorted nature of the

TABLE 1. SELECTED CHEMICAL DATA FOR SAMPLES FROM PERING MINE CORES

| | Spherule-rich samples | | | | Samples from other layers | | |
|--------------------------------|-----------------------|--------|---------|--------|---------------------------|--------|---------|
| | P11-1A | P11-1B | P11-1B1 | P9-1A | P11-27B | P11-31 | P11-14A |
| Location | -- | -- | -- | -- | 46.5 m | 54.8m | 3.0 m |
| SiO ₂ | 52.51 | 62.45 | N.D. | 57.90 | 32.09 | 58.81 | 11.62 |
| TiO ₂ | 2.63 | 1.63 | N.D. | 2.36 | 1.09 | 1.35 | 0.18 |
| Al ₂ O ₃ | 24.94 | 17.17 | N.D. | 20.45 | 8.92 | 14.44 | 2.96 |
| Fe ₂ O ₃ | 0.55 | 2.54 | 6.68 | 0.55 | 8.16 | 6.76 | 15.59 |
| MnO | 0.01 | 0.03 | N.D. | 0.04 | 0.77 | 0.06 | 1.35 |
| MgO | 3.63 | 1.88 | N.D. | 2.42 | 7.37 | 2.74 | 16.11 |
| CaO | 0.35 | 1.76 | N.D. | 0.88 | 13.56 | 0.62 | 25.28 |
| Na ₂ O | 0.055 | 0.069 | 0.05 | 0.095 | 0.064 | 0.11 | 0.031 |
| K ₂ O | 11.09 | 8.56 | 6.29 | 11.50 | 6.55 | 10.33 | 1.05 |
| P ₂ O ₅ | 0.07 | 0.04 | N.D. | 0.05 | 0.20 | 0.14 | 0.03 |
| LOI | 4.38 | 3.59 | N.D. | 3.88 | 21.35 | 4.30 | 25.12 |
| Total | 100.22 | 99.72 | -- | 100.13 | 100.12 | 99.66 | 99.33 |
| Sc | 40.6 | 23.5 | 20.3 | 13.2 | 22.7 | 21.2 | 1.78 |
| V | 545 | 293 | N.D. | 378 | 160 | 177 | 18 |
| Cr | 385 | 264 | 180 | 418 | 104 | 152 | 28.6 |
| Co | 2.39 | 42.7 | 87.1 | 17.1 | 21.2 | 16.7 | 15.4 |
| Ni | 15 | 68 | 167 | 65 | 49 | 99 | 22 |
| Zn | 30 | 640 | 200 | 650 | 592 | 124 | 3 |
| Ga | 6.8 | 6.2 | 26 | 9 | 52 | 12 | 37 |
| As | 0.39 | 8.98 | 21.9 | 1.93 | 4.89 | 7.59 | 22.1 |
| Se | 0.75 | 0.72 | 0.91 | 0.29 | 0.47 | 0.42 | 0.48 |
| Sb | 0.049 | 0.95 | 1.45 | 0.38 | 0.33 | 0.61 | 1.71 |
| Cs | 7.38 | 4.92 | 3.51 | 19.1 | 2.14 | 14.6 | 1.61 |
| Hf | 5.17 | 8.83 | 12.6 | 5.38 | 2.45 | 5.43 | 1.58 |
| Ir (ppb) | 2.5 | 6.4 | 5.6 | 4.8 | 0.6 | <1 | <1.1 |
| Au (ppb) | 4.1 | 3.9 | 8 | 1.2 | 6.1 | 5.3 | 2.5 |
| Th | 4.22 | 5.03 | 5.93 | 5.63 | 1.35 | 6.85 | 2.04 |

Note: Locations of nonspherule samples are given as meters beneath spherule layer in core P11. Samples 27B and 31 consist of interbedded tuff and shale; sample 14A is carbonate with interbeds of pyritic shale. All Fe as Fe₂O₃; major elements in weight percent; trace elements in ppm, except as noted. N.D. = not determined.

quartzose sand that it contains, we infer that the layer originally consisted of well-sorted sand-sized material, so this matrix is probably secondary. Some of the matrix was clearly produced by compaction of the spherules, and some probably originated as pore-filling cement. That K-feldspar is intergrown with muscovite in the matrix clearly indicates that the current minerals are authigenic phases.

GEOCHEMISTRY

The major and trace element compositions (to 46 elements) of all samples from the Pering mine were analyzed by instrumental neutron activation analysis (INAA) and X-ray fluorescence (XRF) spectrometry. An aliquot of the spherule layer was also analyzed by fire assay followed by inductively coupled plasma-mass spectrometry (ICP-MS) to determine the contents of the platinum group elements (PGEs). Details for all analytical methods are given in Koeberl et al. (2000). The four samples from the Monteville spherule layer show considerable variation in major and trace element composition, in agreement with their variation in minerals. For example, the ranges in Fe₂O₃ and K₂O contents are 0.55–6.68 and 6.3–11.5 wt%, respectively (Table 1). The only other samples with K₂O values this high were the two tuffaceous samples, which also contain authigenic K-feldspar. The spherule samples show relatively high values of Ti, Sc, V, As, Se, Sb, Cs, Hf, Ta, and W. For these elements, there seems little difference between the contents measured in the tuff and those measured in the spherule-rich samples. Rare earth elements (REE) in the spherule samples show a pattern that is fairly flat, save for a small Eu anomaly. The pattern and the abundances of the refractory lithophile elements, including the REEs, are typical of normal Archean continental crustal material.

However, the spherule samples are characterized by a significant siderophile element enrichment compared to all other samples analyzed from the drill core, e.g., for Ni (to 167 ppm), Co (to 87 ppm), Cr (to 418 ppm),

TABLE 2. PGES IN A SAMPLE FROM THE MONTEVILLE SPHERULE LAYER

| | Elemental concentrations | | | | | |
|------------------|--------------------------|------|------|------|------|------|
| | Ir | Ru | Rh | Pt | Pd | Au |
| P11-1A | 1.8 | 3.6 | 0.51 | 5.4 | 2.3 | 0.93 |
| Detection limits | 0.02 | 0.05 | 0.02 | 0.14 | 0.10 | 0.02 |

Note: One replicate only; concentrations and detection limits both given in parts per billion. On the basis of previous studies of possible impact layers (Koeberl et al., 2000; Reimold, et al. 2000), the uncertainty on these concentrations is estimated to be 10%.

and, especially, Ir (to 6.4 ppb). The average value of Ir in the 4 samples from the spherule layer is 4.8 ppb, whereas Ir is below detection limits in 14 of the other 16 samples, including both of the tuffaceous samples and samples containing pyritic shale (Table 1). In the only two nonspherule samples where Ir was detected, its concentration is on the order of 0.2 ppb (near the detection limit of the INAA technique), and both of these samples were collected close above the spherule layer, so they could contain reworked material. The Ni/Ir ratios in the spherule samples are near chondritic, whereas the other elements show considerable deviations, perhaps reflecting variations in the indigenous concentrations of these elements in the target rocks. The high Ir values do not correlate directly with abundances of chalcophile elements, ruling out remobilization in hydrothermal and mesothermal processes, as has been proposed for the Barberton spherule layers (Koeberl and Reimold, 1995). The best evidence for an extraterrestrial component in the Monteville spherules comes, however, from the PGE data (Table 2). In a normalized abundance plot (Fig. 2), the interelement ratios of the PGES are near chondritic within the probable error. Detailed electron microscope studies showed no indication of the presence of any sulfides or other obviously siderophile element-bearing accessory minerals. Thus, it is likely that the extraterrestrial component is relatively unaltered and finely dispersed, similar to what has been observed in more recent distal impact deposits (see review by Montanari and Koeberl, 2000).

SUMMARY AND CONCLUSIONS

We studied a set of drill-core samples from the Pering mine area, Griqualand West basin, South Africa. These samples span a range of about 85 m, mostly in the Monteville Formation, and include a thin spherule layer that shows stratigraphic and petrographic resemblances to Late Archean spherule layers in the Hamersley basin of Western Australia. A distinct Ir anomaly was found in samples from the Monteville spherule layer relative to the surrounding strata: Ir concentrations in the spherule layer were as high as 6.4 ppb, and the interelement ratios of the PGES are near chondritic. Typical abundances of Ir are 0.02 ppb and 500 ppb in terrestrial and chondritic materials, respectively (Fig. 1 of Koeberl, 1998). This is consistent with ~0.4%–0.5% (but no more than 1%) of chondritic material in the samples analyzed from the Monteville spherule layer. Samples from two Late Archean spherule layers in Western Australia are likewise enriched in Ir relative to associated strata and show maximum values of 1.69 ppb (Simonson et al., 1998).

The Monteville spherule layer is also enriched in siderophile elements other than Ir relative to the surrounding strata, such as Ni, Co, and the PGES. Concentrations are somewhat variable, but the Ni/Ir ratios and the normalized PGE abundances are very close to chondritic values (Fig. 2). Similar results were obtained from the Hamersley spherule layers, although departures from chondritic ratios were observed, which Simonson et al. (1998) attributed to differential mobility during diagenesis. The ubiquitous K-feldspar in the spherules and the matrix of the Monteville layer is clearly secondary in origin. Some spherules from the Cretaceous-Tertiary (K-T) boundary layer also consist of K-feldspar that at first was thought to be an original phase

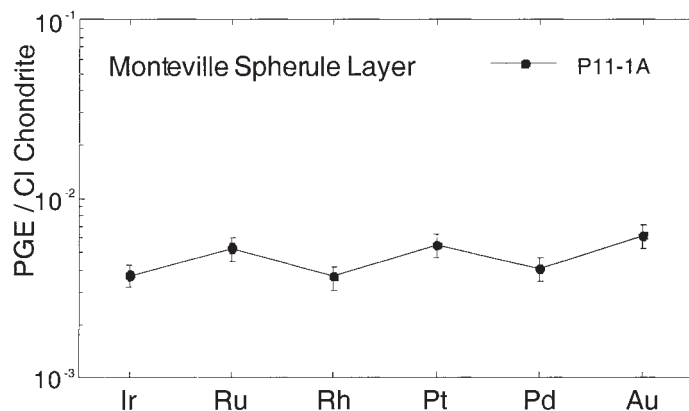


Figure 2. Chondrite-normalized plot of platinum group element (PGE) data from sample P11-1A. CI chondrite PGE concentrations (ppb; from Jochum, 1996) are as follows: Ir, 480; Ru, 683; Rh, 140; Pt, 982; Pd, 560; and Au, 148.

(Smit and Klaver, 1981), but was subsequently found to have formed via diagenetic replacement (DePaolo et al., 1983). As Kyte et al. (1985) noted for the K-T boundary layer, partial mobilization of the siderophiles is to be expected during the conversion of the original phase(s) to authigenic ones.

In addition to having diagenetic phases similar to certain occurrences of the K-T boundary layer, the Ir fluence of the Monteville spherule layer is also similar. A fluence for the Monteville layer can be derived by using the observed thickness of 8 cm for the spherule-rich layer in core P11 and an Ir concentration of 4.8 ppb, which is the average of the four samples analyzed. We further assume that the layer has the density of orthoclase (2.57 g/cm³). This is a conservative assumption, because all of the minor phases observed (e.g., muscovite, apatite, pyrite) have greater densities, which would result in higher calculated fluences. These assumptions yield an Ir fluence of about 100 ng/cm² for the Monteville spherule layer. For comparison, the average Ir fluence of the K-T boundary layer on the Pacific plate is ~90 ng/cm² (Kyte et al., 1996). Despite the similarity of these numbers, the K-T boundary layer is no more than 2–3 mm thick at distal sites (Smit, 1999), whereas the 8 cm thickness of the Monteville spherule layer in core P11 is representative of what we have observed in this layer at most study sites (Simonson et al., 1999).

However, the minerals and geochemistry of the Monteville spherule layer differ markedly from those of four Early Archean spherule layers found in the Barberton greenstone belt of South Africa (Lowe and Byerly, 1986; Lowe et al., 1989; Kyte et al., 1992; Koeberl and Reimold, 1995; Reimold et al., 2000). In contrast to the Monteville spherule layer, most of the samples analyzed from the Barberton spherule layers consist primarily of microquartz with sericite or chlorite (Byerly and Lowe, 1994), and Ir concentrations are dramatically higher; reported values are as much as 450 ppb (Kyte et al., 1992) and 2700 ppb Ir (Koeberl and Reimold, 1995). Kyte et al. (1992) calculated an Ir fluence of >2500 ng/cm² for the Barberton spherule layer known informally as S4. This higher fluence primarily reflects a higher Ir concentration, because the total thicknesses of spherules in layer S4 and the Monteville spherule layer differ by no more than a factor of two. The highest levels of Ir and other siderophile elements are partially associated with secondary sulfides in the Barberton spherule layers (Koeberl and Reimold, 1995) and are not restricted to spherule layers, but also occur in associated strata (Reimold et al., 2000). In contrast, sulfides are scarce in the samples we analyzed from the Monteville spherule layer, and none of the sulfides examined in this study shows any enrichment in the siderophile elements (Table 1).

In summary, the Late Archean Monteville Formation of South Africa contains a single spherule-rich layer that is anomalously enriched in siderophile elements, especially Ir (to 6.4 ppb), relative to enclosing tuffs,

shales, and carbonates. Normalized PGE abundances are near chondritic. The levels and patterns of these anomalies are similar to those of spherule layers in the Hamersley basin of Western Australia, as are other sedimentological and petrographic characteristics. We therefore infer that these spherule layers originated via similar processes; they are probably reworked layers of distal impact ejecta and/or condensate consisting of as much as 1% material from an extraterrestrial impactor and $\geq 99\%$ material from terrestrial target rocks. In contrast, there are significant differences between the Monteville spherule layer and Early Archean spherule layers in the Barberton greenstone belt, most notably much higher levels of siderophile enrichment and greater thicknesses of spherules in the latter (see Reimold et al., 2000, and Simonson and Harnik, 2000, for further discussion). Although the Barberton spherules probably originated as either melt droplets or condensate from a large impact (Shukolyukov et al., 2000), some difference in their formation mechanism is needed to account for the observed differences between the Barberton and Monteville layers. That the Barberton succession is more deformed and metamorphosed than the Monteville Formation may help explain the observed geochemical differences. Whatever the explanation, the data presented here clearly show that each Precambrian spherule layer is unique and needs to be characterized individually, as is true for the impact spherule layers of the Phanerozoic.

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