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Osmium isotopes fingerprint mantle controls on the genesis of an epithermal gold province

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ABSTRACT

The formation of crustal blocks enriched with gold (Au) deposits above subduction zones is intimately bound to the genesis and evolution of magmatic-hydrothermal systems. A long-standing question, however, is whether the metal fertility of these systems stems from distinct sources that are anomalously enriched in Au or from subsequent processes occurring during crustal magma emplacement and hydrothermal activity. The Deseado Massif auriferous province in southern Patagonia (Argentina) is a unique place to test these contrasting hypotheses because Au-bearing mantle xenoliths indicate the presence of an underlying Au-rich lithospheric mantle reservoir. However, direct geochemical links between the Au-rich mantle source and the formation of the Deseado Massif auriferous province in the overlying crust remain to be established. To address this prominent gap in knowledge, we used sulfide Re-Os geochronology to identify the source of Au at Cerro Vanguardia, the largest low-sulfidation epithermal Au-Ag deposit in the Deseado Massif. Pyrite from high-grade Au quartz veins yielded an isochron age of 147.4 ± 2.9 Ma (mean square of weighted deviates = 1.04, n = 8) and an initial $^{187}Os/^{188}Os$ ratio of 0.26 ± 0.01, fingerprinting a dominant mantle control for the source of Os and, by inference, the source of Au. Our data provide a unique geochemical linkage between an Au-rich subcontinental lithospheric mantle source and the genesis of epithermal Au deposits, supporting the hypothesis that pre-enriched mantle domains may be a critical factor underpinning the global-scale localization of Au provinces.

INTRODUCTION

Subduction zones are key sites on Earth where some of the largest concentrations of gold (Au) are found (Sillitoe, 2008). The formation of these Au-rich crustal regions is bound to the onset of highly efficient hydrothermal processes driven by arc- and back-arc–related magmas in the shallow crust (Hedenquist and Lowenstern, 1994). However, large auriferous crustal domains are heterogeneously distributed in space and time, and they form in response to specific tectonomagmatic events, indicating that not all magmatic systems develop the same Au fertility (Sillitoe, 2008). A widely proposed hypothesis posits that significant Au enrichment in Earth's crust is controlled by the distribution of Au-rich sources in the deep lithosphere (e.g., McInnes et al., 1999; Richards, 2009; Muntean et al., 2011; Griffin et al., 2013; Tassara et al., 2017; Holwell et al., 2019; Wang et al., 2020; Schettino et al., 2022). This view holds that pre-existing Au enrichment in source reservoirs such as lower-crustal cumulates or the subcontinental lithospheric mantle (SCLM) may be a key factor underpinning upper-crustal Au enrichment. However, evidence for a genetic link between enriched sources at depth and the formation of overlying Au-rich provinces remains elusive.

The Deseado Massif in southern Patagonia, Argentina (Fig. 1), is an auriferous province (~28.2 million ounces [Moz] in Au equivalent; Guido and Jovic, 2019) that hosts 13 producing, and over 40 nonproducing, low-sulfidation epithermal gold-silver (Au-Ag) deposits formed during the Middle to Late Jurassic (Schalamuk et al., 1997). Subsequent Neogene volcanism brought to the surface peridotite xenoliths that represent the SCLM beneath the Deseado Massif Auriferous Province (DMAP), providing an exceptional opportunity to explore the deep lithosphere directly beneath an Au-rich province. These xenoliths revealed a plume-related metasomatic assemblage that includes native Au particles and sulfides with Au/Ag ratios identical to that of the overlying epithermal deposits, indicating that Au enrichment in the SCLM that may have acted as a major source of Au for the development of the DMAP (Tassara et al., 2017). However, further testing of this hypothesis requires complementary constraints on the source of Au in the epithermal deposits.

The rhenium-osmium (Re-Os) isotopic system is a unique geochemical tool for tracing the source of ore metals in magmatic-hydrothermal systems (e.g., Mathur et al., 2000; Morelli et al., 2007; Saintilan et al., 2021). Whereas Os is a compatible element during partial melting of the mantle, Re behaves as a moderately incompatible element (Mallmann and O'Neill, 2007; Mungall and Brenan, 2014). The time-integrated

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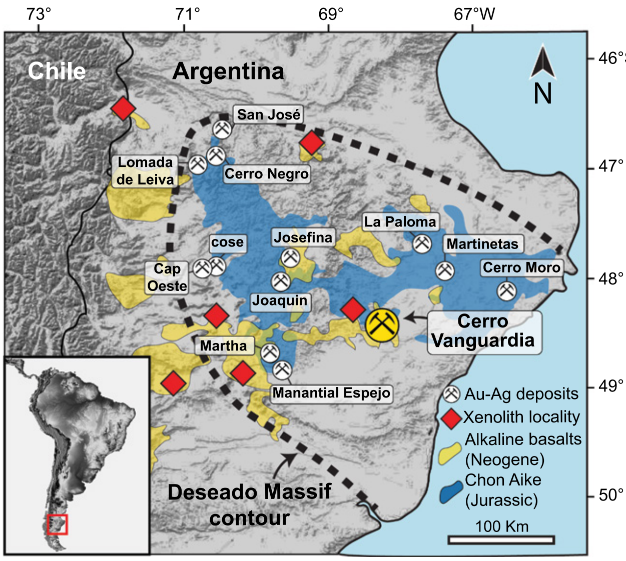

Figure 1. Geologic map of southern Patagonia (Argentina), showing locations of Au-Ag epithermal deposits in the Deseado Massif (labeled) and associated Jurassic volcanism. Mantle xenoliths localities are also shown for reference, along with accompanying Neogene alkaline basalts. Map is modified after Tassara et al. (2017).

result of this contrasting behavior leads to high Re/Os ratios and radiogenic Os isotope compositions in the present-day upper continental crust ($^{187}Os/^{188}Os$ ~1.4; Peucker-Ehrenbrink and Jahn, 2001; Chen et al., 2016) compared to the primitive upper mantle ($^{187}Os/^{188}Os$ ~0.1296; Meisel et al., 2001). Therefore, and because of its highly siderophile and chalcophile nature, the isotopic composition of Os in magmatic-hydrothermal ore systems can readily distinguish between mantle and crustal sources of metals of similar geochemical affinity such as Au (e.g., McInnes et al., 1999; Mathur et al., 2000, 2003; Saintilan et al., 2021).

We used Re-Os geochronology to constrain the source of Au in the DMAP. We focused on the world-class Cerro Vanguardia low-sulfidation epithermal Au-Ag deposit (Fig. 1), the largest mineralized system in the DMAP (~11 Moz Au equivalent; Guido and Jovic, 2019). The purpose of our study was to evaluate the relative contributions of the mantle and crust as the source of gold.

GEOLOGIC BACKGROUND AND SAMPLES

The formation of the DMAP was linked to the development of the Jurassic Chon Aike silicic large igneous province (CA-SLIP; Fig. 1; Pankhurst et al., 1998; Guido and Campbell, 2011). The CA-SLIP formed under an extensional regime in a back-arc position influenced by a subduction zone to the west and the thermal impact of the Karoo mantle plume to the east during the initial stages of the breakup of Gondwana (Navarrete et al., 2019). It is composed of ca. 187 to ca. 144 Ma, medium- to high-K, calc-alkaline basaltic andesites and extensive rhyolites (Pankhurst et al., 1998). The late-stage calc-alkaline intermediate to silicic units of the CA-SLIP constitute the parental magmas of the Deseado Massif Au-Ag deposits (Schalamuk et al., 1997).

The ore bodies at Cerro Vanguardia cover an area over ~350 km^2 and represent one of the most extensive epithermal systems in the world (Schalamuk et al., 1997). The Au-Ag mineralization is hosted by numerous steep-dipping quartz veins that crosscut rhyolitic ignimbrites and volcaniclastic rocks of the Chon Aike Formation. The sample material for this study consists of ~7.5 kg of pyrite-bearing high-grade quartz vein retrieved from underground (32 g t^{-1} Au and 663 g t^{-1} Ag; 120 m below ground level; 48°27′30″S, 68°15′30″W). The sample is dominated by quartz and adularia with abundant pyrite, and sphalerite and galena intergrowths. Gold is typically found as electrum particles included within anhedral pyrite grains (Figs. 2A–2D; Fig. S1 in the Supplemental Material[1]). The electrum-bearing pyrite grains are distributed along Ginguro bands, which are black to dark-gray ore layers found in banded quartz veins from low-sulfidation Au-Ag deposits.

RHENIUM-OSMIUM DATA

Re-Os isotope analyses of eight pyrite samples were conducted by isotope dilution–negative–thermal ionization mass spectrometry (ID-N-TIMS) at the Geochemistry and Geochronology Center, Yale University (USA). Details of sample preparation and analytical procedures are available in the Supplemental Material. Re and Os concentrations in pyrite ranged from 0.25 to 0.56 ng g^{-1} and from 5.7 to 21.2 pg g^{-1}, respectively (Table S1). The Re-Os isotope data yielded an eight-point isochron date of 147.4 ± 2.9 Ma and an initial ($^{187}Os/^{188}Os$)$_i$ ratio, Os$_i$, of 0.26 ± 0.01, calculated using Isoplot version 4.15 (Ludwig, 2011; Fig. 2E). The obtained Re-Os date is consistent with previous K-Ar radiometric ages obtained for the sericitic-argillic alteration zone of the Cerro Vanguardia deposit (152–151 Ma ± 3.5 Ma; Arribas et al., 1996) and biotite from nearby silicic units of the

[1]Supplemental Material. Methods, Figure S1, and Table S1. Please visit https://doi.org/10.1130/GEOL.S.20466240 to access the supplemental material, and contact editing@geosociety.org with any questions.

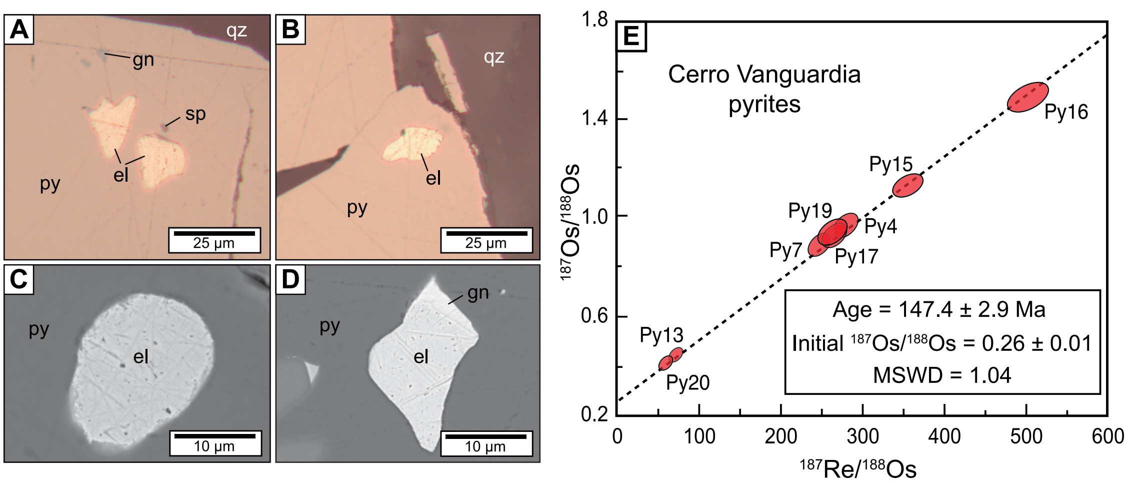

Figure 2. (A–D) Reflected light photomicrographs (A,B) and backscattered electron images (C,D) showing electrum grains included within pyrite. (E) Re-Os isochron plot for Cerro Vanguardia (Au-Ag deposit in the Deseado Massif, southern Patagonia) pyrite samples. MSWD—mean square of weighted deviates, qz—quartz, gn—galena, sp—sphalerite, el—electrum, py—pyrite.

Chon Aike Formation (153.2–148.8 ± 3.6 Ma; Arribas et al., 1996).

DISCUSSION

The bulk $^{187}Os/^{188}Os$ composition of the upper continental crust at ca. 147 Ma is estimated to have been ~0.82 ± 0.41 to ~1.30 ± 0.35, with significant local isotopic heterogeneities (~0.031 to ~0.059 ng g^{-1} Os; Peucker-Ehrenbrink and Jahn, 2001; Chen et al., 2016). In contrast, the $^{187}Os/^{188}Os$ ratio of the primitive upper mantle is estimated to have been ~0.1286 at 147 Ma (~2.6 ± 1 ng g^{-1} Os; Meisel et al., 2001). However, enriched mantle sources may display slightly more radiogenic signatures. For instance, oceanic-island basalts have $^{187}Os/^{188}Os$ <0.175 (Day, 2013), primitive arc basalts have $^{187}Os/^{188}Os$ <0.319 (Alves et al., 2002), and metasomatized SCLM xenoliths can have $^{187}Os/^{188}Os$ ratios of up to ~0.35 (Carlson et al., 2005). Currently available data on the metasomatized xenoliths from the SCLM beneath the Deseado Massif indicate bulk-rock $^{187}Os/^{188}Os$ ratios of up to ~0.21 at 147 Ma (Fig. 3; Tassara et al., 2018). The Os_i for the Cerro Vanguardia pyrites (0.26 ± 0.01) is only slightly more radiogenic and is similar to values for the upper mantle (Figs. 2E and 3). This $^{187}Os/^{188}Os$ composition is comparable to the lower end of values reported for porphyry and high-sulfidation epithermal Au deposits elsewhere (Fig. 3). Furthermore, the obtained Os_i is close to the upper end of values for the underlying SCLM and falls within the range reported for arc basalts (Fig. 3). Because crustal rocks typically have elevated Os isotope signatures ($^{187}Os/^{188}Os$ >> 0.4), (1) the low Os_i in Cerro Vanguardia pyrites suggests little contribution of ore metals from radiogenic crustal sources, and (2) we can assume that the obtained Os_i represents a maximum value for the rocks that sourced metals to the DMAP.

The traditional view for the origin of intermediate to silicic magmas related to the low-sulfidation Au-Ag deposits of the DMAP holds that they formed by partial melting of a 1.6–1.16 Ga hydrous mafic lower crust triggered by the thermal impact of the Karoo mantle plume. These melts subsequently underwent significant assimilation and fractional crystallization (AFC; Pankhurst and Rapela, 1995; Pankhurst et al., 1998). This view implies that the Au budget of the DMAP was largely sourced from the crust. An alternative hypothesis states that voluminous mantle-derived melts related to the Karoo mantle plume mixed with crustal components in a deep crustal hot zone in a back-arc position. This melting led to the formation of the calc-alkaline silicic magmas parental to the epithermal deposits after AFC (Riley et al., 2001). The latter interpretation would imply a significant mantle-derived source for the Au budget of the DMAP.

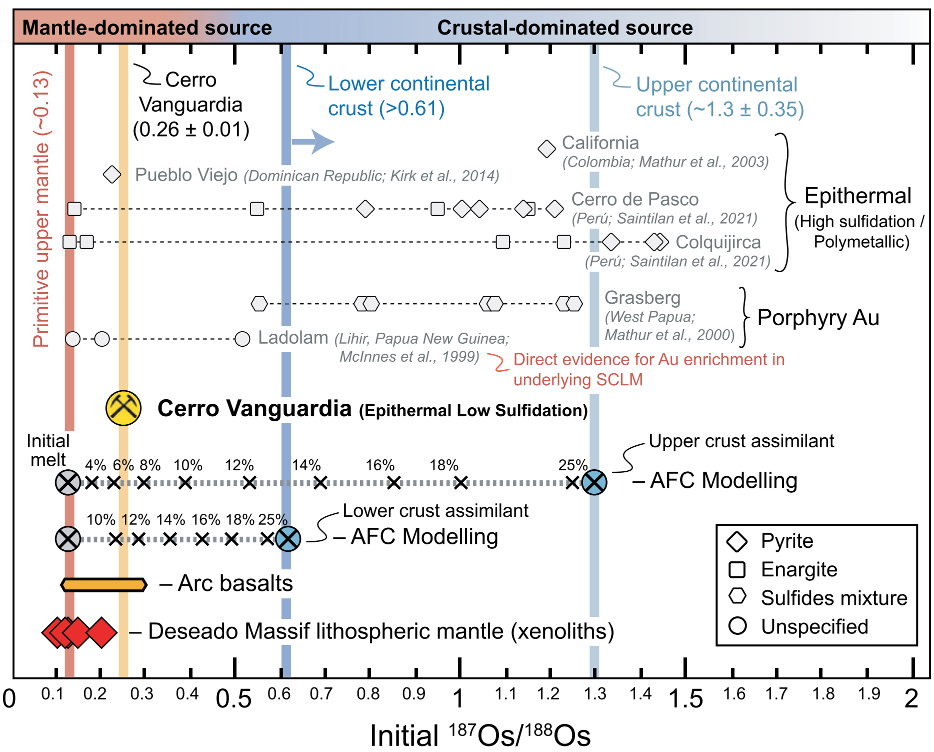

Figure 3. Initial $^{187}Os/^{188}Os$ composition of Cerro Vanguardia (Au-Ag deposit in the Deseado Massif, southern Patagonia) pyrite samples compared to that of lithospheric mantle xenoliths from the Deseado Massif (Tassara et al., 2018), arc basalts worldwide (Alves et al., 2002), and Os_i composition of selected Au deposits (McInnes et al., 1999; Mathur et al., 2000, 2003; Kirk et al., 2014; Saintilan et al., 2021). Primitive upper mantle is after Meisel et al. (2001). Upper continental crust is after Peucker-Ehrenbrink and Jahn (2001). Also shown is an assimilation fractional crystallization (AFC) model following DePaolo (1981). SCLM—subcontinental lithospheric mantle.

The Os_i value from pyrites at Cerro Vanguardia (0.26 ± 0.01) allows us to test these contrasting hypotheses for the source of Au in the DMAP. Given a mean 1.35 Ga age for a hydrous mafic lower crust in the Deseado Massif (based on Nd model ages; Pankhurst and Rapela, 1995), a mantle-like Os_i composition (0.13), and a conservative $^{187}Re/^{188}Os$ ratio of 24 for primitive basalts from Patagonia (Alves et al., 2002), we calculated a $^{187}Os/^{188}Os$ ratio of ~0.61 for the lower crust at the time of Au mineralization (ca. 147 Ma). Therefore, melting of this lower-crustal reservoir would have conveyed a minimum Os_i value of 0.61 to the resulting magmas and the genetically associated epithermal deposits. In reality, this Os_i should have been even higher due to the effects of postmelting AFC and because the chosen $^{187}Re/^{188}Os$ ratio could have been significantly higher, considering values in arc basalts globally (range 24–5796, mean ~797; Alves et al., 2002). Consequently, the Os_i of 0.26 ± 0.01 at Cerro Vanguardia does not support a lower-crustal origin for the source of Os—and by inference Au—to the epithermal deposits. Instead, the low Os_i value indicates the strong input of an unradiogenic Os component (Fig. 3). This scenario is consistent with direct evidence from mantle xenoliths, which record the imprint of Au-rich plume-related metasomatic melts fertilizing the SCLM beneath the Deseado Massif (Tassara et al., 2017).

Because Os behaves compatibly during magmatic differentiation, the $^{187}Os/^{188}Os$ composition of evolving magmas is particularly sensitive to the effects of AFC (Saal et al., 1998). This is illustrated in Figure 3, where the effect of AFC on the Os isotope composition of a differentiating magma was modeled using the equations of DePaolo (1981) for a parental magma with a primitive upper mantle–like Os_i of 0.13 and 0.1 ng g^{-1} Os, and an assimilant with an upper continental crust $^{187}Os/^{188}Os$ ratio of 1.3 and 0.041 ng g^{-1} Os. The mass of assimilated rocks over the mass of crystallized material (*R*) was fixed at 0.6, and the crystal-melt Os partitioning coefficient was 10. In this scenario, only ~7% of AFC is required to increase the mantle $^{187}Os/^{188}Os$ ratio to the values at Cerro Vanguardia (Fig. 3). Consideration of the assimilation of a juvenile basement did not significantly change these results. For instance, assimilation of lower-crustal Patagonian rocks with an estimated minimum $^{187}Os/^{188}Os$ of 0.61 would shift the required AFC degree only up to ~11% (Fig. 3). Thus, the low Os_i of pyrite at Cerro Vanguardia fingerprints a dominant mantle source for Os and, by inference, for Au. The Os isotope data alone cannot discern between a pristine mantle or a

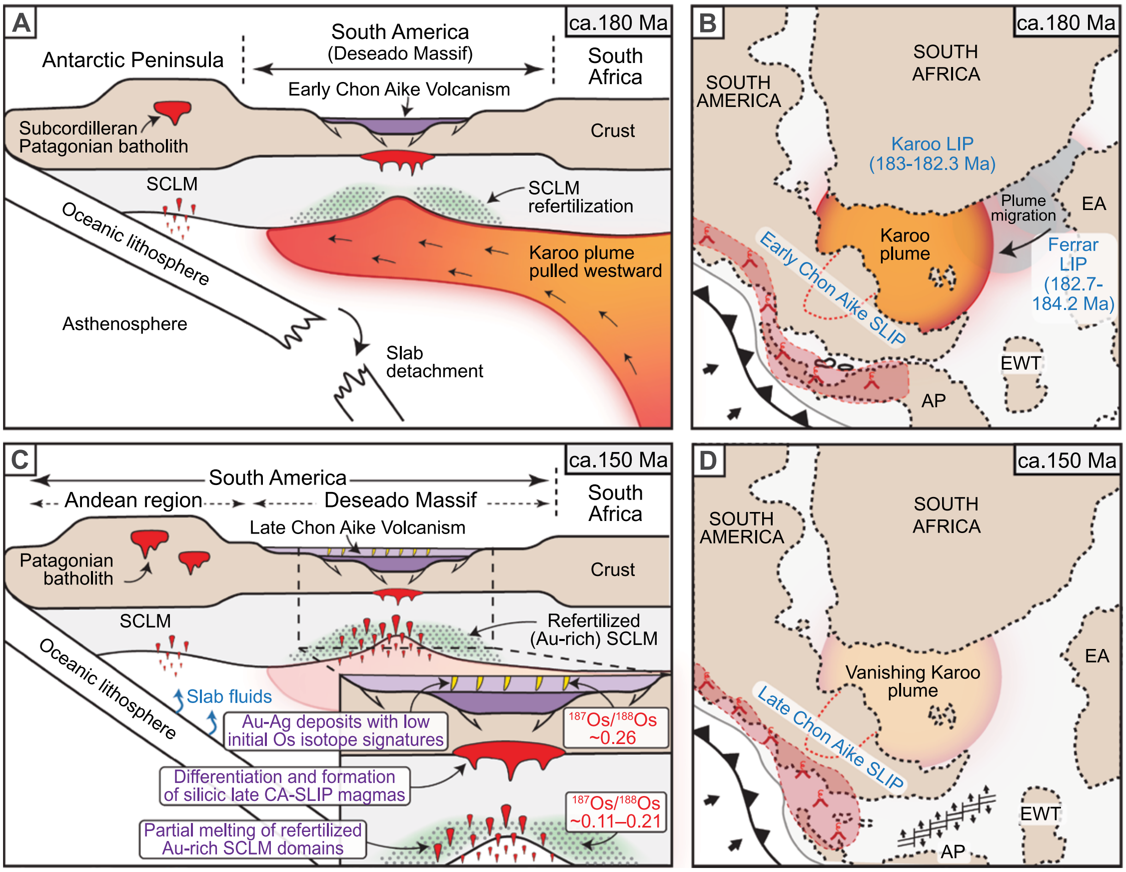

Figure 4. Geologic model and paleogeographic configuration during the formation of the Deseado Massif auriferous province (southern Patagonia). (A,B) Slab detachment during the Early Jurassic resulted in migration of the Karoo plume below the Deseado Massif, which caused early stages of the Chon Aike silicic large igneous province (CA-SLIP) emplacement (coeval with the Karoo and Ferrar large igneous provinces [LIPs]), and formation of Au-rich subcontinental lithospheric mantle (SCLM) reservoirs after plume-related metasomatism. (C,D) After protracted plume activity underneath the Deseado Massif, the late stages of CA-SLIP incorporated partial melts of metasomatized Au-rich SCLM reservoirs, increasing their ore-forming potential. AP—Antarctic Peninsula, EWT—Ellsworth-Whitmore terrane; EA—East Antarctica.

metasomatized SCLM source. However, we favor involvement from the latter because it is consistent with previous independent evidence for an Au-rich SCLM source beneath the DMAP (Tassara et al., 2017).

SUMMARY AND METALLOGENIC MODEL FOR THE DESEADO MASSIF

Pyrite grains from the Cerro Vanguardia Au-Ag deposit, the largest deposit in the DMAP, yielded a Re-Os isochron age of 147.4 ± 2.9 Ma (mean square of weighted deviates = 1.04, $n = 8$). The isochron Os_i (~0.26 ± 0.01) indicates a strong mantle component in the source of Os that we interpret, by inference, to fingerprint the source of Au. Our data are consistent with previous direct evidence from peridotite xenoliths revealing the presence of Au-rich SCLM domains as a source of Au in the DMAP (Tassara et al., 2017). These SCLM reservoirs were interpreted to have formed after plume-related metasomatism caused by the southwestward pull of the Karoo plume underneath the DMAP in response to a slab detachment episode during the early stages of Gondwana breakup in the Early Jurassic (Fig. 4A; Tassara et al., 2017; Navarrete et al., 2019). We suggest that after 20–30 Ma of plume activity beneath the Deseado Massif and coeval subduction on the western margin of Gondwana, incorporation of partial melts of Au-rich SCLM domains by ascending calc-alkaline magmas increased the Au fertility during the late stages of the Jurassic CA-SLIP, ultimately producing the DMAP (Fig. 4B). The similarity between the Os_i of Cerro Vanguardia pyrites and that of peridotite xenoliths that represent the underlying SCLM is a key test for this hypothesis (Fig. 3). As such, the Os_i of pyrite hosting gold at Cerro Vanguardia provides direct evidence for a link between a metal-rich SCLM domain and the formation of Au deposits in the overlying crust, supporting the hypothesis that Au enrichment of the mantle may influence the global-scale distribution of auriferous provinces.

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