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Geochemical Perspectives Letters

^{190}Pt-^{186}Os geochronometer reveals open system behaviour of ^{190}Pt-^{4}He isotope system

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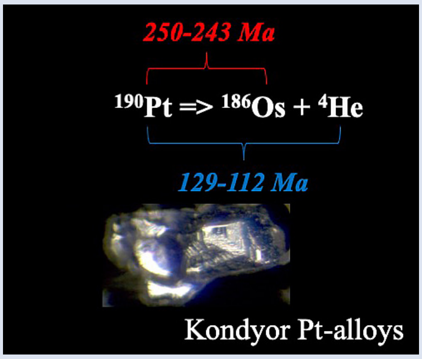
Abstract



Platinum Group Minerals are typically dated using the ^{187}Re-^{187}Os and ^{190}Pt-^{186}Os isotope systems and more recently using the ^{190}Pt-^{4}He geochronometer. The ^{187}Re-^{187}Os and ^{190}Pt-^{186}Os compositions of Pt-alloys from the Kondyor Zoned Ultramafic Complex (ZUC) analysed here reveal overprinting for both geochronometers except in one alloy exhibiting the most unradiogenic ^{187}Os/^{188}Os and most radiogenic ^{186}Os/^{188}Os signatures. These signatures argue for an Early Triassic mineralisation, when silicate melts/fluids derived from the partial melting of an Archean mantle crystallised to form the Kondyor ZUC while the ^{190}Pt-^{4}He chronometer supports an Early Cretaceous mineralisation. We propose that Kondyor ZUC represents the root of an alkaline picritic volcano that constitutes the remnants of an Early Triassic island arc formed during the subduction of the Mongol-Okhotsk ocean seafloor under the Siberia craton. After the Early Cretaceous collision of Siberia with the Mongolia-North China continent, the exhumation of deep-seated structures - such as the Kondyor ZUC - allowed these massifs to cool down below the closure temperatures of the Pt-He and K-Ar, Rb-Sr isotope systems, explaining their Early to Late Cretaceous ages for the Kondyor ZUC.

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Introduction

Platinum group minerals (PGM, *e.g.,* Os-alloys, Pt-alloys, Pt-arsenides) are critical host phases of the Highly Siderophile Elements (HSE; Os, Ir, Ru, Rh, Pt, Pd, Re) in the Earth's mantle and crust. They are typically dated with the ^{187}Re-^{187}Os and/or ^{190}Pt-^{186}Os isotope systems (*e.g.,* Walker *et al.*, 1997; Meibom and Frei, 2002; Pearson *et al.*, 2007; Coggon *et al.*, 2012).

Recently, the ^{190}Pt-^{4}He isotopic system has emerged as an alternative geochronometer for Pt-rich PGM. The ^{190}Pt-^{4}He and ^{190}Pt-^{186}Os geochronometers are both measuring the alpha decay of ^{190}Pt, with the only difference being that one measures the accumulation of the daughter product ^{186}Os and the other the accumulation of the decay particle ^{4}He. The Pt-He geochronometer was so far used to date the Pt-alloys from the Kondyor Zoned Ultramafic Complex (ZUC), which is located in the Aldan Shield on the South-East margin of the Siberian Craton (Fig. S-1 and Supplementary Information) (Shukolyukov *et al.*, 2012a; Mochalov *et al.*, 2016, 2018). The Early Cretaceous Pt-He isochron ages (112 ± 7 Ma and 129 ± 6 Ma, calculated using a ^{190}Pt half-life of 469 Gyr: Begemann *et al.*, 2001) agree well with the Rb-Sr, Sm-Nd and K-Ar ages obtained on the main lithologies (whole rock and mineral phases) but conflict with the Re-Os T_{RD} model ages obtained on erlichmanite (OsS_2), sperrylite ($PtAs_2$), Os-alloys and Pt-alloys (Cabri *et al.*, 1998; Malitch and Thalhammer, 2002) that vary from Neoproterozoic (658-603 Ma) to future ages, when back calculated to the present-day primitive mantle (PM) ^{187}Os/^{188}Os estimate (Meisel *et al.*, 2001).

The combination of multiple isotope systems for dating single mineral phases offers the opportunity to resolve "open system behaviour" and to assess which isotopic signatures provide geologically meaningful information on the age and origin of minerals. Here we report the coupled ^{190}Pt-^{186}Os and ^{187}Re-^{187}Os signatures obtained by Laser Ablation Multi Collector Inductively Coupled Plasma Mass Spectrometry (LA-MC-ICPMS) (Supplementary Information) on 13 sub-millimetric Pt-alloys separated from a chromitite schlieren (sample 1265; Pushkarev *et al.*, 2015) hosted in the dunitic core of the Kondyor ZUC. Our Pt-alloys are a different subset from those investigated for the ^{190}Pt-^{4}He isotope system. Shukolyukov *et al.* (2012a) and Mochalov *et al.* (2016, 2018) dated (i) Pt-alloys from different lithologies of the Kondyor ZUC, including the chromitite lenses of the dunitic core and

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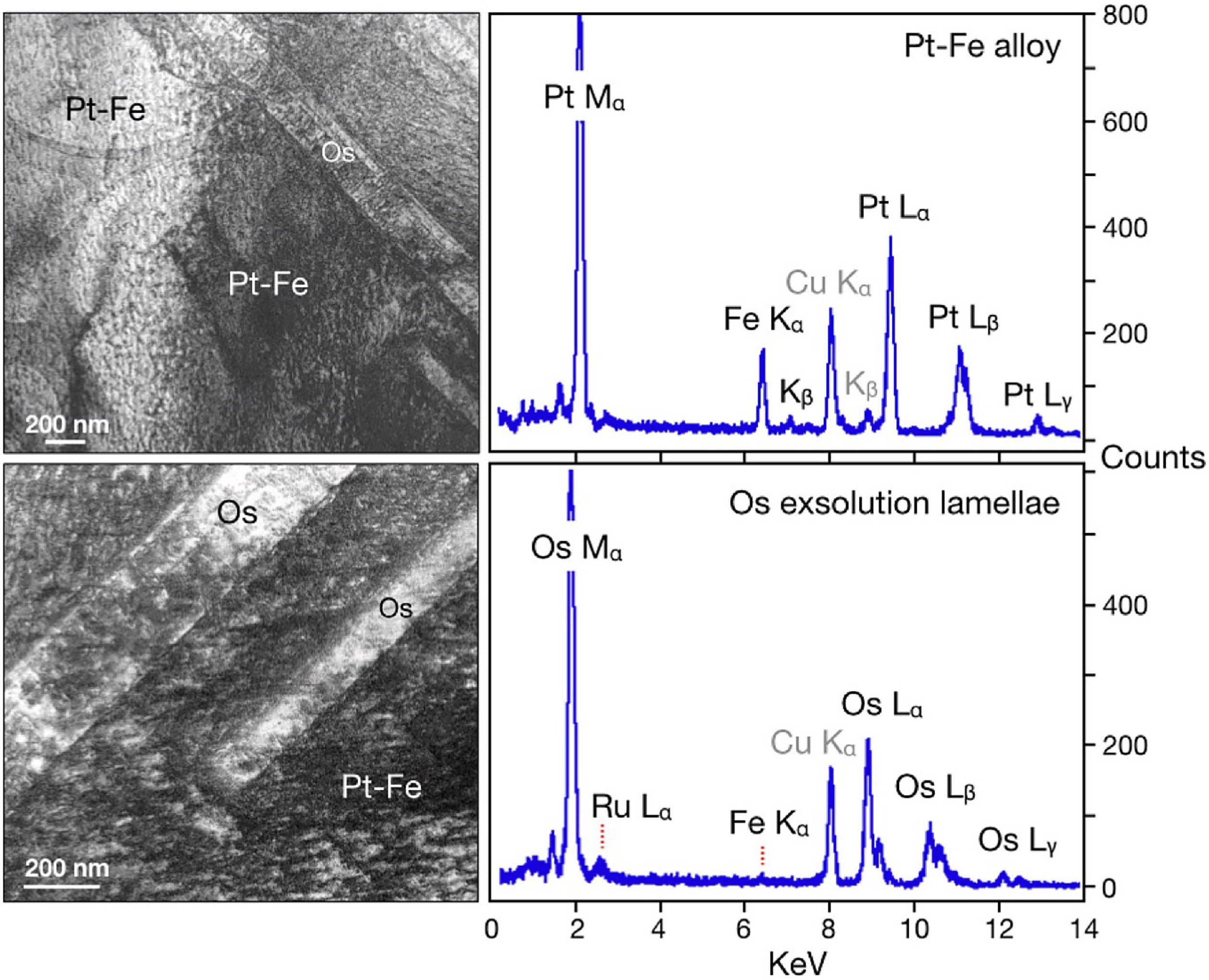

Figure 1 Bright field image and EDS spectra of Kondyor Pt-Fe alloys and their pure Os exsolution lamellae (FIB-TEM image, GFZ, Potsdam, Germany). The Cu peaks on the EDS spectra are due to the Cu grid that carries the FIB section.

(ii) alluvial Pt-Pd PGM. The FIB-TEM investigations on a few of our Pt-alloys revealed a very complex nanoscale exsolution pattern consisting of spinodal exsolutions of Pt-Fe alloys (*e.g.*, Pt_3Fe, PtFe) and pure Os exsolution lamellae (Fig. 1).

Results

The Kondyor Pt-alloys display radiogenic $^{186}Os/^{188}Os$ and unradiogenic $^{187}Os/^{188}Os$ compositions (Fig. 2 a,b). The most radiogenic $^{187}Os/^{188}Os$ signatures (0.1246; alloys L-S2 and E-S2, Table S-2) agree well with those previously obtained on five Kondyor Os-rich alloys (0.1248-0.1252; Malitch and Thalhammer, 2002). Conversely, the least radiogenic $^{187}Os/^{188}Os$ (0.110096 ± 2136; alloy D-S2) is close to the composition of Re-free, least metasomatised peridotite xenoliths of the Tok locality (0.109; estimated for $Al_2O_3 = 0$ wt. % on the $^{187}Os/^{188}Os$ *vs.* Al_2O_3 "aluminochron"; Ionov *et al.*, 2006), which like the Kondyor ZUC is located in the East Aldan Shield (Fig. S-1). Overall, the $^{187}Os/^{188}Os$ compositions are decoupled from the $^{187}Re/^{188}Os$ ratios (Fig. 2a). In contrast, the $^{186}Os/^{188}Os$ compositions define a positive trend with $^{190}Pt/^{188}Os$, which - if considered to represent an isochronous relationship - yields an age of 249.8 ± 12 Ma (Fig. 2b). The $^{187}Os/^{188}Os$ and $^{186}Os/^{188}Os$ signatures are negatively correlated despite the sympathetic variation of both parent/daughter elemental ratios (Fig. 2c).

Robustness of the Re-Os and Pt-Os Isotope Systematics

The decoupling of the $^{187}Os/^{188}Os$ from both $^{187}Re/^{188}Os$ and $^{186}Os/^{188}Os$ signatures demonstrate the open system behaviour of the Re-Os isotope system in the Kondyor Pt-alloys. This is best explained by the overprinting of the Os-poor, least radiogenic $^{187}Os/^{188}Os$ of the Pt-alloy D-S2 by an Os-rich (*ca.* 700 times richer) contaminant with a $^{187}Os/^{188}Os$ of 0.1246 (Fig. 3a), similar to the most radiogenic $^{187}Os/^{188}Os$ of our Kondyor alloys (*e.g.*, points E-S2) and very close to the least radiogenic $^{187}Os/^{188}Os$ compositions previously reported by Malitch and Thalhammer (2002) and Cabri *et al.* (1998) for Kondyor PGM (Fig. 2a). Both the $^{186}Os/^{188}Os$ *vs.* $^{187}Os/^{188}Os$ and $^{186}Os/^{188}Os$ *vs.* 1/Os relationships (Fig. 3b) can be reproduced with such a mixing scenario. Importantly, the negative $^{187}Os/^{188}Os$ *vs.* $^{187}Re/^{188}Os$ and the relationships between the $^{187}Os/^{188}Os$ and the abundance of Os exsolution lamellae (monitored by the ^{188}Os signal) in the Pt-alloys likely suggest that this mixing scenario reflects a gradual overprinting of the mantle source of the Kondyor mineralisation by subduction-related fluids (Supplementary Information).

The Pt-alloy D-S2 is then the least overprinted of our Kondyor subset (Fig. 3a,b). This view is further supported by the closeness of its $^{187}Os/^{188}Os$ and $^{187}Re/^{188}Os$ ratios (0.001196 and 0.00541; Table S-2) to those of the Re-free, least metasomatised Tok peridotite xenoliths (0.109 and 0; Ionov *et al.*, 2006),

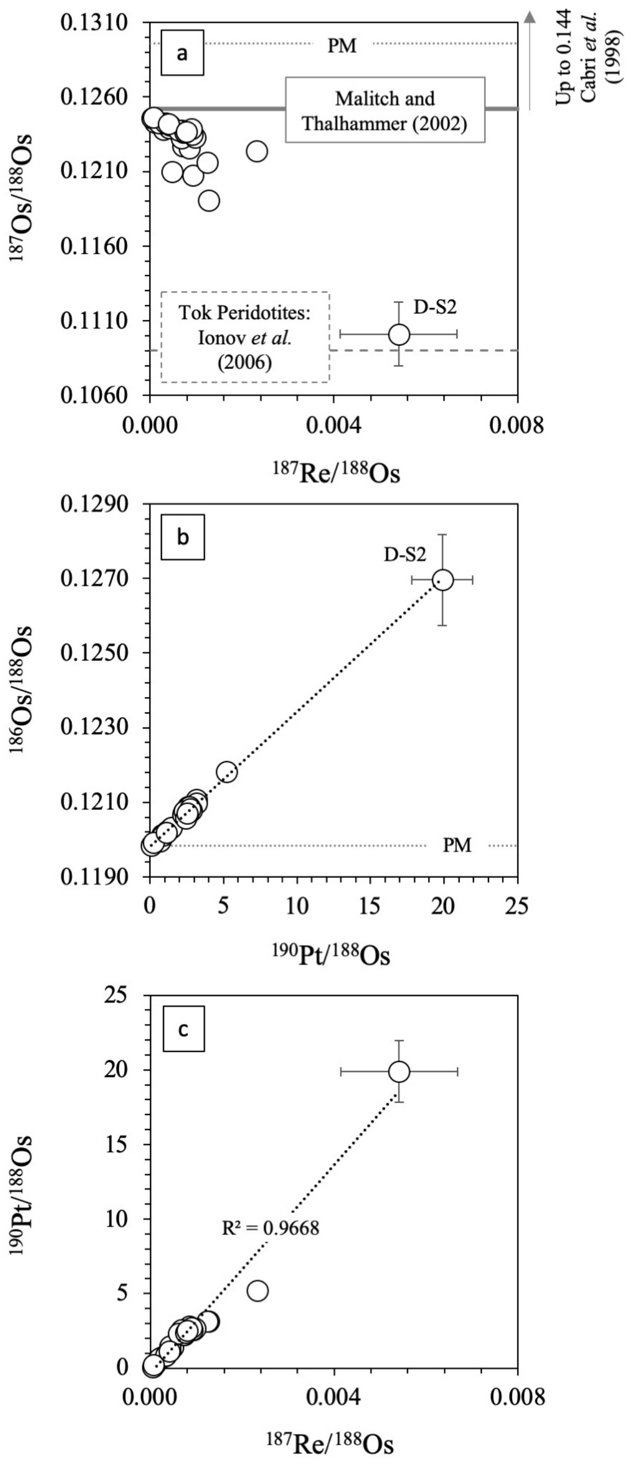

Figure 2 **(a)** Variations of $^{187}Os/^{188}Os$ *vs.* $^{187}Re/^{188}Os$, **(b)** of $^{186}Os/^{188}Os$ *vs.* $^{190}Pt/^{188}Os$ and **(c)** of $^{190}Os/^{188}Os$ *vs.* $^{187}Re/^{188}Os$. The primitive mantle (PM) $^{186}Os/^{188}Os$ and $^{187}Os/^{188}Os$ values are respectively from Day *et al.* (2017) and Meisel *et al.* (2001). If the positive correlation between $^{186}Os/^{188}Os$ *vs.* $^{190}Pt/^{188}Os$ is considered to be an isochronous relationship, it yields an age of 249.8 ± 12 Ma and an intercept of 0.119821 ± 0.000024 (2 sigma) (MSWD = 0.81).

implying that the $^{187}Os/^{188}Os$ composition of alloy D-S2 may still hold geologically meaningful constraints. Its Re-Os T_{RD} model age points at a 2630 Ma old PUM-like mantle source for the Kondyor Pt-mineralisation (the Re-Os T_{MA} model age is 2664 Ma). Occurrence of Archean mantle underlying the Aldan Shield is also supported by the T_{RD} model ages of the Tok peridotites (2770 Ma) and by Pb-Pb isotope systematics of the Mesozoic lamproitic magmatism (~3 Ga; Davies *et al.*, 2006). Considering that the present-day PM has a $^{186}Os/^{188}Os$ of 0.1198388 and a $^{190}Pt/^{186}Os$ of 0.0022 (Day *et al.*, 2017), the 2630 Ma PUM-like mantle source of the Kondyor Pt-mineralisation then had a maximum $^{186}Os/^{188}Os$ of 0.1198303. If we consider such an initial $^{186}Os/^{188}Os$ composition, the D-S2 Pt-alloy would require 242.6 Myr to evolve to its present day $^{186}Os/^{188}Os$ signature. This age is similar within error to that extrapolated from the multi-grain Pt-Os isochron-like trend defined by our Kondyor Pt-alloys (249.8 ± 12 Ma; Fig. 2b).

Ages of ~250-240 Ma are recognised regionally within the Aldan Shield (Lena and Aldan (Palaeo) Rivers: Wang *et al.*, 2011; Miller *et al.*, 2013), the Baikal Lake Region (*e.g.*, Gladkochub *et al.*, 2010) and within basins (*e.g.*, Onon and Mohe-Upper Amur), located South of the Aldan Shield and adjacent to the Mongol-Okhotsk Fold belt (Guo *et al.*, 2017). The Mongol-Okhotsk fold belt (Fig. S-1), which rims the Siberian Craton on its South Margin over *ca.* 3000 km, represents the suture zone left after the closure of the Mongol-Okhotsk Ocean - as its seafloor was subducted under the Siberia craton and under the Mongolia-North China continent (Amur plate) -, and the subsequent collision of the Siberian craton with the Mongolia-North China continent (*e.g.*, Zorin, 1999; Guo *et al.*, 2017). The age distribution along the Mongol-Okhotsk fold belt demonstrates an eastward zip-like closure of the Mongol-Okhotsk ocean (Zorin, 1999) initiated in the Late Palaeozoic in NE Mongolia (Zhao *et al.*, 2017) and in the Early Triassic in the eastern part of the Mongol-Okhotsk belt, south of Aldan Shield Region (Guo *et al.*, 2017). The age of the subsequent collision between the Mongolia-North China continent and Siberia craton also evolves eastwards from Middle Jurassic to Early Cretaceous (Zorin, 1999).

Why are the ^{190}Pt-^{186}Os and the ^{190}Pt-^{4}He "Ages" of the Kondyor Pt-alloys Different?

Both the ^{190}Pt-^{4}He and ^{190}Pt-^{186}Os isotopic systems are based on the radioactive alpha decay of the ^{190}Pt so they should yield identical ages. However, for the Kondyor Pt-alloys, the Pt-He isochronal ages (Shukolyukov *et al.*, 2012a; Mochalov *et al.*, 2016, 2018) are ~110-140 Myr younger than the Pt-Os ages.

Several lines of evidence suggest that the age inconsistency may reflect an open system behaviour of the Pt-He isotopic system. First, Shukolyukov *et al.* (2012a,b) and Mochalov *et al.* (2016) argued that radiogenic ^{4}He is retained in the structure of native metals as vesicles that are only released upon melting of the native metals (>1000 °C). However, the only ^{4}He thermal desorption experiment conducted on Pt-alloys by Shukolyukov *et al.* (2012a) revealed ^{4}He loss ($[^{4}He] \neq 0$) for temperatures as low as ~700 °C (see Fig. 4 in Shukolyukov *et al.*, 2012a). While the ^{4}He loss appears marginal during their experiment, it will be significant if Pt-alloys reside in the lithospheric mantle (with an equilibration temperature >700 °C) for 10s-100s of Myr. It is thus possible that the ^{4}He is not fully trapped in the structure of the Pt-alloys until the ^{4}He closure temperature in these minerals is attained. One can additionally consider how the nanoscale exsolution patterns within the Kondyor Pt-alloys will affect the ^{4}He loss/retention. The grain boundaries proposed as a preferential sink for ^{4}He (Shukolyukov *et al.*, 2012b) may turn out to be preferential ^{4}He

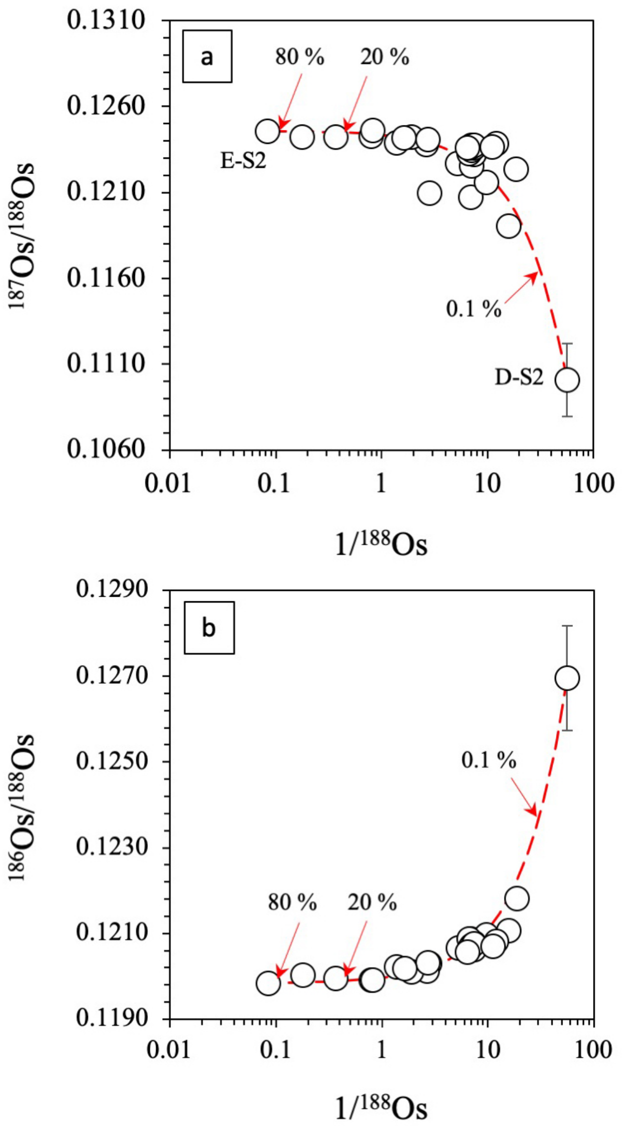

Figure 3 **(a)** $^{186}Os/^{188}Os$ and **(b)** $^{187}Os/^{188}Os$ variations with the Os concentrations ($1/^{188}Os$ beam). Red dotted line represents the overprinting of the most pristine Pt-alloy D-S2 by an Os-rich contaminant characterised by $^{187}Os/^{188}Os$ and $^{186}Os/^{188}Os$ signatures of Pt-alloy E-S2 (0.12457 and 0.119851, respectively).

loss sites when Pt-alloys are intensely exsolved (Fig. 1). The Pt-free nature of the Os exsolution lamellae combined with the extremely Os-poor composition of their Pt-alloy hosts (Fig. 1; Malitch and Thalhammer, 2002; Nekrasov *et al.*, 2005) argues for an equilibration temperature below 500 °C (see Pt-Os phase diagram in Okrugin, 2002), thus well below the 700 °C temperature mark of ^{4}He loss onset observed for Pt alloys (see above). The last evidence suggesting a low closure temperature (<600 °C) of the Pt-He isotopic system comes from the similarity of the Pt-He isochronal ages with the Rb-Sr, Sm-Nd and K-Ar obtained on whole-rock and single minerals (biotite, feldspar) of the dunitic core, pyroxenites and late metasomatic dikes of Kondyor ZUC (149-83 Ma: *e.g.,* Orlova, 1992; Cabri *et al.*, 1998; Pushkarev *et al.*, 2002).

Implication for the Origin and Evolution of the Kondyor ZUC

The combined LA-MC-ICPMS investigation of the Re-Os and Pt-Os isotope signatures demonstrates that the Pt-mineralisation, contemporaneous to the formation of the Kondyor ZUC, originates ~250-240 Myr ago from the melts and fluids produced by partial melting of possibly an Archean PUM-like mantle source, which could be the Siberian cratonic mantle. Considering the orthopyroxene-poor, olivine- and clinopyroxene-rich nature of Kondyor ZUC (Orlova, 1992; Malitch and Thalhammer, 2002) and its extreme Pt-mineralisation, we argue that, rather than being a metasomatised mantle diapir (Burg *et al.*, 2009), Kondyor ZUC represents the root of a ~250-240 Ma old alkaline picritic volcano (Simonov *et al.*, 2011), which together with other Aldan ZUC (*e.g.,* Chad) likely formed part of an Early Triassic island arc at the southeast margin of the Aldan shield due to the subduction of the Mongol-Okhotsk ocean seafloor northwards under the Siberian Craton (see Zorin, 1999; Guo *et al.*, 2017). The uplift associated with the Early Cretaceous collision of the Siberian craton with the Mongolia-North China continent (after the closure of the Mongol-Okhotsk ocean) combined with the subsequent major extensional phase evidenced by the development of Early Cretaceous rift systems may have contributed to the unroofing and exhumation of deep-seated structures such as metamorphic core complexes (Zorin, 1999). In such an unroofing and exhumation scenario, the Kondyor ZUC would attain sub-surface conditions and cool down below the closure temperatures of the K-Ar, Rb-Sr and Pt-He isotope systems, explaining why these geochronometers yield almost exclusively Early to Late Cretaceous ages for the Kondyor ZUC.

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Additional Information

Supplementary Information accompanies this letter at http://www.geochemicalperspectivesletters.org/article1924.

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