Bottom-up mineral exploration: Ore-element upgrading in the upper mantle and tools for its discovery

Christopher J.M. Lawley* and Bruce A. Kjarsgaard

Geological Survey of Canada, 601 Booth Street, Ottawa, Ontario K1A 0E8
*Corresponding author's e-mail: christopher.lawley@canada.ca

ABSTRACT

Mineral exploration targeting typically focuses on the critical pathways and depositional traps of ore systems. The role of pre-enriched source regions on ore deposit genesis is unclear, particularly for systems that involve significant transport of ore components (e.g. Au). Because Au is suggested to be derived from the mid- to lower-crust and/or upper mantle at orogenic deposits, it is not surprising that the deepest and most inaccessible parts of this ore system is also the least studied and understood. Herein we address that knowledge gap by studying the upper mantle directly, using examples of young ophiolitic mantle (Atlin, British Columbia) and ancient cratonic mantle (Superior craton, Ontario; Slave craton, Nunavut). Project results demonstrate two dominant mechanisms for Au upgrading in the lithospheric mantle: (1) base metal sulphide precipitated during melt:rock reactions between peridotitic residues and asthenospheric melts, and (2) hydrous, fluid-inclusions trapped after release from subducting slabs. The remelting of such pre-enriched metasomatized mantle has been previously proposed as a viable source for a variety of ore systems (e.g. mafic to ultramafic rock-hosted platinum-group element deposits). Mapping the architecture of these deep lithospheric metasomatized mantle domains and their association with rising asthenospheric melts is of significant interest for mineral exploration. New geochemical methods and data processing completed as part of the current study, coupled with advances in geophysical imaging, provide additional tools for mapping these pre-enriched lithospheric mantle domains at depth.

INTRODUCTION

Most ore systems are hosted in the crust and represent the focusing and upgrading of ore components (e.g. ore-forming elements and ligands) by fluids or melts along crustal pathways and at their depositional site (Begg et al., 2010; McCuaig et al., 2010; Hronsky et al., 2012; Huston et al., 2016). Multiple physical and/or chemical upgrading processes are required to produce the exceptional concentrations and tonnages of ore at individual deposits (i.e. often 100–10,000x enriched relative to the average continental crust: Boyle, 1979; Fyfe, 1987; Hedenquist and Lowenstern, 1994; Ulrich et al., 1999; Wilkinson et al., 2009; Heinrich and Zurich, 2014). The most important processes for upgrading are specific to each ore system (notwithstanding a number of similar processes across ore systems), but most are thought to occur during transport and/or during deposition (e.g. interaction between hydrothermal fluids and host rocks; crustal sulphur contamination and subsequent sulphide saturation during magmatic differentiation; separation of a magmatic volatile phase: McCuaig et al., 2010; Huston et al., 2016). However, upgrading processes that operate in the source regions of these systems, particularly for ore systems that involve significant transport of ore components (e.g. Au), are less well understood and their impact on metal endowment are more controversial (Richards, 2009; Hronsky et al., 2012; Arndt, 2013; Goldfarb and Groves, 2015).

The conventional view is that the source regions of most ore systems are rather unexceptional, which, if correct, would tend to focus most of the exploration effort on recognizing the critical pathways for ore components and the most favourable trapping environments for ore deposition (Fyfe, 1987; McCuaig et al., 2010; Richards, 2011; Barnes and Fiorentini, 2012; Wilkinson, 2013; Barnes et al., 2016; Huston et al., 2016). The world-class Sudbury Ni-Cu deposits provide perhaps one of the better examples of an ore system that did not involve a particularly Ni-, Cu- and/or PGE-rich source. Because the Sudbury impact structure is so large (≥200 km), the bulk composition of the resulting melt sheet is approximately the same as average and/or lower continental crust (Therriault et al., 2002; Mungall et al., 2004). The transfer and upgrading of ore components from the Ni-poor melt sheet to a Ni-rich sulphide liquid was the primary process responsible for one of the greatest concentrations of base metals on Earth (Therriault et al., 2002; Mungall et al., 2004; Laznicka, 2014). A source enriched in ore components

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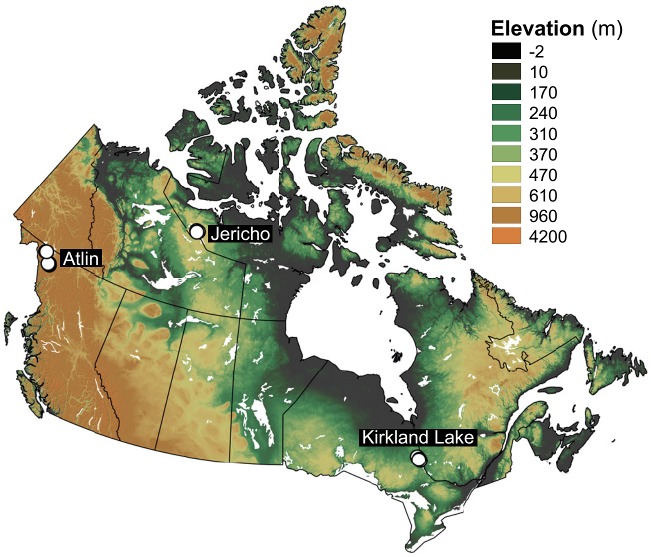

Figure 1. Map of Canada showing research activity locations and samples (white dots). Shuttle radar topography mission (SRTM) elevation data (m) is coloured coded using a percentile scaling.

is therefore not required for ore deposit genesis, at least in some cases.

Kimberlite-hosted diamond deposits, in contrast, require source regions that are pre-enriched in ore components (Richardson et al., 1984; Stachel and Harris, 2008; Griffin et al., 2013). Diamonds are carried to surface by ascending kimberlitic melts that sample diamondiferous lithospheric mantle underlying the continental crust (Fig. 1, 2). Intersecting diamond-bearing mantle lithosphere represents one of the critical controls on the diamond endowment of kimberlite-hosted deposits (Richardson et al., 1984). Accordingly, mineral exploration for diamonds often adopts a bottom-up approach during regional targeting. Advances in deep geophysical imaging of the mantle (e.g. seismic and magnetotelluric methods: Snyder et al., 2004), coupled with detailed petrology studies of kimberlite-derived mantle xenolith and xenocryst mantle cargo (Nimis, 1998; Grütter, 2009), are used to identify favourable lithospheric mantle domains that are prospective for diamond formation and preservation (Morgan, 1995). Crustal pathways (i.e. faults and rock contacts) overlying these fertile mantle source regions are considered more prospective environments for kimberlite ascent and diamond transport (Fig. 2). The bottom-up exploration approach is important because the footprint of fertile lithospheric mantle domains is significantly larger (10s to 100s of km) than the size of an individual kimberlite intrusion (100s of m), which means that the most prospective settings for finding diamond-bearing kimberlites can be prioritized and identified prior to detailed geophysical and surficial sediment discovery surveys and drilling.

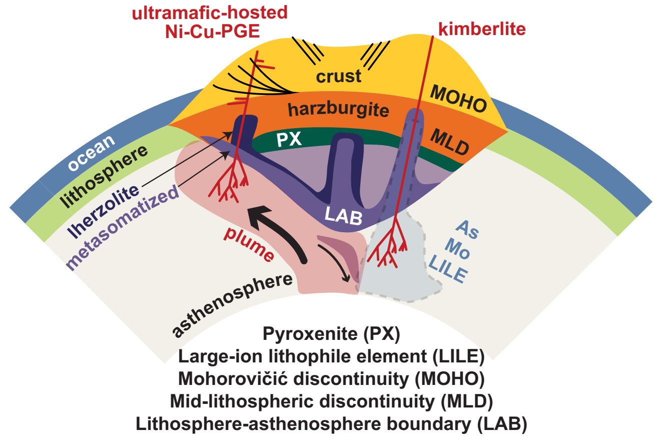

Figure 2. Sketch showing the inferred structure of the cratonic mantle. Fluid- and melt-rock reactions tend to precipitate base metal sulphide within the metasomatized mantle lithosphere. Re-melting of these pre-enriched mantle domains have previously been proposed as favourable source regions for generating ore-forming magmas.

The magmatic precursors of other mafic to ultramafic melts also interact with the lithospheric mantle during accent from their deeper asthenospheric mantle source regions (Fig. 2). Because samples of the lithospheric mantle can be up to 10x enriched in ore components relative to the convecting upper mantle (e.g. containing a few ppb, 10^9, platinum-group elements (PGE): Barnes et al., 2015; Lorand and Luguet, 2016; Saunders et al., 2018), interaction between rising ultramafic melts and refertilized lithospheric mantle provide a possible mechanism to explain the geochemical signature of some ore deposits (Begg et al., 2010; Hronsky et al., 2012; Arndt, 2013; Griffin et al., 2013). The giant Bushveld Complex provides one of the few recognized available examples of a magmatic ore system that owes its precious metal endowment, at least in part, to a pre-enriched lithospheric mantle source (Richardson and Shirey, 2008).

However, the role of a modified lithospheric mantle in the genesis of many other ore systems deposited in the crust remains controversial (McInnes et al., 1999; Richards, 2009; Hronsky et al., 2012; Arndt, 2013). Some of this uncertainty stems from the relatively poor understanding of how the deep mantle roots underlying the continents formed (Pearson et al., 2007; Griffin et al., 2009). The specific mechanisms for upgrading ore components in the lithospheric mantle also remain poorly understood, particularly for ore-forming elements other than PGE. These questions are important to address if bottom-up exploration is also to be applied to other ore-systems (Snyder et al., 2004; Begg et al., 2010; Griffin et al., 2013).

Research completed as part of the Targeted Geoscience Initiative-5 Mantle Metal Mobility activity provides new constraints on the mechanisms that drive upgrading of ore components in the lithospheric mantle and new tools for recognizing where these processes may have occurred in the geological past. Research was focused on Au, but activity results also provided critical new constraints on the behaviour of a highly important suite of ore-forming elements during lithospheric mantle formation, melting, and metasomatism (i.e. Ni, Cu, Zn, As, Se, Mo, Ag, Sn, Sb, Te, W, Pb, and Bi). These results have potential applications to other ore systems (e.g. porphyry Cu, sedimentary-exhalative, volcanogenic massive sulphide, and magmatic Ni-Cu). The present activity was split into separate sub-activities, each studying different lithospheric mantle settings: 1) field- and laboratory-based research on young oceanic mantle as exposed in ophiolite belts from northern British Columbia; and 2) laboratory-based studies of ancient cratonic mantle, which were based on kimberlite-hosted mantle fragments (i.e. xenocrysts and xenoliths) from the Abitibi belt and Slave craton (Fig. 1). Research highlights from both sub-activities are described below.

MANTLE MECHANISMS FOR UPGRADING ORE COMPONENTS

Ophiolite belts preserve tracts of oceanic crust and its underling mantle. The excellent exposure of the upper mantle rocks at the Nahlin ophiolite (Atlin, British Columbia; Fig. 1) provide an opportunity to research the behaviour of ore-forming elements during mantle melting and metasomatism. However, the trace (low ppm) to ultratrace (low ppb) concentration of most elements within unmineralized mantle peridotite poses significant analytical challenges. To address that challenge, a new method was developed as part of the current study to analyze nanopowders by laser ablation - inductively coupled plasma - mass spectrometry (LA-ICP-MS: Lawley et al., 2020). The new wet-milling method was miniaturized to analyze very small sample volumes (1–2 g), which is required for the limited size typical of xenoliths from basaltic and/or kimberlitic volcanism. A research study applying this new approach has been successfully completed by Kroner (2019).

Whole-rock results acquired using the new nanopowders method, coupled with conventional lithogeochemistry and isotopic studies (Os and Pb), demonstrate that Nahlin ophiolite peridotite is the residue of extensive melt extraction, possibly in a suprasubduction zone setting (Lawley et al., 2020). Ore-forming elements are incompatible during partial melting and should, in principle, be transferred to the crust. However, many of the melt-depleted Nahlin ophiolite peridotite samples yield ore-forming element concentrations that approach (e.g. Cu, Se) and/or exceed (e.g. As, Ag, Mo, Te, Bi) the composition of the primitive upper mantle asthenosphere (i.e. mantle that is unaffected by melt extraction associated with crust formation). These unusual concentrations of the ore-forming elements are opposite to their expected behaviour during partial melting. Pyroxenite dykes and high-temperature, Si-metasomatized mantle peridotite (i.e. olivine websterite) also yield anomalous concentrations for these elements, suggesting that interaction between residual peridotite and passing melts (or fluids) re-fertilized Au and other ore components. Refertilized pyroxenite dykes share geochemical similarities with arc theoleiite and/or boninites (Edwards, 1995; Varfalvy et al., 1997), which, if correct, provide a genetic link of Au-upgrading in the lithospheric mantle to arc and back-arc magmatic processes. Anomalous ratios of ore-forming and other incompatible trace elements that are normally unfractionated by mantle melting further suggest that re-fertilization also involved hydrous fluids derived from the subducting plate. Such fluid- and melt-metasomatized mantle domains have previously been described as fertile source regions for generating magmas enriched in ore components, such as Au (Richards, 2011; Hronsky et al., 2012; Griffin et al., 2013). Our new work demonstrates that these same source regions are also enriched in other components associated with Au (e.g. As, Ag, Mo, Te, Bi; Fig. 2).

The refertilization of ore components to melt-depleted peridotite established by the nanopowders data is further recorded at the microscale. Documentation of the association between metasomatic clinopyroxene and base metal sulphide (BMS: pentlandite ±pyrrhotite±chalcopyrite) indicates metasomatic BMS precipitated during melt- and/or fluid-rock reactions (Fig. 3). Where present, BMS represent the dominant mineral host for base- and precious-metals, as demonstrated by high-resolution LA-ICP-MS mapping (Fig. 3: Lawley et al., 2019). Our results suggest that high-temperature mantle metasomatism is the dominant process for enriching melt-depleted peridotite, prior to serpentinization.

New, in situ LA-ICP-MS ore-element results from cratonic mantle olivine grains also point to trapped fluid and/or melt metasomatism that, for melt-depleted peridotite xenoliths devoid of metasomatic BMS, represent a significant host for the whole-rock budget of a subset of ore-forming elements. The composition of these olivine-hosted inclusions provide a new tool for tracing the upgrading of ore components at depth within the lithospheric mantle (*see discussion below*).

NEW TOOLS FOR DETECTING Au UPGRADING IN THE CRATONIC MANTLE

Most of what we know about the cratonic mantle comes from remotely acquired geophysical datasets and/or mantle fragments (1–10s cm; very rarely up to 1

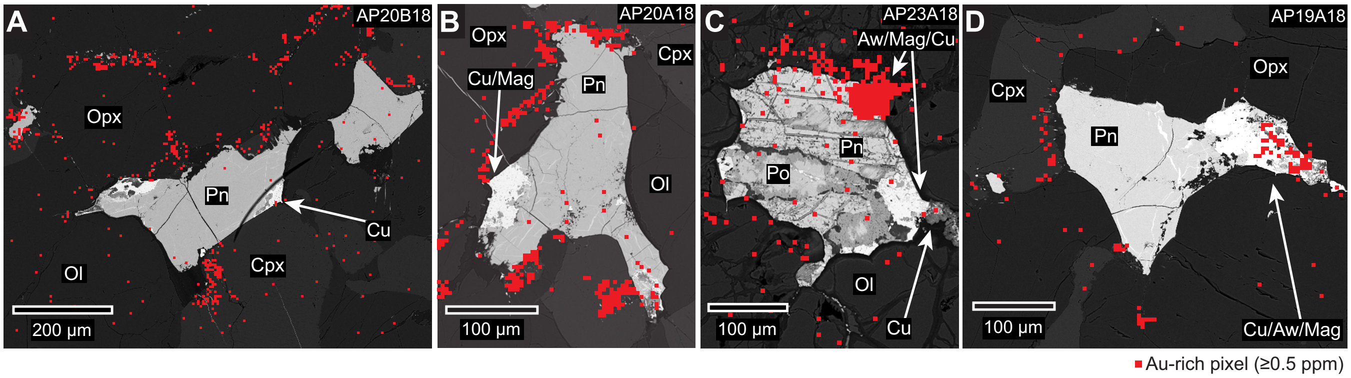

Figure 3. a–d) Scanning electron microscope (SEM) backscatter electron images (BSE) of base metal sulphide (BMS; Pn = pentlandite; Po = pyrrhotite) of the Nahlin ophiolite. Superimposed on the SEM-BSE images are laser alblation - inductively coupled plasma - mass spectrometry (LA-ICP-MS) mapping results (Lawley et al., 2019). Element map pixels with Au concentrations greater than the analytical detection limit (typically 0.5–1 ppm using a 4–6 μm spot) locally reflect trace element substitution into the S-poor mineral assemblage (Aw = awaruite; Cu = native Cu; Mag = magnetite) replacing the BMS **(c)**. Ultrafine electrum also occurs as inclusions within mantle silicates (Cpx = clinopyroxene; Ol = olivine; Opx = orthopyroxene) and fracture-hosted grains **(a, b, d)**. We suggest that ultrafine electrum may represent a significant host for the whole-rock Au budget, but, in this case, likely reflect microscale remobilization from BMS breakdown during serpentinization. Similar Au-rich inclusions have also been observed within fresh, unserpentinized mantle olivine from Kirkland Lake and Jericho.

m) transported to surface by kimberlites (i.e. xenoliths and xenocrysts: Snyder et al., 2004; Pearson et al., 2007; Pearson and Wittig, 2008; Stachel and Harris, 2008; Griffin et al., 2013). With the exception of PGE, very little research has been conducted on the concentration levels of other ore-forming components within the lithospheric mantle, precluding understanding the potential for upgrading processes. To address this knowledge gap, trace to ultratrace element studies using LA-ICP-MS analyses of mantle xenocrysts and xenoliths from the Superior and Slave cratons were studied as part of the Mantle Metal Mobility activity.

New results suggest that some ore-forming elements are readily substituted into the olivine crystal structure due to their size and charge (e.g. Ni, Zn, and to a lesser extent Cu). Other ore-forming element concentrations (including As, Se, Mo, Ag, Sn, Sb, Te, Pb, Bi, Au, Pt, and Pd) within olivine and clinopyroxene xenocrysts were anomalous and are interpreted to reflect trapped inclusions, which, if correct, likely represent the first direct sampling of precious metal modified cratonic mantle beneath the Abitibi greenstone belt (Lawley et al., 2018a,b). Although the concentrations of these ore-forming elements are extremely low (ppb), the association between Na-altered olivine xenocrysts with Au-, Pt-, and Pd-bearing inclusions likely reflect mantle metasomatism. Olivine xenocrysts also contain anomalous concentrations of As, Ag, Mo, Te, and Bi, possibly related to trapped, hydrous fluid-inclusions. A large proportion of the metasomatized mantle xenocrysts from the southern Superior craton were sampled at the base of the lithospheric mantle (~180 km), consistent with upgrading of ore components by rising asenthospheric melts (Fig. 2). Metasomatized mantle domains were also recognized in the mid-lithosphere (~100 km) by xenocrysts with anomalous high-field strength element-rich (HFSE) signatures. Modified mantle domains sampled at this depth correspond to relatively conductive lithosphere, suggesting re-fertilization of cratonic mantle may have also been associated with the crystallization of other metasomatic minerals (amphibole, phlogopite, etc.) in the mid-lithosphere (Fig. 2). Metasomatic clinopyroxene sampled at both of these depths yield variable Pb isotopic compositions and point to metasomatized domains that are up to one billion years older than the timing of kimberlite volcanism (Lawley et al., 2018a,b).

Similar ore-forming element inclusions were also recognized from olivine derived from Slave craton peridotite xenoliths and xenocrysts, indicating the findings are not restricted to the southern Superior craton (Fig. 1). Hence the trace to ultratrace element composition of mantle xenocrysts, coupled with isotopic studies and geophysical imaging, represent powerful tools for mapping the upgrading of ore-forming elements in the lithospheric mantle.

IMPLICATIONS FOR EXPLORATION

The lithospheric mantle represents the cold, melt-depleted residues leftover from the extraction of the continental crust (O'Reilly et al., 2001; Pearson et al., 2007; Griffin et al., 2009). Past melting events have depleted the lithospheric mantle in Fe, which makes it less dense relative to the convecting mantle (Jordan, 1975). The positive buoyancy of lithospheric mantle has played an important role in the stabilization of the continents and the preservation of ore systems hosted within the crust (Griffin et al., 2013). This is particularly critical for ancient cratons, which host a variety of ore systems that would likely have been destroyed and

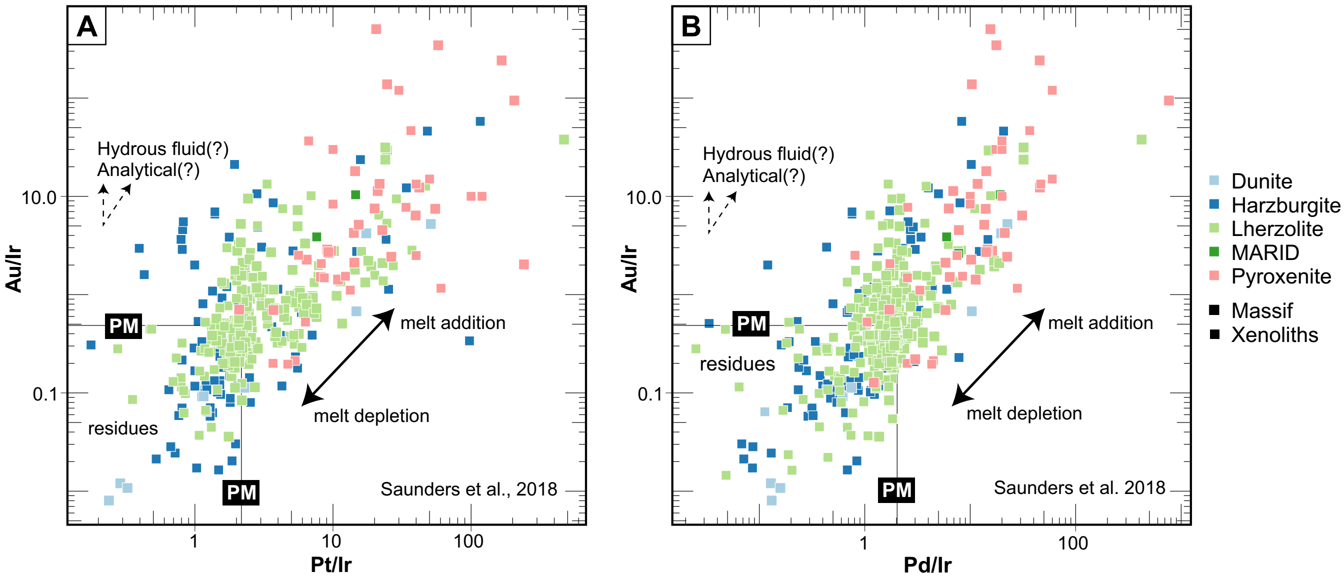

Figure 4. Global compilation of whole-rock platinum group elements (PGEs) and Au concentrations (Saunders et al., 2018). Melt-depleted peridotite samples yield Au/Ir, Pt/Ir, and Pd/Ir ratios that are lower than the primitive mantle (PM) due to their incompatibility during mantle melting. Increasing melt:rock interaction tends to increase incompatible element concentrations as demonstrated by the composition of pyroxenite dykes and melt-impregnated peridotite samples. Hydrous fluid metasomatism yields an Au/Ir ratio greater than the melt-rock array and may represent a second-order control for a smaller subset of mantle samples.

recycled into the mantle during subduction over geological time. The lithospheric architecture that formed during continent building also focuses magmas and their ore components during younger tectonic events (Begg et al., 2010; Mole et al., 2017). The concentration of numerous ore deposit types at the margins of cratonic blocks point to the important role that lithospheric architecture has on the genesis and preservation of ore systems (Begg et al., 2010; Hronsky et al., 2012; Griffin et al., 2013).

The precursors to ore-forming magmas likely interact with the lithospheric mantle during ascent (Fig. 2). Evidence for this process includes the precipitation of metasomatic BMS, which represents the primary mechanism for re-fertilizing melt-depleted mantle residues in precious metals and other ore components (Fig. 3). Because metasomatic BMS are typically enriched in Au, Pt, and Pd relative to other PGE (e.g. Ir), re-fertilized peridotite residues yield elevated Au/Ir, Pt/Ir, and Pd/Ir ratios (Fig. 4). Re-analysis of the global compilation of Au data for mantle rocks (Saunders et al., 2018), completed as part of the current study, suggests that a relatively large proportion of melt-depleted peridotite is re-fertilized (Fig. 4). At the Nahlin ophiolite, metasomatic BMS are also enriched in Ag, Bi, and Te and the BMS were likely precipitated during interaction between pyroxenite dykes with melt-depleted harzburgite in a suprasubduction zone setting. The precious metal signature of the Nahlin ophiolite pyroxenite dykes agrees with the trend towards elevated Au/Ir ratios defined by the global compilation, suggesting that passing melts play an important role in re-fertilizing the lithosphere through precipitation of BMS (Saunders et al., 2018).

New results further suggest that BMS alone are unable to explain the entire Au enrichment. Other Au-bearing phases are required to balance the whole-rock lithospheric budget in at least some cases (Luguet et al., 2004; Ferraris and Lorand, 2015). Olivine compositions and high-resolution LA-ICP-MS mapping suggests that some of this missing Au may be hosted by ultrafine native Au inclusions (Fig. 3). Similar inclusions have previously been reported (Ferraris and Lorand, 2015; Tassara et al., 2017; Zelenski et al., 2018) and were also identified in the current study due to the excellent sensitivity of the LA-ICP-MS mapping method (Fig. 3). Samples with a significant proportion of such Au-bearing inclusions would likely yield elevated whole-rock Au/Ir ratios (Fig. 4). A small subset of whole-rock analyses that deviate from the global trend defined by pyroxenite dykes and re-fertilized harzburgite may reflect the presence of such inclusions or possibly an analytical artefact due to the nugget effect (i.e. Au and other precious metals are heterogeneously distributed, which, coupled with the small sample volume used in some analytical methods, can yield spurious concentrations that are not representative of the rock as a whole; Fig. 4). We speculate that such inclusions, where present, reflect trapping of hydrous fluids, which has previously been suggested as a second important mechanism for upgrading Au within the lithospheric mantle (McInnes et al., 1999; Richards, 2009; Hronsky et al., 2012; Groves and Santosh, 2016). New results from the Nahlin ophiolite demonstrate that such fluid-fluxed samples and their trapped inclusions also yield suprachondritic ratios for a range of ore-forming elements (e.g. Mo/Ce and As/Pb), providing additional tracers for identifying similar metasomatic mantle domains from the rock record (Lawley et al., 2020).

Overall, it would appear that re-fertilization has had a significant impact on some ore-forming element concentrations within the lithospheric mantle. For example, global compilations suggest that the lithospheric mantle contains 1.2 ppb Au (Saunders et al., 2018), which is essentially identical to the expected composition of the depleted mantle (1.0 ppb: Salters and Stracke, 2004) and primitive mantle (i.e. the expected composition of the mantle prior to crustal extraction, 1.7 ppb: Palme and O'Neill, 2014). The bulk continental crust also yields Au concentrations (1.3 ppb: Rudnick and Gao, 2003) that are similar to these mantle estimates. The agreement between all four reservoirs suggest that Au was not fractionated during the formation of the continents or, more likely, that Au was subsequently re-introduced into the lithospheric mantle after crust extraction.

One of the critical remaining questions is whether the metasomatized lithospheric mantle also supplies ore components to asthenospheric melts during ascent (Richards, 2009; Hronsky et al., 2012; Arndt, 2013). As an example of this process, assimilation of lithospheric mantle orthopyroxene by kimberlite melt is well documented (Sharygin et al., 2017). However, direct evidence for scavenging of ore components by rising asthenospheric melts and their interaction with previously metasomatized mantle lithosphere are comparatively scarce (Arndt, 2013). Peridotite xenoliths and corresponding volcanic arc lavas from Lihir Island (Papua New Guinea) provide one of the rare examples where a pre-enriched lithospheric mantle source could be linked directly to a gold deposit at surface (McInnes et al., 1999). The North China craton also represents one of the few examples of a major gold province (Yang et al., 2003) overlying lithospheric mantle that is systematically enriched in ore-forming elements relative to the global average (Saunders et al., 2018). Mesozoic gold deposits in the North China craton are hosted within previously metamorphosed Archean-Proterozoic basement, providing compelling evidence that fluids and ore components of metamorphic origin were not involved in deposit genesis (Yang et al., 2003; Li and Santosh, 2014; Groves and Santosh, 2016). Instead, these deposits have been linked to reworking of the lithospheric mantle underlying the North China craton with subcrustal sources of fluids and metals (Li and Santosh, 2014). Similar settings for Au have since been identified within Paleoproterozoic orogens superimposed on Archean basement in Canada and Tanzania (Lawley et al., 2013, 2015), although the source of gold at most Au ore systems and deposits remains equivocal. Most other studies that advocate for a mantle source region pre-enriched in ore components are based on indirect links, including distinct host rock compositions with a lithospheric mantle signature (Zhang et al., 2008), atypical ore-forming element ratios (Kepezhinskas and Defant, 2001), and/or isotopic evidence for interaction between ore-hosting rocks and an enriched mantle source (Richardson and Shirey, 2008).

Whether pre-enriched sources are required for ore deposit genesis is important because fertile source regions have the potential to significantly broaden the footprint of Au ore systems and provide a possible mechanism to explain the variable endowment observed between cratons (Sillitoe, 2008; Hronsky et al., 2012). Re-melting the kind of metasomatized, pre-enriched lithosphere documented herein, could, in principle, produce fertile melts that go on to form ore systems in the crust (McInnes et al., 1999; Sillitoe, 2008; Richards, 2009; Hronsky et al., 2012; Griffin et al., 2013). If correct, exploration should target lithospheric pathways that connect pre-enriched mantle with deep crustal fault networks. Unfortunately, the mechanisms for transferring ore components to rising asthenospheric melts remain very poorly understood and obvious examples of this process occurring are rare. Future research should focus on the composition of melts that have interacted with fertilized peridotite, particularly with ore-forming elements other than PGE. More research is also required on the interaction of lithospheric mantle-derived melts and their associated hydrothermal fluids once they reach the crust, since there is often very little evidence for such rocks at individual deposits. Finally, advances in geophysical and geochemical imaging technologies will continue to improve our understanding of the lithospheric mantle and its controls on ore systems.

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