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The ruthenium isotopic composition of the oceanic mantle

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ABSTRACT

The approximately chondritic relative, and comparatively high absolute mantle abundances of the highly siderophile elements (HSE), suggest that their concentrations in the bulk silicate Earth were primarily established during a final ~0.5 to 1% of “late accretion” to the mantle, following the cessation of core segregation. Consequently, the isotopic composition of the HSE Ru in the mantle reflects an amalgamation of the isotopic compositions of late accretionary contributions to the silicate portion of the Earth. Among cosmochemical materials, Ru is characterized by considerable mass-independent isotopic variability, making it a powerful genetic tracer of Earth’s late accretionary building blocks. To define the Ru isotopic composition of the oceanic mantle, the largest portion of the accessible mantle, we report Ru isotopic data for materials from one Archean and seven Phanerozoic oceanic mantle domains. A sample from a continental lithospheric mantle domain is also examined. All samples have identical Ru isotopic compositions, within analytical uncertainties, indicating that Ru isotopes are well mixed in the oceanic mantle, defining a $\mu^{100}\text{Ru}$ value of 1.2 ± 7.2 (2SD). The only known meteorites with the same Ru isotopic composition are enstatite chondrites and, when corrected for the effects of cosmic ray exposure, members of the Main Group and sLL subgroup of the IAB iron meteorite complex which have a collective CRE corrected $\mu^{100}\text{Ru}$ value of 0.9 ± 3.0 . This suggests that materials from the region(s) of the solar nebula sampled by these meteorites likely contributed the dominant portion of late accreted materials to Earth’s mantle.

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1. Introduction

Ruthenium is a highly siderophile element (Re, Os, Ir, Ru, Pt, Rh, Pd, Au) comprising seven stable isotopes ($^{96}\text{Ru} = 5.54\%$; $^{98}\text{Ru} = 1.87\%$; $^{99}\text{Ru} = 12.8\%$; $^{100}\text{Ru} = 12.6\%$; $^{101}\text{Ru} = 17.1\%$; $^{102}\text{Ru} = 31.6\%$; $^{104}\text{Ru} = 18.6\%$). There are two short-lived chronometers associated with Ru, ^{98}Tc – ^{98}Ru and ^{99}Tc – ^{99}Ru ($t_{1/2} = 4.2$ – 10 Myr and $t_{1/2} = 0.21$ Myr, respectively; Kobayashi et al., 1993), however, no evidence for the presence of Tc in the early Solar System has yet been discovered. The HSE are highly soluble in metallic phases and are, therefore, characterized by very high metal–silicate partition coefficients under low pressure conditions ($D^{\text{Met/Sil}} > 10,000$, where D refers to the concentration ratio of an element in liquid metal to liquid silicate; Kimura et al., 1974; Mann et al., 2012). As a result, the putative lunar forming impact and associated final core segregation event likely largely stripped the bulk silicate Earth (BSE) of HSE (Touboul et al., 2015). The projected absolute abundances of HSE in the BSE, however, are only ~200 times lower than bulk CI chondrites abundances,

and the HSE are in approximately chondritic relative abundances (Morgan, 1986; Becker et al., 2006). Additionally, the projected $^{187}\text{Os}/^{188}\text{Os}$ and $^{186}\text{Os}/^{188}\text{Os}$ ratios for the BSE are within the range defined by chondritic meteorites (where $^{187}\text{Re} \rightarrow ^{187}\text{Os}$: $t_{1/2} = 42$ Gyr, $^{190}\text{Pt} \rightarrow ^{186}\text{Os}$: $t_{1/2} = \sim 490$ Gyr; Morgan, 1985; Walker et al., 1997; Meisel et al., 2001; Brandon et al., 2006). These characteristics are not predicted from metal–silicate partitioning experiments, making it unlikely that the HSE budget of the BSE was established as a result of core formation (Chou, 1978; Morgan, 1986; Becker et al., 2006). A more plausible scenario is that the HSE were added to the BSE during a late accretion phase, that is, after cessation of core formation by the accretion of ~0.5 to 1 wt.% of Earth’s mass to the mantle, in the form of planetesimals with chondritic bulk composition (Kimura et al., 1974; Chou, 1978). Further, late accretion may have been dominated by the impact of several large projectiles (2500–3000 km in diameter) stochastically delivered and emulsified into the BSE (Bottke et al., 2010; Dahl and Stevenson, 2010). Recent studies based on N body simulations and Pt isotope constraints support the contention that late accretion played a dominant role in establishing the abundances of HSE in the mantle (Rubie et al., 2016; Creech et al., 2017).

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Given the likely late accretionary origin of the HSE in the BSE, these elements are well suited to probing the characteristics of late accretionary impactors. For example, the $^{187}\text{Os}/^{188}\text{Os}$, $^{186}\text{Os}/^{188}\text{Os}$, and the relative abundances of the HSE estimated from the BSE are most similar to some enstatite chondrites (e.g., Meisel et al., 1996, 2001; Becker et al., 2006; Brandon et al., 2006). Formulating accurate and precise projections of BSE compositions from mantle and mantle-derived materials, however, requires accurate assessment of the effects of prior mantle processing, such as melting, crystallization, and metasomatism (e.g., Alard et al., 2000; Becker et al., 2006). These are inherently difficult variables to constrain, and thus, their associated uncertainties have led to substantial conjecture in the projections of BSE compositions and the composition of late accreted materials (Lorand et al., 2009).

Ruthenium isotopes are an important additional tool for constraining the genetics of late accretionary contributions to the BSE. Here, the term “genetic” refers to the isotopic compositions of the primary nebular materials from which an object was built (Walker et al., 2015). The utility of Ru isotopes for genetic study of late accreted materials is based on the assumption that most of the Ru present in the BSE was added by late accretion, as well as the observation that Ru is isotopically highly heterogeneous among different meteorite groups, with $^{100}\text{Ru}/^{101}\text{Ru}$ ratios varying by up to 140 ppm among chondrites and achondrites (Chen et al., 2010; Fischer-Gödde et al., 2015; Fischer-Gödde and Kleine, 2017). The large, mass-independent isotopic variations, termed anomalies, are presumed to result from the incorporation of variable proportions of presolar components, which were produced via nucleosynthetic reactions in stars (e.g., *s*-, *r*-, *p*-process; Zinner, 1998), into planetary building blocks. This heterogeneity is a product of either a poorly homogenized distribution of presolar materials in the disk, or from thermal processing of presolar material in the disk (e.g., Qin and Carlson, 2016). Nucleosynthetic Ru isotope anomalies observed in meteorite groups most likely reflect variable incorporation of *s*-process materials in the feeding zones of their respective parent bodies (Chen et al., 2010; Fischer-Gödde et al., 2015; Fischer-Gödde and Kleine, 2017). By comparing the Ru isotopic compositions of different meteorites with the isotopic composition of the terrestrial mantle, it should be possible to identify the dominant signature of late accretionary impactors.

To successfully apply genetic testing, precise determination of the Ru isotope compositions of both cosmochemical and terrestrial mantle endmembers is required. Presently, the Ru isotopic composition of the BSE is poorly constrained. Two prior studies have reported limited data for Phanerozoic materials sampling two mantle sources, the Shetland Ophiolite Complex and the Josephine Ophiolite Complex (Fischer-Gödde et al., 2015; Bermingham et al., 2016), and one study has reported Ru isotope composition data for a Bushveld chromitite (Fischer-Gödde and Kleine, 2017). To more fully investigate the Ru isotopic composition of the oceanic mantle, we report isotopic data for Archean and Phanerozoic materials which were collected from eight oceanic mantle reservoirs. One sample, which likely samples the continental lithospheric mantle (Guli; Malitch and Lopatin, 1997; Merkle et al., 2012), is also included. The Ru isotopic compositions of the IAB iron meteorite complex have previously been reported to match that of the BSE (e.g., Chen et al., 2010; Fischer-Gödde et al., 2015), however, it was subsequently suggested that some IAB iron meteorites had a distinct Ru isotopic composition from the BSE (Fischer-Gödde and Kleine, 2017). As a result, we also conduct a detailed examination of six meteorites from this complex.

2. Samples

Terrestrial samples were selected based on their Ru concentration, age, and location, and include three whole rock chromitites and ten Os–Ir–Ru alloy grains (Table 1). Chromitites (C1, C2, C3) were provided by B. O’Driscoll (University of Manchester) as hand samples (~10 g). These chromitites were collected from serpentinized chromitite seams in the Cliff area of the Shetland Ophiolite Complex (~492 Ma; Spray and Dunning, 1991). The chromitites are characterized by high Ru concentrations (~23 ppm) and a narrow range in $^{187}\text{Os}/^{188}\text{Os}$ (0.12922 ± 3 , 2σ , $n = 6$; O’Driscoll et al., 2012). The Ru isotopic compositions of the chromitites likely reflect the composition of their source region given the restricted mobility of HSE during serpentinization, the chemically robust nature of chromite, and the high concentration of the HSE in these samples (O’Driscoll et al., 2012). Furthermore, the Cr contents of the samples indicate that they formed as a result of high melt/rock interactions, implying that they provide a well-averaged composition of the oceanic mantle from which the rocks originated.

Multiple Os–Ir–Ru alloy grains from nine different worldwide locations were also analyzed (Table 1). The grains were obtained from the Natural History Museum, London (UK), I. Puchtel (University of Maryland, USA), and K.N. Malitch (A.N. Zavaritsky Institute of Geology and Geochemistry, Russia). Osmium–iridium–ruthenium alloys are commonly found in association with ultramafic rocks. They normally occur as inclusions within minerals such as chromite, or as detrital grains in streambeds draining peridotitic terrains. Such grains are commonly interpreted to form as the result of either the release and subsequent reduction of Os, Ir and Ru during alteration of sulfides contained in upper mantle rocks, or by direct precipitation in upper mantle magma chambers (e.g., Cabri et al., 1996; Peck et al., 1992). By either mechanism, the Ru isotopic compositions of these grains likely provide an accurate reflection of the isotopic composition of their mantle source regions, given their high resistance to modification by subsequent thermal events and moderate to very high Ru concentrations (1–30 wt.%; Malitch, 2004; Malitch et al., 2011).

Samples examined here include three Os–Ir–Ru grains found in placer deposits near Port Orford, Oregon (USA). These grains are associated with the Josephine Ophiolite Complex (Walker et al., 2005). The $^{187}\text{Os}/^{188}\text{Os}$ ratios of these grains are consistent with the Os isotopic composition of chromitites sampled from the Josephine Creek, indicating formation in the Mesozoic upper mantle (~162 Ma; Walker et al., 2005). The grains have between 43–73 wt.% Os, 15–37 wt.% Ir, 5.8–15 wt.% Ru, and display homogeneous intra- and inter-sample $^{187}\text{Os}/^{188}\text{Os}$ ratios (Table 1).

One Os–Ir–Ru alloy grain each from Bald Hills and Savage River, Tasmania, Australia, were analyzed. Grains collected from these locations have ^{187}Os -isotopic model ages of ~130 Ma (Walker et al., 1997; Brandon et al., 1998). One grain from Yodda, Papua New Guinea was analyzed. Spatially related grains from this area have ^{187}Os -isotopic model ages of ~80 Ma (Brandon et al., 2006). No information about HSE abundances of the Bald Hills, Savage River, or Yodda alloy grains is currently available. Two Os–Ir–Ru alloy grains from the Nizhny Tagil and Kushva regions in the Urals, Russia were analyzed. No information about HSE abundances or Re–Os isotope systematics of the Kushva alloy grain is currently available, however, laurite and Os–Ir–Ru grains alloys from the neighboring Nizhny Tagil massif have Os-isotope model ages of ~870 ± 50 Ma (Anikina et al., 2014; Tessalina et al., 2015).

Two bulk samples comprising several small (<500 μm) Os–Ir–Ru alloy grains were analyzed. One was collected from Quaternary sediments in an area of the Ingaranga River (prospecting line L-365, Fig. 2 in Merkle et al., 2012) associated with the Guli clinopyroxene–dunite massif of the Maimecha–Kotui Province in Polar Siberia, Russia (Malitch and Lopatin, 1997). Osmium–

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