



Nature of late accretion to Earth inferred from mass-dependent Ru isotopic compositions of chondrites and mantle peridotites

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ABSTRACT

Elevated abundances of highly siderophile elements in Earth's mantle are thought to reflect the late accretion of primitive material after the cessation of core formation, but the origin of this material, and whether or not it can be linked to specific types of meteorites remain debated. Here, mass-dependent Ru isotopic data for chondrites and terrestrial peridotites are reported to evaluate the chemical nature and type of the late-accreted material. After correction for nucleosynthetic Ru isotope anomalies, enstatite, ordinary and carbonaceous chondrites all have indistinguishable mass-dependent Ru isotopic compositions. Thus, neither distinct formation conditions in the solar nebula nor parent body processes resulted in significant mass-dependent Ru isotope fractionation. All five terrestrial peridotites analyzed have mass-dependent Ru isotopic compositions that are indistinguishable from each other and from the composition of chondrites. The chondritic mass-dependent Ru isotopic composition of Earth's mantle is difficult to reconcile with prior suggestions that the late accretionary assemblage was a mixture of chondrites with a chemically evolved metal component. Although this mixture can reproduce the suprachondritic Ru/Ir inferred for Earth's mantle, it consistently predicts a heavy Ru isotopic composition of Earth's mantle with respect to chondrites. This is because metal components with elevated Ru/Ir are also enriched in heavy Ru isotopes, resulting from isotope fractionation during core crystallization. Thus, if late accretion involved impacts of differentiated protoplanetary bodies, then the projectile cores must have been either homogenized upon impact, or added to Earth's mantle completely, because otherwise Earth's mantle would have inherited a non-chondritic mass-dependent Ru isotopic composition from the unrepresentative sampling of core material.

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1. Introduction

Late accretion is defined as continued accretion onto Earth following the putative Moon-forming impact and the cessation of core formation, resulting in the addition of ~0.5 wt% of broadly chondritic material to the Earth's mantle (e.g., Walker et al., 2015). Evidence for late accretion primarily comes from abundances of the highly siderophile elements (HSE: Os, Ir, Ru, Rh, Pt, Pd, Re and Au) in Earth's mantle, which are much higher than expected for metal-silicate equilibration during Earth's core formation (e.g., Chou, 1978; Kimura et al., 1974; Mann et al., 2012). This observation, together with the broadly chondritic relative HSE abundances inferred for Earth's mantle (e.g., Becker et al., 2006; Fischer-Gödde et al., 2011; Day et al., 2017), is most easily explained by late accretion after core formation was complete. However, the nature of the late-accreted material and whether it con-

sisted of specific meteorite types or of material not represented by meteorites remains a matter of debate (e.g., Walker et al., 2015). For instance, on the basis of Os isotopes and HSE abundances it has been suggested that the late-accreted material had an enstatite or ordinary chondrite-like composition (Meisel et al., 1996; Walker et al., 2002) or consisted of a mixture of carbonaceous chondrite-like material with a chemically evolved metal component, similar in composition to some iron meteorites (Fischer-Gödde and Becker, 2012). Distinguishing between these interpretations is important not only for understanding the late stages of terrestrial planet formation, but also because addition of carbonaceous chondrite-like material during late accretion may have been the major source of Earth's water and highly volatile species (Marty, 2012; Wang and Becker, 2013).

Ruthenium is well suited to examine these issues and constrain the nature of the late-accreted material. As a HSE, most or all of the Ru in Earth's mantle derives from late accretion, and so its elemental and isotopic composition predominantly reflects that of the material added after the cessation of core formation. Of note, HSE systematics of continental peridotites suggest that Earth's mantle

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is characterized by slightly suprachondritic Ru/Ir and Pd/Ir (Becker et al., 2006). The elevated Pd/Ir is not seen in oceanic peridotites, however, suggesting that the Pd/Ir of continental peridotites has been modified by refertilization and is, therefore, not a signature of the bulk silicate Earth (Day et al., 2017). By contrast, the suprachondritic Ru/Ir is observed for both continental and oceanic peridotites, indicating that the bulk of Earth's mantle is characterized by higher Ru/Ir compared to known chondrites (Becker et al., 2006; Day et al., 2017). It has been suggested that the elevated Ru/Ir reflects the late accretion of a fractionated iron meteorite-like component (Fischer-Gödde and Becker, 2012), or is the result of sulfide segregation within the Earth's mantle, during which Ru was less chalcophile than the other HSE (Laurenz et al., 2016; Rubie et al., 2016). Thus, identifying the process by which the elevated Ru/Ir was produced would provide important information on the nature of the late-accreted material, and the relative roles of late accretion and core formation in establishing HSE abundances in Earth's mantle.

In this study, *mass-dependent* Ru isotope variations among meteorites and the Earth's mantle are used to constrain the nature and type of the late-accreted material. Until now, most Ru isotope studies have utilized *mass-independent* isotope anomalies as genetic tracers (Bermingham and Walker, 2017; Bermingham et al., 2018; Chen et al., 2010; Fischer-Gödde et al., 2015; Fischer-Gödde and Kleine, 2017). These Ru isotope anomalies arise from the heterogeneous distribution of presolar components at the bulk meteorite and planetary scale. All meteorites analyzed to date are characterized by a deficit in *s*-process Ru nuclides (Fischer-Gödde et al., 2015; Fischer-Gödde and Kleine, 2017), with the possible exception of some non-magmatic iron meteorites (Bermingham and Walker, 2017). Thus, the late accretionary assemblage does not seem to be represented by meteorites, but more likely derives from bodies that were initially located closer to the Sun, in the terrestrial planet region (Fischer-Gödde and Kleine, 2017). Unlike the *mass-independent* nucleosynthetic Ru isotope anomalies, *mass-dependent* Ru isotope variations reflect the physico-chemical processes affecting material in the solar nebula or on parent bodies. Thus, although the nucleosynthetic Ru isotope anomalies indicate that meteorites are not representative of the actual bodies comprising the late accretionary assemblage, mass-dependent Ru isotope variations among meteorites can nevertheless be used to constrain the chemical nature of the late-accreted material. For instance, the distinct classes of chondrites formed under very different conditions, which may have led to mass-dependent Ru isotope variations that could then potentially be used to link the late accretionary assemblage to a specific formation location within the protoplanetary disk. Furthermore, most iron meteorites are characterized by heavy Ru isotope compositions, resulting from Ru isotope fractionation between coexisting solids and liquids during core crystallization (Hopp et al., 2018). Thus, if the suprachondritic Ru/Ir of Earth's mantle results from late accretion of a chemically evolved metal component, then the Ru isotopic composition of Earth's mantle would likely be heavy with respect to the composition of chondrites. This makes Ru isotopes uniquely useful to examine as to whether the late accretionary assemblage consisted of material with chondrite-like compositions or also contained other material, such as a differentiated metal component.

Here, mass-dependent Ru isotopic data for chondrites and terrestrial mantle peridotites are reported. These data are used to assess the magnitude of any Ru isotope fractionation between Earth's mantle and chondrites, with the ultimate goal to constrain the nature of the late-accreted material, and more generally the process of late accretion.

2. Samples and analytical methods

2.1. Sample preparation

Chondrites investigated in this study include eighteen samples from the three major chondrite classes (enstatite, ordinary and carbonaceous chondrites). Samples were cut from larger slices using a diamond saw, cleaned with SiC abrasives and de-ionized water in an ultrasonic bath, and crushed to fine powders in an agate mortar. About 0.3 to 1 g of each chondrite powder was digested using reverse *aqua regia* inside sealed borosilicate Carius tubes at 220 °C for 2 days (Shirey and Walker, 1995; Hopp et al., 2016).

Chondrites have variable nucleosynthetic Ru isotope anomalies (Chen et al., 2010; Fischer-Gödde et al., 2015; Fischer-Gödde and Kleine, 2017), which have to be corrected when determining mass-dependent Ru isotope variations. This is necessary because the double spike inversion used to calculate the isotope fractionation assumes that the measured isotope variations are purely mass-dependent (Burkhardt et al., 2014). As the Carius tube digestion employed here does not dissolve all presolar phases present in primitive chondrites (Fischer-Gödde et al., 2015), we also determined the nucleosynthetic Ru isotopic composition on unspiked digestion aliquots. After Carius tube digestion, the sample solutions were transferred into 50 ml centrifuge tubes and centrifuged for 20 minutes to separate and remove any undissolved silicates. After centrifuging, the solution was split into two aliquots (~35% and ~65%, respectively), which were transferred into two separate 60 ml Savillex PFA beakers. The ~65% aliquot was used for nucleosynthetic Ru isotope analyses, whereas the ~35% aliquot was spiked with an appropriate amount of ^{98}Ru – ^{101}Ru double spike for the measurement of mass-dependent Ru isotope variations. To avoid any cross contamination, the spiked and unspiked aliquots were processed and measured separately, using different lab ware that was strictly kept separate.

The spiked solutions were equilibrated for 24 h in 6 M HCl at 120 °C on a hotplate. This procedure results in complete spike-sample equilibration, as demonstrated for iron meteorites (Hopp et al., 2018). To ensure that complete spike-sample equilibration is also achieved for chondrites, seven aliquots of a homogeneous ~100 g Allende (CV3) powder prepared in Münster (MS-A) were processed using different digestion and spike-sample equilibration methods. Whereas five Allende samples were processed as described above, two Allende samples were spiked prior to digestion using the Ni sulfide fire assay method (e.g., Creech et al., 2017a). This method results in complete melting of the sample and therefore accesses all Ru from presolar phases. The mass-dependent Ru isotopic compositions of all seven Allende samples, after correction for nucleosynthetic isotope variations, agree very well, demonstrating that complete spike-sample equilibration was achieved (Table 1).

Equilibrated chondrites (petrologic types 5–6) no longer contain presolar phases, and all samples from a given chondrite group, when digested in Carius tubes, have homogeneous nucleosynthetic anomalies (Fischer-Gödde et al., 2015; Fischer-Gödde and Kleine, 2017). This is because any pre-existing nucleosynthetic Ru isotope heterogeneity within these samples has been equilibrated and homogenized during thermal metamorphism on the parent bodies. For these samples, no unspiked aliquots were analyzed in this study, and measured mass-dependent Ru isotope variations were corrected using previously reported nucleosynthetic Ru isotopic data (Fischer-Gödde et al., 2015; Fischer-Gödde and Kleine, 2017). For these samples, an appropriate amount of ^{98}Ru – ^{101}Ru double spike was added prior to digestion in Carius tubes.

In addition to the chondrites, five terrestrial peridotites from three different locations (Balmuccia Massif, Ligurides, Beni Bousera) were also investigated. These peridotites are well characterized

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