



# Rubidium isotopic composition of the Earth, meteorites, and the Moon: Evidence for the origin of volatile loss during planetary accretion



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## ARTICLE INFO

### Article history:

Received 3 April 2017

Received in revised form 18 May 2017

Accepted 23 May 2017

Available online 15 June 2017

Editor: M. Bickle

### Keywords:

volatile depletion  
rubidium isotopes  
the Moon  
chondrites

## ABSTRACT

Understanding the origin of volatile element variations in the inner Solar System has long been a goal of cosmochemistry, but many early studies searching for the fingerprint of volatile loss using stable isotope systems failed to find any resolvable variations.

An improved method for the chemical purification of Rb for high-precision isotope ratio measurements by multi-collector inductively-coupled-plasma mass-spectrometry. This method has been used to measure the Rb isotopic composition for a suite of planetary materials, including carbonaceous, ordinary, and enstatite chondrites, as well as achondrites (eucrite, angrite), terrestrial igneous rocks (basalt, andesite, granite), and Apollo lunar samples (mare basalts, alkali suite). Volatile depleted bodies (e.g. HED parent body, thermally metamorphosed meteorites) are enriched in the heavy isotope of Rb by up to several per mil compared to chondrites, suggesting volatile loss by evaporation at the surface of planetesimals. In addition, the Moon is isotopically distinct from the Moon in Rb. The variations in Rb isotope compositions in the volatile-poor samples are attributed to volatile loss from planetesimals during accretion. This suggests that either the Rb (and other volatile elements) were lost during or following the giant impact or by evaporation earlier during the accretion history of Theia.

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## 1. Introduction

Although the abundance of volatile elements in the inner Solar System has important implications for the accretion and evolution of the terrestrial planets, the origin of volatile element variations in inner Solar System bodies is debated. For example, although carbonaceous chondrites (CC) constitute the most chemically primitive meteorite group, they display differing degrees of volatile element depletions (i.e. depletions in elements with a 50% condensation temperature,  $T_c$ , between 250 K and 1250 K). The CI-type CC have elemental abundances that closely match the composition of the solar photosphere (excluding Li, H, C, N, O, and the noble gases; [Palme et al., 2014](#)). The other CC classes show increasing depletions in volatile elements corresponding to increasing elemental volatility in the order CI–CM–(CO, CV)–CK ([Palme et al., 2014](#)). Hypotheses to account for these volatile element abundance variations in bulk chondrites are varied and include nebular effects (e.g. condensation or evaporation; [Wasson and Chou, 1974](#);

[Ringwood, 1966](#)), or mixing of distinct chemical and isotopic reservoirs ([Larimer and Anders, 1967](#)).

Similarly, volatile element depletions in achondrites and in the terrestrial planets relative to CI are variable, and the origins of these variations are debated. Like undifferentiated meteorites, differentiated bodies may inherit their volatile element abundances from the processes of condensation and/or the early stages of accretion. However, differentiated bodies may also experience volatile loss through high-energy processes during the later stages of accretion and subsequent planetary evolution (e.g. evaporative loss during impacts, magma ocean degassing). The signature of these and other geochemical processes may be recorded in the isotopic compositions of planetary materials, making isotopes a key tracer of processes in the early solar system.

Rubidium is one element that is particularly well-suited to study the questions outlined above. Rubidium is a moderately volatile lithophile element with a  $T_c$  of 800 K ([Lodders, 2003](#)). Due to the similar incompatibility of Rb and Sr, the Rb/Sr ratio is generally constant in planetary materials and is a useful indicator of volatile depletion since Sr ( $T_c = 1455$  K) is much more refractory than Rb. The Rb/Sr variations in bodies from the inner solar system span a range of nearly four orders of magnitude ([Halliday and Porcelli, 2001](#)). Rubidium is present at a level of 1–2 ppm

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in bulk chondrites (e.g. [Lodders and Fegley, 1998](#)). During partial melting, Rb is relatively enriched in crustal materials, such as terrestrial granites (~200 ppm Rb), compared to the mantle (cf. <0.2 ppm Rb in terrestrial peridotite) (e.g. [Lodders and Fegley, 1998](#)). Multiple bodies in the inner Solar System exhibit significant depletions in Rb compared to chondrites, including the Earth, the Moon, and achondrites from the Howardite–Eucrite–Diogenite (HED) and angrite groups. The large variations in Rb/Sr ratio and Rb concentration (denoted [Rb]) in these planetary materials may hold important clues to the mechanism of volatile element depletion in the inner Solar System.

Rubidium is comprised of two isotopes: the stable isotope  $^{85}\text{Rb}$  (72.165%) and the long-lived radioactive isotope  $^{87}\text{Rb}$  (27.835%), which decays to  $^{87}\text{Sr}$  with a half-life of  $49.76 \times 10^9$  y ([Nebel et al., 2011b](#)). Mass-dependent Rb isotope variations in planetary materials are likely limited in most cases for several reasons. First, Rb is heavy compared to traditional stable isotope systems (e.g. C, N, O, S) so the relative mass difference between the two Rb isotopes is small. Second, Rb exists only as  $\text{Rb}^+$  and therefore no isotopic fractionation occurs during partitioning between phases with differing valence states (e.g. as for Fe between Fe(II) and Fe(III)). Third, equilibrium isotope fractionation due to partitioning between phases is generally larger for species among covalent bonds, but Rb has an ionic bonding character. Despite these reasons, large mass-dependent variations may occur during evaporation due to the volatile nature of Rb. The pioneer study on Rb isotopes in chondrites ([Nebel et al., 2011a](#)) found generally similar Rb isotope compositions between the Earth and chondrites, but with a relatively large analytical uncertainty of 0.2‰. It is possible that improvements in precision may allow the identification of small variations in Rb isotope compositions. Finally, the geochemical relationship between Rb and K makes the further study of Rb timely: although early studies on K isotope cosmochemistry did not find resolvable K isotope variations ([Humayun and Clayton, 1995](#)), recent work has uncovered a small difference between the K isotope composition of the Earth and the Moon ([Wang and Jacobsen, 2016a, 2016b](#)).

Here we present a new method for the chemical separation of Rb for high-precision isotope ratio measurements by multi-collector inductively-coupled-plasma mass-spectrometer (MC-ICP-MS). This method is used to report new Rb isotope data for a broad suite of planetary materials, including chondrites from the Carbonaceous (CC), Ordinary (OC), and Enstatite (EC) groups, achondrites (eucrite, angrite), terrestrial igneous rocks (basalt, andesite, granite), and Apollo lunar samples (basalt, alkali suite). The goal of this work is to present an updated overview of Rb isotope variations in the inner Solar System and to provide an initial assessment of the possible causes of any variations.

## 2. Samples and methods

### 2.1. Samples

A variety of terrestrial samples were selected in order to gain insight into the behavior of Rb during igneous differentiation. The terrestrial rocks used in this study include one granite (GS-N), one andesite (AGV-2), and four basalts. The basalts include samples from a variety of settings: one Columbia River continental flood basalt (BCR-2), one Mid-Ocean Ridge Basalt (MORB) from the mid-Atlantic ridge (EW9309 10D), and Ocean Island Basalts (OIB) from Hawaii (BHVO-2) and Galapagos (AHANEMO2 D20B).

Meteorite samples were selected from a variety of groups to achieve an overview of Rb isotope variations in the inner Solar System. This study focuses on observed meteorite falls when possible in order to avoid the effects of terrestrial weathering on Rb isotope compositions. The chondrites used in this study include

the CC Orgueil (CI1), Murchison (CM2), Cold Bokkeveld (CM2), Felix (CO3.3), Lancé (CO3.3), Allende (CV3, oxidized type), Vigarano (CV3, reduced type), and Karoonda (CK4). Other chondrites analyzed include the unequilibrated OC Krymka (LL3.2) and the EC Abebe (EH4), Indarch (EH4), and Khaipur (EL6). In addition, two thermally metamorphosed CM2 were selected (PCA 02012 and PCA 02010). These samples display indicators of significant volatile loss suggesting that they have been heated to temperature of ~900 °C ([Nakato et al., 2013](#); [Beck et al., 2014](#)).

A primary goal of this work was to investigate the Rb isotope composition of volatile-depleted bodies, including HED meteorites, angrites, and lunar basalts. The achondrites analyzed include two HED meteorite falls: the monomict eucrites Juvinas and Stannern. One angrite, SAH99555, was also measured for Rb isotope composition.

The comprehensive suite of Apollo mission lunar basalts includes two low-Ti olivine basalts (12012, 15555), one low-Ti ilmenite basalt (12016), one low-Ti low-K ilmenite basalt (10003), two low-Ti high-K ilmenite basalts (10017, 10057). One Alkali suite cataclastic norite (77215) from Apollo 17 was also selected for Rb isotope analysis.

### 2.2. Chemical purification of Rb

Whole rock samples were crushed by hand using an agate mortar until a fine powder was obtained. A minimum of 0.5 g of terrestrial rock or meteorite and 100 mg of lunar samples was crushed in order to avoid non-representational sample analysis. An aliquot of  $\leq 125$  mg of powdered sample was weighed depending on the Rb concentration of the sample; masses were calculated to yield  $>20$  ng Rb for isotope ratio measurements. Sample powders were digested using a mixture of concentrated HF/HNO<sub>3</sub> and heated at 130 °C in closed Teflon bombs for  $>48$  h. After evaporation of the HF/HNO<sub>3</sub>, 6N HCl was added and the samples were again heated at 130 °C to achieve dissolution of fluoride complexes. Samples were then evaporated to complete dryness and were ready for chemical purification.

Previous work on Rb isotopes utilized Zr doping for correction of mass bias on Rb ([Nebel et al., 2005, 2011a](#)). The goal of this study was to develop an improved chemical purification procedure to avoid the necessity of doping and improve analytical precision. To that end, a multi-step column chemistry procedure was developed in order to ensure complete separation of Rb from matrix elements and quantitative recovery of Rb (see Supplementary materials for additional discussion of the methods developed by this work). This chemistry achieves efficient separation of Rb from elements with isobaric and molecular interferences (in particular  $^{87}\text{Sr}$  from  $^{87}\text{Rb}$ , but also double-charged Er or Yb isotopes), on mass/charge of interest (84, 85, 86, 87, 88; Sr isotopes are monitored for the correction of  $^{87}\text{Sr}$  on  $^{87}\text{Rb}$ ). A major focus was sufficient removal K to avoid any potential matrix effects on the measured Rb isotope compositions (see supplementary materials).

Samples were first subjected to a Ca removal step using Eichrom normal DGA resin (50–100  $\mu\text{m}$ ). Following digestion, samples were dissolved in 1N HNO<sub>3</sub> and loaded on BioRad Poly-Prep chromatography columns containing 1.8 mL conditioned DGA resin; Rb was collected by washing with 1N HNO<sub>3</sub> while  $>90\%$  Ca was effectively retained by the resin. Our tests have shown that this is an important step to avoid two adverse effects caused by Ca: first, Ca may change the shape and placement of the Rb peak on cation exchange resin; this can lead to either loss of Rb (i.e. not collecting full Rb cut) or the presence of Ca in Rb cut (see supplementary materials).

After the Ca removal step, the collected sample was evaporated to dryness and the residue was dissolved in 3N HCl. The Rb was separated from matrix elements using cationic resin in three suc-

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