

Solution chemistry controls multisite sorption of ^{137}Cs on micaceous soils

ADAM J. FULLER^{1*}, SAMUEL SHAW¹, CAROLINE L. PEACOCK¹, DIVYESH TRIVEDI², AND IAN T. BURKE¹

¹School of Earth & Environment, University of Leeds, UK
eeajf@leeds.ac.uk (*presenting author)

²National Nuclear Laboratory, UK

Accidental releases of radioactive material have led to elevated levels of the hazardous radionuclide ^{137}Cs at a number of sites, notably: Fukushima, Japan; Chernobyl, Ukraine; Hanford, USA and Sellafield, UK. Once present in soils the mobility and bioavailability of ^{137}Cs is governed by its sorption to clay minerals [1]. This behaviour is influenced by the soil's mineralogy, solution pH, nature of competing cations, and the Cs concentration. We investigated the multi-site sorption behaviour of Cs on micaceous soils, as a function of Cs concentration and solution chemistry (pH and ionic strength).

This work used batch experiments to investigate sorption of Cs onto bulk 1 mm sieved micaceous soils from near Sellafield, W. Cumbria, UK. The amphoteric sorption behaviour of Cs is strongly controlled by its solution concentration (Fig. 1). At very low concentrations (10^{-11} M) Cs sorption is unaffected by pH. However as the concentration of Cs is increased pH becomes an important controlling factor on sorption.

At low concentrations, where sorption is pH independent, Cs selectively sorbs to the highly specific Frayed Edge Sites (FES) of illite. At increased concentrations (10^{-4} M) the FES become saturated and the excess sorbs to the basal plane [2]. This is a cation exchange process and is controlled by solution pH. At excessive Cs concentrations (10^{-1} M) the clay's sorption sites are saturated and any amphoteric behaviour is masked.

This study shows the complexity of the Cs sorption process on micaceous soils. It is governed by an interrelationship between solution chemistry and the availability of multiple sorption sites, as controlled by Cs solution concentration. It is important therefore to ensure that experiments are conducted at environmentally relevant Cs concentrations and that chemical variables are carefully controlled.

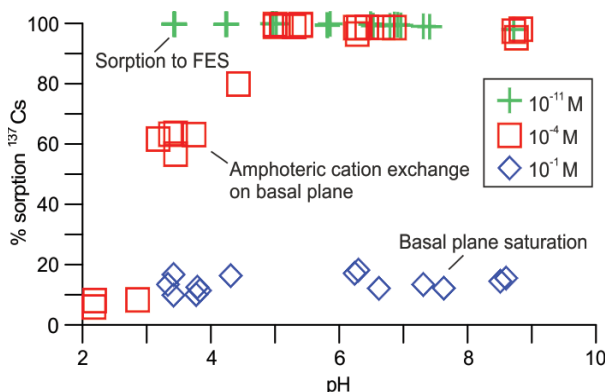


Figure 1 Sorption of Cs as a function of pH and Cs concentration

[1] Cornell (1993) *J. Radioanal. Nuclear Chem.* **171**, 483-500. [2] Poinsot, et al. (1999) *Geochim. Cosmochim. Acta.* **63**, 3217-3227

Isotope dilution analysis of Se and Te in chondritic meteorites

CLAUDIA FUNK^{1*}, FRANK WOMBACHER¹, HARRY BECKER², AND ADDI BISCHOFF³

¹Institut für Geologie und Mineralogie, Universität zu Köln, Germany. cfunk0@uni-koeln.de (*presenting author)

²Institut für Geologische Wissenschaften, Freie Universität Berlin, Germany

³Institut für Planetologie, Westfälische Wilhelms Universität Münster, Germany

Introduction

The moderately volatile elements Se and Te have similar equilibrium (50%) condensation temperatures (697 and 709 K, respectively) at 10^{-4} bar for a gas of solar-system composition [1]. While the purely chalcophile element Se is exclusively hosted in sulfides [e.g. 2], the siderophile element Te can be incorporated in alloys, but also in sulfides due to its strong chalcophile affinity [1]. In CK and R chondrites and refractory inclusions in CV chondrites Te additionally forms noble metal-rich tellurides such as chengbolite (PtTe_2) and moncheite (PtTe_2) [e.g. 3, 4]. To further constrain early cosmochemical processes that led to (primary) depletion and fractionation processes affecting these two elements, precise Se and Te abundances were determined for different groups of primitive chondrites.

Analytical approach

^{77}Se and ^{125}Te enriched spikes were added to typically 50 mg of meteorite sample powder which was subsequently dissolved by HF-HNO₃ acid digestion. Chemical separation of Se and Te followed by using thiol cotton fiber (TCF). Mass spectrometric measurements were performed by multi collector ICP-MS, hydride generation and background subtraction.

First Results

The presented method provides accurate and precise Se and Te abundances for chondritic samples. Concentrations were so far determined for a number of bulk carbonaceous chondrites (CI, C2 ungr., CM1, CM2, CO3, CV3, CR2, CH2), enstatite chondrites (EL3), ordinary chondrites (H3.3, L3.0), and Rumuruti chondrites (R3). Selenium and tellurium abundances are in general agreement with literature values [e.g. 2, 5]. Uniform Se/Te ratios were found for CI, C2 ungr. (Tagish Lake), CR2, CM1, and in one of the CM2 chondrites (Nogoya), whereas other CM2s, CV3, CO3.2 and CH2 chondrites show slightly lower Se/Te ratios. Ordinary, enstatite, and Rumuruti chondrites have consistently higher Se/Te ratios than carbonaceous chondrites. The data suggests that the CI chondrite Se/Te ratio is slightly higher than previously estimated by [1], and that Se/Te ratios discriminate ordinary, enstatite and Rumuruti chondrites from carbonaceous chondrites. Furthermore a weathered piece of Allende appears to confirm the suggestion that Se in chondrites is easily affected by terrestrial weathering [e.g. 2, 4].

[1] Lodders (2003) *Astrophys. J.* **591**, 1220-1247. [2] Dreibus et al. (1995) *Meteoritics* **30**, 439-445. [3] Geiger and Bischoff (1995) *Planet. Space Sci.* **43**, 485-498. [4] Bischoff et al. (2011) *Chemie der Erde* **71**, 101-133. [5] Brown et al. (2000) *Science* **290**, 320-325.