Ba isotope composition of Tagish Lake meteorite. G. Srinivasan, A. Ali, I Jabeen and S. Srinivasan.

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Introduction: The study of Ba isotopes is useful tool to understand the variations in s-process and r-process produced isotopes in the whole rock fractions of primitive meteorites [1]. Isotopic anomalies in Ba, Sm and Nd have been established in whole rock samples through high precision measurements using TIMS [1-3]. These measurements have shown that while there are p process deficiencies in Sm, and there is no r or s process anomaly in Nd or Sm. Ba isotope anomalies in Murchison acid residues have been known since early 1990s. Furthermore it has also been shown that acid leaches of whole rock fractions of meteorites show variable anomalies in Ba, suggesting different carrier phases for the different anomalies [4]. We report here preliminary results from similar leaching experiments of Tagish Lake meteorite and Ba isotope composition in the leach fractions and residue.

Analytical techniques: One gram of Tagish lake meteorite powder obtained from American Meteorite Laboratory was sequentially leached using acetic acid, HNO_3 and HCl. The residue from each step was the source material for the next step. 1) The first leach fraction was from a 4 ml, 1:1 mixture of water and 18M acetic acid at room temperature for one day. 2) The second leach fraction was from 4 ml of 6M HNO_3 at room temperature five days. 3) The final leach was obtained with 6M HCl at room temperature for 3 hours and then cooked at 75^oC for 24 hours. The supernatant and residue in all steps were separated by centrifugation at 3000 rpm for 15 minutes. The three leach fractions were dried at 110^oC and then treated with Aqua regia and HCl. The final residue was digested by HF and HNO_3 mixture by heating at 110^oC for 2 days. Later the residue was treated by Aqua regia and 6M HCl. The leached fractions and residue were finally dissolved in 4M HCl and pure fractions of Ba were separated using cation exchange resin following the methods developed in our laboratory [5].

Ba isotope analyses were carried out on Isoprobe-T TIMS. The methods were developed and calibrated using Ba standard obtained from Alfa Aesar and Spec Pure. Using a Re-triple filament geometry, ~ 0.750 μg of Ba in dilute HCl was loaded on the outer filament. The Ba load was sandwiched with 1 μl of 0.1 M phosphoric acid. After the final load the filament was heated to dull red glow for few seconds. The ionization (center) filament was heated to about 4Amperes and the outer and inner filaments were heated to about 2Amperes. A typical signal of $^{138}Ba \sim 6$-7 volts was obtained on 10^{11} ohm resistor. Ba was measured in multi-dynamic mode using 4 sequences. The counting time for each sequence was 10 seconds. A cycle consisted of 4 sequences and a measurement consisted of 400 cycles. The ^{136}Ba and ^{138}Ba signals were corrected for ^{136}Ce, and ^{138}Ce and ^{138}La interference respectively using ^{140}Ce and ^{139}La signal as a proxy and abundance ratio of $^{136}Ce/^{140}Ce = 0.002091$; and $^{138}La/^{139}La = 0.00900$ and $^{138}Ce/^{140}Ce = 0.0028377$. The Ba isotope ratios were corrected for fractionation was using the measured $^{134}Ba/^{136}Ba$ and using a reference value of 0.3078. ^{130}Ba signal was not measured in this measurement matrix. The $^{132}Ba/^{136}Ba$ was measured in a static mode while $^{135}Ba/^{136}Ba$, $^{137}Ba/^{136}Ba$ and $^{138}Ba/^{136}Ba$ were measured in dynamic mode. The precision achieved using this measurement matrix is about few ppm (internal precision) and for ^{135}Ba, ^{137}Ba and ^{138}Ba; and for ^{132}Ba it was ~ 40 ppm (internal). Since the number of measurements is too few we do not report external precision.

Results & Discussion: We studied Tagish Lake carbonaceous chondrite - a pristine organic-rich meteorite [6]. This is the first reported attempt to study Ba isotopes in this meteorite. We have previously reported Ba isotopic composition for bulk eucrite (Piplia Kalan) for Ba isotopes [7]. Tagish Lake meteorite has 2.94 wt.% [6] total organic carbon (TOC), three-times more than Murchison (1.03 wt.%). Our meteorite sample digestion experience shows that dissolution of eucrite is not as challenging as carbonaceous chondrites because of presence of organic matter. There are a number of methods available to dissolve carbonaceous chondrites such as alkaline fusion, HCl+boric acid method, and

repeated acid dissolution and Aqua Regia treatment. Even though alkaline fusion is capable of complete digestion [8] unfortunately it results in very high Ba blank [1]. The acid leaching technique offers the potential to isolate the Ba in different carrier phases without physically isolating the carrier phases.

The preliminary results from Tagish Lake are: The leach fractions from steps one and three had insufficient Ba for reliable isotopic analyses. Ba obtained from leach of Step 2 and residue was analyzed for 80 cycles and 200 cycles respectively. The signal strength from these samples was lower than the typical strengths obtained from standards. The $^{134}Ba/^{136}Ba$ measured values from these samples did not show any significant fractionation from standard values and are comparable in ranges with the values obtained from Ba standards. The leach fraction #2 showed excess in ^{135}Ba and ^{137}Ba of 276±4ppm and 176±5 ppm respectively. The ^{138}Ba and ^{132}Ba have normal values. The residue showed a complementary deficit, -345±2ppm in ^{135}Ba of and -167±2ppm in ^{137}Ba. The ^{138}Ba has normal value within errors, and ^{132}Ba has a deficit of -280±45ppm.

The results obtained from Murchison [9] offers a parallel for comparison. Both Murchison and Tagish Lake leach fractions display a positive anomaly in Ba isotopes 135, 137 and near normal values in 138. The excess in Tagish Lake leach is 50 times less than in Murchison, and it does not show excess in in ^{132}Ba. The Tagish Lake residue and Murchison residue show negative anomaly in 132, 135 and 137 Ba isotopes, and ^{138}Ba is normal in case of Tagish Lake. The trends of the anomaly are similar to earlier reported results for Murchison [4]. The composition of residue can be modeled as < 1% excess in s-process isotopes, while the leach can be modeled as a < 0.02% deficit in s process component.

Based on experimental data of leach and residue from Allende CAIs and assuming that the production ratio of a late input r-process isotopic component is constant the ratio of deviations of ^{135}Ba and ^{137}Ba isotopic ratios is estimated to be = 0.81 [10]. The excess in ^{135}Ba after subtracting the r process component can be attributed to sources other than r process. One possibility is the decay of short-lived radionuclide ^{135}Cs (half-life ~ 2.3 Ma). Assuming the Cs/Ba ratio measured in bulk rock Tagish Lake samples ~ 0.03 [6] as representative for the leach, and that initial $^{135}Ba/^{136}Ba$ was normal we estimate a $^{135}Cs/^{133}Cs$ model value of ~ 2×10^{-3}. This value is much higher than the value of ~ $(4.82 \pm 0.79) \times 10^{-4}$ reported earlier [10]. The estimates of $^{135}Cs/^{133}Cs$ for Tagish Lake are limited by the lack of specific measurements of elemental abundances of Cs and Ba in individual leach fractions. If the estimate of Cs and Ba abundances of leach using the bulk sample values underestimates the value of Cs/Ba then the ^{135}Cs abundance would be overestimated. To address some of the above issues we are leaching larger additional fractions of Tagish Lake fractions to reproduce and Ba isotope data and accurately estimate Cs/Ba ratio in individual fractions.

References : [1] Carlson et al. 2007 Science 316, 1175-1178. [2] Ranen & Jacobsen (2006) *Science, 314*, 809-812. [3] Andreasan and Sharma (2006) *Science 314* 806-809. [4] Hidaka et al. (2003) EPSL 214, 455-466 [5] Ali & Srinivasan (2011) IJMS, 299, 27-34 [6] Brown et al. (2000) Science 290, 320-325. [7] Srinivasan & Ali (2010) 73 MS meeting, #5344. [8] Yokoyama et al. (2007) *EPSL*, 259, 567-580 [9] Carlson et al. (2010) *LPS* #2415. [10] Hidaka et al. (2001) *EPSL* 193, 459-466.