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Insight into the Evolution of the Tagish Lake Carbonaceous Chondrite by Analysis of the Oxygen Isotopic Composition of Extracted Water and Mössbauer Spectroscopy.


Introduction: Elucidating the processes that have occurred on primitive meteorite parent bodies and understanding the relationship between different classes of primitive meteorites has been dominated in recent years by research into the processes of low temperature alteration [1-3]. The results of such alteration are exhibited particularly clearly in the highly altered matrix material in carbonaceous chondrites [4]. Tagish Lake, a carbonaceous chondrite consisting of over four hundred recovered fragments, was observed to fall in British Colombia on Jan 18th 2000 [5]. Initial analyses of this meteorite have resulted in its classification (based mainly upon mineralogy, oxygen isotopes and bulk chemical composition) as being intermediate between CM and CI meteorites [3] and suggest that Tagish Lake may be one of the most primitive Solar System materials yet studied. We have measured the oxygen isotopic composition of water extracted from samples of Tagish Lake using a recently developed stepped pyrolysis/fluorination system [6] and have found that the resulting yield and isotopic data more closely resembles patterns derived from CI meteorites than those from CM samples. However, Mössbauer spectroscopy suggests that anhydrous silicates may be much more abundant in Tagish Lake than in CI meteorites.

Analytical Technique: Measurements of $\delta^{17}$O and $\delta^{18}$O were made on water released by pyrolysis of crushed, bulk meteorite samples of 1-6 mg, using a continuous-flow, isotope ratio mass spectrometer (Delta C, Finnigan Mat). The samples used in this study were from a ~5g piece of pristine material which had been maintained at sub zero temperatures since initial impact. Sub-samples were removed while still frozen and were loaded within one hour of warming to room temperature. Water evolved from the samples was swept by a flow of purified helium directly onto the in-line fluorinating agent (CoF$_3$), releasing oxygen. Unwanted reaction products, mainly HF, were removed from the flow by chemical and cryogenic traps. After cryofocussing, the effluent passed through a capillary GC column for purification before mass spectrometer analysis. Samples were heated in a step-wise manner from room temperature to 800°C, with steps of either 33 or 50°C depending upon the anticipated yield. The heating duration at each step was five minutes. The 1σ precision of the data, as determined by repeat analysis of standard waters and an internal solid standard, are ±0.30 ‰ ($\delta^{18}$O) and ±0.14 ‰ ($\Delta^{17}$O). A Mössbauer spectrum was recorded from a sample of ~100mg sealed in a teflon bag at 298K [7].

Results: The Mössbauer spectrum showed a number of interesting features. We observe components consistent with magnetite, anhydrous silicates, sulphides and Fe-bearing clays. Given that Tagish Lake contains 19.3wt% Fe [5], these absorptions may be converted into wt% of iron bearing phases. Based on this analysis, Tagish contains 8.4wt% magnetite, similar to CIs (Orgueil has 10.6wt%, Alais 7.7wt% magnetite [8]) and the CM-like meteorite Essebi (8.9wt% [8]), but much higher than other CMs, typically ~1wt%. Anhydrous silicates are much more abundant in Tagish than in CI meteorites, approaching CM values, whilst Fe-bearing clays are proportionately lower in abundance than either CM or CI. Although the absorption is strongly overlapped, sulphides appear to be significantly more abundant in Tagish compared with either CM or CI meteorites, in contrast to the findings of [5], suggesting that the bulk of the sulphides may be present as a fine-grained matrix phase.

The water yield found during our stepped pyrolysis analyses was 3.5-4 wt%, approximately half that of CM meteorites and only about 1/3 of that from CI samples. The lower yield is in good agreement with the Mössbauer data suggesting that a larger proportion of the matrix consists of anhydrous silicate material rather than hydrated silicates. While releasing a much smaller quantity of water than the CIs the release profile (see figure) is otherwise similar to CI samples with two peaks in yield at about 150-200°C and 550°C. CM meteorites also exhibit 2 peaks in yield, however, the second peak is at the distinctly lower temperature of ~350°C. The range of $\delta^{18}$O values measured in Tagish Lake is far greater than that found in either CM or CI samples, spanning about 27‰ and having a plateau value at around +22‰. Despite the range, the majority of water was enriched in heavy isotopes and the bulk mean of +13.5‰ is approximately 10‰ heavier than for either CI or CMs [6].

Discussion: The apparent similarities between the CI meteorites and Tagish Lake perhaps suggest a genetic link between the two. Typical CI samples produce complex $\delta^{18}$O patterns suggesting multiple water reservoirs. In contrast Tagish Lake produces a much simpler...
pattern that may indicate mixing between two components, one dominant at low temperature with a $\delta^{17}$O between $-4$ and $0\%$e and a second, dominant at higher temperatures with a $\delta^{17}$O close to $+22\%$e. The lack of a complex $\delta^{17}$O release pattern may result from significantly less terrestrial weathering of the lower temperature minerals. In Tagish Lake samples the second peak in yield is somewhat larger than in CIs while the first peak is noticeably smaller, perhaps again, reflecting the relatively short period of time available for terrestrial alteration. The greatest similarity between Tagish Lake and the CI class of meteorites is in the $\Delta^{17}$O values. These were found to be very similar to those of CI samples, with lower temperature steps lying close to the terrestrial fractionation line and higher temperature steps rising to a poorly defined plateau of about $+1\%$e (see figure, top). The presence of terrestrial $\Delta^{17}$O values for steps up to ~250°C in CI samples had been presumed to be the result of isotopic exchange of low temperature phases with terrestrial water. However, the presence of this effect in Tagish Lake samples that had remained frozen since impact suggests that values close to the TF line may in fact be indigenous or that exchange with terrestrial oxygen occurs very quickly and extensively, possibly even at sub-zero temperatures.

A significant difference between CIs and Tagish Lake is the offset in the $\Delta^{17}$O values of Tagish Lake water and silicates. These display a large difference approaching 2\%e with high temperature water rising to +1\%e, while the bulk silicate values are -0.7 to -1\%e [5]. Models developed in recent years to account for the composition of silicates and fluids on the parent bodies of these meteorites have assumed isotopic starting compositions for both rock and fluid reservoirs. Fluids were assumed to have large positive values (both for $\Delta^{17}$O and $\delta^{18}$O) and pre-cursor silicates to have large negative values for both parameters [1-3]. Subsequent mixing and hydrothermal activity within the nebula and as part of a parent body then produced the range of isotopic values observed today. During reaction between the two reservoirs extreme values were moderated and final isotopic compositions were largely controlled by the different proportions of oxygen present in fluid and silicate reservoirs and how near to isotopic equilibrium the final mineral assemblage was. The parent material of CI meteorites appears to be have been dominated by fluids during alteration, with the final assemblage approaching isotopic equilibrium. This resulted in the final silicate and water isotopic compositions possessing positive values of $\Delta^{17}$O separated by only ~0.7\%e. CM meteorites suggest that parent material was apparently further from isotopic equilibrium with a difference of about 1.4\%e between the $\Delta^{17}$O values of the silicate and water components. The relatively large negative $\Delta^{17}$O values retained by the silicates (~2.5\%e) suggests that rock was the dominant source of oxygen during parent body hydrothermal activity. If such models are to successfully account for the isotopic composition of the silicate and water portions of Tagish Lake then reactions between the two must have fallen far short of attaining equilibrium.

References:

![Figure 1. Water yield and oxygen isotopic data from a stepped pyrolysis experiment with a 4mg sample of Tagish Lake.](1703.pdf)