CARRIERS OF HYDROGEN IN THE CM CARBONACEOUS CHONDRITES. M. R. Lee[1], B. E. Cohen[1,2] and A. Boyce[3], [1]School of Geographical & Earth Sciences, University of Glasgow, U.K. (Martin.Lee@Glasgow.ac.uk), [2]School of GeoSciences, University of Edinburgh, U.K., [3]Scottish Universities Environmental Research Centre, East Kilbride G75 0QF, U.K.

Introduction: Carbonaceous chondrite meteorites delivered water and other volatiles to the terrestrial planets early in their history [1]. The CMs are of particular importance in this context because they contain ~10 wt. % structural water [2], most of which is hosted by phyllosilicates and organic matter [3, 4]. The phyllosilicates are relatively rich in H that is isotopically light, whereas organic matter has a lower concentration of H that is isotopically heavier. The abundance of these two components is therefore a first order control on the H content of the CMs. Falls and finds also contain terrestrial H that is hosted by adsorbed molecular water and H_2O/OH-bearing alteration products such as goethite and gypsum [5, 6]. This terrestrial component can comprise a significant proportion of the total H budget of a CM [2, 7].

An important determinant of the contributions of the three carriers of H to the bulk abundance of isotopic composition of H is the nature and extent of parent body processing [1, 8]. In order characterise and quantify these effects we have analysed eleven meteorites with different degrees of aqueous alteration, and two that have undergone post-hydration heating. Using stepwise pyrolysis we have sought to distinguish and quantify the contributions of phyllosilicates, organic matter and adsorbed terrestrial water to the total H budget of each meteorite.

Materials and methods: Eleven CMs were analysed, and results are described in the context of three subsets. Subset A have been mildly aqueously altered: D'Angelo Bluff (DNG) 06004, Dominion Range (DOM) 08013, LaPaz Icefield (LAP) 02239, Lewis Cliff (LEW) 85311, MacKay Glacier (MCY) 05230, Queen Alexandra Range (QUE) 97990. Subset B samples have been highly aqueously altered: Allan Hills (ALH) 83100, Cold Bokkeveld, Scott Glacier (SCO) 06043. Subset C have been aqueously altered and undergone post-hydration heating: Elephant Moraine (EET) 96029, Pecora Escarpment (PCA) 91084.

The analytical work was undertaken at SUERC and using 33–79 mg chips. Samples were not preheated, but held under vacuum overnight prior to analysis. Each chip was incrementally heated at seven steps: 100, 200, 300, 400, 500, 700 and 1000/1100 °C (±5 °C) and held at each temperature for a minimum of 15 minutes. Hydrogen yields for each step were measured using the Pirani vacuum pressure gauge, and isotopic compositions using a VGI Optima mass spectrometer with reproducibility better than ± 2 ‰ for δD.

Results: The average amount of H evolved by each subset was: A = 0.85 ±0.08 wt. %; B = 1.19 ±0.05 wt. %; C = 0.73 ±0.26 wt. %. Figures 1-3 show the average proportion of total H measured in each temperature step (Figs 1a, 2a, 3a), and its isotopic composition (Figs 1b, 2b, 3b).
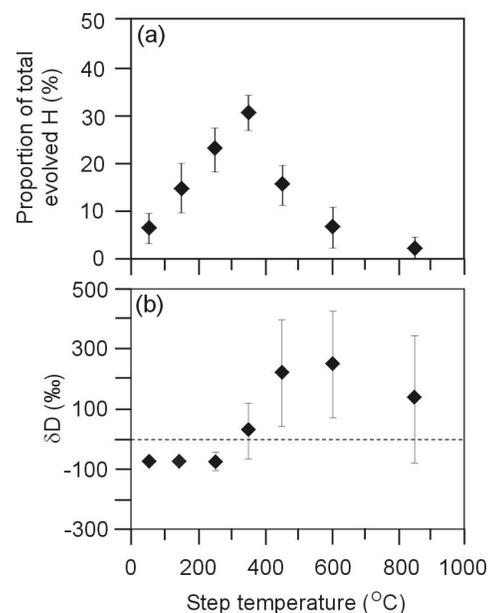

Fig. 1. Stepwise pyrolysis results for the mildly aqueously altered CMs of subset A. Averages of six samples are plotted. Error bars 1σ.

For the mildly aqueously altered meteorites of subset A the proportion of total H evolved increases in each step to 300–400 °C, then falls at higher temperatures. Average δD varies little in the first three steps but becomes significantly heavier as temperatures rise (Fig. 1). The three highly aqueously altered CMs of subset B show a progressive increase in the amount of H evolved with temperature, reaching a peak in the 500–700 °C step; δD becomes progressively lighter over all seven steps (Fig. 2). The two subset C samples show no systematic pattern in the proportion of H that is evolved with increasing temperature. δD varies over a relatively narrow range (-70 to 34 ‰), becoming heavier over the first six steps (Fig. 3).
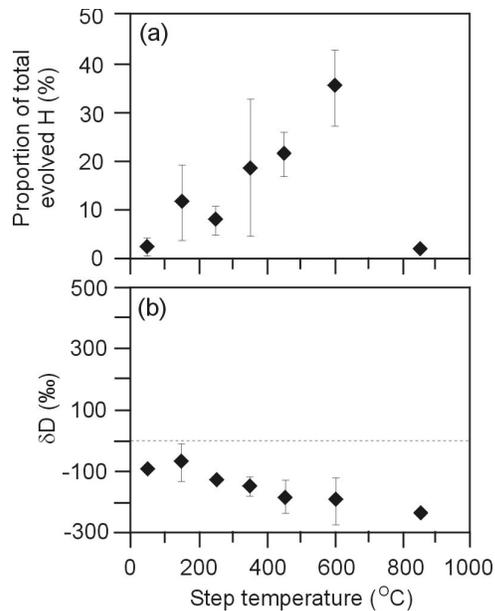
Fig. 2. Stepwise pyrolysis results for the heavily aqueously altered CMs of subset B. Averages of three samples are plotted. Error bars 1σ.

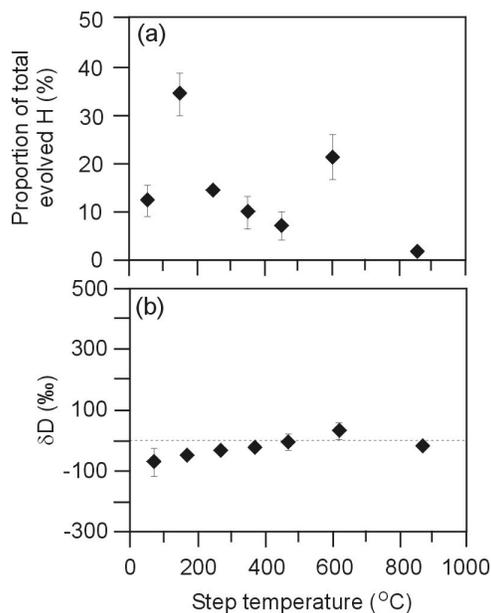

Fig. 3. Stepwise pyrolysis results for post-hydration heated CMs of subset C. Averages of two samples are plotted. Error bars 1σ.

Discussion: The three subsets are distinct in the abundance and isotopic composition of evolved H. The proportion of H evolved from subset A is highest at 300–400 °C, which is consistent with phyllosilicate dehydroxylation, as is the relatively light δD. Hydrogen evolved at higher temperatures is isotopically

heavier, showing that organic matter makes a greater contribution. The largest release of H from subset B, again inferred to represent phyllosilicate dehydroxylation, is at 500–700 °C. Differences in dehydroxylation temperature between subsets A and B may in part relate to contrasts in phyllosilicate chemical composition and mineralogy. The progressive fall in δD across the whole temperature range shows that organic-matter derived H comprises a small proportion of the H budget of highly aqueously altered CMs (Fig. 2).

The largest proportion of H evolved from the two CMs of subset C was at 100–200 °C (δD -26 ‰), which is consistent with being sourced from loosely bound molecular water [9]. δD is ~0 ‰ at all steps indicating that much of the phyllosilicate- and organic matter-hosted H was removed by post-hydration heating so that terrestrial water is a major carrier of H.

Quantifying adsorbed terrestrial water: Most of the H evolved at <200 °C by the eleven CMs is likely to be from terrestrial water, which would be consistent with its isotopic composition (average δD -75 ±21 ‰ and δD -76 ±22 ‰ for the 100 °C and 200 °C steps, respectively). This terrestrial H comprises 4–33 % (average 19 %) of the total H of subsets A and B. These results agree well with 11–26 % hydration water (average 18 %) measured in five CMs by [2]. By contrast, this terrestrial component comprises 46% and 47% of total H measured from the two post-hydration heated samples of subset C.

Conclusions: Stepwise pyrolysis of CM carbonaceous chondrites can effectively distinguish H that is hosted by: (i) phyllosilicates, (ii) organic matter, (iii) adsorbed terrestrial water. The latter comprises ~20 % of the total H of aqueously altered CMs, and ~50% of those that have been post-hydration heated. This high proportion of terrestrial H underscores the importance of samples returned from Ryugu and Bennu for determining the H isotopic composition of carbonaceous chondrite material uncontaminated by terrestrial water.

References: [1] Alexander C.M.O'D et al. (2012) *Science* 337, 721–723. [2] Jarosewich E. (1990) *Meteoritics* 25, 323–337. [3] Robert F. and Epstein, S (1982) *Geochim. Cosmochim. Acta* 46, 81–95. [4] Eiler J. M. and Kitchen N. (2004) *Geochim. Cosmochim. Acta* 68, 1395–1411. [5] Velbel M. A. (1988) *Meteoritics* 23, 151–159. [6] Lee et al. (2021) *Meteoritics & Planet. Sci.* (in press). [7] Vacher L. G. et al. (2020) *Geochim. Cosmochim. Acta* 281, 53–66. [8] Alexander C.M.O'D et al. (2013) *Geochim. Cosmochim. Acta* 123, 244–260. [9] Garenne A. et al. (2014) *Geochim. Cosmochim. Acta* 137, 93–112.