Oxygen and carbon isotopic ratios of calcite in the Nogoya CM chondrite

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CM chondrites exhibit evidence for aqueous alteration to variable degrees. Carbonate is a secondary mineral of aqueous alteration. Detailed petrological and mineralogical observations along with O-isotope measurements have been carried out by previous studies. These studies have suggested that carbonates in CM chondrites did not form in a single event but formed intermittently (Tyra et al., 2012; Lee et al., 2013). In this study, we conducted in-situ O- and C-isotope measurements on calcite grains in the Nogoya CM 2.2-2.3 chondrite. Isotope measurement on carbonates in multiple "generations" could shed light on the evolution of O and C isotopic compositions during aqueous alteration.

We found many calcite grains by SEM observation. As reported in the previous studies (e.g., Lee et al., 2014), two types of calcite grains with distinct mineralogical characteristics were found. Following the definition by Lee et al. (2014), we describe these calcite grains as type 1 and 2 grains. Most type 1 grains are single crystals and have serpentine/tochilinite rims. On the other hand, type 2 grains are polycrystalline and microporous, and do not have rims.

Oxygen isotopic compositions are highly different between type 1 and 2 grains, but similar within each type ( $d^{18}O = 34.7$  %(type 1) and 19.3 %(type 2) on average). The average  $D^{17}O$  values are -2.5 % (type 1) and -5.4 %(type 2). The  $d^{18}O$  and  $D^{17}O$  values indicate that type 2 calcite formed later than type 1 calcite, because progressive alteration led to 0-isotope exchange between water and anhydrous silicate with lower  $d^{18}O$  and  $D^{17}O$  values than water. Carbon isotopic ratios of type 1 grains are similar ( $d^{13}C = 31.8$  %on average), whereas type 2 grains have variable  $d^{13}C$  values ranging from 28.8 to 61.2 %. These observations indicate an increase in  $d^{13}C$  values in later stages of aqueous alteration.

It has been suggested that the Rayleigh-type isotopic fractionation driven by the escape of  $^{13}$  C-poor CH $_4$  could have led to  $^{13}$ C enrichments of the dissolved inorganic C (Guo and Eiler, 2007). The observed d $^{13}$ C increase of  $^{\sim}30$  &can be explained if  $^{\sim}28$  % of the dissolved C was reduced to produce CH $_4$  and was lost at 28  $^{\circ}$ C. If true, aqueous alteration in CM chondrites would have occurred in an open system at least in later stages.

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