Implications of Meteoritic $^{36}$Cl Abundance for the Origin of Short-Lived Radionuclides in the Early Solar System. L. A. Leshin$^{1,2}$, Y. Guan$^3$, and Y. Lin$^3$, Department of Geological Sciences, $^1$Center for Meteorite Studies, Arizona State University, Tempe AZ 85287-1404, USA, $^2$Inst. of Geology and Geophysics, Chinese Academy of Sciences, Beijing 100029, China. (laurie.leshin@asu.edu).

Introduction: Short-lived, now extinct radionuclides detected in primitive meteorites have revealed important information about events in the earliest history of our solar system. They can be used as fine-scale chronometers to trace processes in the solar nebula and on meteorite parent bodies [1]. The relative abundances of different short-lived isotopes in the early solar system can also be used to constrain the local galactic environment of solar system formation because the different production mechanisms (i.e., supernova or AGB stars vs. “local production” scenarios) result in different abundance patterns of short-lived radionuclides. Finally, the different chemical affinities (e.g., volatile vs. refractory) of short-lived radionuclides provide an opportunity to track unique chemical processes in the early solar system. Thus, investigation of short-lived radionuclides, including the search for evidence of “new” short-lived isotopes (those not positively detected previously) has the potential to shed light on many important questions in cosmochemistry. Recently, we reported the first direct evidence for the presence of $^{36}$Cl in meteorites by detecting its in situ decay product – $^{36}$S excess, in alteration assemblages of a Ca-Al-rich inclusion (CAI) from the Ningqiang carbonaceous chondrite [2]. $^{36}$Cl has a half-life of 0.3 million years (My) and decays to either $^{36}$Ar (98.1%, $\beta^-$) or $^{36}$S (1.9%, $\varepsilon$ and $\beta^+$) [3]. Here we combine the evidence for live $^{36}$Cl with $^{26}$Al-$^{26}$Mg systematics and petrologic observations of the CAI to report the “canonical” $^{36}$Cl abundance, and we explore the implications of the presence of live $^{36}$Cl for models for the origin of short-lived radionuclides in our solar system.

Results and Discussion: Alteration assemblages, consisting of sodalite (Na$_8$Al$_6$Si$_6$O$_{24}$Cl$_2$) and nepheline (Na$_4$Al$_4$Si$_4$O$_{16}$), were studied in three CAIs from the Ningqiang carbonaceous chondrites and two EH3 enstatite chondrites – EET87746 and ALH 77295. Sulfur isotopes were analyzed in the ASU GeoSIMS lab, along with correlated investigation of the $^{26}$Al-$^{26}$Mg system. The sodalite contains high Cl (7.5 wt%) with no detectible sulfur (<0.06 wt% from the electron probe), and therefore represents the best available phase to search for evidence of now decayed $^{36}$Cl.

The fine-grained alteration assemblages in the two CAIs from the enstatite chondrites showed no resolvable $^{36}$S excess because of their low $^{35}$Cl/$^{34}$S ratios (<100). Ion images revealed the presence of fine-grained hot spots of $^{36}$S, suggesting the presence of tiny sulfides, which would mask evidence of $^{36}$Cl. Four alteration assemblages in the Ningqiang CAI (NQJ1-1#1) with high $^{35}$Cl/$^{34}$S ratios (up to 57,000) showed clear $^{36}$S excesses that correlate with $^{35}$Cl/$^{34}$S ratios. The inferred ($^{36}$Cl/$^{35}$Cl)$_0$ ratios ($\pm 2\sigma$) are: (4.6 $\pm$ 0.6) $\times$ 10$^{-6}$, (5.1 $\pm$ 1.0) $\times$ 10$^{-6}$, (7.7 $\pm$ 2.5) $\times$ 10$^{-6}$, and (1.1 $\pm$ 0.2) $\times$ 10$^{-5}$, respectively. The two assemblages with the highest Cl/S ratios yield the best determination of the initial ($^{36}$Cl/$^{35}$Cl)$_0$ ratios of $\sim$5$\times$ 10$^{-6}$ in sodalite.

The inferred ($^{36}$Cl/$^{35}$Cl)$_0$ ratios ([5–11] $\times$ 10$^{-6}$) of the alteration assemblages of the Ningqiang CAI are about 4 to 8 times higher than a previous estimate (1.4 $\pm$ 0.2) $\times$ 10$^{-6}$, which derived from measurements of $^{36}$Ar excess in matrix of the Efremovka carbonaceous chondrite [4]. There are two possible explanations for the difference. First, the vast majority (94-96%) of $^{36}$Ar in meteorites is a mixture of a trapped component, a spallation component, and the decay of cosmogenic $^{36}$Cl (i.e., $^{36}$Cl produced by recent irradiation by cosmic rays). Estimation of $^{36}$Ar excess from the decay of short-lived $^{36}$Cl is highly uncertain, because it only can be done after subtraction of these components, especially the most abundant trapped component. Furthermore, recent study of isotopic compositions of noble gases in meteorites showed a large variation of $^{38}$Ar/$^{36}$Ar ratios due to experimental artifact, requiring a reassessment for the entire reported $^{36}$Ar excess [5]. Thus, the most likely explanation is that the previous report of $^{36}$Cl decay products was inaccurate. However, it is possible that the difference may represent a time interval between the alteration of the CAI and formation of Cl-bearing phases in Efremovka matrix. Assuming homogeneous distribution of $^{36}$Cl, the difference corresponds to a time interval of 0.6-1.2 My.

A previous petrographic and mineralogical study of the Ningqiang CAI [6] showed that sodalite and nepheline are the products of secondary processes that altered the primary minerals in the inclusion. Therefore, the ($^{36}$Cl/$^{35}$Cl)$_0$ ratios in-
ferred from the sodalite analyses cannot represent the initial value at the time when CAIs formed. To deduce the “canonical” ($^{36}\text{Cl}/^{35}\text{Cl}$)$_0$ value, the time interval between the formation and alteration of the Ningqiang CAI was examined using the $^{26}\text{Al}/^{26}\text{Mg}$ system in each individual mineral phase, and petrographic relationships among the CAIs in Ningqiang [6]. Although “canonical” $^{26}\text{Al}$ levels were detected in the unaltered melilite-spinel-rich crust, consistent with previous observations in Ningqiang CAIs, neither anorthite nor the alteration assemblages in the mantle of NQJ1-1#1 show resolvable $^{26}\text{Mg}$ excess, providing a maximum inferred ($^{26}\text{Al}/^{27}\text{Al}$)$_0$ ratio of $\sim 0.7 \times 10^{-4}$. Assuming the ($^{26}\text{Al}/^{27}\text{Al}$)$_0$ ratios of melilite and sodalite reflect a temporal difference, then sodalite formed at least 1.5 – 2 Myr after the formation of melilite. Using this time difference and the best ($^{36}\text{Cl}/^{35}\text{Cl}$)$_0$ value ($\sim 5 \times 10^{-6}$) of sodalite, the “canonical” ($^{36}\text{Cl}/^{35}\text{Cl}$)$_0$ ratio at the time when CAIs first formed is then inferred to be $\geq 3 \times 10^{-4}$.

This ($^{36}\text{Cl}/^{35}\text{Cl}$)$_0$ value can be used to constrain the source of short lived radionuclides and the setting of solar system formation. If from a stellar source, supernova model predictions of the ($^{36}\text{Cl}/^{35}\text{Cl}$)$_0$ value range from $3 \times 10^{-6}$ to $2 \times 10^{-5}$ [7], whereas low-mass AGB star models suggest much lower values ($\leq 7.85 \times 10^{-7} - 2.4 \times 10^{-6}$)[8,9]. The initial ($^{36}\text{Cl}/^{35}\text{Cl}$)$_0$ ($\geq 3 \times 10^{-4}$) at CAI formation estimated in this study is clearly significantly higher than the suggestion from the AGB star models, but consistent with the predicted values from a supernova source.

Alternatively, in a local irradiation model an upper limit of ($^{36}\text{Cl}/^{35}\text{Cl}$)$_0 = 1.3 \times 10^{-4}$ has been predicted [10], which, although below our lower limit, is at least broadly consistent with the observation from this study. However, the volatile nature of chlorine and the observation of $^{35}\text{Cl}$ in sodalite are probably not compatible with the theory of local irradiation origin. According to the models of Shu et al. [e.g., 11, 12], intense irradiation of the proto-Sun evaporated Mg-Fe-silicate dustballs to form CAIs, and simultaneously produced short-lived radionuclides (e.g. $^{10}\text{Be}$, $^{26}\text{Al}$, $^{41}\text{Ca}$ and $^{53}\text{Mn}$) through the bombardment of CAI materials by energetic particles from the Sun. After their formation, CAIs bearing short-lived radionuclides were ejected to distant locations where chondrites accreted. Sodalite, a product of secondary alteration at relatively low-temperature, formed after the CAI primary phases, and in a different environment. Therefore, in the local irradiation model, $^{36}\text{Cl}$ itself would have to be produced in a gaseous phase very close to the proto-Sun by intense irradiation, and then a mechanism to transport the gaseous $^{36}\text{Cl}$, coupled with solids, from the CAI formation region to distant chondrite forming regions must be invoked. Finally, the transported gaseous $^{36}\text{Cl}$ would have to be incorporated later into alteration assemblages. According to the local irradiation model [11], however, gases and small particles would be thrown into interstellar space with finite escape speeds, decoupled from the large CAIs that fell back to the disk. Therefore, the observation of $^{36}\text{Cl}$ in this study presents a serious challenge to the local irradiation models in which the irradiation takes place, and the CAIs form, very near the young Sun. Rather, it is more consistent the hypothesis that the solar system formed in proximity to a massive star which supplied the short-lived isotopes to the forming solar system when it went supernova [e.g., 13].

Unlike other short-lived radionuclides (e.g. $^{26}\text{Al}$, $^{41}\text{Ca}$, and $^{53}\text{Mn}$), chlorine is a volatile element, and it is closely associated with secondary alteration observed in CAIs, chondrules and matrix. The timing (and location) is an important issue in understanding volatile-rock interactions commonly observed in most major components of primitive meteorites. Because of its short half-life, $^{36}\text{Cl}$ may serve as a new chronometer with fine resolution especially for low temperature events in the nebula and/or on asteroidal bodies. The mere observation of $^{36}\text{Cl}$ in sodalite suggests that secondary alteration of at least this CAI occurred relatively shortly after CAI formation. However, new work to follow up this discovery, analyzing Cl-bearing phases from other meteorite groups and in other petrographic settings, is needed. The data could reveal an important record of nebular and parent body alteration, help trace the earliest history of volatiles in our solar system, and shed light on the origin of the short-lived radionuclides in the solar system.