THE LITHIUM AND BORON ISOTOPIC COMPOSITIONS OF CHONDRULES. M.-C. Liu⁴ and M. Chaus-ssidon², ¹Dept. of Earth, Planetary, and Space Sciences, UCLA, Los Angeles, USA (mcliu@ucla.edu), ² Institut de Physique du Globe de Paris, Sorbonne, Paris Cité, Univ. Paris Diderot, UMR 7154 CNRS, F-75005 Paris, France (chaussidon@ipgp.fr).

Introduction: Spallation reactions between energetic (GeV) protons and alpha particles in the Galactic Cosmic Rays and C-N-O nuclei in the interstellar medium are the main pathway to synthesize boron isotopes in the universe [1]. However, this process produces a characteristic $^{11}$B/$^{10}$B $\approx 2.5$ [2,3], too low to account for the Solar System $^{11}$B/$^{10}$B $= 4.04$, inferred from the measurements of CI chondrites [4]. Two hypotheses aiming to solve this long standing issue have been proposed. The first one involves spallation of hydrogen in the molecular cloud by low (MeV) energy $^5$He and $^5$Li over that of $^{11}$B. While none of the above two hypotheses could be confirmed by astronomical observations, isotopic measurements of chondrules showing large $^{11}$B-excesses (up to 50‰) correlating with the B contents have led to the idea that low-E spallation would have been responsible for additional $^{11}$B incorporated into the Solar System [7]. Interestingly, a similar study by [8] found no $\delta^{11}$B variations in chondrules outside the analytical uncertainties of $\pm 25%$. These controversial results beg the question of how widespread the boron isotope heterogeneity is, as this signature could shed light on the origin of boron in the Solar System.

Lithium isotopes could potentially help to discern which mechanism most likely accounted for the extra $^{11}$B in the Solar System. Despite multiple astrophysical sources for lithium isotopes, a significant portion of this element in the universe was also derived from charged particle spallation on C-N-O, with the production $^{7}$Li/$^{6}$Li ratio being $\approx 1$–2 (c.f. solar $^{7}$Li/$^{6}$Li $= 12.02, [9])$, regardless of the irradiation energy [10]. Therefore, if solar $^{11}$B/$^{10}$B $= 4.04$ truly resulted from mixing between GCR and low-E irradiation components, sub-solar $^{7}$Li/$^{6}$Li should be associated with non-solar $^{11}$B/$^{10}$B. In contrast, if the Solar System acquired extra $^{11}$B from neutrino spallation during supernova explosion, components enriched in both $^{11}$B and $^{7}$Li would be expected because lithium synthesized by this process is essentially all $^{7}$Li [6].

To better constrain the origin of boron in the Solar System, we have performed coordinated lithium and boron isotope measurements of 5 chondrules on the UCLA CAMECA ims-1290 ion microprobe. Here we report the new data and discuss their implications.

Experimental: Chondrules analyzed here are typical Type-I porphyritic olivine chondrules ($\approx 700$ µm in size) found in the Allende meteorite. The interior of olivine crystals is characterized by a Mg# > 97. All chondrules have been previously measured for oxygen isotopes, and $\Delta^{17}$O in olivine within individual chondrules was found to be homogeneous to the level of 0.15‰ [2,11].

Lithium and boron isotope measurements were conducted on the UCLA CAMECA ims-1290 ion microprobe. The polished thick meteorite section was sputtered with a 23 keV, 10 nA $^{16}$O$^-$ ion beam (ϕ = 5µm) generated by the Hyperion-II oxygen source. Mass resolution was set at 2,500 to separate $^{18}$Mg$^{++}$ and $^{28}$Si$^{++}$ from the peaks of interest. Secondary ions were measured by using the axial electron multiplier (EM) in peak hopping mode with the following mass sequence: $^{5}$Li, $^{7}$Li, $^{9}$Be, $^{10}$B, $^{11}$B, and $^{28}$Si$^{++}$. Instrumental mass fractionation was corrected for by using an NBS 616 glass. Final lithium and boron isotopic ratios were calculated by using the total counts [12], and expressed in delta notation ($\delta^{7}$Li and $\delta^{11}$B, normalized to chondritic $^{7}$Li/$^{6}$Li = 12.02 [9] and $^{11}$B/$^{10}$B = 4.04 [4]).

We focused on the data from the interior of olivine; therefore, spots overlapping grain boundaries were excluded from data reduction. This is a major improvement compared to [7], where many spots were mixture of olivine and mesostasis, the latter of which being possibly prone to secondary alteration that affected initial lithium and boron isotopic signatures.

Result and discussion: Large $\delta^{7}$Li and $\delta^{11}$B variations were found in chondrule olivine. All spots are isotopically heavy in $\delta^{7}$Li (up to +40‰), which appears to be correlated negatively with the Li/Si ratio (Fig. 1a). In contrast, $\delta^{11}$B ranges from $\pm 40%$ to $+70%$, although most data points are consistent with being chondritic within errors. The variation range increases as the B/Si ratio decreases, with the highest $\delta^{11}$B value being $73\pm 44%$, similar to that found by [7] (Fig. 1b). Interestingly, $\delta^{7}$Li and $\delta^{11}$B are not obviously correlated with one another, as was also observed previously [13]. Given the very low beryllium abundances, radiogenic $^{7}$Li and $^{10}$B from the decay of short-lived $^{7}$Be ($t_{1/2} = 53$ days) and $^{10}$Be ($t_{1/2} = 1.3$ Myr) are negligible.

There are three possible explanations for the observed lithium and boron isotope variations: (1) Isotope fractionation associated with evaporation during melting, (2) mixing of chondritic and solar energetic particle (SEP) produced lithium and boron recorded by chondrule precursors, and (3) mixing between $\nu$-process and GCR-irradiation components recorded by chondrule precursors. In the following sections, we examine each
mechanism and argue that our findings here can be best understood in scenario (3).

Isotope fractionation during melting: It has been experimentally determined that evaporation under vacuum or in a low-pressure hydrogen-dominated gas during melting could fractionate isotopes, leaving isotopically heavy compositions in the residues [14]. This process can explain the relationship between $\delta^7$Li and Li/Si quantitatively, but fails to account for that between $\delta^{11}$B and B/Si. In addition, the lack of correlation between $\delta^7$Li and $\delta^{11}$B argues against evaporation processes being the main mechanism resulting in the observed isotopic variations. Lithium diffusion into olivine from the melt (or the mesostasis) is another potential source of lithium isotopic variation [15]. However, this would not produce the trends shown in Fig. 1a. In addition there is no relationship observed in the olivine between $\delta^7$Li and distance to the mesostasis.

Mixing of chondritic and SEP-produced lithium and boron recorded by chondrule precursors: This process can account for both positive and negative $\delta^{11}$B values in chondrule olivine if low-E and high-E irradiated material, respectively, mixed with a chondritic component to form chondrule precursors. However, as stated previously, the irradiation production ratio of $^7$Li/$^6$Li is $\sim$1–2, independent of the energy. If chondrule precursors represent a mixture of irradiated and chondritic components, they should be characterized by sub-chondritic $^7$Li/$^6$Li. The absence of negative $\delta^7$Li values in the data implies that irradiation components, if any, would have been largely diluted or erased during the formation of chondrule precursors.

Mixing between $\nu$-process and GCR-irradiation components recorded by chondrule precursors: This scenario requires that chondrule precursors represent a mixture of material derived from the $\nu$-process, GCR irradiation, and a chondritic reservoir. Components enriched in $^7$Li and $^{11}$B would have originated from the $\nu$-process, and the former could easily erase GCR-produced $^7$Li/$^6$Li isotopic signatures since lithium produced in the $\nu$-process is all $^7$Li. The slightly negative $\delta^{11}$B values observed here could have been a result of mixing between GCR-produced and $\nu$-process $^{11}$B/$^{10}$B. Although this scenario qualitatively explains the observed lithium and boron isotope variations in chondrules, one caveat is that the nucleosynthetic yields of lithium and boron isotopes in the $\nu$-process are still highly model-dependent and sensitive to the input parameters. More detailed understandings of physics of supernova explosion are certainly required for more accurate modeling of lithium and boron isotope evolution in the Solar System.


Figure 1. (a) $\delta^7$Li vs Li/Si. A strong negative correlation between the isotopic composition and elemental abundance is observed. (b) $\delta^{11}$B vs B/Si. Although the larger isotope variations are seen at lower elemental abundances, the two are not obviously correlated. The yellow circles represent chondritic compositions.