



**Supplementary Fig. S3** Plot of the REE + Y atoms vs. Na atoms (both per formula unit (apfu), based on 56 oxygen atoms) in the analyzed merrillite grains, relative to literature data. REE substitution of Ca within the merrillite crystal structure requires charge balance, which then in turn leads to vacancies that were initially occupied by Na in the ideal merrillite structure (Jolliff et al. 2006). Hence, eucritic and lunar merrillite high in REE, show very low Na atoms per formula unit. While mesosiderites also show very low Na content (<0.75 apfu.), the remaining, non-Martian meteorites exhibit higher Na abundances close to the ideal formula of 2 atoms per 56 oxygens. Figure after Jolliff et al. (2006; Figs. 5 and 6) and Shearer et al. (2015; Fig. 8).

**References:**

- Jolliff, B.L., Hughes, J.M., Freeman, J.J., and Zeigler, R.A. (2006) Crystal chemistry of lunar merrillite and comparison to other meteoritic and planetary suites of whitlockite and merrillite. *American Mineralogist*, 91 (10), 1583–1595.
- Shearer, C.K., Burger, P.V., Papike, J.J., McCubbin, F.M., and Bell, A.S. (2015) Crystal chemistry of merrillite from Martian meteorites: Mineralogical recorders of magmatic processes and planetary differentiation. *Meteoritics & Planetary Science*, 50 (4), 649–673.