This study investigates the use of the $^{40}$K-$^{40}$Ca system as a tracer to better quantify the contributions of silicate and carbonate lithologies in the dissolved load of major Himalayan rivers. Previous work using Sr isotopes as a proxy for silicate weathering has been complicated by the redistribution of radiogenic $^{87}$Sr between silicate and carbonate lithologies, particularly in the Lesser Himalaya, where dolomites exhibit $^{87}$Sr/$^{86}$Sr ratios as high as 0.85. The $^{40}$Ca signature of carbonates, on the other hand, appears to be remarkably resistant to metamorphism and dolomitization [1]. It was therefore anticipated that the $^{40}$K-$^{40}$Ca system could circumvent issues associated with such secondary events, and yield more robust constraints on the relative contribution of silicate vs. carbonate lithologies in dissolved river loads.

The main difficulty in applying the $^{40}$K-$^{40}$Ca decay scheme as a tracer lies in the analytical precision required to measure small variations ($\sim 1 \varepsilon$-unit) on the large $^{40}$Ca isotope (96.9%). This difficulty can now be overcome using the Finnigan Triton TIMS, which allows measurements of the $^{40}$Ca/$^{44}$Ca ratio with external precision of 0.35 $\varepsilon$-unit in multidynamic mode. Using this method, we generated high-precision $^{40}$Ca data on carbonates/dolomites, bedload sediments, dissolved load, and clay samples originating from and representing the main litho-tectonic units of the Himalaya. Our results show that metamorphosed dolomites from the Lesser Himalaya (LH) exhibit no radiogenic $^{40}$Ca excess despite highly variable $^{87}$Sr/$^{86}$Sr signatures (0.73-0.85). Thus, all Himalayan carbonates appear to be characterized by a homogeneous $\varepsilon^{40}$Ca=0. In contrast, silicate material is radiogenic, with $\varepsilon^{40}$Ca averaging +1 in the Tethyan Sedimentary Series (TSS), +1.6 in the High Himalaya crystalline (HHC) and +4 $\varepsilon$-units in the LH. Results obtained from a series of 35 Himalayan rivers (including the Brahmaputra, Ganga and its main tributaries) show that $\varepsilon^{40}$Ca in the dissolved load is significantly influenced by silicate weathering, with $\varepsilon^{40}$Ca ranging from +0.1 in rivers draining carbonate dominated catchments to +1.6 $\varepsilon$-units in rivers draining predominantly gneissic catchments of the High Himalaya. No simple relation exists between $^{87}$Sr and $^{40}$Ca systematics, which likely reflects the decoupling of Rb-Sr and K-Ca systems in LH dolomites. In contrast, $^{40}$Ca signatures correlate well with proxies of carbonate weathering such as Ca/Na or Mg/Na ratios. Overall, our results indicate that the $^{40}$Ca signature of Himalayan rivers primarily reflects the lithological nature of their erosional source, and highlight the significant contribution of HHC gneisses to the dissolved calcium budget of the Ganga and Brahmaputra.