

# Non-conservative Behavior of Lithium during Estuarine Transport: Impact on Seawater Li budget

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We report significant non-conservative behavior of lithium, in both concentration ( $[Li]_{Dissolved}$ ) and isotope space ( ${}^7Li_{Dissolved}$ ), during low flow in the Ganga River Estuary. Li demonstrates a seasonally (monsoon vs non-monsoon) contrasting behavior, with non-monsoon samples demonstrating +0.8–3.1‰ deviation of estuarine  ${}^7Li_{Dissolved}$  from conservative mixing line. This observation, if true for global rivers will have significant implication on the riverine mass and isotope flux of Li to seawater, and thus the seawater Li mass budget.

The secular changes in  ${}^7Li_{Seawater}$  over the Cenozoic is primarily linked to variations in continental and seafloor chemical weathering vs marine reverse weathering processes. This inference relies on the assumption that fluvial lithium is conservatively transported across estuaries. However, variation in Eh-pH, ionic strength, and SPM load in large river estuaries results in: (i) precipitation/solubilization of metals, (ii) adsorption-desorption, and (iii) secondary clay formation/dissolution. Benthic boundary fluxes can also impact the fluid phase composition in estuaries. We determined the  $[Li]$  and  ${}^7Li$  of seasonally and spatio-temporally resolved samples from the Hooghly River Estuary, spanning a salinity range of 0.1–29 psu. Samples from 80 different locations covered dissolved phase, suspended and bedload sediments including their exchangeable, oxide, carbonate, clay, and residue fractions. Groundwater and shallow (0-50 cm) porewater samples were analysed to constrain possible benthic boundary flux.

During low flow (non-monsoon), the  ${}^7Li_{dissolved}$  is enriched in  ${}^7Li$  by 0.8-3.1‰ in the low-mid salinity range of 0.3–3.0 psu. Whereas  $[Li]_{dissolved}$  demonstrates a slightly muted non-conservative behavior with -6 to -14% offset from the mixing line. Uptake of Li in the water column by secondary clays, with a fractionation factor ( $\alpha_{clay - fluid}$ ) of  $0.96849 \pm 0.0006$  can account for the non-conservative behavior. Relatively high non-monsoon residence time of materials in the estuary enables such fluid-particle interactions at low SPM load (150-200 mg/L). Whereas a conservative behavior of Li in both concentration and isotope space is observed during monsoon, despite extremely high SPM load (~2 g/L). We hypothesize that the lack of manifestation of fluid-particle interaction on the fluid phase chemistry of estuary is due to significantly short residence time during monsoonal high freshwater discharge.