

# **Lithium concentrations and isotopic compositions of ancient shales trace continental silicate weathering across the Archean-Proterozoic boundary**

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Li concentrations and isotopic compositions and major and trace elements were measured for 34 shale samples from the Witwatersrand Supergroup and Transvaal Supergroup, Kaapvaal craton, South Africa, with depositional ages ranging from 2960 to 2250 Ma, to investigate the compositional evolution of continental crust and weathering conditions across the Archean-Proterozoic boundary. The  $\delta^7\text{Li}$  values of the shales range from -4.3 to +4.8 ‰ and are mostly lower than those of average MORB ( $+3.7 \pm 2$  ‰,  $2\sigma$ ). The [Li] values of the shales, ranging from 6 to 72  $\mu\text{g/g}$  are higher than those of average MORB ( $6.2 \pm 2.4$   $\mu\text{g/g}$ ,  $2\sigma$ ), whereas mostly lower than those of average PAAS ( $63.0 \pm 25.0$   $\mu\text{g/g}$ ,  $2\sigma$ ). The CIA average of 81 for the shales is significantly higher than the average upper continental crust (59-65) and PAAS (69). Combining these data with data from other sedimentary rocks (i.e., shales, tillites, pelites, etc) from different geologic periods, it appears that the low  $\delta^7\text{Li}$  values are generally accompanied by high CIA and  $\text{Al}_2\text{O}_3/\text{SiO}_2$  values, reflecting a continental silicate weathering trend. The  $\delta^7\text{Li}$  and CIA values of the Archean sedimentary rocks (with an average of  $-1.0 \pm 4$  ‰ ( $2\sigma$ ) and  $81 \pm 18$  ( $2\sigma$ ), respectively) are systematically lower and higher, respectively, than those of the Paleoproterozoic sedimentary rocks (with an average of  $0.3 \pm 3.6$  ‰ ( $2\sigma$ ) and  $75 \pm 24$  ( $2\sigma$ ), respectively). These results demonstrate that the Archean crust experienced more intense weathering than Paleoproterozoic crust, supporting a mainly warm and humid Archean climate condition. Although the combined [Li] values generally increase with the increase of CIA values and the decrease of  $\delta^7\text{Li}$  values, indicating uptake of lithium by clay minerals during silicate weathering, the Archean sedimentary rocks exhibit lower [Li] values (with an average of  $27.0 \pm 31.6$   $\mu\text{g/g}$ ,  $2\sigma$ ) than those of the Paleoproterozoic sedimentary rocks (with an average of  $42.1 \pm 43.6$   $\mu\text{g/g}$ ,  $2\sigma$ ). This may suggest clay mineral surface-adsorbed lithium experienced continuous flushing and leaching by acid rainfall during the Archean period.