

## The onset of oxidative weathering traced by uranium isotopes

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The invention of photosynthesis was a key moment in Earth history, initiating major changes in the evolution of the oceans, atmosphere and life. An increasing number of studies provide evidence that enhanced oxygen levels already existed before the Paleoproterozoic Great Oxidation Event (GOE), likely generated by photosynthesis [1,2]. However, the onset of photosynthetic oxygenation, as well as spatial and temporal oxygen fluctuations are highly debated. Here, we present U isotope data from black shales, carbonates and iron-rich sedimentary rocks with a deposition age of 3.2 to 2.2 Ga. Uranium (U) is a redox-sensitive trace metal whose redox changes induce characteristic isotope fractionations that may be preserved in the rock record. The analysed samples are mainly drill core and some outcrop samples from the Barberton (3.23 to 3.15 Ga) and Transvaal Supergroups (Ghaap and Chuniespoort Groups, 2.58 to 2.52 Ga; Pretoria Group 2.42 to 2.25 Ga) in South Africa. Sub-recent U mobilisation was monitored by simultaneous analyses of  $\delta^{234}\text{U}$ , and detrital contribution was monitored with Th/U and Al/U. Only those samples with significant U enrichment ( $U_{\text{EF}} > 2$ , relative to average continental crust) were considered. They show variations in authigenic  $\delta^{238}\text{U}$  ranging from -0.83 to 0.05 ‰, which significantly deviate from typical  $\delta^{238}\text{U}$  of the continental crust (-0.2 to -0.4 ‰). Remarkably, we observed a significant increase in  $\delta^{238}\text{U}$  variability from samples of the Ghaap Group and those of the Pretoria Group (Duitschland Formation). The predominantly light U isotope composition of the latter samples may best be explained by the onset of partial weathering of uraninite, with oxidative mobilisation of  $^{235}\text{U}$  just before the GOE. Low  $\delta^{238}\text{U}$  values were not observed in the overlying Timeball Hill Formation, deposited after the GOE, which may indicate the onset of quantitative weathering of uraninite and other U-bearing minerals.

[1] Kendall et al. (2013) Chem. Geology 362, 105 – 114.

[2] Wille et al. (2007) GCA 71, 2417 – 2435.