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Cycling of the elements of life within the Archean crust-mantle system

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The major elements of life are carbon, hydrogen, oxygen and nitrogen, and constraining their cycles between the inner and outer geochemical reservoirs is critical for improving our understanding of the evolution of a life-supporting atmosphere. Archean Earth was almost certainly characterized by fundamental changes in the dynamics of the crust-mantle system [1], as recorded in the pronounced growth of felsic crust and complementary depleted mantle lithosphere, which led to stabilization of the first continents. Although volatile element abundances, including the activity of oxygen, have been deduced for the Early Archean mantle using products of major melting such as komatiites, direct evidence for the origins of carbon- and nitrogen-bearing fluids/melts and the redox-controlled speciation of these volatiles in the Earth's upper mantle before 3 Ga is scarce and poorly constrained.

We measured the C- and N-isotope compositions of the oldest confirmed gem diamonds, which were recovered as placer stones from 3.1-2.9 Ga old conglomerates of the Witwatersrand Supergroup, Kaapvaal craton. Utilizing N-aggregation and C-isotope systematics, we obtained firm evidence that the Wits diamonds formed between 3.5 and 3.1 Ga within Earth's upper mantle. However, the N-isotope compositions suggest that recycled sedimentary components contributed to the diamond-forming fluids/melts. High-precision SIMS C-isotope profiles across the Wits diamonds reveal that the growth medium contained oxidized carbon species in the form of CO_3^{2-} and CO_2 (as opposed to CH_4). These findings are among the most robust evidence for the presence of highly oxidized and recycled components within Earth's mantle by 3.5-3.1 Ga, and possibly before [2].

New isotope evidence from the earliest recorded CO_2 -rich surface magmatism on the North Atlantic craton supports the observation from the Wits diamonds that critical elements of life were vigorously exchanged between the deep Earth and its surface reservoirs prior to 3 Ga. These constraints on the deep extension of the Archean volatile cycle suggest that modern-style plate tectonics operated before 3 Ga and that atmospheric oxygenation after 2.5 Ga was unrelated to the onset of subduction-recycling on Earth.

[1] Dhuime et al. (2012), *Science* 335, 1334-1336

[2] Smart et al. (2016), *Nature Geoscience* 9, 255-259