

# Composition of continental crust altered by the emergence of land plants

Christopher J. Spencer<sup>1\*</sup>, Neil S. Davies<sup>2</sup>, Thomas M. Gernon<sup>3</sup>, Xi Wang<sup>1</sup>, William J. McMahon<sup>2</sup>, Taylor Rae I. Morrell<sup>1</sup>, Thea Hincks<sup>3</sup>, Peir K. Pufahl<sup>1</sup>, Alexander Brasier<sup>4</sup>, Marina Seraine<sup>1</sup>, Gui-Mei Lu<sup>1,5</sup>

\*Corresponding author. Email: [c.spencer@queensu.ca](mailto:c.spencer@queensu.ca)

<sup>1</sup>Department of Geological Sciences and Geological Engineering, Queen's University; Kingston K7L 2N8, Ontario, Canada

<sup>2</sup>Department of Earth Sciences, University of Cambridge; Downing Street, Cambridge CB2 3EQ, United Kingdom

<sup>3</sup>School of Ocean & Earth Science, University of Southampton; Southampton SO14 3ZH, United Kingdom

<sup>4</sup>School of Geosciences, University of Aberdeen, King's College; Aberdeen AB24 3UE, United Kingdom

<sup>5</sup>State Key Laboratory of Geological Processes and Mineral Resources, School of Earth Sciences, China University of Geosciences; Wuhan 430074, China

**The evolution of land plants during the Palaeozoic Era transformed Earth's biosphere. Because the Earth's surface and interior are linked by tectonic processes, the linked evolution of the biosphere and sedimentary rocks should be recorded as a near-contemporary shift in the composition of the continental crust. To test this hypothesis, we assessed the isotopic signatures of zircon formed at subduction zones where marine sediments are transported into the mantle, thereby recording interactions between surface environments and the deep Earth. Using oxygen and lutetium-hafnium isotopes of magmatic zircon that respectively track surface weathering (time-independent) and radiogenic decay (time-dependent), we find a correlation in the composition of continental crust after 430 Myr ago, which is coeval with the onset of enhanced complexity and stability in sedimentary systems related to the evolution of vascular plants. The expansion of terrestrial vegetation brought channelled sand-bed and meandering rivers, muddy floodplains, and thicker soils, lengthening the duration of weathering before final marine deposition. Collectively, our results suggest that the evolution of vascular plants coupled the degree of weathering and timescales of sediment routing to depositional basins where they were subsequently subducted and melted. The late Palaeozoic isotopic shift of zircon indicates that the greening of the continents was recorded in the deep Earth.**

35 Plants are the dominant kingdom of life on Earth, accounting for  $\approx 450$  gigatons of a total  $\approx 550$   
36 Gt of extant living biomass <sup>1</sup>, and successfully colonizing  $\sim 84\%$  surface area of the presently  
37 subaerially exposed continental crust <sup>2</sup>. Yet Earth's status as a 'green planet' is geologically  
38 recent, and other than some millimetre-thick microbial mats <sup>3</sup>, terrestrial vegetation was absent  
39 for approximately 90% of the planet's history. Recently reported palynological evidence  
40 indicates that the first embryophytes (land plants) likely evolved from aquatic charophyte algae,  
41 that had begun to adapt to terrestrial settings by at least the early Ordovician (c. 480 Myr ago) <sup>4,5</sup>.  
42 The earliest empirical fossil evidence for vegetated continents comes from cryptospores that  
43 appear recurrently in strata onwards from the Middle Ordovician (c. 467-470 Myr ago) <sup>6</sup>,  
44 attesting to relative evolutionary stasis in which early land plants were restricted to wet coastal  
45 environments, and maintained diminutive, low diversity, simple forms for the first 30-40 Myr of  
46 their history <sup>7,8</sup>. Eventually this primitive vegetation became outcompeted by vascular plants, and  
47 both spores and plant megafossils attest to a major late Silurian-Early Devonian adaptive  
48 radiation of land plants (diversification, morphological innovation and increasing size) that  
49 initiated at the end of the Llandovery Epoch (c. 433 Myr ago) <sup>9</sup>.

## 50 **Land Plants and Riverine Sediments**

51 Early Palaeozoic land plants had profound impacts on Earth surface processes and geological  
52 products. Fossil evidence for the advent of land plants is coeval with a step increase in the  
53 amount of mudrock deposited in continental settings <sup>10,11</sup>. Throughout the Precambrian,  
54 terrigenous mudrocks were primarily the product of physical erosion with limited pedogenic clay  
55 mineral formation <sup>12</sup>. However, geochemical data suggest that in the early Palaeozoic, the  
56 primary locus of the global 'clay mineral factory' shifted from the oceans to the land <sup>13</sup> as early  
57 land plants induced the inception of new pathways in (micro)biologically mediated weathering.  
58 Fossil evidence supports this notion, as Silurian plants—and their bacterial and fungal

59 symbionts—formed early plant ground covers that would have intensified clay mineral  
60 weathering<sup>14</sup>.

61 The Early Devonian evolution of advanced rooting systems with root axes and meristems<sup>15</sup>  
62 would have caused a further increase to the volume of the critical zone for terrigenous  
63 weathering, promoting a greater root surface area and amplified connectivity with the reactive  
64 near-surface environment<sup>16</sup>. The increased volume of mud and clay on the continents was likely  
65 also boosted by the disruption of seaward transport of fine sediments by the retentive influences  
66 of land plants as obstacles and stabilizers<sup>10,11,17</sup>, and possibly by the capacity of their organic  
67 matter to heighten mud flocculation and deposition<sup>18</sup>.

68 Physical sedimentary processes and products on the continents were also influenced by the  
69 evolutionary adaptations of plants. Advances such as the evolution of vascular plant roots,  
70 around 411 Myr ago<sup>15</sup>, and the earliest trees at 390-388 Myr ago<sup>19,20</sup>, bound channel margins  
71 and increased complexity in river geomorphology at a variety of spatio-temporal scales<sup>10</sup>. A  
72 prominent impact is seen in the rise in abundance of mud-rich fluvial deposits during the Silurian  
73 and Devonian, distinguishing them from more ancient mud-poor alluvium and attesting to an  
74 increase in meandering river planforms<sup>10</sup>. Although rapidly migrating sandy meandering  
75 systems were almost certainly in existence before the evolution of land plants<sup>10</sup>, the Silurian and  
76 Devonian ushered in the first abundant mesoscale, muddy meandering channels<sup>21</sup>, which would  
77 have both fragmented and lengthened source-to-sink sediment conduits and maintained stable  
78 floodplains as long-lived sediment staging areas. Incorporation of the plant organic matter into  
79 floodplain sediments also affected carbon cycling, driving enhanced production of calcrete  
80 (calcium carbonate) in terrestrial sediments from the Silurian onwards<sup>22,23</sup>. It seems likely that  
81 the development of plants with rhizospheres in the Siluro-Devonian will also have directly  
82 enhanced chemical rock weathering<sup>24</sup>: a hypothesis that demands checking of suitable proxies.

83 The oxygen isotopic composition of the continental crust is one such proxy <sup>25</sup>. From the bulk  
84 Earth average  $\delta^{18}\text{O}$  of  $\sim 5.5\text{‰}$  (as compared to Vienna Standard Mean Ocean Water) <sup>26</sup>, isotopic  
85 fractionation during low-temperature weathering and water-rock interaction led to elevated  
86 whole-rock  $\delta^{18}\text{O}$  values in siliciclastic sedimentary rocks. Even in sparsely vegetated  
87 environments, the weathering of silicate material results in higher  $\delta^{18}\text{O}$  in sediments and minimal  
88 post-depositional isotope exchange with ground or surface waters <sup>27</sup>. Whilst high  $\delta^{18}\text{O}$  ( $> 15\text{‰}$ )  
89 were present throughout the Proterozoic Eon and exhibit a marked increase across the Archean–  
90 Proterozoic transition <sup>28</sup>, the distribution of  $\delta^{18}\text{O}$  and chemical index of alteration of non-  
91 glaciogenic mudrocks does not dramatically change throughout the Phanerozoic (Supplementary  
92 Figure 1) <sup>28</sup>.

93 Conversely, empirical geologic evidence shows that the volume of continental mudrock  
94 preserved in the geologic record expanded dramatically during the early Palaeozoic <sup>11</sup>. Models  
95 have also predicted that the advent of land plants increased silicate weathering rates by a factor 2  
96 to 10 (ref. <sup>24</sup>) albeit in a transitory way <sup>29</sup>, and an increase in silicate weathering and increased  
97  $\text{CO}_2$  fixation via photosynthesis led to substantive cooling during the late Ordovician <sup>30</sup> and  
98 Devonian <sup>31</sup>. These lines of evidence suggest that associated signals could be found in under-  
99 explored archives, including Earth's zircon  $\delta^{18}\text{O}$  record.

## 100 **Sediment Recycling in Magmatic Systems**

101 Sedimentary deposition along continental margins and within ocean basins ultimately leads to a  
102 global fraction of that material becoming incorporated into magmatic systems during subduction  
103 <sup>32,33</sup>. The most common loci of such activity are subduction zones where sedimentary material is  
104 either subducted into the asthenosphere <sup>34</sup>, or melted in the upper crust along the periphery of  
105 rising plutons <sup>35</sup>. While other convergent margins also form sediment-derived magmas (e.g.,

106 continental orogens<sup>36</sup> and back-arc basins<sup>37</sup>), given the length of global subduction zones, these  
107 are likely the largest contributor of magma influenced by sedimentary material<sup>38</sup>.

108 The influence of sediment on magmatic systems is most commonly tracked through geologic  
109 time using  $\delta^{18}\text{O}$  and U-Pb geochronology of the mineral, zircon<sup>39</sup>. It is understood that any  
110 igneous rocks with  $\delta^{18}\text{O}$  values divergent from the average mantle (~5.5‰) must have been in  
111 part or wholly derived from supracrustal material<sup>39</sup> and that the presence of a sedimentary  $\delta^{18}\text{O}$   
112 signature in magmatic zircon can be traced from some of the oldest terrestrial zircon (~4.27 Gyr  
113 ago)<sup>40,41</sup> to the youngest exposed granites (818.5±9.6 kyr ago)<sup>42</sup>. Another powerful isotopic tool  
114 in zircon is Lu-Hf, where the radioactive  $\beta$ -decay of  $^{176}\text{Lu}$  to  $^{176}\text{Hf}$  allows for comparison of the  
115 Lu-Hf isotopic composition of zircon at formation with the chondrite uniform reservoir and  
116 depleted mantle.  $^{176}\text{Lu}/^{177}\text{Hf}$  is displayed as  $\epsilon\text{Hf}$  that is normalized to the chondrite uniform  
117 reservoir.<sup>43</sup>

### 118 **Tracing Sedimentary Components in Zircon**

119 We developed a new compilation of U-Pb, Lu-Hf, and O isotopic data from detrital zircon<sup>5</sup> to  
120 evaluate secular change in their correlations (Fig. 1). To assess the correlation of  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}$   
121 over time, we use a rolling window to visualize changes in the slope,  $r^2$ , Pearson's correlation,  
122 and Spearman's Rank correlation (Figs. 1F, 2)<sup>5</sup>. We find no statistically reliable correlation  
123 between  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}$  prior to 450 Myr ago. After ~430 Myr ago, however,  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}$  show a  
124 strong correlation as well as a statistically significant step change between 450-430 Myr ago  
125 (Fig. 2; Extended Data Fig. 2)<sup>5</sup>. Covariance in independent zircon O and Lu-Hf isotopic datasets  
126 (analyses not from the same zircon grains) has previously been linked to deglaciation and  
127 sediment subduction following the Cryogenian global glaciation<sup>44</sup>. In contrast, this study  
128 evaluates the correlation of O and Lu-Hf isotopes in individual zircon thus providing a unique

129 view at the grain scale <sup>5</sup>. In addition, we expand upon the work of ref. <sup>11</sup> through the application  
130 of a step change algorithm to the percentage of mudrocks in sedimentary successions, which  
131 reveals a significant increase at 430 Myr ago (Fig. 3; Extended Data Fig. 3) in tandem with the  
132 late Silurian adaptive radiation of land plants <sup>9</sup>. Assessment of tectonic subdivisions using the  
133 timing of supercontinent assembly confirms that none of the periods of substantial collisional  
134 orogenesis yield a statistically significant correlation between zircon  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}$  (Extended  
135 Data Fig. 4) meaning that supercontinent tectonics can be ruled out as the driver of this signal <sup>5</sup>.

136 The U-Pb and Lu-Hf isotope systems track magmatism and crystallization at temperatures  
137  $>700^\circ\text{C}$  and thus document geologic processes occurring deep in Earth's crust and recording the  
138 chronology of magmatic processes (i.e., mantle extraction and zircon crystallization). While the  
139 oxygen isotopic compositions of zircon are also imparted within the magmatic system at depth,  
140 elevated  $\delta^{18}\text{O}$  indicates the melting of material that was once exposed at Earth's surface (i.e.,  
141 (meta)sedimentary material; Fig. 1A). Given that the melting of (meta)sedimentary material  
142 occurs in a multitude of magmatic rocks with sediments of varying ages <sup>45</sup> and different tectonic  
143 settings (e.g., young sediments in oceanic arcs; <sup>32</sup>; old sediments in collisional orogens; <sup>36</sup>), there  
144 is no prima facie expectation for Lu-Hf and O isotopes in zircon to be correlated.

145 The post-430 Myr ago correlation between these two magmatic proxies implies some secular  
146 change in the rates of sediment transport from source-to-sink, with the sediment sink  
147 subsequently facilitating melting of sedimentary material. Importantly, the  $\epsilon\text{Hf} - \delta^{18}\text{O}$  correlation  
148 requires a diversity of processes including the formation of zircon with mantle-derived isotopic  
149 signatures (both in Lu-Hf and O isotopes) along with the melting of progressively older and  
150 more highly weathered sedimentary rocks. As mantle-derived magmatism forms the endmember  
151 from which all primary crustal magmas form, the secular change discussed herein is assumed to  
152 primarily represent sedimentary processes.

153 Due to the time-integrated nature of  $\epsilon\text{Hf}$ , the comparison of  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}$  through time is  
154 problematic. However, when dealing with narrow windows of time (<250 Myr) the amount of  
155 radiogenic decay in the chondrite uniform reservoir (CHUR) is minimized. An alternative  
156 assessment can be done by using crustal residence ages (depleted mantle model age minus  
157 crystallization age). This however comes with an assumption of the  $^{176}\text{Lu}/^{177}\text{Hf}$  applied globally.  
158 Nevertheless,  $\epsilon\text{Hf}$  versus  $\delta^{18}\text{O}$  show broadly the same patterns as crustal residence versus  $\delta^{18}\text{O}$   
159 (Extended Data Fig. 5, Extended Data Fig. 6). We compare secular change of zircon  
160 compositions with sedimentological and paleontological data to quantitatively understand  
161 potential linkages between terrestrial biosphere evolution—including the expansion of vascular  
162 land plants and their influence on source-to-sink sediment transfer—to the melting of sediments  
163 in convergent margins and the composition of magmatic zircon and by proxy the continental  
164 crust.

165 The widespread development of post-Archean continental freeboard <sup>46,47</sup> and the concomitant  
166 increase in continental weathering is inferred from an increase of mudrock  $\delta^{18}\text{O}$  throughout the  
167 Proterozoic <sup>28</sup>; no equivalent secular change is apparent in post-Proterozoic mudrocks (Extended  
168 Data Fig. 1). Therefore, the importance of progressive weathering and mudrock formation (and  
169 thus facilitation of the increase in zircon  $\delta^{18}\text{O}$  when (meta)sedimentary material is melted) must  
170 also be in some way tied to the duration of time between progressive weathering (i.e., increasing  
171  $\delta^{18}\text{O}$ ) and sediment deposition in loci available for incorporation and melting in magmatic  
172 systems (e.g., along a subduction trench).

### 173 **Connecting Land Plants and Zircon**

174 The duration of surface weathering influences the radiogenic ingrowth of Lu-Hf isotopes and  
175 mass-dependent fractionation of oxygen isotopes, and is dependent on sediment residence time

176 within the critical zone <sup>48</sup>. Sediment delivered to rivers at source is then routed through a  
177 sediment transfer system. The residence time of a pulse of sediment within this system increases  
178 with: 1) the length of the river system; 2) the number of buffers within that system where  
179 sediment undergoes transient storage and remains static (e.g., floodplains); and 3) the relative  
180 abundance of suspended bed-material load relative to bedload <sup>49,50</sup>. The greater sinuosity of  
181 meandering channels (compared with braided river networks) means that they are inherently  
182 longer routing systems. The associated long-lived floodplains of meandering rivers provide more  
183 robust buffers than those in laterally-mobile braided rivers <sup>51</sup>, and sediment transport is more  
184 regularly dominated by mixed- or suspended bed-material load. Each of these considerations  
185 imply that the time taken for source-to-sink sediment transport is greater in meandering than in  
186 braided rivers. Even though individual particles of suspended sediment can be transported more  
187 rapidly than bedload within a channel <sup>52</sup>, individual sediment particles can transit almost five  
188 times slower in meandering rivers (9.6 yr km<sup>-1</sup>) than in braided rivers (1.9 yr km<sup>-1</sup>) <sup>53</sup>. This  
189 discrepancy is largely accounted for by residence of particles within hillslope <sup>54</sup> and floodplain <sup>55</sup>  
190 staging areas, which are only re-entrained after intervals on the order of 10<sup>4</sup>-10<sup>5</sup> kyr <sup>56,57</sup>, within  
191 the timeframe of mature soil development and chemical weathering <sup>54,55</sup>.

192 Relevantly, at 430 Myr ago, the empirical shift towards correlated O and Hf-Lu isotopes in  
193 zircon is coincident with a global unidirectional shift towards an increased abundance of  
194 meandering river channel morphologies (Fig. 3), as tracked by the frequency distribution to  
195 physical characteristics of alluvium archived in continental basins <sup>10,58</sup>. River sediments  
196 deposited after this juncture exhibit a jump in the abundance of mudrock <sup>11</sup> and heterolithic  
197 lateral accretion sets <sup>21</sup>, attesting to a rise in long-lived floodplains, channel stability, and  
198 mesoscale sinuosity, that can be mechanistically explained by the novel influence of vegetation  
199 acting as a geomorphic agent <sup>11,21</sup>. While the specific impact of vegetation manifests at the local



200 scale, the global aggregation of a multitude of local effects (which did not exist prior to c. 430  
201 Myr ago) increased the probability of prolonged continental weathering, with downstream  
202 impacts on the isotopic composition of the marine sediment <sup>13</sup> that could be incorporated into  
203 magmatic systems <sup>59</sup>.

204 Based on the considerable post-430 Myr ago increase of the zircon isotopic correlation (Lu-Hf  
205 and  $\delta^{18}\text{O}$ ), we hypothesize a connection between the evolution of land plants and the final fate of  
206 sediment in magmatic systems. Following deposition of sedimentary successions along passive  
207 margins, these successions were then in prime placement to be subducted along accretionary  
208 margins or alternatively get caught in collisional orogens. In both scenarios, sedimentary units  
209 are metamorphosed and typically assimilated into arc magmas or melted in collisional systems.  
210 The zircon in the resulting magmas exhibit elevated  $\delta^{18}\text{O}$  values due to incorporation of  
211 sediments. While elevated zircon  $\delta^{18}\text{O}$  occurs as early as 4.27 Gyr ago <sup>40,41</sup>, critically the  
212 coupling between radiogenic isotope systematics (Lu-Hf, U-Pb) and  $\delta^{18}\text{O}$  in zircon is absent until  
213 after 430 Myr ago.

214 The unidirectional shift in both the zircon and stratigraphic record likely rules out an origin owed  
215 to cyclical or episodic geological processes (e.g., tectonic or climatic) as such phenomena have  
216 persisted across the planet from the Archean onwards <sup>60</sup>. The studied interval, from 1200 – 0 Myr  
217 ago includes multiple orogenies, numerous alternations between icehouse and greenhouse  
218 conditions and the assembly of two supercontinents yet, similar to the unidirectional upsurge in  
219 alluvial mudrock <sup>11</sup>, none of these extrinsic events appear to have exerted any influence on the  
220 observed oxygen and lutetium-hafnium isotopes in zircon. The singular observable shift that  
221 does occur, and that we describe, is both coincident with—and explicable by—the onset of more  
222 complex vegetation in the late Silurian.

223 We propose that the evolution of land plants led to the stabilization of fluvial systems, resulting  
224 in substantial upland weathering and the formation of larger volumes of mud. The resulting  
225 higher degrees of weathering along decelerated source-to-sink sediment pathways led to higher  
226  $\delta^{18}\text{O}$  signatures in the mud deposited as mudrocks. The eventual melting of this high  $\delta^{18}\text{O}$  mud  
227 led to higher  $\delta^{18}\text{O}$  magmas, providing a direct link between the evolution of the nascent  
228 terrestrial biosphere and composition of the lithosphere.

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### 237 **Author contributions:**

238 CJS conceived of the idea and with the help of XW, TRM, MS, NSD, and WJM compiled and  
239 interpreted data. CJS, XW, TH, TMG, and GML assisted with the statistical analysis. CJS, XW,  
240 WJM, and TH constructed the figures. CJS, NSD, and TMG wrote the manuscript with input  
241 from XW, WJM, TRM, TH, PKP, AB, MS, and GML.

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### 243 **Competing interests:**

244 Authors declare that they have no competing interests.



246 **Figure Captions**

247 **Fig. 1:  $\epsilon\text{Hf}$  versus  $\delta^{18}\text{O}$  in zircon since 720 Myr ago (A-E).** It is assumed here that all primary  
248 magmas are initially derived from the mantle with a  $\delta^{18}\text{O}$  of  $\sim 5.5\text{‰}$  and a crustal residence time  
249 less than  $\sim 250$  Myr (approximating the depleted mantle compositions <sup>61</sup>). The degree of  
250 correlation between  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}_{\text{zircon}}$  is markedly different in the latter two panels (A & B), with  
251 the panels covering pre-430 Myr ago showing greater degrees of scatter and weak correlations.  
252 F)  $r^2$  versus slope of the regression from 700 Myr ago to 0 Myr ago in 10 Myr steps using a  
253 rolling window <sup>5</sup>.

254 **Fig. 2: Statistical relationship between  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}$  of zircon.** A step-change algorithm  
255 (conjugate partitioned recursion <sup>5</sup>) demonstrates a statistically valid step change in the slope,  $r^2$   
256 and correlation coefficients at either 450 Myr ago (linear regression slope and Pearson's  
257 Correlation) or 430 Myr ago ( $r^2$  and Spearman's Rank Correlation). The increase in vascular  
258 plants at 450 Myr ago and the increase in mudrock percentage at 430 Myr ago are shown as  
259 vertical dashed lines.

260 **Fig. 3: Synthesis of paleontological and sedimentological data from the early Palaeozoic.**  
261 Bottom panel: percentage of mudrocks in sedimentary successions from the late Ediacaran  
262 through the Carboniferous (data from ref. <sup>11</sup>). We find a step change in the abundance of  
263 mudrocks at 430 Myr ago <sup>5</sup>. It is after this point that the correlation between zircon  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}$   
264 increases considerably (Fig. 2). Middle panel: cumulative classification of fluvial rock units  
265 through the early Palaeozoic <sup>58</sup> showing a shift in the type and diversity of fluvial deposit

266 characteristics at ~430 Myr ago. Top panel: Number of vascular plant species <sup>62</sup> and approximate  
267 timing of the first land plants, vascular plants, deeper roots, arborescence, and seeds <sup>63</sup>.

268 **Fig. 4: Schematic model of fluvial systems both before and after the development of land**  
269 **plants.** Prior to the Devonian Period, minimal land plants, rapid channel migration, and river  
270 systems dominated by braided rivers led to rapid sediment transfer, minimal development of  
271 mud-sized particles, and minor deposition of mudrocks along continental margins. In contrast,  
272 after the Devonian, the expansion of vascular land plants led to the development of meandering  
273 rivers and the dramatic slowing of sediment transfer to marine environments. These diamic  
274 depositional systems influenced the isotopic composition of sediment available for reworking  
275 and melting in magmatic systems along destructive plate boundaries.

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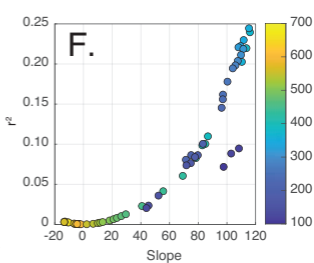
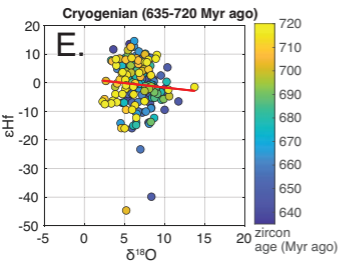
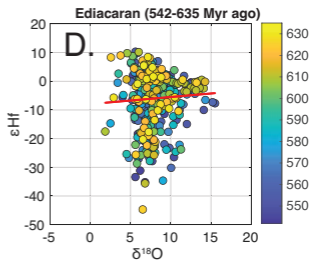
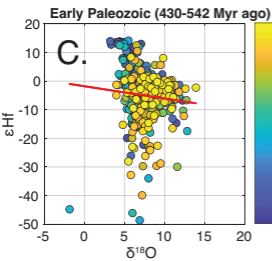
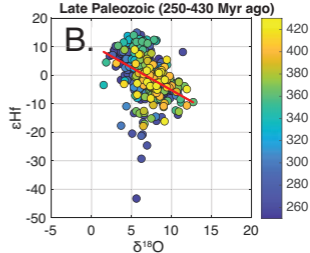
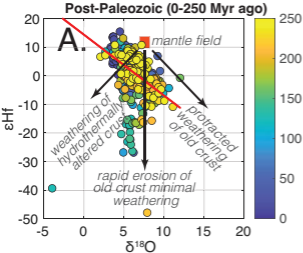
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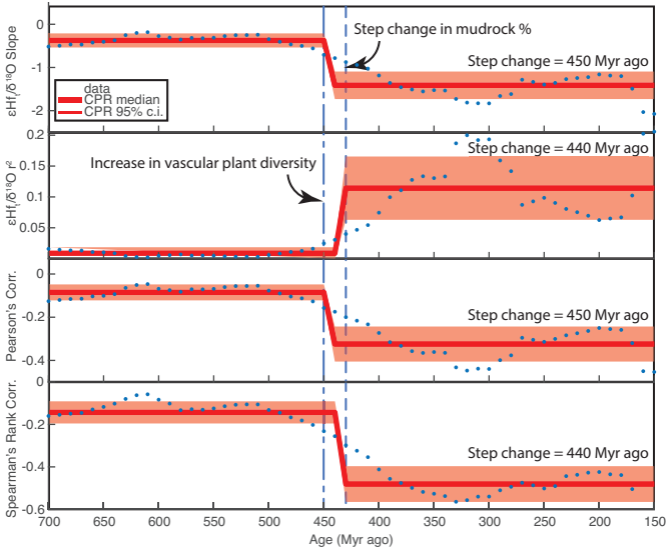
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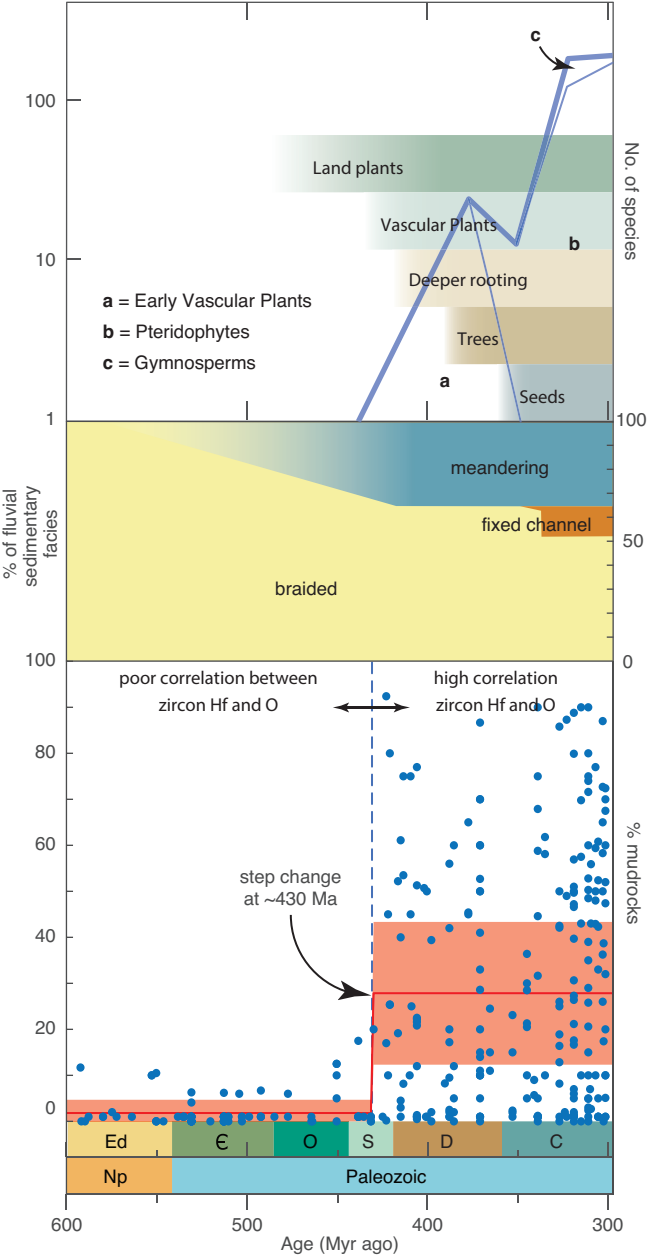
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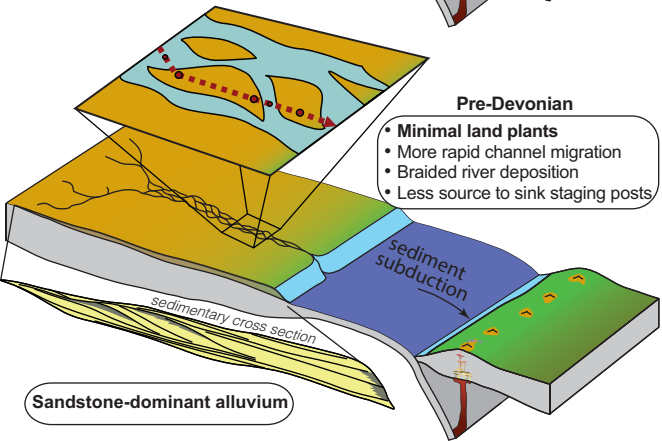
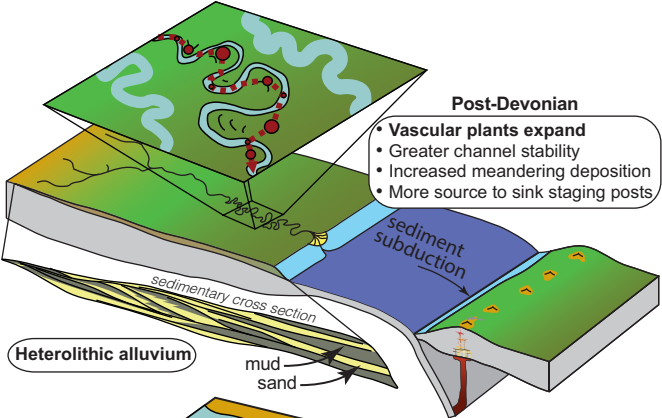
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466 Correspondence and requests for materials should be addressed to CJS.











467 **Methods**

468 Data Compilation

469 Data were compiled from 183 publications (Data S1) that included U-Pb, Lu-Hf, and O  
470 isotopic analyses of detrital zircon. Only data that passed strict quality control (concordant U-Pb  
471 ages <sup>64</sup>, accurate stable Hf isotope ratios <sup>65</sup>, and reliable  $\delta^{18}\text{O}$  analyses <sup>66</sup>) were included in our  
472 analysis. U-Pb ages,  $\epsilon\text{Hf}(t)$ , and 2-stage depleted mantle model ages were recalculated using  
473 published isotopic ratios using unified decay constants and initial chondritic and depleted mantle  
474 values <sup>67</sup>. Crustal residence times (CR) were calculated by subtracting the U-Pb age from the  
475 depleted mantle model age.

476 Statistical Methods

477 We used a rolling window to visualize the change over time in the linear regression slope  
478 and  $R^2$  values for CR and  $\delta^{18}\text{O}$ , and similarly the change in correlation coefficient (Pearson and  
479 Spearman's Rank) between CR and  $\delta^{18}\text{O}$ , and in a separate test, between zircon  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}$   
480 through time (Extended Data Fig. 6).

481 For our purposes, the input time series do not need to be regular; the only requirement is  
482 that values for both parameters (CR and  $\delta^{18}\text{O}$ ) exist at any given time step in the time series. We  
483 define a window width “w” (e.g., 100 Myr), an increment “i” (e.g., 5 Myr; this is the interval at  
484 which we repeat the calculations), and a start and end time.

485 This gives an output timeseries  $t$ , from start to end at increments  $i$ . At every time step  $t$ , we  
486 select the data for CR and  $\delta^{18}\text{O}$  that lie within the time interval  $[t-(w/2), t+(w/2)]$ ; i.e., a  
487 symmetric window centred on  $t$ . We then simply calculate the linear regression slope and  $R^2$   
488 value for CR and  $\delta^{18}\text{O}$  (and separately for zircon  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}$ ) using the R function “lm”:

489  $linear\_model = lm(CR \sim \delta^{18}O)$

490 The correlation is calculated using the R function, “cor”:

491  $Pearson\_correlation = cor(\delta^{18}O, CR)$

492  $Spearman\_correlation = cor(\delta^{18}O, CR, method = "spearman")$

493 This process is repeated for all time steps. Note the windows overlap and are centred on the  
494 time given on the x axis in the plots shown in figure 2.

495 We next used conjugate partitioned recursion (CPR) to evaluate the potential presence of  
496 step changes in the linear regression slope,  $r^2$  and correlation coefficients (Fig. 2). This iterative  
497 algorithm uses binary partitioning by marginal likelihood and conjugate priors (conjugate  
498 partition recursion) to identify an unknown number of change-points<sup>68</sup>. If the marginal  
499 likelihood favours a change-point model, then the algorithm defines a change point and two-  
500 sigma uncertainty bounds of the two averages before and after the change point. In applying the  
501 CPR algorithm to the correlation statistics data, we identify change-points at 450 Myr ago for  
502  $\epsilon Hf$  and  $\delta^{18}O$  slope and Pearson’s correlation, and 430 Myr ago for  $\epsilon Hf$  and  $\delta^{18}O$   $r^2$  and  
503 Spearman’s rank correlation. We also performed CPR to demonstrate the presence of a change-  
504 point in the percentage of mudrocks in fluvial sequences using the data of ref.<sup>11</sup> (Fig. 3).

505 To further demonstrate the robustness of the change points described above, we also applied  
506 a Bayesian change point algorithm using transdimensional Markov chain Monte Carlo<sup>69</sup>. This  
507 method infers probability distributions based on the number and locations of changepoints, and  
508 the mean values between changepoints. It uses not just a single set of model parameters, but is  
509 computed using 10E6 simulations from the probability distribution to evaluate the position of the  
510 change points through time using the slope of the regression,  $r^2$  of the regression, Pearson’s  
511 correlation, and Spearman’s rank correlation together in its identification of potential change  
512 points (Extended Data Fig. 3).

513  *$\epsilon Hf$  versus crustal residence ages*

514 As  $\epsilon\text{Hf}$  is time integrative, comparing  $\epsilon\text{Hf}$  values at different ages result is strictly invalid (e.g.  
515  $\epsilon\text{Hf} = 0$  at 200 Ma is not equivalent to  $\epsilon\text{Hf} = 0$  at 100 Ma). However, given the narrow windows  
516 of time used in this study (<250 Myr), the difference is negligible within our interpretive  
517 framework. To address this, we also applied our statistical methods to crustal residence ages  
518 compared to  $\delta^{18}\text{O}$  through time. Crustal residence time is calculated here as the difference  
519 between the U-Pb age and the projected “model age” where the radioactive decay trajectory  
520 (controlled by  $^{176}\text{Lu}/^{177}\text{Hf}$ ) intersects with the depleted mantle <sup>70</sup>. This provides an estimate of  
521 how much time elapsed between the initial extraction (or melting) of the mantle and  
522 crystallization of zircon. It is important to note that crustal residence times are not geologic ages  
523 and are directly tied to depleted mantle model ages. The crustal residence time is generally low  
524 when magmas are extracted directly from mantle with zircon forming relatively rapidly, as in  
525 subduction zones. In contrast, the crustal residence time is high when ancient crust is remelted  
526 100’s of Myrs to ~4 Gyr after initial extraction from the mantle <sup>71</sup>.

### 527 Timing of plant evolution

528 Here we pinpoint the timing of land plant colonization primarily based on the fossil record.  
529 While alternative suggestions of the onset of greening, arising from molecular phylogenies of  
530 living plants, suggest an earlier start to the process, there are reasons to be cautious for the  
531 statistical biases inherent in these modelling approaches <sup>72</sup>. A counterargument has been made  
532 that molecular phylogenies are closer to the truth for estimating timing because the paucity of  
533 non-marine strata in the Cambrian and Ordovician renders its fossil record unreliable; either  
534 because that record has been subducted <sup>73</sup> or because there was a limited continental area during  
535 the interval <sup>74</sup>. Both of these arguments are problematic: firstly, because empirically there is no  
536 exponential decay of non-marine strata with age, as sometimes envisaged by molecular  
537 phylogenists <sup>75</sup>, and secondly because the abundance of small continents with wide continental

538 shelves during that interval is well suited for preserved fossil evidence for land plants, as the best  
539 early record of such comes from dispersed spores preserved in shallow marine strata <sup>6</sup>. The fossil  
540 record thus likely provides a robust estimate of the onset of greening and, regardless, molecular  
541 clock analyses and fossil evidence are increasingly convergent on the timing of embryophyte  
542 (land plant) origins. Molecular clocks suggest that embryophytes emerged in a mid-Cambrian to  
543 Early Ordovician interval <sup>76</sup>. Recent palynologic evidence favours the end of this bracket and  
544 indicates that ancestral embryophytes began evolving from freshwater aquatic charophyte algae  
545 around the Early Ordovician (c. 480 Myr ago) <sup>4</sup>.

546 *Covariance and correlation of Lu-Hf and O isotopes*

547 Keller et al. (2019) reported the covariance between Lu-Hf and O isotopes in zircon. This  
548 study used two separate databases that respectively reported Lu-Hf and O isotopes in zircon.  
549 They identified a large covariance peak that coincided with the timing of Cryogenian global  
550 glaciation and argued in favour of a connection between glaciogenic erosion and the composition  
551 of zircon that came from magmas influenced by the assimilation of the resulting flux of  
552 glaciogenic sediment into magmatic systems. Their numerical analysis also displayed a second  
553 peak of covariance that coincides with the evolution and proliferation of land plants that is the  
554 topic of the present study. Although this second Palaeozoic peak in zircon Lu-Hf and O  
555 covariance is more prominent in the Keller et al. (2019) study, it was not discussed. Importantly,  
556 covariance, as used in the Keller et al. (2019) study, refers to the extent to which two random  
557 variables change in tandem. For example, when the slope of secular change in Lu-Hf isotopes  
558 and  $\delta^{18}\text{O}$  are changing at the same rate then covariance is high. In contrast, correlation measures  
559 strength and direction of the linear relationship between two variables. Importantly, it is not  
560 possible to measure the strength (or quality) of the linear relationship when using covariance.  
561 The Keller et al. study evaluates how Lu-Hf isotopes and  $\delta^{18}\text{O}$  change in tandem using

562 independent datasets whereas our study aims to evaluate how strongly Lu-Hf isotopes and  $\delta^{18}\text{O}$   
563 are directly correlated through time. Interestingly, our study also shows a minor increase in the  
564 strength of correlation within the timeframe of the Snowball Earth – however an increase that is  
565 a factor of five smaller than the increase in correlation associated with the rise of land plants.

566 Keller et al. (2019) use independent  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}$  datasets, that is, they use two separate  
567 databases for the two isotopic systems. This means their interpretation is limited to a comparison  
568 between the secular changes in each dataset and they are unable to directly compare Lu-Hf  
569 isotopes and  $\delta^{18}\text{O}$  for specific zircon grains. In contrast, our dataset uses only Lu-Hf isotopes and  
570  $\delta^{18}\text{O}$  from the same zircon grain providing improved discriminating power in discerning secular  
571 changes in the data, which we argue is essential for our purposes. It is assumed each dataset is  
572 independently representative, which has previously been demonstrated as not being the case for  
573 Lu-Hf isotopes<sup>77</sup>. Further, a later compilation of zircon  $\delta^{18}\text{O}$  has shown a significant bias  
574 between detrital and igneous zircon<sup>78</sup> which are indiscriminately combined in Keller et al.  
575 (2019). We seek to overcome these issues by comparing Lu-Hf and O isotopes from specific  
576 zircon grains.

577 The Keller et al. (2019) study reports two significant peaks in  $\delta^{18}\text{O}$  and  $\epsilon\text{Hf}$  covariance, one  
578 at ~650 Ma and the other with higher covariance at ~420 Ma. While they show both peaks in  
579 their figure 2, they omit the ~420 Ma peak from their discussion. They posit the ~650 Ma  
580 covariance\*slope peak is associated with “increasing crustal reworking”, while the large negative  
581 covariance\*slope trough at ~420 Ma is associated with “decreasing crustal reworking”. N.B. that  
582 while they state in the figure caption that a large negative covariance\*slope is associated with  
583 decreased crustal reworking, they do not state the age of this trough, nor speculate on a potential  
584 cause. We posit this ~420 Ma covariance peak is supportive of our hypothesis that the evolution  
585 of land plants influenced the covariance of Lu-Hf and O isotopes in zircon.





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# Composition of continental crust altered by the emergence of land plants

Christopher J. Spencer<sup>1\*</sup>, Neil S. Davies<sup>2</sup>, Thomas M. Gernon<sup>3</sup>, Xi Wang<sup>1</sup>, William J. McMahon<sup>2</sup>, Taylor Rae Morrell<sup>1</sup>, Thea Hincks<sup>3</sup>, Peir K. Pufahl<sup>1</sup>, Alexander Brasier<sup>4</sup>, Marina Seraine<sup>1</sup>, Gui-Mei Lu<sup>1,5</sup>

\*Corresponding author. Email: [c.spencer@queensu.ca](mailto:c.spencer@queensu.ca)

<sup>1</sup>Department of Geological Sciences and Geological Engineering, Queen's University; Kingston K7L 2N8, Ontario, Canada

<sup>2</sup>Department of Earth Sciences, University of Cambridge; Downing Street, Cambridge CB2 3EQ, United Kingdom

<sup>3</sup>School of Ocean & Earth Science, University of Southampton; Southampton SO14 3ZH, United Kingdom

<sup>4</sup>School of Geosciences, University of Aberdeen, King's College; Aberdeen AB24 3UE, United Kingdom

<sup>5</sup>State Key Laboratory of Geological Processes and Mineral Resources, School of Earth Sciences, China University of Geosciences; Wuhan 430074, China

## Extended Data Figures and Tables

### This PDF includes:

Extended Data Figures 1-6

Extended Data Table 1

### Extended Data Figure Captions

**Extended Data Fig. 1: Secular plot of  $\delta^{18}\text{O}$  in mudrocks and zircon during different Periods in the Palaeozoic.** Note that no systematic or long-term change in shale composition is present throughout the Phanerozoic. Mudrock  $\delta^{18}\text{O}$  data are from <sup>34,79,80</sup>. Uncertainty bars are 2 S.D.

**Extended Data Fig. 2: Transdimensional Markov chain Monte Carlo simulation of the zircon crustal residence versus  $\delta^{18}\text{O}$  slope,  $r^2$ , and correlation coefficients demonstrates a statistically valid change point between 450-410 Myr ago with a maximum likelihood at 440 Myr ago (using one million simulations).**

31 **Extended Data Fig. 3: Transdimensional Markov chain Monte Carlo (MCMC) simulation**  
32 **and conjugate partitioned recursion (CPR) of the percentage of mudrocks through time.**  
33 Data from ref. <sup>17</sup>. MCMC yields a statistically valid change point between 430-420 Myr ago with  
34 a maximum likelihood at 423 Myr ago (using one million simulations) whereas CPR shows a  
35 change point at 430 Myr ago.

36 **Extended Data Fig. 4: Crustal residence time vs  $\delta^{18}\text{O}$  in zircon through time.** This includes  
37 (A) the Archean Eon (pre-2500 Myr ago), and major supercontinent assembly events including  
38 (B) Nuna from 2200 to 1700 Myr ago, (C) Columbia from 1700 to 1200 Myr ago, (D) Rodinia  
39 from 1200-900 Myr ago, Pangea from 400-250 Myr ago, and (F) post-Pangea assembly from 250  
40 Myr ago to present.

41  
42 **Extended Data Fig. 5: Crustal residence versus  $\delta^{18}\text{O}$  in zircon since 720 Myr ago (A-E).** It  
43 is assumed here that all primary magmas are initially derived from the mantle with a  $\delta^{18}\text{O}$  of  
44  $\sim 5.5\%$  and a crustal residence time less than  $\sim 250$  Myr (approximating the depleted mantle  
45 compositions <sup>61</sup>). The degree of correlation between  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}_{\text{zircon}}$  is markedly different in  
46 the latter two panels (A & B), with the panels covering pre-430 Myr ago showing greater degrees  
47 of scatter and weak correlations. F)  $r^2$  versus slope of the regression from 700 Myr ago to 0 Myr  
48 ago in 10 Myr steps using a rolling window.

49  
50 **Extended Data Fig. 6: Statistical relationship between zircon  $\epsilon\text{Hf}$  and  $\delta^{18}\text{O}$  through time.** A  
51 step-change algorithm (conjugate partitioned recursion <sup>5</sup>) demonstrates a statistically valid step  
52 change in the slope,  $r^2$  and correlation coefficients at either 450 Myr ago (linear regression slope  
53 and Pearson's Correlation) or 430 Myr ago ( $r^2$  and Spearman's Rank Correlation). The increase  
54 in vascular plants at 450 Myr ago and the increase in mudrock percentage at 430 Myr ago are  
55 shown as vertical dashed lines.

56  
57 **Extended Data Table 1. (Separate file)**

58  
59 Compiled zircon U-Pb, Lu-Hf, and  $\delta^{18}\text{O}$  database.

