1	The miniature paleo-speleothems record of the 'greening' Earth
2	in the early Ediacaran (635 Ma)
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## 18 Abstract

19 A rapid transition from the snowball Earth to the 'greening' Earth in the early Ediacaran, triggered by enhanced terrestrial weathering and then by elevated primary 20 productivity, has been speculated by isotope researches<sup>3</sup>. However, direct geological 21 22 evidence of continental weathering in the early Ediacaran is still lacking. This study 23 examines paleo-speleothems related to the karstic dissolution surface at the upmost cap 24 dolostone of the early Ediacaran Doushantuo Formation (635 Ma). Observation of 25 sheet-crack thin-sections from platform to slope facies in South China suggests that 26 plentiful speleothem-like structures in chalcedony should be interpreted as low-27 temperature silicified calcareous paleo-speleothems. Furthermore, isopachous dolomite, speleothems, chalcedony, and quartz should have filled sheet-cracks during the uplift, karstification, and subsequently hydrothermal processes before the secondary transgression. Thus these widely distributed paleo-speleothems, which are direct geological evidence for the 'greening' Earth in the early Ediacaran, might represent an initial formation of the soil-ecosystem after the barren snowball Earth.

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# **1. Introduction**

34 Paleo-climate evolution in the Neoproterozoic (1,000-542 Ma), the most notable of which was the abrupt climate transition in the aftermath of the Marinoan deglaciation 35 (~635 Ma<sup>4</sup>) recorded in the 3-5 m cap dolostones and its idiosyncratic sedimentary 36 structure, was the prelude to the Cambrian explosion. Isotopic studies <sup>1-3</sup> have described 37 a rapid evolution from a snowball Earth to a 'greening' Earth, which could greatly 38 39 enhance groundwater influx of photosynthetic carbon from phytomass and promote the 'clay mineral factory', and then subsequently increase the atmosphere's O<sub>2</sub> content <sup>5</sup> and 40 trigger the expansion of multicellular life. This rapid evolution centered on a drastic 41 42 change of the continental weathering mode which elevated the bio-available P flux and 43 promoted the marine primary productivity. Therefore, geological records of 44 contemporaneous continental weathering are important clues to understanding the 45 evolution of life in the Neoproterozoic and to illuminating the Cambrian explosion.

46 Karstification is the most important continental weathering process in carbonate 47 distribution zones, of which the environment change could be record by speleothems 48 (stalagmites, stalactites, flowstones etc.). Studies of modern karst caves suggest that 49 speleothem deposition is mostly controlled by evolution of CO<sub>2</sub> contents in drip-water, 50 which originate from precipitation (atmospheric CO<sub>2</sub>) and supersaturation in soil zone (CO<sub>2</sub> from bio-respiration and organic decomposition) and then degassing in caves  $^{6-8}$ . 51 Thus paleo-speleothems in paleo-karst, such as dripstone <sup>9</sup>, micro-stalactite <sup>10,11</sup> and 52 stromatolitic laminae coatings <sup>12</sup>, are significant evidence for subaerial exposure and 53 54 paleo-pedogenesis. The widespread discontinuous karstic surface at the top of cap dolostones (635 Ma) in Africa, Canada and China has disclosed a global karstic dissolution event <sup>13</sup> caused by uplifting of continental shelf responding to deglacial isostatic rebound <sup>14</sup>. Hence, more detailed geological evidence such as paleospeleothems are expected to be preserved in fissures, voids and sheet-cracks related to the karstic dissolution event <sup>13</sup>.

In this paper, we report a series of miniature but delicate paleo-speleothems (including stalagmites, stalactites and coatings) preserved in the sheet-crack of Marinoan cap dolostone (~635 Ma) in South China. These paleo-speleothems further confirm the possibility of a broad and transient continental uplift with exposure and continental weathering due to the deglacial isostatic rebound.

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# 2. Geological setting and observations

The Doushantuo Formation of basin, slope and platform facies is widely deposited 66 on the Yangtze Block of South China (see Fig.1 in ref.<sup>13</sup>), and is underlain by Marinoan 67 deglacial tillite unit. This shows a transition from purple diamicite (<100 m) in platform 68 69 facies to grey diamicite (>1000 m) in basin facies. The 3-5 m cap dolostone in basal 70 Doushantuo Formation from platform to slope facies is significant with disrupted massive dolomicrite and unique structures (such as giant wave ripples <sup>15</sup>, teepee-like, 71 sheet-cracks etc. <sup>16</sup>). A broadly karstic dissolution surface, caused by uplift by isostatic 72 73 rebound, has been confirmed by geological observation in South China. The total 74 duration (<1.0 Ma) from deposition to exposure and dissolution of the cap dolostone 75 has been constrained by high-precision U-Pb zircon age of  $634.57 \pm 0.88$  Ma at the topmost of Nantuo diamictite <sup>4</sup> and  $635.23 \pm 0.57$  Ma at the topmost of cap dolostone 76 77 <sup>17,18</sup>, respectively. Remarkably, sheet-cracks have a uniform mineral paragenetic 78 sequence across the entire Yangtze Block: they start with isopachous dolomites 79 (sometimes with minor barite), followed by siliceous minerals (chalcedony and quartz), and ending with later stage calcite and barite <sup>19</sup>. 80

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Miniature but perfect stalactites and stalagmites, which are the most typical

calcareous speleothems (gravitational dripping water forms) (Fig.1, Fig.2), have been
gradually disclosed in chalcedony cements from thick (more than 2-3 cm) sheet-cracks
of cap dolostones and distributing from slope (Wenghui and Daping) to platform facies
(Xiaofenghe and Beidoushan) sections on the Yangtze Block. Based on this discovery,
flat and thinner laminae, partly with botryoidal structures, that extensively encrust the
ceiling and floor of the sheet-cracks or breccias (Fig.3), have been interpreted here as
coatings (non-gravitational water-film forms).

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## 2.1 stalactite

Many stalactites, hanging downwards from the ceiling of sheet-cracks and exhibiting as single one or conjoined by multi-stalactites, have been found in the Beidoushan and Wenghui sections (Fig. 1, 2a, 2e and 2d). The most common individual stalactites are elongated columns, ranging from less than 0.4 cm to about 1.0 cm in diameter, and from less than 1 cm to more than 3 cm in length.

95 Three growth stages could be identified by laminae rhythm in two vertical profiles 96 of stalactites from the Beidoushan section (Fig.1). The first is the straight soda-straw 97 with a central channel. The channel is about 100 µm in diameter and 1-2 cm in length 98 and is lined by brown organisms and filled with cryptocrystalline chalcedony. The wall 99 of the soda-straw is comprised of fibrous chalcedony with about 400-500 µm thickness 100 and it is also coated by brown organisms. The second stage is distinguished by density 101 rippling lamina couplets in flank and botryoid structures in the tip, which reflect stable 102 and slow feeding. The final stage is composed of relative broader lamina couplets which 103 reflect continuous and affluent feeding.

The cross-profiles of stalactites are distinguished by multilayer concentric circularity structures with alternations from dark to light. The significant difference, however, is the soda-straw structure, which is generally present in stalactites but absolutely absent in stalagmites. The three growth stages described above can be clearly observed in the cross-profiles of stalactites from the Beidoushan section. In the Wenghui section, however, only two growth stages are displayed in the cross-profiles of stalactites (Fig.2a, 2e and 2d) and in the vertical-profile of stalagmites. These
differences suggest that the two sections have different paleo-environments.

112 **2.2 stalagmite** 

113 Some stalagmites, which grow upwards from the floor of the sheet-crack, were 114 discovered in Wenghui, Xiaofenghe and Beidoushan sections. They are mainly 115 composed of translucent chalcedony, which makes them obviously distinguished from 116 the surrounding white crystalline quartz in hand-specimens of the Wenghui and 117 Beidoushan sections (Fig.2b, 2d and 2f). Most of them are cylindric in shape, slightly 118 wider in the root and narrower in the tip, with length concentrated around 1-3.5 cm and diameters of about 0.5-1.3 cm. This thin diameter style, classified as "Minimum-119 diameter" stalagmite<sup>20</sup>, is coincident with the short drip fall height<sup>21</sup> in the sheet-cracks. 120

121 A perfect vertical profile of stalagmite from the Wenghui section shows a clear 122 transition of growth style (Fig.2b) under a reflecting light microscope, while, such a 123 transition is relatively blurred under transmitted light. The early growth style is 124 distinguished by a stacked botryoid structure, which could be observed in the bottom part of modern stalagmites <sup>22-25</sup> (Fig.2b) and which represent turbulence of the dripping 125 126 water at the beginning of stalagmite deposition or indigent feeding and slow precipitation<sup>26</sup>. The later growth is significant with continuous and smooth rhythmical 127 128 laminae couplets by a dark and a light lamina, which is similar to modern calcareous stalagmites <sup>25,27,28</sup> and which represent affluent feeding and stable precipitation <sup>22,29-31</sup>. 129 130 The rhythmical laminae are about 350 µm thick and contain about 20-30 lamina couplets. 131

There is one complete stalagmite and three complete stalactite cross-profiles in the same slide, the latter of which are characterized by a central channel texture (Fig.2d). There are two growth stages. The first of these is typical of unity cryptocrystalline chalcedony and the latter of them is features concentric fibrous chalcedony laminae, corresponding to the two-growth style in the vertical profile as mentioned above (Fig.2b). There are about 20-30 laminae couplets within the 2100 µm thick outer zone and this is rich in organics as seen by the obvious increase in fluorescence (Fig.2c, 2e
and 2f) when compared to the inner zone. Significantly, residue calcite core and laminae
have been observed in one stalagmite cross-profile from the Xiaofenghe section
(Fig.3h).

142 **2.3 coatings** 

143 In almost all of the cap dolostone sections in South China, the wall (mainly composed of ceiling and floor) of the sheet-cracks and the breccias in them are 144 145 extensively covered with less than 0.1 cm to 1 cm thick chalcedony coatings, which 146 tend to have fairly continuous layers, and are characterized by visible rippled growth 147 morphology and stacked layering (Fig. 3). Remarkably, partly silicified calcite coatings, 148 which could be distinguished under reflected light and scanning electron microscope 149 (SEM) (Fig.3e and 3f), was preserved in the Daping section. The coatings may be 150 botryoidal (Fig. 3b), or even spiral (looks like vermiform helictites) (Fig.3c and 3d), 151 but in most cases they are smoothly curving along the wall of the sheet-crack and the 152 breccias with stable thickness (Fig. 3a). On the whole, the coatings comprise of 15-30 153 lamina couplets, which are much more obvious in ultraviolet fluorescent (Fig.2c and 3g), with single couplet thickness ranging from 20 µm to 60 µm. These morphology 154 155 and laminae structure indicate that the smooth coatings periodically precipitated from 156 adhesive water-films condensed from humid caves, the botryoidal structure are 157 produced by surface tension dividing water-films into drops and the vermiform 158 helictites produced by the addition of drip water to already present water-films.

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## 3. Discussion

Protogenetic siliceous speleothems are commonly developed in caves or lava tunnels overlain by silicate rocks (such as quartzites, sandstones, granites etc.) <sup>32</sup>. Although platform-wide black shale overlaid on the cap dolostone <sup>33</sup> seems to be a potential silica source, lack of karstic surface and weathering dissolution textures in the black shale suggests that the sheet-crack speleothems were not protogenetic siliceous 165 speleothems and were constrained before the deposition of the black shale. Indeed, the 166 sheet-crack speleothems are confined below the widespread paleo-karstic surface the age of which has been previously determined by two ash beds with zircon U-Pb age of 167  $634.57 \pm 0.88$  Ma and  $635.23 \pm 0.57$  Ma  $^{17,18}$  respectively. Additionally, the coatings in 168 the Daping section are mostly consisted of calcareous laminae (Fig. 3e and 3f) and the 169 170 silicified stalactites in Xiaofenghe section still retain a few calcareous laminae (Fig. 3h). 171 Therefore, the sheet-crack speleothems deposited at ca. 635Ma were originally calcareous speleothems, which is akin to modern silicification-preserved speleothems 172 formed by low-temperature metasomatism of primary calcareous speleothems <sup>34,35</sup>. 173

174 Three successive events associated with the Marinoan cap dolostone in South China have been summarized as such <sup>13</sup>: (1) the first postglacial transgression and 175 deposition of the cap dolostone; (2) isostatic rebound, uplift and karstification of the 176 177 cap dolostone; and (3) the second postglacial transgression, multiphase cave fillings 178 and post-cap deposition. Multiple mineral generations on walls of sheet-cracks are attributed to the beginning of the second postglacial transgression <sup>13</sup> or a low-179 temperature hydrothermal episode <sup>19,36</sup>, however, minerals corresponding to the uplift 180 181 and karstification event have not been depicted. No obvious dissolution phenomena by 182 later erosion have been observed on the surface of the paleo-speleothems, indicating 183 that the deposition of the paleo-speleothems has been quickly terminated by the low temperature hydrothermal process. Given that the hydrothermal episode developed 184 185 after the beginning of the second transgression, chert lens (as siliceous tufa) should be 186 observed upon the karstic surface, nevertheless, siliceous cements and veins have been 187 strictly confined beneath the karstic surface. Thus we interpret the deposition and 188 hydrothermal silicification of the sheet-crack calcareous speleothems as successive 189 processes during exposure and karstification.

190 Modern karst studies indicate that necessary condition for karstic dissolution is the 191 soil-ecosystem (soil, plant, microbial, etc.), which afford organic matter and plentiful 192  $CO_2$  <sup>37,38</sup> in the water of an epikarst zone and hence the relative speleothems could

partly record the overlying ecosystem information. Karst dissolution in some high-193 altitude and cold-climate regions occurs in the absence of soil <sup>7</sup>, however, and so 194 obviously a temperature gradient caused by a huge altitude drop is needed to form 195 196 speleothems in these conditions. Paleo-karsts are defined as karsts developed largely or entirely during past geological periods <sup>39</sup>. Freytet (2002) refers to karsts or vugs that 197 are centimeters to decimeters as microkarsts<sup>11</sup>, and these are an important evidence of 198 ancient subaerial exposure and paleosols formation <sup>9,11,40</sup>. Like pseudomicrokarsts <sup>11</sup>, 199 200 we define the microkarsts developed in past geological periods as paleomicrokarsts, in 201 which the speleothems are defined as paleomicrospeleothems. The Precambrian 202 palaeosols are habitats for early terrestrial life. The paleomicrospeleothems in the 203 corresponding carbonate strata are important geological evidence that record the early 204 biological evolution of the Earth. Paleomicrospeleothems (fibrous flowstone lining grike system) found in the Mesoproterozoic in Canada<sup>41</sup> and U.S.A<sup>35</sup> and the exquisite 205 206 paleomicrospeleothems (icicle-like pendants, hemispherical protrusions and ground-up columns) reported in the Dengying Formation <sup>42</sup> may represent contemporaneous 207 pedogenesis processes. In the early Ediacaran, although recovery of ocean-ecosystem 208 209 from the brutal snowball Earth had been confirmed by vase-shaped fossils in tillite, the 210 geological evidence for terrestrial-ecosystem revival are still expected yet. Here, the 211 silicified paleomicrospeleothems preserved in the 3-5 m cap dolostone suggest that the 212 soil-ecosystem had been broadly established in South China just after uplifting and 213 exposing of the cap dolostone.

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# 4. Summary and Implication

This paper reports the widely distributed miniature silicified paleospeleothems in sheet-cracks in Marinoan cap dolostone from South China, which depict specific karstic process of the cap dolostone during uplifting and exposure caused by isostatic rebound. These are 1) miniature speleothem growth during karstification; 2) speleothem termination and silicification by low-temperature fluid before the second transgression. These paleo-speleothems have recorded the rapid recovery of the soil-ecosystem after a snowball Earth during cap dolostone rebound and karstic dissolution, which is key geological evidence for the 'green' Earth model.

The karstic dissolution surface may have been widely distributed on a global scale at early Ediacaran, implying that the coincident silicified calcareous paleo-speleothems are also global distributed. The silicification preservation process has destroyed some original geochemical information such as carbon/oxygen isotopes, but the plentiful organic-rich laminae are preserved. Thus, the bio-markers in these organic-rich laminae are expected to further document the evolution of soil-ecosystem.

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**Author Contributions** 

T.L., G.Z., and T.G. designed the research. T.L., G.Z., and T.G. collected the samples. G.Z and T.G. conducted experiments. G.Z., T.G., and T.L. developed the interpretation and prepared the manuscript with contributions from M.Z.

241 Methods

Three sheet-crack samples (14XFH-1, 14XFH-3 and 14XFH-5) from Xiaofenghe section (N30°48′54″, E111°03′20″), Hubei Provinces, three sheet-crack samples (14DPc1-1, 14DPc1-2 and 14DPc1-3) from Daping section (N28°59′01″, E110°27′42″), Hunan Provinces, four sheet-crack samples (16WH-1, 16WH-2, 16WH-3 and 16WH-

4) from Wenghui section (N27°49'55", E109°01'32"), Guizhou Provinces and four 247 samples (18BDS-2, 18BDS-4, 18BDS-7 and 18BDS-9) from Beidoushan section 248 (N27°01'40", E107°23'22"), Guizhou Provinces were collected from the cap dolostone 249 of the Doushantuo Formation in South China. Petrographic slices (100 µm and 200 µm 250 in thickness) and polished slabs of the sheet-crack samples were cut both perpendicular 251 and horizontal to bedding plane and investigated under transmitted light microscopy (TLM), reflected light microscopy (RLM) and fluorescent light microscopy (FLM). 252 253 254 Reference 255 1 Kennedy, M., Droser, M., Mayer, L. M., Pevear, D. & Mrofka, D. Late Precambrian 256 oxygenation; inception of the clay mineral factory. Science 311, 1446-1449 (2006). 257 2 Knauth, L. P. & Kennedy, M. J. The late Precambrian greening of the Earth. Nature 460, 728 258 (2009). 259 3 Kump, L. R. 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360	Figure Legends
361	Figure 1   Polished slab and micrographs of stalactite from Beidoushan

section. a, Polished slab shows stalactites in a sheet-crack scanning by a HP ScanJet,
the white solid rectangle highlights a column (connective bodies of stalactite and

364 stalagmite), white dotted rectangles highlight conjuction stalactites, the yellow arrow 365 denotes a single complete stalactite with "soda straw" drip channel. **b**, Petrogrphic slice shows transverse and vertical sections of stalactite under TLM (transmission light 366 367 microscope), the white and red arrows denote the vertical and transverse sections of 368 "soda straw" drip channel, respectively. c, Enlarged view of the stalactite vertical 369 section in **b** (rectangle) under FLM (fluorescent light microscope), the white arrow 370 denotes the vertical section of "soda straw" drip channel. d, Enlarged view of stalactite 371 transverse in b (rectangle) under FLM, red arrows denote the transverse of "soda straw" 372 drip channel.

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374 Figure 2 | Polished slabs and micrographs of stalagmite, stalactite and coating 375 from Wenghui section. a, Polished slab showing stalagmite, stalactite and coating in a 376 sheet-crack, white dotted lines highlight the coating areas in the sheet-crack, white 377 arrows denote stalactites, yellow arrows denote stalagmites. b, Petrographic slice shows 378 a vertical section of stalagmite and coating under RLM, white dotted lines highlight the 379 coating areas. c, Enlarged view of coating vertical section shows the organic-rich 380 laminae in **b** (rectangle) under FLM. **d**, Petrographic slice shows stalagmite and 381 stalactite transverses under TLM, red arrows and yellow arrows denote a single "soda straw" drip channel and the aggregation of "soda straw" drip channel, respectively. e, 382 383 Enlarged view of a stalactite vertical section in d (rectangle) under FLM, showing the 384 organic-rich laminae and "soda straw" drip channel. f, Enlarged view of a stalagmite 385 transverse section in **d** (rectangle) under FLM, showing the organic laminae but lack of 386 "soda straw" drip channel.

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Figure 3 | Polished slab, hand specimen and micrographs of coatings and a special stalactite. a, Polished slab shows coatings in a sheet-crack, white arrows denote the coating of a dolostone breccia. b, Hand specimen shows a coating lining in a sheetcrack, white dotted line highlights the coating boundary, black and red arrow denote

botryoidal and mold structure of the coating. c, Petrographic slice shows a coating with 392 a vermiform-like helictite under FLM. **d**, Enlarged view of the helictite in **c** (rectangle) 393 under SEM (scanning electron microscope). e, Petrographic slice shows a partly 394 395 silicified organic-rich calcareous coating under TLM. f, Enlarged view of the coating in e (rectangle) under SEM, showing the silicified calcareous coating, white arrows 396 397 highlight the siliceous cements. g, Enlarged view of the coating in e (rectangle) under 398 FLM, showing the organic-rich laminae. h, Petrographic slice shows a silicified 399 stalactite under TLM, white arrows highlight the residual calcite laminae. a, c and d 400 from Beidoushan section, **b** from Zhangcunping section, **e-g** from Daping section, **h** 401 from Xiaofenghe section.



Fig. 1

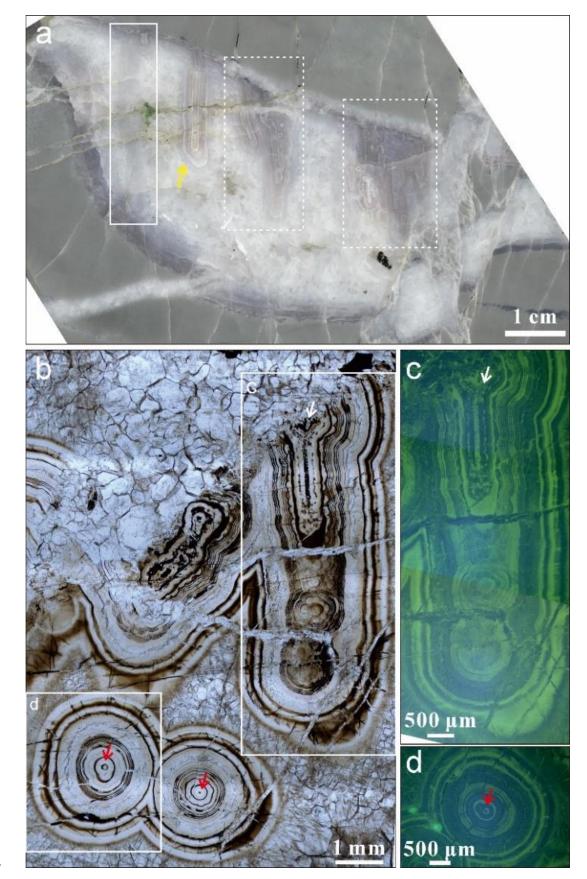




Fig. 2





Fig. 3

