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Article:

Zhang, L, Wang, C, Wignall, PB et al. (4 more authors) (2018) Deccan volcanism caused coupled pCO₂ and terrestrial temperature rises, and pre-impact extinctions in northern China. *Geology*, 46 (3). pp. 271-274. ISSN 0091-7613

<https://doi.org/10.1130/G39992.1>

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1 Deccan volcanism caused coupled $p\text{CO}_2$ and terrestrial
2 temperature rises, and pre-impact extinctions in northern
3 China

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16 **ABSTRACT**

17 Evaluating the terrestrial climate record provides a critical test of the roles of
18 Chicxulub impact and Deccan Traps volcanism during the Cretaceous-Paleogene (K-Pg)
19 mass extinction. Most evidence has come from marine records, but our new clumped
20 isotopes data from paleosol carbonates in the Songliao Basin provide a terrestrial climate

21 history from northern China. This reveals there was a pre-impact warming caused by the
22 onset of Deccan Traps volcanism, whereas the following short-term cooling then another
23 warming episode were likely caused by Chicxulub impact and post-boundary volcanism.
24 Our study suggests the $p\text{CO}_2$ levels were probably the main control on the latest
25 Cretaceous cooling and the climatic fluctuations across the K-Pg boundary interval in
26 northern China. In the Songliao Basin, the pre-impact Deccan volcanism links to losses of
27 half of the lacustrine algae species (charophytes) and almost all the lacustrine ostracodes;
28 this suggests that the Deccan Traps volcanism had already destabilized the ecosystem and
29 caused extinctions prior to the Chicxulub impact.

30 **INTRODUCTION**

31 The cause of the Cretaceous-Paleogene (K-Pg) mass extinction has been one of the
32 most intense scientific debates of past decades, with the relative roles of Chicxulub impact
33 and Deccan Traps volcanism providing the main discussion (Keller, 2014). The key issues
34 are whether Deccan eruptions caused coincident $p\text{CO}_2$ and paleotemperature rises (e.g.,
35 Nordt et al., 2002; Huang et al., 2013); and whether these pre-impact climate changes were
36 already imposing stresses on the global biota (e.g., Keller, 2014; Petersen et al., 2016a;
37 Witts et al., 2016). Detailed terrestrial climatic trends over the boundary interval could help
38 to evaluate the effects of these two closely timed events, but poor temporal resolution of
39 often fragmentary sections and ambiguous proxies usually restrict the significance of
40 terrestrial records (Tobin et al., 2014), making it difficult to evaluate the global picture.

41 By applying clumped isotope (Δ_{47}) paleothermometry to paleosol carbonates, we
42 present a relatively continuous K-Pg terrestrial climatic record with high-temporal
43 resolution in northern China that includes paleotemperatures, $\delta^{18}\text{O}_{\text{water}}$ values (soil water),
44 and $p\text{CO}_2$ from ca. 76 Ma to ca. 65 Ma. These new data extend the spatial coverage of
45 paleoclimatic estimates during the K-Pg interval and demonstrate that pre-impact climate
46 changes, caused by Deccan Traps volcanism, had already imposed stresses on the global
47 biota.

48 **MATERIALS AND AGE CONSTRAINTS**

49 Late Cretaceous–early Paleogene stratigraphy was recovered in the SK-In (north
50 core) borehole ($44^\circ 12' 44.22''\text{N}$, $124^\circ 15' 56.78''\text{E}$; Fig. 1) in the central part of the Songliao
51 Basin, northern China (Wang et al., 2013). In the Sifangtai and Mingshui Formations,
52 many distinctive calcareous paleosols, or calcisols, were identified and consist of
53 carbonate nodules, slickensides, mottled colors, and fossil root traces (Huang et al., 2013;
54 Gao et al., 2015). In this study, 51 paleosol carbonates (diameters range from 1.0 to 3.0 cm)
55 were collected from 44 paleosol Bk horizons (Fig. DR2; Tables DR4 and DR5 in the GSA
56 Data Repository¹). All of the samples come from shallow burial depths (no deeper than 1
57 km), suggesting that they have, at most, only been slightly influenced by burial diagenesis
58 or solid-state C-O bond reordering (Passey and Henkes, 2012). After petrographic vetting,
59 based on optical and cathodoluminescence properties (Fig. DR3), all samples were found

60 to be dense micrite except for sample SK-31, which has been excluded from the following
61 discussion.

62 By using thorium (Th) data from the Sifangtai and Mingshui Formations, an
63 astronomical time scale was established by tuning filtered 405 k.y. eccentricity cycles to
64 the astronomical solution La2010d, which calibrates the timing of the polarity chron
65 C29r-C30n boundary (342.1 ± 1.4 m in depth) to ca. 66.30 Ma and the K-Pg boundary (318
66 ± 1.2 m in depth) to ca. 66.00 Ma (Wu et al., 2014) (Fig. DR2).

67 **METHODS**

68 The clumped isotope analyses were conducted at Johns Hopkins University (the
69 laboratory has now moved to University of Michigan, Ann Arbor, USA) following the
70 methods described in Passey et al. (2010), and at Heidelberg University (Germany)
71 following the methods described in Kluge et al. (2015). The Δ_{47} temperatures are
72 calculated using the calibration of Passey and Henkes (2012) with an acid temperature
73 correction of 0.082%. The $\delta^{18}\text{O}_{\text{water}}$ (soil water) values are calculated from the Δ_{47}
74 temperatures and $\delta^{18}\text{O}$ of paleosol carbonates using the calibration of Kim and O'Neil
75 (1997). The paleo-atmospheric CO_2 ($p\text{CO}_2$) is calculated following the methods described
76 by Breecker and Retallack (2014) (Table DR3). The $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ are reported relative to
77 either the Vienna Peedee belemnite (mineral) or the Vienna standard mean ocean water
78 scales. The Δ_{47} values are reported relative to the absolute reference frame (Dennis et al.,
79 2011) (Tables DR1 and DR2).

80 RESULTS

81 The Δ_{47} temperatures range from 15.2 °C to 42.1 °C, with an average value of 24.9
82 °C (Fig. 2; Table DR4). Initially, temperatures were relatively high (~35 °C at ca. 76 Ma)
83 before decreasing to ~15 °C at the Campanian-Maastrichtian boundary (ca. 72 Ma). After
84 that, temperatures increased to ~30 °C ca. 71 Ma before decreasing again to a low point of
85 16.7 °C ca. 67.24 Ma (except for a short warming between 68 and 67 Ma). After ~1 m.y. of
86 low temperatures, a rapid warming of ~6 °C occurred between ca. 66.39 Ma and ca. 66.31
87 Ma, ~300 k.y. before the K-Pg boundary. Immediately before the K-Pg boundary (~100
88 k.y.), the temperature dropped more than 10 °C ca. 66.11 Ma. Finally, temperatures rapidly
89 increased once again by ~10 °C ca. 65.9 Ma before decreasing to ~22 °C ca. 65.5 Ma (Fig.
90 2; Table DR5). The general cooling trend is consistent with temperature trends both from
91 marine (e.g., Friedrich et al., 2012; Linnert et al., 2014; Petersen et al., 2016a) and
92 terrestrial (e.g., Kemp et al., 2014; Tobin et al., 2014) sections, indicating that this a global
93 signal.

94 We note that the Δ_{47} temperatures of the majority of soil carbonates were summer
95 biased (e.g., Passey et al., 2010; Snell et al., 2014); although a few represent other seasons
96 (e.g., Peters et al., 2013; Gallagher and Sheldon, 2016). In a monsoon climate, the soil
97 carbonate likely formed immediately before the cooling effects of the monsoon rains and
98 after the hottest part of the summer (Breecker et al., 2009). In the Songliao Basin,
99 monsoonal rainfall immediately followed the hottest part of summer (Chen et al., 2013).

100 We thus speculate that the carbonate nodules in SK-In were formed in summer. It is
101 noteworthy that our values are consistent with terrestrial climate records from similar
102 paleolatitudes across the Late Cretaceous-early Paleogene in North America (Fig. DR6)
103 based on temperature estimates from fossil plants (annual temperatures + 15 °C) and
104 clumped isotopes of fossil bivalves and paleosol carbonates (summer temperatures) (Snell
105 et al., 2014; Tobin et al., 2014).

106 The $p\text{CO}_2$ values range from 348 ppmv to 2454 ppmv (Fig. 2; Table DR5) with an
107 out-of-range value of 3460 ppmv that is excluded from the following discussion. The
108 average $p\text{CO}_2$ is 1575 ppmv for the Campanian, 1180 ppmv for the Maastrichtian, and
109 1058 ppmv for the Danian, generally showing a decreasing trend. The lowest levels of
110 ~600 ppmv occurred ca. 67.5–66.5 Ma, and then showed a rapid ~500 ppmv increase ca.
111 66.4–66.3 Ma. Levels decreased again just before the K-Pg boundary and increased back to
112 previous values immediately after the K-Pg boundary (Fig. 2).

113 Previous studies have predicted that $p\text{CO}_2$ levels underwent a long-term decline,
114 from ~1975 ppm to 450 ppm, during the Late Cretaceous (Wang et al., 2014). The
115 paleo- CO_2 reconstructed from pedogenic carbonates from North America rose
116 dramatically from 780 ppm in the Maastrichtian to 1440 ppm near the K-Pg boundary, but
117 declined sharply to 760 ppm at the boundary (Nordt et al., 2002). The pattern is consistent
118 with ranges and trends predicted in this study. Maastrichtian $p\text{CO}_2$ levels based on $\delta^{13}\text{C}$ of
119 paleosol carbonates from the Songliao Basin have been previously estimated to be between

120 277 ± 115 and 837 ± 164 ppmv during the K-Pg boundary interval (Huang et al., 2013).

121 However, these estimates assume a mean annual air temperature instead of summer

122 temperature (Δ_{47} temperature), and thereby underestimate the temperatures.

123 The $\delta^{18}\text{O}_{\text{water}}$ values range from -10.9‰ to -5.0‰ , and show a bimodal
124 distribution with $\sim 3\text{‰}$ shifts (Fig. 2; Tables DR4 and DR5). Similar bimodality in $\delta^{18}\text{O}$
125 values of fresh water in the Western Interior during the Late Cretaceous was attributed to
126 changes of water sources in the study areas (e.g., Tobin et al., 2014; Petersen et al., 2016b).
127 The soil water from which the soil carbonates formed mainly comes from meteoric water
128 (Quade et al., 1989). For two main moisture sources of northeast China, precipitation of the
129 westerly cold air masses from the Arctic Ocean and Central Asia are $\sim 3\text{‰}$ lower in $\delta^{18}\text{O}$
130 than warm air masses from the Pacific Ocean transported by the East Asian summer
131 monsoon (Gao et al., 2015). Therefore, we assume that the periodic excursions of the
132 $\delta^{18}\text{O}_{\text{water}}$ values may represent periodic fluctuations of relatively warm and cool climate
133 leading to periodic shifting of either the warm air masses with more ^{18}O -enriched
134 precipitation or cold air masses with more ^{18}O -depleted precipitation to the Songliao Basin,
135 reflecting the sensitivity of mid-latitudes terrestrial climates in a greenhouse world (Gao et
136 al., 2015).

137 **DISCUSSION AND CONCLUSIONS**

138 Both the Δ_{47} temperatures and the $p\text{CO}_2$ records across the K-Pg boundary interval
139 in northern China shows a decreasing trend with several fluctuations (Fig. 2); this suggests

140 that $p\text{CO}_2$ is the main driving factor forcing Late Cretaceous climatic fluctuations. The Δ_{47}
141 temperatures and $p\text{CO}_2$ levels ca. 67 Ma, ca. 69 Ma, and ca. 72 Ma were close to the
142 modern levels. During these periods, the Δ_{47} temperatures are generally 5–8 °C lower than
143 intervening periods and the $p\text{CO}_2$ are close to 750 ppmv (Fig. 2), which is the threshold for
144 Antarctic glaciation according to climate models (DeConto et al., 2008; Ladant and
145 Donnadieu, 2016). Antarctic records also showed near freezing sea surface temperatures
146 and accompanying glacioeustatic sea-level lowstands at 66.8 Ma and 68.8 Ma (Petersen et
147 al., 2016a).

148 Near the polarity chron C30n-C29r boundary, ~300–400 k.y. before the K-Pg
149 boundary, the temperature increased from ~22 °C to ~28 °C ca. 66.3 Ma (Fig. 3). Around
150 the same time, the $\delta^{18}\text{O}_{\text{water}}$ values increased from ~-9‰ to ~-6‰ (Fig. 3). Terrestrial
151 summer temperatures in North America similarly rose, although by a more modest 5 °C,
152 and stabilized at ~30 °C prior to the K-Pg boundary (Tobin et al., 2014). In addition,
153 marine records also show a pre-boundary warming in the latest Cretaceous (e.g., Li and
154 Keller, 1998; MacLeod et al., 2005; Petersen et al., 2016a). These climatic changes broadly
155 coincide with the onset of main Deccan eruptions (66.288 ± 0.085 Ma or 66.38 ± 0.05 Ma
156 based on different dating methods) (Renne et al., 2015; Schoene et al., 2015). Based on the
157 estimations of lava erupted during the main Deccan eruptions and CO_2 emitted per cubic
158 kilometer of lava, Petersen et al. (2016a) suggested that the pre-boundary volcanism
159 emitted 270–900 ppmv CO_2 onto a background atmospheric concentration of ~360–380

160 ppmv (Beerling et al., 2002). According to our results, the $p\text{CO}_2$ increased by ~400–500
161 ppmv from a background atmospheric concentration of ~348–870 ppmv in northern China
162 (Fig. 3); this is broadly consistent with the prediction. Therefore, we suggest that the onset
163 of Deccan volcanism likely caused the temperature and $p\text{CO}_2$ rise ca. 66.4–66.3 Ma in
164 northern China and Antarctica.

165 After the latest Maastrichtian warming, ~100 k.y. before the K-Pg boundary,
166 temperatures dropped sharply by more than 10 °C then recovered to the previous warming
167 temperature level at the beginning of the Paleogene (Fig. 3). Simultaneously, the $p\text{CO}_2$
168 records also show a drastic fall then rise across the K-Pg boundary. This trend was also
169 identified in North America immediately before the K-Pg boundary, when temperatures
170 fell by ~8 °C (Tobin et al., 2014). In the marine records, a rapid short-term sea surface
171 temperatures decrease of 7 °C immediately after the Chicxulub impact was recognized
172 using TEX_{86} paleothermometry of sediments from Texas and New Jersey (USA)
173 (Vellekoop et al., 2016). In contrast, clumped isotope paleothermometry of well-preserved
174 bivalve shells from Seymour Island, Antarctica, showed that sea surface temperatures
175 decreased immediately before the K-Pg boundary and then rose rapidly (Petersen et al.,
176 2016a), although these marine temperature changes are more modest compared to the
177 terrestrial ones. Petersen et al. (2016a) suggested that the post-boundary volcanism
178 possibly emitted another 825–900 ppmv. However, our records suggest only an ~300–400
179 ppmv increase onto a background atmospheric concentration of ~700–800 ppmv, lower

180 than this estimate but similar to the changes during pre-boundary volcanism; this may
181 suggest a lower volatile component in these eruptions. The duration of the ~6–8 °C
182 increase is also comparable to the pre-boundary rise (Fig. 3).

183 The lacustrine and palynological fossils from SK-In reveal distinct phases of
184 turnover across the K-Pg boundary interval (Li et al., 2011; Scott et al., 2012; Wan et al.,
185 2013) (Fig. 3; Fig. DR8). Although the sample density is low (10–25 m spacing),
186 palynological data show that major losses occurred amongst pollen taxa ~500 k.y. prior to
187 the boundary and left an impoverished assemblage that persisted across the boundary (Li et
188 al., 2011). One study has suggested that this palynological change may be due to
189 lithological variation rather than extinction (Wan et al., 2013). Ostracodes show major
190 extinctions around the K-Pg boundary (Scott et al., 2012; Wan et al., 2013) with losses
191 beginning ~200 k.y. (ca. 66.21 Ma) before the boundary in the Songliao Basin (Fig. 3; Fig.
192 DR8): 11 species disappeared before the boundary and 3 after it. Thus, the ostracode
193 extinctions in northern China show a good temporal link with the onset of the Deccan
194 volcanism. Abundant charophytes occur from ~340–317 m in SK-In (Wan et al., 2013),
195 and they also show losses (20 of 40 species) beginning after the onset of main Deccan
196 eruptions, and ~150 k.y. (ca. 66.15 Ma) before the K-Pg boundary. This extinction episode
197 is followed by appearance of several short-lived abundant (disaster) taxa (Scott et al., 2012;
198 Wan et al., 2013) (Fig. 3; Fig. DR8). Further charophyte losses (18 of 40 species) occur

199 after the K-Pg boundary. Therefore, like the ostracodes, the charophytes losses begin
200 around the onset of the main Deccan eruptions.

201 In total, two-thirds of the extinctions occurred before the Chicxulub impact but
202 after onset of eruption of the Deccan Traps and are thus solely linked to Deccan Traps
203 volcanism. However, it seems unlikely that the high temperatures and the rate of warming
204 led to the extinctions in northern China. The losses (ca. 66.15 Ma for ostracode and ca.
205 66.21 Ma for charophytes) occurred hundreds of thousands years after the onset of
206 warming (ca. 66.4–66.3 Ma). Similar levels of warmth and phases of rapid warming and
207 cooling had occurred before the extinctions. Therefore, we suggest that it is possible other
208 Deccan-linked environmental effects, i.e., acid rains or emission of toxic substances, led to
209 the pre-boundary extinctions in northern China. The remaining one-third of extinction
210 losses took place at the K-Pg boundary, at the time both the Chicxulub impact and the
211 post-boundary Deccan Traps volcanism occurred. Therefore, we cannot strictly
212 discriminate the relative role of these two events played in the post-boundary warming and
213 extinctions, but it is clear that Deccan Traps volcanism had already destabilized the
214 Songliao Basin ecosystem prior to the impact.

215 **ACKNOWLEDGMENTS**

216 We thank B.H. Passey for the use of the laboratory facilities in Johns Hopkins
217 University (Baltimore, Maryland, USA) and for performing a quality assurance of the data.
218 We thank Z. Zhou, H. Wu, and C. Koeberl, who gave us many useful comments. This

219 study was financially supported by National Natural Science Foundation of China (grants
220 41790450, 41702107), the National Key R&D Plan of China (grant 2017YFC0601405),
221 and the China Geological Survey Program (grant DD20160207).

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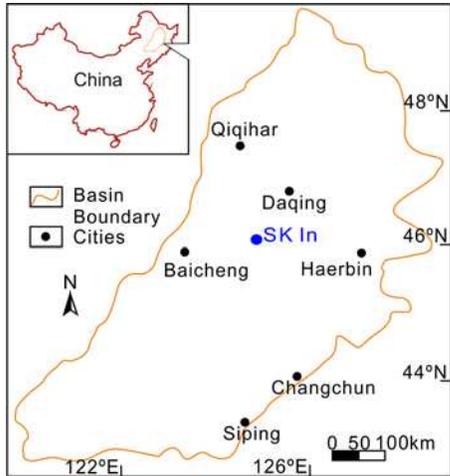
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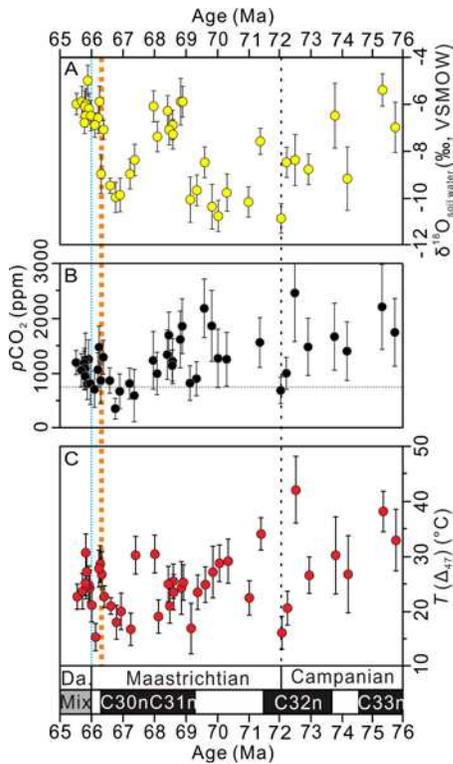
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363 references, Figure DR1-DR8, and Table DR1-DR5, is available online at
364 <http://www.geosociety.org/datarepository/2018/> or on request from
365 editing@geosociety.org.



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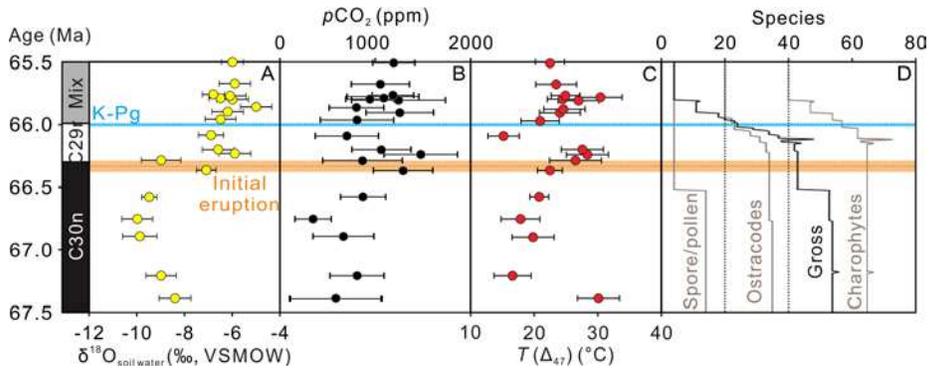
367 Figure 1. The Songliao Basin in northern China (orange outline), showing location of the

368 SK-In (north core) borehole (Wang et al., 2013).



369

370 Figure 2. The paleoclimate across the Cretaceous-Paleogene (K-Pg) boundary in northern
 371 China. A: The $\delta^{18}\text{O}_{\text{water}}$ (Vienna standard mean ocean water, VSMOW) record. B: The
 372 $p\text{CO}_2$ (ppmv) record. C: The Δ_{47} temperature (T) record. The 1σ standard errors are shown
 373 as black vertical bars. Dotted black line marks boundary between the Campanian and
 374 Maastrichtian at 72.1 Ma. Dotted orange line marks onset of the main Deccan eruptions at
 375 66.288 ± 0.085 Ma (Schoene et al., 2015) or 66.38 ± 0.05 Ma (Renne et al., 2015). Dotted
 376 blue line marks the K-Pg boundary at ca. 66.00 Ma (Wu et al., 2014) or 66.043 ± 0.086 Ma
 377 (Renne et al., 2013) and the Chicxulub impact occurred at 66.038 ± 0.098 Ma (Renne et al.,
 378 2013). Dotted gray line marks the threshold for Antarctic glaciation (750 ppmv) according
 379 to climate models (DeConto et al., 2008). Da—Danian; C—polarity chron.



380

381 Figure 3. Climatic records and ranges of selected fossil groups across the

382 Cretaceous-Paleogene (K-Pg) boundary interval. C—polarity chron. A: The $\delta^{18}\text{O}_{\text{water}}$

383 values (Vienna standard mean ocean water, VSMOW) record. B: The $p\text{CO}_2$ (ppmv) record.

384 C: The Δ_{47} temperature (T) record. D: The biotic data in the Songliao Basin, northern China

385 (Scott et al., 2012); note this column is separated into three sub-columns for spore and/or

386 pollen, ostracodes, and charophytes. The 1σ standard errors are shown as black horizontal

387 bars. See Figure 2 for legends.