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Abrupt chemical weathering increase across the Permian–Triassic boundary

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Abstract

Previous studies have suggested a variety of causes for the end-Permian extinction event including a bolide impact, flood basalt volcanism, methane clathrate dissociation, or some combination of catastrophic processes. One common feature of these hypotheses is the prediction of an enhanced earliest Triassic greenhouse. New high-resolution geochemical results from Graphite Peak, Antarctica support an abrupt increase in chemical weathering in the earliest Triassic among otherwise genetically similar paleosols of similar provenance. Relative to the latest Permian paleosols, the earliest Triassic paleosol exhibits greater leaching, greater accumulation of immobile REEs, and evidence of lower soil pO_2. With no evidence of an erosional unconformity between the paleosols, which are separated by <15 cm stratigraphically, these results support a rapid shift (perhaps $<10{,}000$ years) to an earliest Triassic greenhouse and a role for methane release in the extinction event and its aftermath.

Keywords: Methane; Antarctica; P-T boundary; Paleosols; Greenhouse

1. Introduction

The mass extinction that occurred at the end of the Permian was the largest in the history of life, with more than 90% of species going extinct (Hallam and Wignall, 1997). The extinction event decimated both marine and terrestrial ecosystems, with whole ecotypes disappearing for millions of years (e.g., coal-forming swamps; Retallack et al., 1996). It has been suggested that the extinction event was either accompanied or followed by a greenhouse event (Retallack, 1999, 2001; Berner, 2002; Kidder and Worsley, 2003), possibly in response to a bolide impact (Retallack et al., 1998; Becker et al., 2001; Basu et al., 2003), flood volcanism (Renne et al., 1995), H_2S poisoning (Kump et al., 2005) or methane clathrate release (Krull and Retallack, 2000; Sheldon and Retallack, 2002; de Wit et al., 2002). The object of this study was to look for evidence of a "post-apocalyptic greenhouse" (Retallack, 1999) and to assess the tempo of any observed shift. The terrestrial Permian–Triassic boundary exposed at Graphite Peak, Antarctica (Retallack and Krull, 1999; Krull and Retallack, 2000) was chosen for resampling because it contains a continuous section of paleosols spanning the boundary that have previously been subject to a variety of sedimentological and chemostratigraphical studies, allowing for a more thorough discussion of the new results.

2. Field site

The Permian–Triassic boundary at Graphite Peak, Antarctica is located at S85.05°, E172.37°, and an

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elevation of 2744 m. The stratigraphy and sedimentology of the section have been described elsewhere (Retallack and Krull, 1999; Krull and Retallack, 2000), as well as its δ^{13}C chemostratigraphy (Krull and Retallack, 2000). Previous studies have also documented shocked quartz (now questioned by Langenhorst et al., 2005) and an iridium anomaly (Retallack et al., 1998), putative extraterrestrial ^{3}He in fullerenes (Poreda and Becker, 2003; for opposing view see Farley and Mukhopadhyay, 2001), chondritic meteorite fragments (Basu et al., 2003), and evidence for methane clathrate release (Krull and Retallack, 2000; Sheldon and Retallack, 2002).

3. Methods

Field work consisted of stratigraphic logging with special attention to sedimentary and pedogenic features (e.g., looking for root traces; Retallack, 1997). Samples were collected from the Permian–Triassic boundary interval identified previously on the basis of δ^{13}C chemostratigraphy and the last occurrence of coal (Retallack and Krull, 1999; Krull and Retallack, 2000). The original trench at Graphite Peak (of Retallack et al., 1998; Retallack and Krull, 1999; Krull and Retallack, 2000) was re-excavated, widened, and resampled for geochemical analysis. Sixteen new geochemical analyses were performed by ALS Chemex of Vancouver, B.C. using X-ray fluorescence (XRF) for major elements, Ba, and Sr, using inductively coupled plasma mass spectrometry (ICP-MS) for rare earth elements (REE; variable uncertainties listed in the Supplemental Data), and using potassium dichromate titration to determine FeO (analytical uncertainty of 0.01%). Twelve additional geochemical analyses from Retallack and Krull (1999) were included, giving an average sample spacing <11 cm for the Permian–Triassic boundary interval (Fig. 1). Samples with GP-XX labels are new, and those with 20XX are from Retallack and Krull (1999). The analyzed suite of trace elements is more limited for the earlier analyses and in some cases it was not possible to analyze FeO, so there are a variable number of analyses for a given paleosol depending on the element of interest (see Supplemental Data).

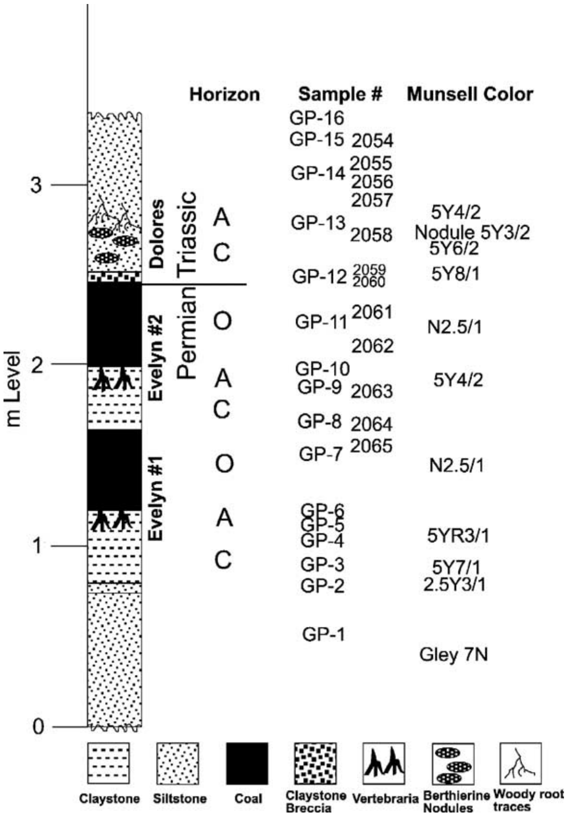

Fig. 1. Stratigraphic column and sample levels.

4. P-T boundary paleosols

The boundary interval studied here consists of three paleosols. The stratigraphic sequence is two Evelyn pedotype coal-bearing paleosols, a "claystone breccia" previously identified as the Permian–Triassic boundary, and a Dolores pedotype Inceptisol-like paleosol (Fig. 1; Retallack and Krull, 1999; Krull and Retallack, 2000). As in other Permian–Triassic boundary sequences in South Africa (Ward et al., 2000) and Russia (Newell et al., 1999), there is a shift to coarser grain sizes across the Permian–Triassic boundary, with the Evelyn paleosols both having well-developed underclays and the Dolores paleosol being silty (Fig. 1). The shift to braided streams observed elsewhere in Gondwana (e.g., Ward et al., 2000; Michaelson, 2002) is higher in the section, ~24 m above the Permian–Triassic boundary (Retallack and Krull, 1999).

4.1. Comparisons between paleosols

The obvious difference between the paleosols is the presence of coals (soil O horizons) preserved at the top of the Evelyn paleosols. The early part of the Triassic is marked globally by a stratigraphic coal gap and a major floral turnover event (Retallack et al., 1996; Looy et al., 1999). Thus, the Dolores pedotype does not simply represent an Evelyn paleosol that has had its coal erosionally removed, because in

Table 1
Comparison of Latest Permian and Earliest Triassic paleosols at graphite peak

Feature	Late Permian	Earliest Triassic
Pedotype	Evelyn (coal measure)	Dolores (gleyed inceptisol)
Root traces	Carbonaceous	Non-carbonaceous
Vegetative covering	Coal swamp	Early successional
$\delta^{13}C_{org}$[a]	−24‰ to −21‰	−45‰ to −24‰
Nodular berthierine	no	yes
Ba/Sr	~2	>6
Major element ratios (Al/Si, Ti/Al, etc.)	Steady	Steady
LREE	Relatively depleted	Relatively enriched
HREE	Relatively depleted	Relatively enriched

[a] Late Permian values are very steady, while early Triassic values are extremely variable, but generally much lighter. The mean Triassic value is ~−31‰ while the mean Permian value is >−24‰ (Krull and Retallack, 2000).

the next 120+ m of section above the boundary at Graphite Peak, none of the Dolores paleosols have an associated coal. It is only after Dolores paleosols disappear from the stratigraphic sequence that coal-bearing paleosols re-appear. The ecological shift from coal swamps and forests to early successional vegetation that accompanied end-Permian mass extinction was also accompanied by a shift from meandering to braided streams in Antarctica (Retallack and Krull, 1999). In spite of these floral and sedimentation style changes, the Evelyn and Dolores paleosols occupied the same lowland, stream proximal position on the landscape and the Dolores paleosol is essentially an Evelyn paleosol without coal (Retallack and Krull, 1999). Thus, the differences between the paleosols in horizonation, (O-A-C for the Evelyn paleosols, A-C for the Dolores paleosol), root traces, berthierine nodules, and chemostratigraphy (Table 1) are attributable to changing environmental conditions and formation time rather than to physiographic setting.

4.2. Major element geochemistry

A common method of discerning between paleosol types (e.g., Sheldon et al., 2002) and of addressing the conditions under which they formed is to look at molar ratios of major elements that are related to soil formational processes (e.g., Sheldon, 2003). The molar ratio of K and Na to Al is a function of the salinization of a paleosol (Retallack, 1997). Similarly, the molar ratio of Al to Si is a function of the clayeyness of a paleosol (Retallack, 1997). Finally, the molar ratio of two common fairly immobile refractory elements (Ti and Al) should be nearly constant through a paleosol if the parent material weathered to form the soil was consistent. Fig. 2 depicts these three element ratios and demonstrates that the parent materials for all three paleosols spanning the Permian–Triassic boundary were similar. This result supports the hypothesis that the primary difference between the Evelyn and Dolores paleosols is environmental or climatic.

4.3. Trace element geochemistry

Trace element ratios are also useful for assessing formational processes in paleosols. The molar ratio of Ba to Sr is an indicator of leaching (Retallack, 1997), because though both elements have similar atomic radii and similar chemical affinities, Sr is significantly more soluble than Ba (Vinogradov, 1959). Retallack (1997) suggests that Ba/Sr ratio values in excess of 2 indicate acidic/leached conditions. An increase in elemental leaching can be attributed to either increasing chemical weathering or changing redox conditions for redox sensitive elements such as Fe or Mn. The earliest Triassic Dolores paleosol shows considerably more subsurface leaching than either of the latest Permian Evelyn paleosols (Fig. 3A; Ba/Sr). This result is further supported by REE element data from all three paleo-

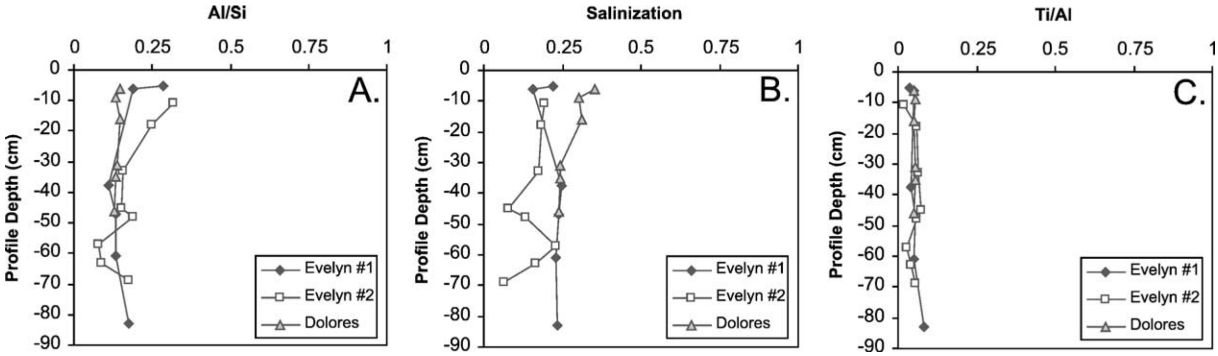

Fig. 2. Major element chemistry across the Permian–Triassic boundary.

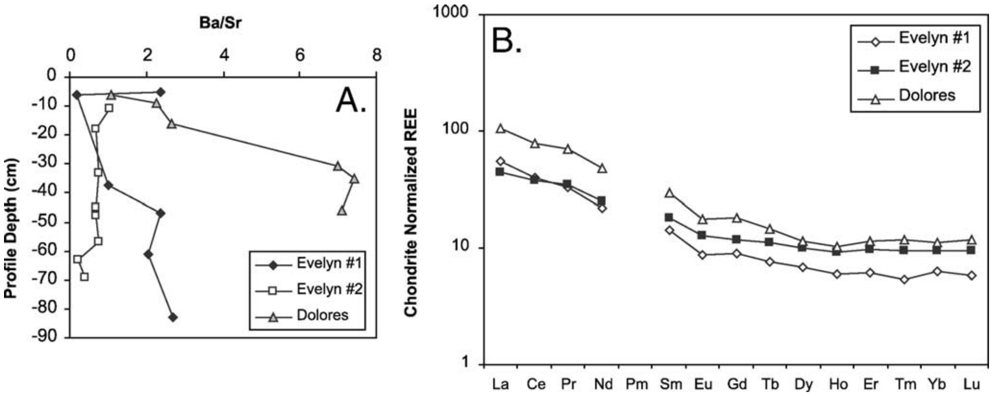

Fig. 3. Trace element comparisons between the latest Permian and earliest Triassic paleosols. (A) Molar ratio of Ba/Sr; B) mean REE values from the penultimate ($n=5$) and last ($n=4$) Permian Evelyn paleosols and the earliest Triassic Dolores paleosol ($n=2$). All values are chondrite normalized using the data of Nakamura (1974).

sols. The earliest Triassic Dolores paleosol is characterized by both light and heavy REE enrichment relative to the latest Permian Evelyn paleosols (Fig. 3B).

5. Discussion

5.1. Consistent provenance?

The change in REE element chemistry could potentially be attributable to a change in sediment provenance if the sediments that constituted the parent material of the Dolores paleosol were REE-enriched relative to those of the Evelyn paleosols. However, a number of trace element and REE ratios are consistent for all three paleosols (Fig. 4; others not depicted). These data are evidence for a common provenance for all three paleosols and comparable weathering processes. Furthermore, the nearly constant Ti/Al ratio of the paleosols (Fig. 2) supports the hypothesis that the parent materials for the paleosols were consistent across the Permian–Triassic boundary in spite of the changes in grain size.

5.2. Post-apocalyptic greenhouse

In Antarctica there is a wholesale change in the Permian compared with Triassic paleosols from coal-measure paleosols to weakly developed and unusual chemically reduced fluvial paleosols (Sheldon and Retallack, 2002) and paleo-Ultisols (Retallack and Krull, 1999). This change has previously been attributed to a “post-apocalyptic greenhouse,” which was proposed as an explanation for deeply weathered paleo-Ultisols in the Early Triassic in Antarctica (Retallack and Krull, 1999) and Australia (Retallack, 1999) at paleolatitudes for which there is no modern analogue. The greater degree of leaching in the earliest Triassic, accompanied by enrichment of typically immobile REEs suggests that weathering intensity increased significantly.

Evelyn paleosols formed over tens of thousands of years on the basis of coal accumulation rates (20–50 ka; Retallack and Krull, 1999, using accumulation rates of 0.5–1.0 mm year^{-1} from Falini, 1965 and assuming compaction to 10% of the original thickness, cf. Shel-

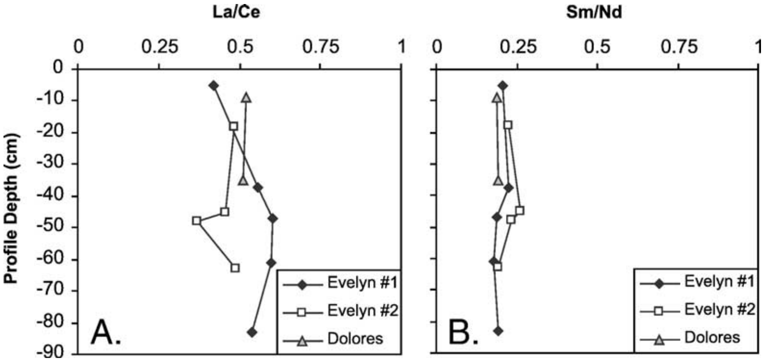

Fig. 4. Trace element ratios for latest Permian and earliest Triassic paleosols. (A) molar ratio of La/Ce; (B) molar ratio of Sm/Nd.

don and Retallack, 2001), whereas the Dolores paleosol formed over hundreds to perhaps a couple of thousand years based on immature horizonation and comparison to modern analogues (0.5–2 ka; Retallack and Krull, 1999). In spite of this discrepancy in formation times, the Dolores paleosol exhibits significantly more leaching (Ba/Sr ratio; Fig. 3A) and significantly more residue of immobile REE (Fig. 3B) indicating that it experienced stronger chemical weathering than the Evelyn paleosols. Stronger overall chemical weathering within an order of magnitude less time is evidence of an abrupt increase in chemical weathering rates across the Permian–Triassic boundary.

An abrupt shift to greenhouse conditions or to an enhanced greenhouse could account for this change. High-latitude paleo-Ultisols (Retallack, 1999; Retallack and Krull, 1999) and augmented chemical and physical weathering (Retallack and Krull, 1999; Newell et al., 1999; Ward et al., 2000; Michaelson, 2002) are also consistent with an enhanced "post-apocalyptic" greenhouse in the earliest Triassic.

A rapid enhancement of chemical weathering rates could occur in response to at least one other type of environmental change: acid rain. While a greenhouse shift would lower rainwater pH and increase leaching, significant addition of H_2S to the atmosphere from a bolide impact or flood volcanism could significantly enhance this effect. Changes in sulfur geochemistry across the Permian–Triassic boundary in South Africa have been invoked as evidence of acid rain (Maruoka et al., 2003), and recent model results (Kump et al., 2005) suggest that oceanic anoxia (e.g., Hallam and Wignall, 1997) at the Permian–Triassic boundary could have resulted in a significant release of H_2S to the atmosphere, while also increasing atmospheric CH_4 levels. Given that the Antarctic Permian–Triassic boundary paleosols and their associated sediments are carbonate-free, they would have had limited buffering capacity to overcome a shift to very acidic rain ($pH < 4.5$), and so should show any evidence of acid rain if it occurred. In modern soils affected by acid rain and with limited buffering capacity, there is extensive leaching of Ca and Mg (Tomlinson, 2003). This is not observed in the Permian–Triassic boundary paleosols in Antarctica, which all have gained Ca relative to their parent material and which have little change in Mg (see Supplemental Data). Thus, at present there is no strong evidence to invoke H_2S-acid rain in Antarctica. Instead, all of the evidence (see above), is consistent with an enhanced earliest Triassic greenhouse, which could have been caused by a number of different mechanisms.

5.3. Tempo of the greenhouse shift

As discussed above, the shift to greenhouse conditions occurs between the last Evelyn paleosol of the Permian and the first Dolores paleosol of the Triassic. With no evidence for an unconformity (Retallack and Krull, 1999; Krull and Retallack, 2000) between the two soils, the tempo of the greenhouse shift must have been very rapid. The Dolores paleosol is derived from a sandstone that rests almost immediately on the end-Permian Evelyn paleosol, separated only by a thin (<15 cm) claystone "breccia" (Retallack and Krull, 1999), unique in the section to this level. The term claystone "breccia" is a bit of a misnomer as most of the claystone clasts are not angular, and are thought to represent individual soil peds that have been deposited as a disturbance layer (Retallack, 2004) as in a modern clear-cut or forest fire, and it represents an aggradational rather than erosional feature (i.e., there is no unconformity; Retallack et al., 2005). Average sedimentation rates (Retallack and Krull, 1999) have been estimated for both the Late Permian (0.36–0.74 mm $year^{-1}$) and the earliest Triassic (0.88–2.24 mm $year^{-1}$), so given no evidence for erosive scouring by the deposition of the claystone, it is highly unlikely that it represents much geologic time, probably much less than 10,000 years based on the thin nature of the claystone. Though it is not possible to restrict estimates of timing any better than that in this section, it is nonetheless clear that the greenhouse shift occurred very rapidly.

In addition to being consistent with the globally rapid shift from meandering to braided streams (Retallack and Krull, 1999; Newell et al., 1999; Ward et al., 2000; Michaelson, 2002), this result is consistent with the pattern of vertebrate extinction that has been documented in South Africa, where a gradual Late Permian extinction was punctuated by an abrupt extinction pulse at the Permian–Triassic boundary, coincident with the negative $\delta^{13}C$ anomalies (Ward et al., 2005). Siberian Traps flood volcanism (e.g., Maruoka et al., 2003) occurred both before and after this rapid greenhouse event, and so is unlikely to have been the primary cause because it represents a geologically gradual process. Thus, while it is tempting to invoke a bolide impact because it is a geologically instant phenomenon, recent results (Farley and Mukhopadhyay, 2001; Langenhorst et al., 2005) have questioned some of the lines of evidence suggestive of an impact event. Nonetheless, some geologically rapid phenomenon is necessary to explain the observed rapidity of the shift to greenhouse conditions.

5.4. A role for methane?

The idea that elevated atmospheric CH_4 levels played a role in end-Permian and earliest Triassic soil formation processes is supported by mass balance results that suggest the extreme $\delta^{13}C$ fluctuations (Table 1; Krull and Retallack, 2000; de Wit et al., 2002; Ward et al., 2005) across the Permian–Triassic boundary can only be attributed to the injection of isotopically light CH_4 into the atmosphere (Berner, 2002). The source of methane could be marine or terrestrial (permafrost) clathrates (e.g., Krull and Retallack, 2000) or possibly be a response to oceanic anoxia and atmospheric H_2S build-up (Kump et al., 2005). Temporarily elevated methane levels would result in low soil-oxygen levels that could have contributed to or exacerbated the extinction of rooted plants (Sheldon and Retallack, 2002). Previous work identified the unusual serpentine-group mineral berthierine at five Gondwanan terrestrial Permian–Triassic boundary sites in Antarctica and Australia including Graphite Peak, and suggested that elevated atmospheric and soil pCH_4 and depleted soil pO_2 played a role in its formation (Sheldon and Retallack, 2002, 2003). New field work associated with this project has identified three more earliest Triassic occurrences of berthierine in Antarctica at Coalsack Bluff, Portal Mountain, and Shapeless Mountain, suggesting that its occurrence is widespread (Retallack et al., 2005). At all of the Antarctic sites, berthierine-bearing Dolores paleosols are unique to the earliest Triassic. A final line of evidence that supports low oxygenation levels and a role for methane is the shift in the gleization ratio (Fe^{2+}/Fe^{3+}; Retallack, 1997) of the paleosols across the Permian–Triassic boundary. The Dolores paleosol was significantly more chemically reduced than the underlying Evelyn paleosols (higher gleization ratio; Fig. 5). This is particularly impressive given that coal-forming environments are often at least mildly chemically reducing (e.g., Retallack, 1997). Taken together, these various lines of evidence suggest that elevated atmospheric and soil CH_4 levels were an important contributing factor to the end-Permian extinction and the earliest Triassic recovery interval.

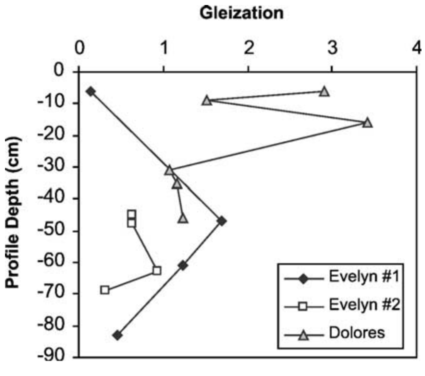

Fig. 5. Gleization ratio across the Permian–Triassic boundary. Gleization is the molar ratio of Fe^{2+}/Fe^{3+}.

6. Conclusions

High-resolution geochemical results from the Permian–Triassic boundary interval at Graphite Peak, Antarctica indicate an abrupt enhancement of chemical weathering in the earliest Triassic even though the formation time for otherwise genetically similar earliest Triassic paleosols was an order of magnitude shorter. This change, accompanied by altered river morphology due to land plant extinctions (Retallack and Krull, 1999; Newell et al., 1999; Ward et al., 2000) and high latitude paleo-Ultisols without modern analogues (Retallack, 1999; Retallack and Krull, 1999), indicates a significantly enhanced earliest Triassic greenhouse (Berner, 2002; Kidder and Worsley, 2003). Also indicated is a role for methane clathrate dissociation, possibly as a cause of the greenhouse (Krull and Retallack, 2000) or as a significant oxygen sink (Sheldon and Retallack, 2002, 2003; Berner, 2002). In addition to methane clathrate dissociation, recent evidence for a meteorite impact and flood volcanism (Retallack et al., 1998; Becker et al., 2001; Basu et al., 2003; Maruoka et al., 2003) and mass-balance modelling examining a variety of causal mechanisms (Berner, 2002) suggest a complex set of causes, perhaps acting synergistically, rather than a singular cause. Furthermore, the evidence of a rapid transition to greenhouse conditions presented here precludes extinction mechanisms that only act gradually (e.g., flood volcanism) from being the sole cause of the event. The contemporaneity of a variety of extinction mechanisms may instead account for the severity of end-Permian extinction.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.palaeo.2005.09.001.

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