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SPECIAL

Carbon isotope excursions and sea-level change: implications for the Frasnian–Famennian biotic crisis

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New carbon and oxygen isotope data from carbonates spanning the Frasnian–Famennian (F–F) boundary in the Guilin area, South China, show a broad positive $\delta^{13}C$ rise and fall, with sharp, short-lived negative $\delta^{13}C$ events; this pattern is comparable to that in Europe and North America. The integration of the isotope stratigraphy with high-resolution sequence stratigraphy corroborates the onset of the positive $\delta^{13}C$ excursion during a third-order sea-level fall in the latest Frasnian. This can best be explained through increased burial of C_{org} during the sea-level fall, brought about by increased organic productivity caused by increased continent-derived nutrient flux to the ocean due to enhanced weathering through the proliferation of land plants in the Devonian. This scenario would have resulted in anoxic and eutrophic conditions over epicontinental seas and blooms of cyanobacteria, creating a highly stressful and fragile ecosystem for oligotrophic normal-marine benthic organisms and leading to their massive decline. The global third-order sea-level fall near the end of the Frasnian may have led to gas hydrate dissociation (giving the negative $\delta^{13}C$ events), and caused wild climatic fluctuations. The subsequent short-term events of sea-level rise, anoxia and eutrophication in the latest part of the F–F transition would have placed additional environmental stresses on the already weakened biota, leading to their further demise.

Keywords: South China, Devonian, mass extinction, ^{13}C, sequence stratigraphy.

The Frasnian–Famennian (F–F) mass extinction has been recognized as one of the five greatest biotic crises in the Phanerozoic (Sepkoski 1986). A stepwise rather than a sudden decline of organisms is more evident on the basis of studies of different faunal groups (e.g. Copper 1986; McGhee 1989). The biotic decline was largely dependent on the palaeolatitude and bathymetric level. Low-latitude shallow-marine organisms were severely affected, whereas those living in high-latitude, deep-water and terrestrial environments were only slightly affected (e.g. Copper 1986; Joachimski & Buggisch 1993). The cause of this biotic event has been extensively debated, and several hypotheses have been proposed including bolide impact (McLaren 1970), oceanic anoxia (e.g. Joachimski & Buggisch 1993) and global cooling (Copper 1986; McGhee 1989). The bolide impact hypothesis derives from studies of F–F sections in South China (Wang *et al.* 1991; Yan *et al.* 1993). However, geochemical studies in Europe and North America have suggested oceanic anoxia and eutrophication, related to long-term transgression, as more likely (e.g. McGhee *et al.* 1986; Joachimski & Buggisch 1993; Wang *et al.* 1996; Joachimski 1997; Murphy *et al.* 2000; Joachimski *et al.* 2001).

This study attempts to constrain the environmental changes across the F–F boundary in South China through the integration of sequence stratigraphy with carbon and oxygen isotope records. This multidisciplinary approach provides more realistic information of the F–F mass extinction.

Geological location and depositional sequences. The studied section is located in a quarry at Baisha between Guilin and Yangshuo. Palaeogeographically, it was located in the spindle-shaped Yangshuo basin, which was surrounded by shallow-water carbonate platforms (Fig. 1; Chen *et al.* 2001*a*).

Two depositional sequences (Sfr, Sfa1), arising from third-order cycles of relative sea-level change, are recognized in the F–F transitional strata (Fig. 1), and these can be correlated in platform and basin successions across South China and elsewhere (Chen *et al.* 2001*a, b*; Fig. 2). The sequence Sfr terminates very close to the top of the Frasnian, and the overlying sequence (Sfa1) straddles the F–F boundary (Figs 1 and 2). In this case, a third-order sea-level highstand and fall occurred near the end of the Frasnian (within the *P. linguiformis* Zone), as indicated by calciturbidites with peloids and calcispheres at the top of the Gubi Formation and a major palaeokarst on adjoining platforms. Four smaller-scale parasequence sets of coarsening-up beds within the calciturbidites (Fig. 1) were caused by higher-frequency (fourth-order) sea-level fluctuations. Sequence Sfa1 also has an upper calciturbidite unit. Three thin layers of dark to black shale (beds a–c, Fig. 1), each 2–5 cm thick, at the base of Sfa1, suggest three rapid, short-lived anoxic events in the very latest stage of the F–F transition. The F–F boundary is placed at the base of the second shaly layer (bed b), based on conodont data (see Fig. 1; GMRBG 1994).

Analytical methods. Samples for analysis were collected from approximately 40 m of section from sequence Sfr to Sfa2 at Baisha, and high-resolution sampling was carried out in the section spanning the F–F boundary (Fig. 1). The C- and O-isotope ratios are reported in per mil relative to PDB, with analytical reproducibility better than 0.1‰ for both.

Results and evaluation. Figure 1 shows the carbon- and oxygen-isotope variations across the F–F boundary. The background $\delta^{13}C$ values are in the range of +0.5 to +1.5‰, as shown in the lower *c.*15 m and upper *c.*20 m of the section. A rapid increase in $\delta^{13}C$ to values around +2.5‰ begins *c.*4 m below the F–F boundary, roughly coinciding with the first occurrence of calciturbidites of Sfr. High $\delta^{13}C$ values are observed (average

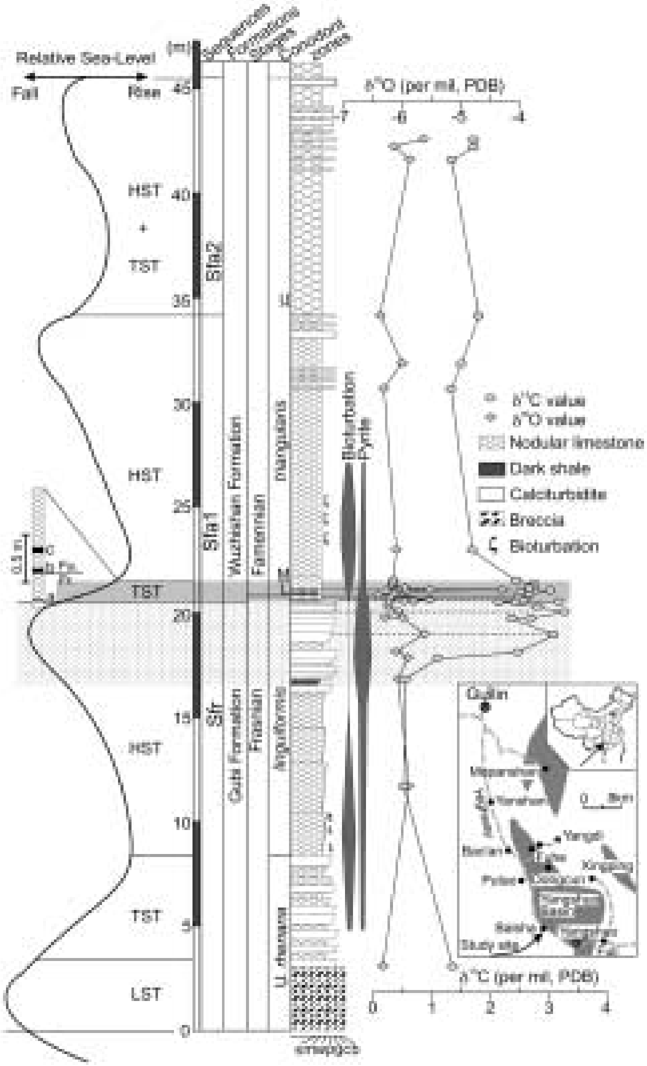

Fig. 1. Lithological logs and δ^{13}C and δ^{18}O variations across the F–F boundary at Baisha, Guilin, South China. Lithological scale: s, shale; m, mudstone; w, wackestone; p, packstone; g, grainstone; c, conglomerate; b, boulder. Depositional systems tracts: LST, TST and HST are lowstand, transgressive and highstand systems tracts respectively. Vertical spindle bars represent relative abundance. Dotted and shaded horizons represent the latest HST and the transgressive deposits of sequences Sfr and Sfa1, respectively. Inset map shows the location of studied section and palaeogeographical setting, in which shaded and non-ornamented areas indicate the basins and platforms in the Devonian respectively.

c.+2.7‰; maximum +3.3‰) upwards through the F–F boundary to *c.*1 m above the boundary, whence δ^{13}C rapidly decreases to values around +1.5‰ within an interval less than 1 m. Within the zone of more positive δ^{13}C, there are two visible negative shifts in δ^{13}C at *c.*0.3 m and *c.*1 m below the F–F boundary, at the top of and within the calciturbidites.

The δ^{18}O values remain relatively similar (*c.*−6.0‰) throughout the section, and compare well to δ^{18}O values measured on diagenetically unaltered brachiopods (e.g., Veizer *et al.* 1986), but slightly lighter than those of abiotic marine cements (e.g., −4.5 to −5.3‰; Carpenter *et al.* 1991). An exceptional enrichment in ^{18}O of +1.5‰ occurs in the third organic-rich shaly layer (bed c, Fig. 1), which precisely coincides with a secondary positive excursion of δ^{13}C (*c.*+3 ‰). In general, secondary δ^{13}C peaks are correlated positively with δ^{18}O peaks in the heavy C interval (Fig. 1), although the latter are not as pronounced as the former.

There are no visible diagenetic effects in thin-sections of our samples, and the homogenous lime mudstones chosen for analysis are non-luminescent. This is further supported by the very stable δ^{18}O values across the section and the low organic carbon content. Even in the relatively organic-rich shaly beds (e.g. beds a–c), there are no ^{13}C- and ^{18}O-depletions in the carbonates (Fig. 1), which would indicate diagenetic alteration through anaerobic oxidation of organic matter (rich in ^{12}C) (Coniglio 1989; Joachimski *et al.* 2001) and/or reaction of burial diagenetic fluids (elevated temperature) with the sediments (e.g. Gao 1993). On the contrary, secondary positive shifts occur for both the carbon and oxygen isotopes of these horizons, in comparison to those of pure limestones (Fig. 1). In addition, apparent covariance between δ^{13}C and δ^{18}O, an indicator of diagenetic alteration (e.g. Rosales *et al.* 2001), is not present. Thus, the isotopic compositions are considered minimally altered diagenetically, and the δ^{13}C values obtained here are assumed as (or nearly) primary signals of coeval seawater.

Global comparisons. The data presented here clearly show a broad positive δ^{13}C anomaly (a heavy-C excursion) across the F–F boundary, with two short-lived negative events (Fig. 1). A similar synchronous heavy-C excursion was reported at Liujing farther south in China (Chen *et al.* 1995), and at Xiangtian a short-lived negative δ^{13}C at the F–F boundary was reported (Wang *et al.* 1991; Yan *et al.* 1993). A broad positive δ^{13}C rise and fall across the F–F section with a negative δ^{13}C event just below the boundary has also been identified in Canada (Geldsetzer *et al.* 1993; Wang *et al.* 1996). The broad positive δ^{13}C excursion stratigraphically corresponds to the top of sequence Sfr and the base of sequence Sfa1, the time of a third-order sea-level fall and rise respectively, spanning approximately 0.5 Ma (Chen & Tucker 2003).

Positive δ^{13}C excursion, third-order sea-level fall and the major biotic crisis. A third-order sea-level highstand and fall occurred in the uppermost part of the Frasnian, as indicated by calciturbidites and palaeokarst in China. This exposure event has also been recognized worldwide (e.g. Johnson *et al.* 1985; Sandberg *et al.* 1988; Muchez *et al.* 1996; Van Buchem *et al.* 1996; House *et al.* 2000; see Fig. 2). Most importantly, the beginning of the sea-level fall coincides with the onset of the heavy-C excursion of the F–F boundary interval (Fig. 1). The simplest interpretation is that this heavy excursion is due to increased burial of C_{org} resulting from an increase in primary bioproductivity. This could have been brought about by increased riverine nutrient flux to the ocean, induced by the sea-level fall. In Late Devonian times, there would have been enhanced chemical weathering through the proliferation of land plants (France-Landord & Derry 1997; Gröcke *et al.* 1999). Enhanced nutrient supply would have stimulated blooms of marine algae and cyanobacteria and resulted in anoxia and eutrophication in semi-restricted epicontinental seas and basins (Algeo *et al.* 1995; Algeo & Scheckler 1998). The strontium isotope data (high radiogenic ^{87}Sr/^{86}Sr ratios) from well-preserved brachiopod shells in F–F boundary strata also suggest a higher continental input into the ocean at this time (Veizer *et al.* 1997).

Sedimentological evidence from the basinal deposits, i.e., black laminated sediments rich in pyrite and lack of bioturbation in the top of Sfr (Fig. 1), supports anoxic intermediate to bottom

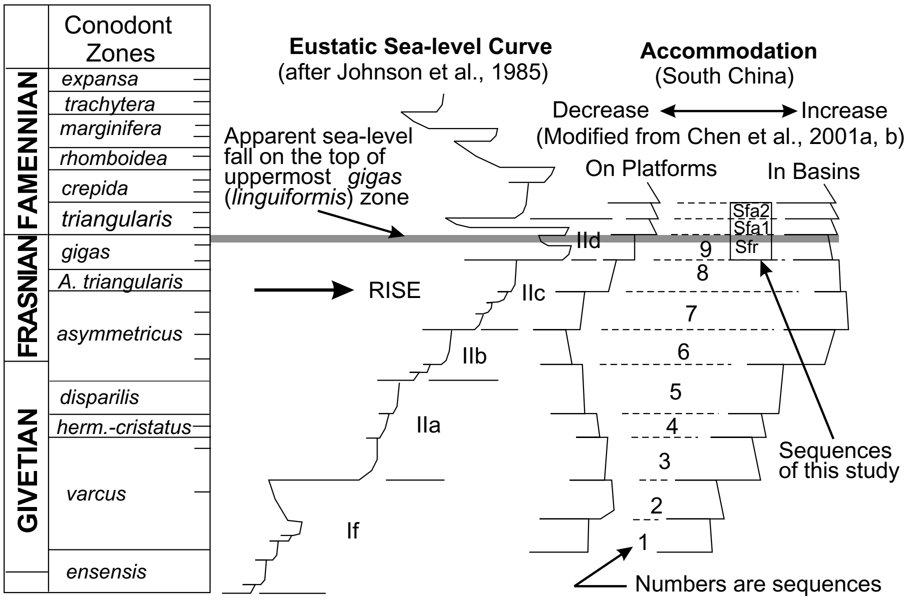

Fig. 2. Comparison of changes in accommodation space for platforms and basins in South China in the Upper Devonian with the eustatic sea-level curve for Euramerica from Johnson *et al.* (1985). The shaded bar indicates the timing of the eustatic sea-level fall in the latest Frasnian.

waters during the sea-level fall. These conditions may have been enhanced through over-consumption of oxygen in surface waters by decaying phytoplankton and cyanobacteria and expansion of the oxygen-minimum zone. Anoxic conditions would have enhanced the burial of C_{org} (e.g. Arthur *et al.* 1988), leading to the positive δ^{13}C excursion in the latest Frasnian.

The simultaneous expansion of cyanobacterial microbes such as *Renalcis* and *Epiphyton* on platform margins in the latest Frasnian supports eutrophic conditions (see Chen *et al.* 2001*a*; Chen *et al.* 2002). The abundant calcispheres derived from the platform and present in the calciturbidites also suggest abnormal salinity and nutrient levels. Carbon isotopic compositions of organic matter in F–F strata determined by Joachimski *et al.* (2001) implicate a major contribution from cyanobacteria under eutrophic conditions.

Observations on the platform successions indicate that the shallow-water, normal-marine benthic fauna (e.g. stromatoporoids, rugose corals and most brachiopods) were in massive decline at this time of flourishing cyanobacterial colonies and calcispheric phytoplankton during the third-order sea-level fall, although a small number of species did survive (Chen & Tucker 2003). This scenario implies that the onset of eutrophication reduced the ecological diversity among reef-builders and other oligotrophic-adapted marine taxa, probably mainly through the deterioration of surface-water clarity and anoxic bottom waters, as respiratory demands exceeded O_2 supply in the presence of excess nutrients (Hallock 1987; Murphy *et al.* 2000).

Third-order sea-level rise and final extinction. Following the third-order relative sea-level fall near the end of the Frasnian, a rapid third-order sea-level rise with three short-term sea-level fluctuations superimposed, is recorded in the carbonates at the base of Sfa1 (Figs 1 & 2); heavy δ^{13}C values are still apparent here, although they rapidly become lighter upwards (Fig. 1). The rapid sea-level rise could have elevated anoxic bottom waters and spread eutrophic surface waters over shallow-water platforms; relatively oxygenated basinal facies were deposited at this time (Fig. 1). This situation would have provided ideal conditions for the continued blooming of cyanobacteria in the upper water-levels (see Chen *et al.* 2001*a*; Chen & Tucker 2003), and sustained the burial of C_{org} during the transgressive stage, as recorded in the upper part of the heavy-C interval (Fig. 1). However, the progressive sea-level rise into the early highstand stage would have reduced the continental nutrient flux to the oceans through the flooding of land areas; this would have lead to decreased burial of C_{org} and accordingly a shift of δ^{13}C values to the background level. Within this zone, a secondary positive δ^{13}C anomaly (i.e. in shaly bed c, Fig. 1) likely resulted from a short-term anoxic event produced by an increased continental nutrient flux during a short-term sea-level fall.

The frequent eustatic fluctuations and anoxia/eutrophication during the lowest Famennian third-order sea-level rise, however, would have exerted further serious environmental stresses on the already-weakened benthic fauna which had survived the earlier major biotic crisis, leading to their further demise. After this, only a few species (e.g. gastropods, ostracodes and small mud-adapted brachiopods) survived, to recover later in shallow-water regions (Chen & Tucker 2003).

Negative δ^{13}C events: gas hydrate dissociation? As noted earlier, many F–F sections around the world show sharp negative δ^{13}C events, within the broad positive rise and fall across the boundary. One such isotope anomaly has been interpreted as the result of a sudden decrease in organic productivity triggered by a meteorite impact (e.g. Wang *et al.* 1991; Yan *et al.* 1993), although the data have been contested. However, negative δ^{13}C events could also be caused by the dissociation of gas hydrates. These mixtures of water, ice and methane (clathrates) usually occur within outer shelf and slope sediments at water depths in excess of 300 m (see Haq 1998). Sea-level falls, bottom-water temperature rises and tectonic activities can cause gas hydrate dissociation, which typically leads to slope instability and collapse, and the release of massive volumes of methane into the atmosphere. The sea-level fall in the latest Frasnian (locally over 100 m; e.g. Van Buchem *et al.* 1996) may have triggered gas hydrate dissociation. Local huge platform collapse recorded close to the top of the Frasnian (e.g. Chen *et al.* 2001*a*) could have been the result. Gas hydrate dissociation events would cause a negative shift of δ^{13}C in carbonate precipitated at the time (e.g. Hesselbo *et al.* 2000), and such events may account for the negative δ^{13}C anomalies within the heavy-C interval (Fig. 1). Methane release could also have caused rapid global warming, since methane is an extreme greenhouse gas, and associated rapid sea-level rises and flooding of anoxic water over the platforms, as deduced for the black shale beds and nodular limestones at the base of sequence Sfa1 in the upper F–F transition strata (Fig. 1).

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