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Key Points:

- Four carbon isotope excursions are recognized in a terrestrial coal seam
- Carbon cycle perturbations during the early Eocene were global in extent
- The four hyperthermals share a similar generic cause

Supporting Information:

- Readme
- Figure S1 and Tables S1 and S2

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Early Eocene carbon isotope excursions: Evidence from the terrestrial coal seam in the Fushun Basin, Northeast China

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Abstract A series of transient global warming events between 56 and 50 Ma are characterized by a pronounced negative carbon isotope excursion (CIE). However, the documents of these hyperthermals, such as Eocene Thermal Maximum 2 and H2 events, have come chiefly from marine sediments, and their expression in terrestrial organic carbon is still poorly constrained. Here we yield a high-resolution carbon isotope record of terrestrial organic material from the Fushun Basin, which displays four prominent CIEs with magnitudes larger than 2.5‰. Based on age constraint and comparisons with deep-sea records, our data provide the first evidence of the four hyperthermals in coal seams and suggest a global significance of these events. Moreover, the difference of CIE magnitudes between marine and terrestrial records shows a significant linear correlation with the marine carbonate CIE, implying that these events are likely attributable to recurring injections of ^{13}C-depleted carbon from submarine methane hydrates and/or permafrost.

1. Introduction

During the late Paleocene and early Eocene, about 59 to 50 Ma, the Earth's surface experienced an interval of progressive warming, peaking in the early Eocene climatic optimum [*Zachos et al.*, 2008]. Superimposed on the long-term warming trend are a series of rapid and transient global warming events, termed hyperthermals, including the Paleocene-Eocene thermal maximum (PETM), ETM2/H1, H2, I1, and I2 [*Nicolo et al.*, 2007; *Zachos et al.*, 2010; *Stap et al.*, 2010; *Abels et al.*, 2012]. All the hyperthermals are characterized by a prominent carbon isotope excursion (CIE) and distinct carbonate dissolution horizons in deep-sea sediments [*Lourens et al.*, 2005; *Nicolo et al.*, 2007; *Zachos et al.*, 2010], signifying a massive input of ^{13}C-depleted carbon into the ocean-atmosphere system [*Dickens et al.*, 1995, 1997]. Among the CIEs, only the PETM has been well studied in marine and terrestrial sediments based on different substrates [*Bowen and Zachos*, 2010], whereas other CIEs have been recognized mostly from marine sediments as terrestrial records are scarce [*Clementz et al.*, 2011; *Abels et al.*, 2012; *Samanta et al.*, 2013]. A fundamental issue regarding these CIE events is whether they share global characteristics and a common cause similar to that defined by the PETM or whether they are documented only locally in marine sediments. If they are all truly global in extent, what are the patterns, expression, and evolution of these events in terrestrial organic material? It is crucial to answer these questions to gain a global picture and to explore the driving mechanisms of the hyperthermals. To address this, we generated a high-resolution carbon stable isotope record of bulk organic matter across an exposed lower Eocene coal seam sequence in the Fushun Basin, Northeast China.

2. Geological Setting and Age Framework

The Fushun basin, located on the eastern limb of the Tanlu Fault zone in China (Figure 1), was formed during the rifting cycle of the Paleocene Period [*Wu et al.*, 2002]. It belongs to a small strike-slip fault basin that trends east-northeast and is approximately 18 km long and 3 km wide. The early Paleogene deposits in the basin are mostly developed with clear stratigraphic sequences from the bottom to the top, including the Lizigou, Guchengzi, Jijuntun, and Xilutian Formations [*Hong et al.*, 1980]. The Lizigou Formation is marked by gray-green thick tuff intercalated with thin coal seams. The Guchengzi Formation consists of a thick coal seam intercalated with carbonaceous shale, indicative of a swampy environment. The Jijuntun Formation is composed of carbonaceous shale, mudstone with interbedded silty mudstones, and thick oil shale beds. Overlying it is a set of thick green mudstones intercalated with marlstones of the Xilutian Formation. This

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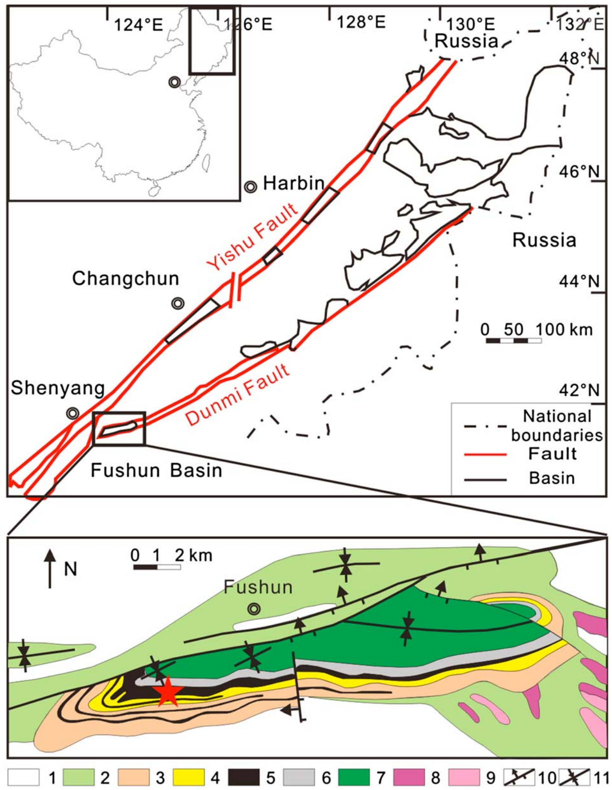

Figure 1. Geological map of Fushun Basin, Liaoning Province, Northeast China, showing the main structures and the location of the Xilutian section (red star). The map is modified from *Meng et al.* [2012]. (1) Proterozoic gneiss. (2) Cretaceous. (3) Paleocene Laohutai Formation. (4) Paleocene Lizigou Formation. (5) Eocene Guchengzi Formation (coal). (6) Eocene Jijuntun Formation (shale). (7) Eocene Xilutian Formation (mudstone). (8) Andesite. (9) Diabase. (10) Fault. (11) Synclinal axis.

study focused principally on the 45 m thick sequence of the Guchengzi Formation coal seam at the Xilutian section (41°50′35.0″N, 123°55′01.1″E), located at the west of the Fushun Basin (Figure 1). Our field observations suggest that the coal seam represents a homogeneous swampy facies and no detectable hiatus exist through the Guchengzi Formation.

Previous studies of the Guchengzi Formation have showed that their pollen assemblage is dominated by tricolpate pollen, especially with the first appearances of some pollen, including *Tricolporollenites hoshyamaensis*, *Ludwigia trilobapollenites*, *Rutaeoipollenites sp.*, and *Tricolpopollenites parmularis* [*Hong et al.*, 1980]. This assemblage reflects the characteristic features of the early Cenozoic plant community [*Hong et al.*, 1980]. Specifically, the first appearance of the diagnostic pollen, *Pistillipollenites mcgregorii*, further assigns the Guchengzi Formation to an age of early Eocene [*Hong et al.*, 1980]. In addition to the palynological evidence, the age of the sediment on the top of the coal seam has been constrained by the paleomagnetic determination of the overlying Jijuntun Formation, which yielded an age of ~52 Ma [*Zhao et al.*, 1994]. On the other hand, our new Ar-Ar radiometric dating of a volcanic tuff in the Lizigou Formation yields the oldest possible age of 55.07 ± 1.18 Ma for the bottom of the coal seam in the Guchengzi Formation (Figure S1 in the supporting information), further constraining the coal seam to the interval between 55.07 ± 1.18 and ~52 Ma. Details of the analytical method and results for Ar-Ar dating are presented in the supporting information.

3. Materials and Methods

A total of 359 bulk samples were collected for stable isotope analyses of total organic carbon (TOC). They were powdered and treated with 1 mol/L HCl for 24 h to remove carbonate, then rinsed with distilled water, and dried at 50°C. Isotopic measurements were carried out on bulk samples using a Flash EA 1112 elemental analyzer, connected to a Finnigan MAT 253 mass spectrometer. For each sample a small pellet of a few milligrams was loaded into a tin capsule. Carbon dioxide for mass spectrometric analysis was generated by O_2 and He flash combustion at 960°C. $\delta^{13}C_{TOC}$ values are reported relative to the international Pee Dee belemnite (PDB) standard (Vienna PDB). Measurement precision for individual analyses is better than ±0.2‰ based on replicate measurements of standards.

4. Results

The $\delta^{13}C_{TOC}$ values of the coal seam vary between −22‰ and −29‰ (Figure 2), within the carbon isotope range of C_3 plants. Our data show that there are four clearly defined negative shifts in $\delta^{13}C_{TOC}$ within the Guchengzi Formation. The first negative shift of $\delta^{13}C_{TOC}$ occurs at 21.7 m with a magnitude of ~6‰, followed by a slow return to ~ −24‰ at 23.2 m, which is 2‰ negative in comparison with the pre-excursion values.

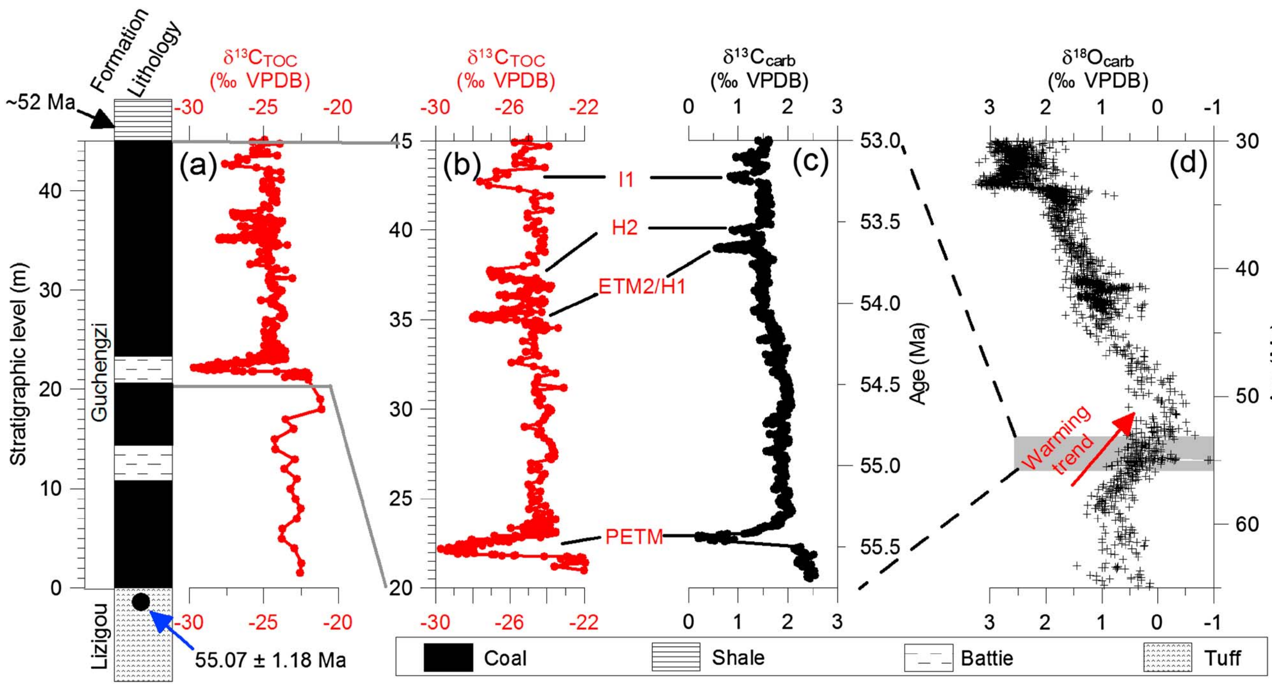

Figure 2. Organic carbon isotope stratigraphy for the Xilutian section and its comparison with deep-sea carbonate δ^{13}C records, highlighting the PETM, H1/ETM2, H2, and I1 carbon isotope excursions. (a) Organic carbon isotope record of the coal seam from the Xilutian section. (b) The details of carbon isotope in Figure 2a, from 20 m to 45 m. (c) Deep-sea carbonate δ^{13}C record from the ODP Site 1262 [*Zachos et al.*, 2010]. (d) The benthic δ^{18}O stack illustrating the climatic context [*Zachos et al.*, 2008]. The blue arrow indicates the radiometric age of a volcanic tuff dated by the Ar-Ar radiometric method. The black arrow indicates the top age of the coal seam from the paleomagnetic determination [*Zhao et al.*, 1994].

The $\delta^{13}C_{TOC}$ then fluctuates around −24‰ with a less variable signal until another two negative $\delta^{13}C_{TOC}$ excursions with magnitudes of ~3.5‰ and ~2.5‰ appear around 35 m and 37.5 m, respectively. The fourth/final negative shift of ~3‰ occurs around 43 m. All the CIEs occur within a homogeneous swampy facies, suggesting that they cannot be explained by a change of lithology or in the sedimentary environment.

5. Discussion

5.1. Interpretation of $\delta^{13}C_{TOC}$

There are three potential scenarios to explain these striking negative shifts in $\delta^{13}C_{TOC}$ within the Guchengzi Formation.

One explanation is that the diagenetic alteration is responsible for the CIEs. It is widely known that during diagenetic processes plant organic matter is subject to physical and chemical variations with each moiety having its own δ^{13}C value and that this may affect the average isotopic composition of plant remains [*van Bergen and Poole*, 2002]. However, some researchers investigated the effects of artificial coalification under controlled laboratory conditions and as a result suggested that the coalification process results in insignificant isotope fractionation, commonly less than 1‰ [*Schleser et al.*, 1999]. In fact, the amount of methane and other higher hydrocarbons liberated during coalification is quite small compared to the huge carbon pool of the coal seam: it can thus be inferred that diagenetic alteration should have little impact on the average isotopic composition of fossil plants and is not likely the cause of the carbon isotope anomalies. The relatively stable background values of $\delta^{13}C_{TOC}$ (~−24‰) between CIEs closely approach the average level of modern C3 plants, further strengthening the argument. Therefore, our $\delta^{13}C_{TOC}$ data are believed to represent the isotopic composition of the pristine living plants.

The second explanation could attribute these CIEs to an increased abundance of angiosperm flora, given potentially more negative δ^{13}C values for angiosperms relative to gymnosperms [*Smith et al.*, 2007]. However, this explanation can be ruled out because palynological data show that angiosperm taxa are the dominated palynomorph, with only a little gymnosperm taxa, and that there is no noticeable change in the taxonomic composition or relative abundance of plant groups through the coal seam [*Hong et al.*, 1980] (cited by *Quan et al.* [2011]).

Third, as land plants fix atmospheric CO_2 by photosynthesis, they discriminate against ^{13}C in favor of ^{12}C, leading to characteristic differences between atmospheric CO_2 and the carbon isotopic composition of terrestrial plants [*Arens et al.*, 2000]. Thus, land plants should track secular changes in the $\delta^{13}C$ of atmospheric CO_2. We can therefore come to the conclusion that the CIEs recorded in the coal seam represent a series of short-term carbon cycle perturbations in the atmospheric carbon pool.

5.2. Comparison of the CIEs Between Terrestrial and Marine Records

Our high-resolution carbon isotope record, coupled with the constraints inherent in the biostratigraphy and in radioactive and paleomagnetic dating, allows a direct comparison with the marine carbonate $\delta^{13}C$ record ($\delta^{13}C_{carb}$) from the Ocean Drilling Program (ODP) Site 1262 [*Zachos et al.*, 2010] and thus helps us to decipher the global and regional characteristics of the hyperthermals. The result shows apparent similarities in pattern and details of the CIEs across the early Eocene (Figure 2). Given the chronological constraints and lack of evidence for any significant stratigraphic break in the coal seam of the Guchengzi Formation, the four CIEs recorded at the Xilutian section are correlated to the PETM, ETM2/H1, H2, and I1 that are clearly defined in the deep-sea sediments [*Zachos et al.*, 2010]. The Xilutian section is the first record of terrestrial organic matter from a coal seam to show clear, isotopic representations of the four Paleogene hyperthermals. These results suggest that the mechanisms producing perturbations in marine $\delta^{13}C_{carb}$ will also produce similar changes in the carbon isotope composition of plants, reflected ultimately in their $\delta^{13}C_{TOC}$ values. In addition, our $\delta^{13}C_{TOC}$ data also show a distinct peak between the ETM2/H1 and H2, which has been documented in the high-resolution benthic $\delta^{13}C$ records from the Walvis Ridge [*Stap et al.*, 2010]. However, whether it represents another hyperthermal event remains an open question.

The discovery of terrestrial equivalents of the three post-PETM hyperthermals demonstrates that these events are globally recognizable and certainly represent a peculiar feature of short duration that repeatedly interrupted the long-term warming trend of the early Eocene, thus reinforcing the suggestion that the PETM was not a unique event but was an extreme example of a series of Paleogene hyperthermals [*Nicolo et al.*, 2007; *Zachos et al.*, 2010; *Abels et al.*, 2012]. A distinguishing feature characterizing these events as described in terrestrial and marine records is that the magnitudes of terrestrial CIEs are generally larger than in deep-sea sections (Figure 2). The exaggeration of terrestrial CIEs has been ascribed to an increase in relative humidity and pCO_2 in the atmosphere [*Bowen et al.*, 2004; *Schubert and Jahren*, 2013] that occurred in direct or indirect response to carbon input to the ocean-atmospheric system. Furthermore, all CIEs experienced a much more gradual onset, with transitional values rather than the single-step or pulsed onset of the PETM recorded in deep-sea sections [*Giusberti et al.*, 2007; *Thomas et al.*, 2002; *Bains et al.*, 1999], meaning that our $\delta^{13}C_{TOC}$ data might indeed reflect the true feature of carbon isotope shifts in the atmosphere.

To assess whether all the CIEs share a similar generic cause, we compared the covariance of differences in the CIEs (ΔCIEs) between terrestrial organic carbon and marine carbonate with the marine CIEs (Figure 3). Our results show that ΔCIEs seem to be significantly correlated ($r = 0.996$, $p < 0.01$) to the CIE magnitudes in $\delta^{13}C_{carb}$. This coherent relationship implies that the terrestrial amplification and/or oceanic dampening of the CIEs are tightly coupled with the amount of carbon added to the ocean-atmosphere system [*Abels et al.*, 2012]. Different carbon sources would mean that significantly different amounts of carbon are required to account for the observed carbon isotope anomaly [*Dickens et al.*, 1995]; further, they would have caused marked differences in atmospheric pCO_2 and in the environment. Assuming that climate sensitivity did not change significantly during the Early Eocene and that these CIEs did not share a similar origin, the ΔCIEs are unlikely to be related to the amount of carbon revealed by the CIE magnitudes; this would also be inconsistent with our results. We therefore conclude that the isotope composition of the carbon source was similar for the four CIEs, as suggested by previous results [*Nicolo et al.*, 2007; *Stap et al.*, 2010; *Abels et al.*, 2012]. The observed correlation through the CIEs does not go through the origin, likely implying either a nonlinear response of local environmental change to carbon release or the existence of further environmental forcing that did not contribute to the CIEs.

5.3. Implications

It is widely accepted that rapid input of ^{13}C-depleted carbon into the ocean-atmosphere system is responsible for the CIEs [*Dickens et al.*, 1997], despite the exact sources of the carbon fuelling the hyperthermals remaining unknown [*Dickens et al.*, 1995; *DeConto et al.*, 2012; *Sexton et al.*, 2011]. Here our

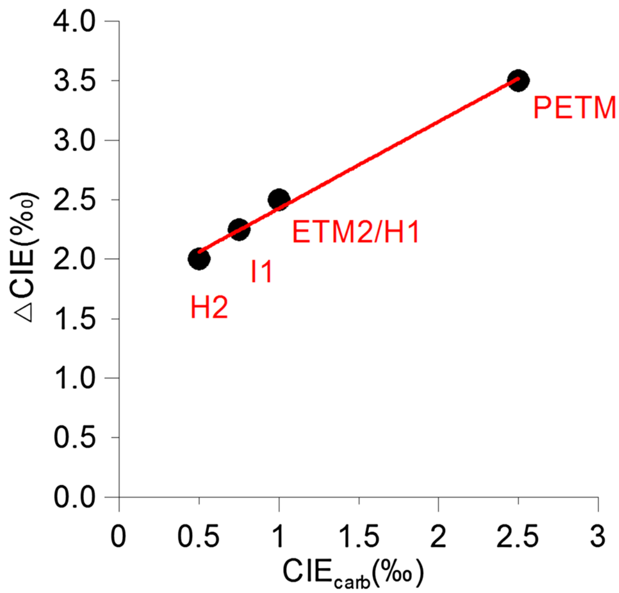

Figure 3. Comparison of differences in carbon isotope excursions (CIEs) between our $\delta^{13}C_{TOC}$ and marine $\delta^{13}C_{carb}$ record (ΔCIE) and the CIE in $\delta^{13}C_{carb}$ from the ODP Site 1262. The significant linear correlation suggests that the four events share a similar cause. $\Delta CIE = CIE_{TOC} - CIE_{carb}$.

high-resolution continental $\delta^{13}C_{TOC}$ record, together with marine records, indicates that the four hyperthermals are of global significance and that they may share a common origin, meaning that they are not unique or stochastic events, such as a comet impact or thermogenic CH_4 production due to magma intrusion to carbon-rich sediments [*Kent et al.*, 2003; *Svensen et al.*, 2004]. This thus demonstrates that they may represent a series of recurring carbon cycle perturbations derived from organic carbon reservoirs on Earth's surface, likely related to orbital forcing [*Lourens et al.*, 2005; *Nicolo et al.*, 2007; *Zachos et al.*, 2010]. These results have three important implications. First, all the events occurred in the context of the early Eocene global warming (Figure 2), implying that the organic carbon reservoirs are sensitive to temperature under high-temperature conditions. Second, the magnitudes of CIEs generally display a tapered trend along with the rise in temperatures (Figure 2), indicating that available amount of organic carbon sources responsible for the CIEs will decrease along with an increase in temperature. The potential candidates for organic carbon sources include submarine methane hydrates and permafrost [*Dickens et al.*, 1997; *DeConto et al.*, 2012], since both are sensitive to temperature and their carbon pools will decrease along with temperature rise. Finally, short-term carbon cycle perturbations recorded in marine $\delta^{13}C_{carb}$ could be one to one mirrored in the $\delta^{13}C_{TOC}$ in the coal seam via the atmosphere (Figure 2); provided that the biostratigraphy is sufficiently well constrained, the CIEs might provide valuable chemostratigraphic markers to correlate marine and terrestrial sites, with a resolution potentially higher than that available from biostratigraphy and magnetostratigraphy.

6. Conclusions

High-resolution $\delta^{13}C_{TOC}$ data display four distinct CIEs in a terrestrial coal seam for the first time. Based on a chronological framework and $\delta^{13}C_{TOC}$ signatures, they correlate to the PETM, ETM2/H1, H2, and I1 already well-defined in marine sections [*Nicolo et al.*, 2007; *Zachos et al.*, 2010]. The recognition of these hyperthermals in a terrestrial coal seam together with their good correspondence with the marine records provides support for the notion that a tight linkage exists between the ocean, atmosphere, and biosphere, confirming that carbon cycle perturbations occurring between 56 and 52 Ma were global in extent and in nature. Because all the events present similar characteristics in the CIEs with different magnitudes, they are likely to be examined by recurrent inputs of ^{13}C-depleted carbon derived from organic carbon reservoirs on the Earth's surface [*Nicolo et al.*, 2007], such as submarine methane hydrates and/or permafrost [*Dickens et al.*, 1997; *DeConto et al.*, 2012]. In addition, the close correspondence between our $\delta^{13}C_{TOC}$ and marine carbonate $\delta^{13}C$ values strengthens the opinion that the $\delta^{13}C_{TOC}$ values of coal seams could be a potentially reliable tool for paleoclimatic interpretation and for stratigraphic correlations between marine and terrestrial sequences.

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Data supporting Figure 2 are available as in the supporting information Table S2. We thank J. Sun for a review of the initial manuscript, Li Chaohui for his fieldwork support, and S. Feng for sampling. We also thank two anonymous reviewers for their helpful comments. This work was supported by the Strategic Priority Research Program of CAS (grant XDB03020503), the (973) National Basic Research Program of China (grant 2013CB956404), and the National Natural Science Foundation of China (grant 40730208).

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