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Carbon isotope fluctuations of terrestrial organic matter for the Upper Cretaceous (Cenomanian–Santonian) in the Obira area of Hokkaido, Japan

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Abstract – Stratigraphic fluctuations of carbon isotope values of terrestrial organic matter within the Upper Cretaceous (Cenomanian–Santonian) sequence in the Obira area of Hokkaido, Japan, record distinctive $\delta^{13}C$ fluctuations for the Cenomanian–Turonian boundary, the Middle Turonian, the upper Turonian–lower Coniacian, and the Santonian. A biostratigraphic framework of the age-diagnostic taxa (ammonoids, bivalves and planktic foraminifers) indicates that these $\delta^{13}C$ fluctuation events are comparable with those recorded in $\delta^{13}C$ data of terrestrial organic matter in Japan and marine carbonates in Europe. These correlations reinforce the utility of these $\delta^{13}C$ events in terms of global chemostratigraphy. In particular, the $\delta^{13}C$ patterns within the overall positive interval of the Cenomanian–Turonian boundary event are highly conformable between marine and terrestrial records. The consistent nature of these different records of $\delta^{13}C$ fluctuation patterns demonstrates that the terrestrial organic $\delta^{13}C$ data mirror the global-scale $\delta^{13}C$ patterns in the carbon reservoir of ocean–atmosphere–terrestrial biosphere during the Cenomanian–Turonian boundary event. In addition, global correlation of short-term marine and terrestrial organic $\delta^{13}C$ fluctuations of the Upper Cretaceous sequence indicate that the magnitude of several terrestrial organic $\delta^{13}C$ events appears more amplified than that of coeval marine carbonate $\delta^{13}C$ events. This correlation is interpreted to mean that the effects of local CO_2 emission into the atmosphere by release of terrestrial methane hydrate or biomass burning of terrestrial vegetation in the hinterland of the NE Asian region have been superimposed on the global $\delta^{13}C$ trend and resulted in the terrestrial organic $\delta^{13}C$ records of the Yezo Group.

Keywords: carbon isotope stratigraphy, Hokkaido, Japan, terrestrial organic matter, Upper Cretaceous.

1. Introduction

Striking stratigraphic fluctuations in the carbon isotopes of carbonates, and marine and terrestrial organic matter, have provided an essential time-control reference for the Upper Cretaceous marine strata. The chemostratigraphic framework is critical as a basis for the clarification of the spatio-temporal climatic and biotic evolution on a global scale, thus detailed Upper Cretaceous $\delta^{13}C$ stratigraphy has been established intensively for marine carbonates in Europe and North America (e.g. Scholle & Arthur, 1980; Gale *et al.* 1993; Jenkyns, Gale & Corfield, 1994; Mitchell, Paul & Gale, 1996; Voigt & Hilbrecht, 1997; Stoll & Schrag, 2000; Tsikos *et al.* 2004; Bowman & Bralower, 2005; Jarvis *et al.* 2006). Recently, Jarvis *et al.* (2006) compiled the $\delta^{13}C$ curve for marine carbonates of the Upper Cretaceous sequence in Europe, and documented the use of 72 carbon isotope events for interregional correlation through the Cenomanian–Santonian sequence. More marine and terrestrial $\delta^{13}C$ data from different regions should be accumulated and these data correlated with the reference $\delta^{13}C_{carbonate}$ profile for the establishment of high-resolution event stratigraphy of the Upper Cretaceous sequence.

The forearc siliciclastic sequence of the Aptian–Paleocene Yezo Group is exposed along a 1200 km long belt that extends from southern Hokkaido in Japan, northward to Sakhalin Island in Russia, along the NW Pacific margin (Okada, 1983; Takashima *et al.* 2004; Shigeta & Maeda, 2005). Because of the extensive distribution with abundant marine macro- and microfossils, the Yezo Group has been employed as a chronostratigraphic reference unit for the circum-Pacific region. Over the past decade, various carbon isotope stratigraphies of terrestrial organic matter have been proposed for the Yezo Group (Hasegawa & Saito, 1993; Hasegawa, 1997, 2003*a*; Ando *et al.* 2002, 2003; Hasegawa *et al.* 2003; Ando & Kakegawa, 2007; Uramoto *et al.* 2007). These studies compared marine and terrestrial carbon isotopes, and established the $\delta^{13}C$ stratigraphy by assuming an isotopic linkage between marine and terrestrial carbon reservoirs within the earth surface subsystems of ocean–atmosphere–terrestrial biosphere; thus, time-stratigraphic terrestrial organic carbon isotope fluctuations for Cretaceous time are

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assumed to reflect δ^{13}C variations in atmospheric CO_2 (Hasegawa, 1997; Ando *et al.* 2002; Gröcke *et al.* 2005). For the Upper Cretaceous sequence of the Yezo Group, several studies have outlined the overall pattern of the δ^{13}C record (e.g. Hasegawa, 1997; Hasegawa *et al.* 2003), and global correlations of the chemostratigraphic events have been documented for the Cenomanian and Turonian sequences (Hasegawa, 2003*a*; Ando & Kakegawa, 2007; Uramoto *et al.* 2007). Further examination of the stratigraphic δ^{13}C record for the Yezo Group will contribute to improvement of the Upper Cretaceous chemostratigraphic framework in the NW Pacific.

The Albian–Campanian sequence of the Cretaceous Yezo Group is widely present throughout the Obira area of Hokkaido (Fig. 1). Several faults recognized in the area record only a minor offset (Fig. 1b), meaning that the complete stratigraphic sequence is preserved. For this reason, many studies have analysed the stratigraphy, palaeontology and geochemistry of the Yezo Group in the Obira area (e.g. Igi *et al.* 1958; Tsushima *et al.* 1958; Tanaka, 1963; Tanabe *et al.* 1977; Matsumoto *et al.* 1981; Sekine, Takagi & Hirano, 1985; Hasegawa & Saito, 1993; Hasegawa, 2001; Nishi *et al.* 2003; Takashima *et al.* 2004; Funaki & Hirano, 2004; Kaneko & Hirano, 2005; Oizumi *et al.* 2005; Takahashi, 2005; Nishimura, Maeda & Shigeta, 2006; Uramoto *et al.* 2007). In terms of carbon isotope stratigraphy, Hasegawa & Saito (1993) presented the first report of the δ^{13}C record around the Cenomanian–Turonian boundary within the Yezo Group, along with the δ^{13}C data for the Oyubari area of central Hokkaido. Uramoto *et al.* (2007) established global chemostratigraphic correlations for the Cenomanian Stage of the Yezo Group, based on the recently refined macro- and microfossil biostratigraphy of the Upper Cretaceous in the Obira area (Nishi *et al.* 2003; Funaki & Hirano, 2004; Oizumi *et al.* 2005). However, stratigraphic δ^{13}C data have yet to be presented and analysed in terms of global chemostratigraphy, for the rocks in the Obira area that overlie the Turonian sequence.

The purpose of this study is to provide a global correlation of the Upper Cretaceous δ^{13}C records between Japan and Europe. We will establish a carbon isotope stratigraphy of sedimentary organic matter for the Upper Cretaceous sequence of the Obira area and then, δ^{13}C patterns are correlated within the Upper Cretaceous marine sequences in NW Pacific and Europe, using the available biostratigraphic framework for the calibration.

2. Geological setting

The middle to upper part of the Yezo Group is widely distributed in the Obira area (Fig. 1b). The sedimentary succession consists of a series of siliciclastic marine sequences up to about 6000 m in maximum thickness. The succession is unconformably overlain by the Tertiary Jugosenzawa Formation.

The lithostratigraphic framework for the Obira area has been presented in previous works (Igi *et al.* 1958; Tsushima *et al.* 1958; Tanaka, 1963; Funaki & Hirano, 2004; Oizumi *et al.* 2005). In this study, we employ the recent lithostratigraphic divisions for the Obira area defined by Funaki & Hirano (2004) and Oizumi *et al.* (2005).

The studied successions of the Yezo Group consist of the conformable Tenkaritoge, Saku and Haborogawa formations, in ascending stratigraphic order. The Tenkaritoge Formation is mainly represented by dark grey siltstone and occasional intercalated thin sandstone. The Saku Formation consists of alternating beds of sandstone and siltstone. The Haborogawa Formation is mainly characterized by dark grey massive siltstone with intercalated sandstone. Occurrences of thick-bedded sandstone (> 1 m) and slump deposits characterize the lower part of the Haborogawa Formation, making it useful for regional correlations (Kamikinenbetsu Sandstone Member: Funaki & Hirano, 2004).

The ages of study units range from Cenomanian to Santonian, based on macro- and microfossil biostratigraphy, and carbon isotope stratigraphy (Fig. 1c). Uramoto *et al.* (2007) recognized the first occurrence of the Late Cenomanian indicator *Inoceramus pictus minus* Matsumoto in the basal part of the study succession within the Tenkaritoge Formation. Hasegawa & Saito (1993) presented the age-diagnostic microfossil biostratigraphy and carbon isotope stratigraphy of terrestrial organic matter within the upper part of the Tenkaritoge Formation, and reported a conspicuous positive δ^{13}C excursion that encompassed the Cenomanian–Turonian boundary. Occurrences of global age-diagnostic macrofossils such as *Vascoceras* cf. *durandi* (Thomas & Peron), *Collignoniceras woollgari* Mantell, *Subprionocyclus* cf. *neptuni* (Geinitz), *Barroisicers* spp. and *Didymotis costatus* (Fric) characterize the Turonian–Coniacian sequences (Tanabe *et al.* 1977; Matsumoto *et al.* 1981; Funaki & Hirano, 2004). Oizumi *et al.* (2005) reported the first occurrence of the Santonian ammonoid *Texanites (Plesiotexanites) kawasakii* (Kawada) from the uppermost part of the study succession in the Haborogawa Formation.

3. Materials and methods

A total of 62 fresh mudstone samples were collected along the Kanajirizawa, Obirashibegawa, Okufutamatazawa and Akanosawa rivers in the Obira area (Fig. 2).

For the evaluation of organic matter, we conducted total organic carbon (TOC) content analyses and Rock-Eval pyrolysis on five selected samples for the Upper Turonian–Santonian sequence. Previous studies have described the compositional and geochemical characteristics of the kerogen for the Cenomanian–Middle Turonian sequence in the Obira area (Hasegawa & Saito, 1993; Hasegawa, 2001; Uramoto *et al.* 2007).

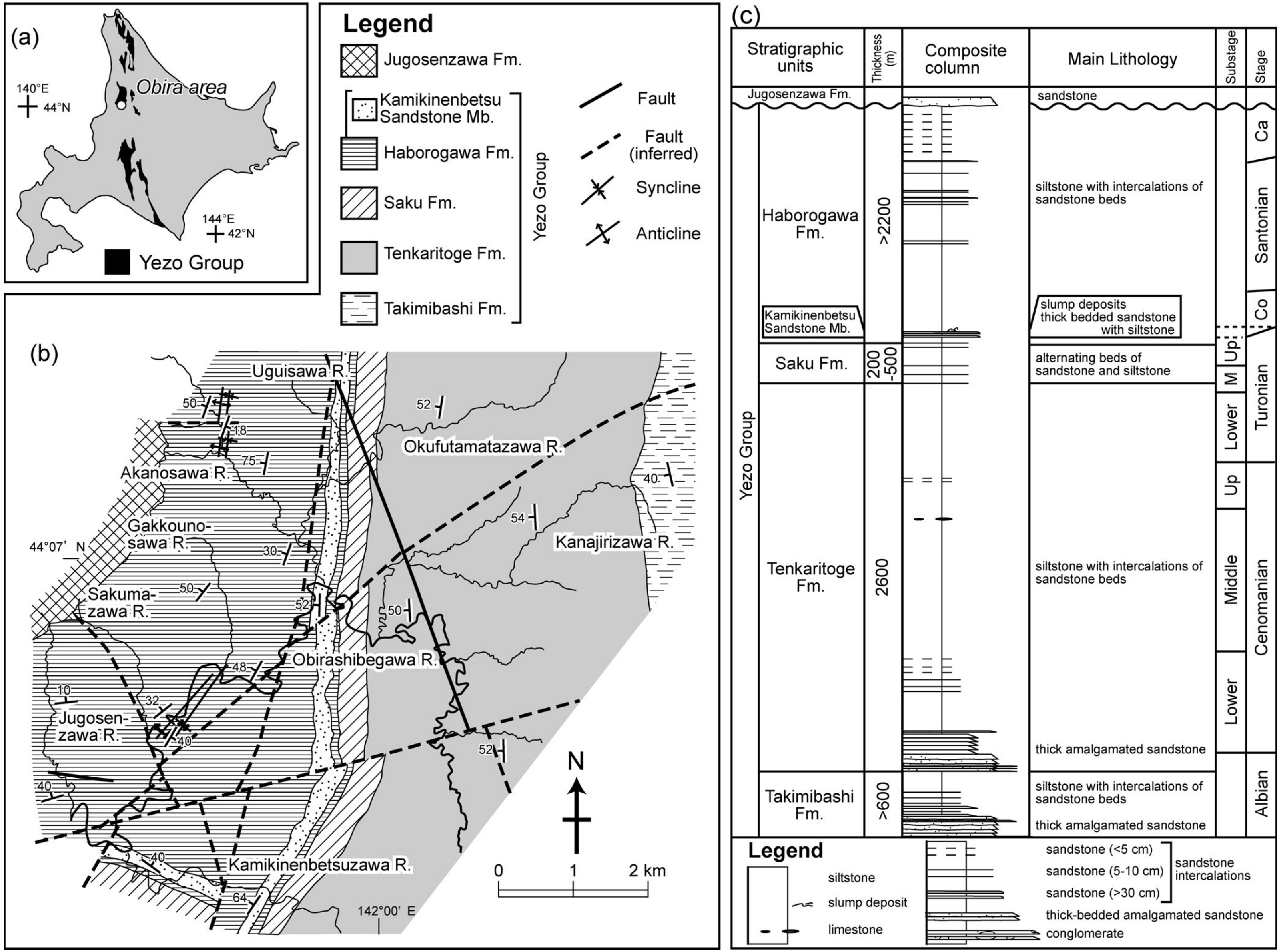

Figure 1. (a) Index map showing the distribution of the Yezo Group (Takashima *et al*. 2004) and locality of the Obira area in Hokkaido, Japan. (b) Geological map of the Obira area (modified after Funaki & Hirano, 2004; Oizumi *et al*. 2005; Uramoto *et al*. 2007). (c) Schematic stratigraphy of the Obira area. Lithostratigraphic divisions are after Funaki & Hirano (2004) and Oizumi *et al*. (2005). Age assignments are from macro- and microfossil biostratigraphy, and carbon isotope stratigraphy by Tanabe *et al*. (1977), Matsumoto *et al*. (1981), Sekine, Takagi & Hirano (1985), Hasegawa & Saito (1993), Nishi *et al*. (2003), Funaki & Hirano (2004), Oizumi *et al*. (2005) and Uramoto *et al*. (2007). Co – Coniacian; Ca – Campanian.

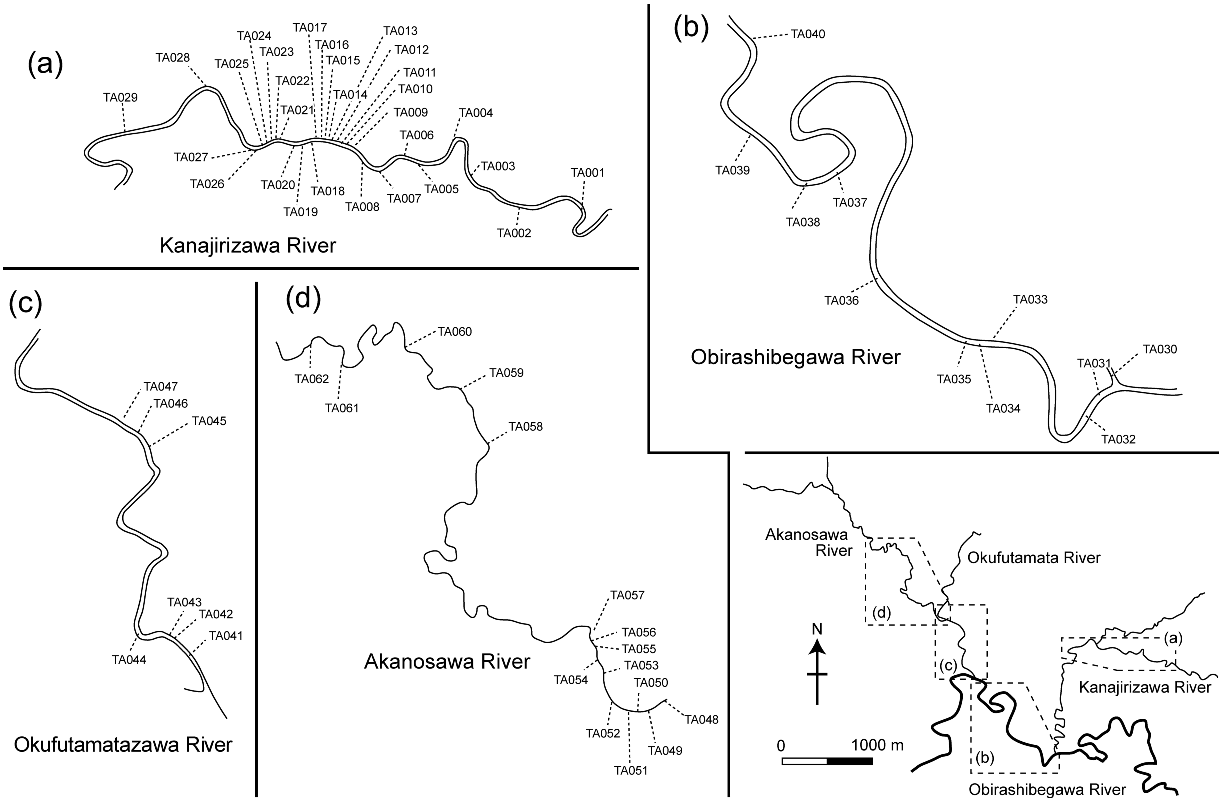

Figure 2. Map showing the sample locations of mudstone in this study.

Powdered bulk samples were acidified with 6N solution of HCl for 24 hours to decompose carbonates. The elemental composition of 20–30 mg of each subsample was then analysed using a Yanaco CHN-Corder MT-3 at JAPEX Research Centre (Chiba, central Japan), employing antipyrine ($C_{11}H_{12}N_2O$) as the standard. The elemental composition of each acid-processed subsample was corrected based on the weight-percent of removed carbonates, and then the TOC content of the whole rock was obtained. We used the TOC values in calculating the Hydrogen Index and Oxygen Index for the following Rock-Eval pyrolysis.

Rock-Eval pyrolysis was conducted using a VINCI Technologies model 6 device at JAPEX Research Centre. From each powdered bulk subsample, 100 mg were pyrolysed from 300 to 650 °C in a nitrogen atmosphere. The amount of free hydrocarbons (S1), hydrocarbons released during heating (S2), CO_2 released during pyrolysis to 390 °C (S3), the temperature at which the maximum amount of S2 hydrocarbons were generated (T_{max}), the Hydrogen Index ($=$ 100·S2/TOC), and the Oxygen Index ($=$ 100·S3/TOC) were recorded.

For $\delta^{13}C$ analyses of organic matter, acid-processed bulk subsamples were treated with a mixture of benzene and methanol (7:3) to eliminate free hydrocarbons. Analyses of $\delta^{13}C$ ratios were performed using a VG Isotech SIRA series II mass spectrometer (precision: ±0.02 ‰) and a GV Instruments Isoprime EA mass spectrometer (precision: ±0.10 ‰) at the JAPEX Research Centre. Carbon isotope ratios were expressed as permil deviation from the PeeDee belemnite (PDB) standard.

4. Results

4.a. Type and maturity of sedimentary organic matter

Table 1 shows the results of total organic carbon content analysis and Rock-Eval pyrolysis, and Figure 3 shows a plot of Hydrogen Index versus T_{max}. The Hydrogen Index values vary from 21 to 34 mg HC/g TOC and T_{max} values range from 433 to 438 °C (Table 1). These ranges correspond to type III/IV kerogen, and the degree of organic maturity is comparable to vitrinite reflectance values between 0.5 and 0.9 %R_o (Fig. 3) (Mukhopadhyay, 1994; Hunt, 1996).

4.b. Carbon isotope records

The obtained $\delta^{13}C$ values of organic matter ($\delta^{13}C_{org}$) are -25.3 ‰ to -21.8 ‰ for the Upper Cretaceous sequence in the Obira area (Table 2), and we plotted our data with the previously reported $\delta^{13}C$ data from the Obira area (Hasegawa & Saito, 1993; Uramoto *et al.* 2007) (Fig. 4). The overall patterns of the Upper Cretaceous $\delta^{13}C_{org}$ profile in the Obira area are independent of lithological variations.

The Upper Cretaceous $\delta^{13}C_{org}$ profile for the Obira area shows remarkable variations at specific horizons (Fig. 4). The most notable feature of the $\delta^{13}C_{org}$ profile in the Obira area is characterized by a $+3.5$ ‰ positive interval from the uppermost part

Table 1. Results of total organic carbon content analysis and Rock-Eval pyrolysis of selected samples

Sample	TOC (%)	S1 (mg/g)	S2 (mg/g)	S3 (mg/g)	HI (mg HC/g TOC)	OI (mg OC/g TOC)	T_{max} (°C)
TA062	0.76	0.00	0.21	0.41	28	54	434
TA061	0.91	0.00	0.27	0.41	30	45	433
TA047	1.15	0.00	0.39	0.47	34	41	436
TA037	1.01	0.00	0.28	0.38	28	38	434
TA036	0.75	0.00	0.16	0.42	21	56	438

TOC – total organic carbon content; HI – Hydrogen Index; OI – Oxygen Index

Table 2. Carbon isotope values in study samples

Sample	δ^{13}C (‰)	Sample	δ^{13}C (‰)	Sample	δδ^{13}C (‰)	Sample	δ^{13}C (‰)
TA016*	−22.27	TA032	−24.14	TA048	−24.44	TA062	−24.64
TA015*	−21.81	TA031	−24.14	TA047	−24.69	TA061	−24.54
TA014*	−22.90	TA030	−24.19	TA046	−24.34	TA060	−24.67
TA013*	−22.52	TA029	−24.56	TA045	−24.00	TA059	−24.83
TA012*	−22.79	TA028	−24.66	TA044	−24.93	TA058	−24.02
TA011*	−23.84	TA027	−23.93	TA043	−24.77	TA057	−23.93
TA010*	−23.49	TA026	−22.47	TA042	−24.70	TA056	−24.06
TA009*	−22.39	TA025*	−22.07	TA041	−24.28	TA055	−23.89
TA008*	−22.29	TA024*	−23.19	TA040	−24.44	TA054	−24.33
TA007	−25.26	TA023*	−23.07	TA039	−24.32	TA053	−24.32
TA006	−25.19	TA022*	−22.93	TA038	−24.90	TA052	−24.63
TA005	−24.36	TA021*	−22.51	TA037	−25.20	TA051	−25.10
TA004	−24.45	TA020*	−21.93	TA036	−25.25	TA050	−24.63
TA003	−24.81	TA019*	−22.41	TA035	−24.42	TA049	−24.64
TA002	−24.61	TA018*	−22.42	TA034	−24.82		
TA001	−23.96	TA017*	−22.03	TA033	−24.67		

Samples with asterisk were analysed with a GV Instruments Isoprime EA mass spectrometer, and other samples with a VG Isotech SIRA Series II mass spectrometer.

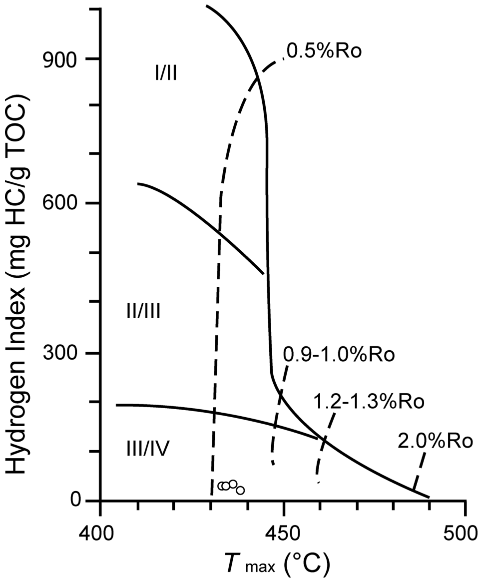

Figure 3. A plot of Hydrogen Index versus T_{max} of selected mudstone samples in the Obira area.

of the planktonic foraminifer *Rotalipora cushmani* Zone to the middle part of the *Whiteinella archaeocretacea* Zone around the Cenomanian–Turonian boundary.

Following the positive excursion, δ^{13}C$_{org}$ values vary between ∼−25 ‰ and ∼−23.5 ‰, superimposed by several short-term positive anomalies of ∼1 ‰ through Turonian–Santonian sequences.

5. Evaluation of sedimentary organic matter

The sedimentary organic matter in the present samples is the type III/IV kerogen (Fig. 3), indicating that sedimentary organic matter with an origin from terrestrial plants predominates in the modal composition of the kerogen (Hunt, 1996).

Based on the estimated vitrinite reflectance values of about 0.5–0.9 %R$_o$ (Fig. 3), the organic maturation level of the samples corresponds to the catagenesis stage (Mukhopadhyay, 1994; Hunt, 1996). It is known that significant changes in the δ^{13}C values of kerogen do not occur below the metamorphic stage (Teerman & Hwang, 1991; Whiticar, 1996). Our results demonstrate that the maximum maturity of the kerogen in our samples did not reach the metamorphic stage; thus, we can rule out any influence of the maturity on the δ^{13}C$_{org}$ values of our samples.

The kerogen character for the Upper Turonian–Santonian sequence in the Obira area is compositionally and geochemically identical to the kerogen for the underlying Cenomanian–Middle Turonian sequences (Hasegawa & Saito, 1993; Hasegawa, 2001; Uramoto *et al.* 2007). Therefore, the δ^{13}C values of the kerogen in study samples represent fluctuations in the carbon

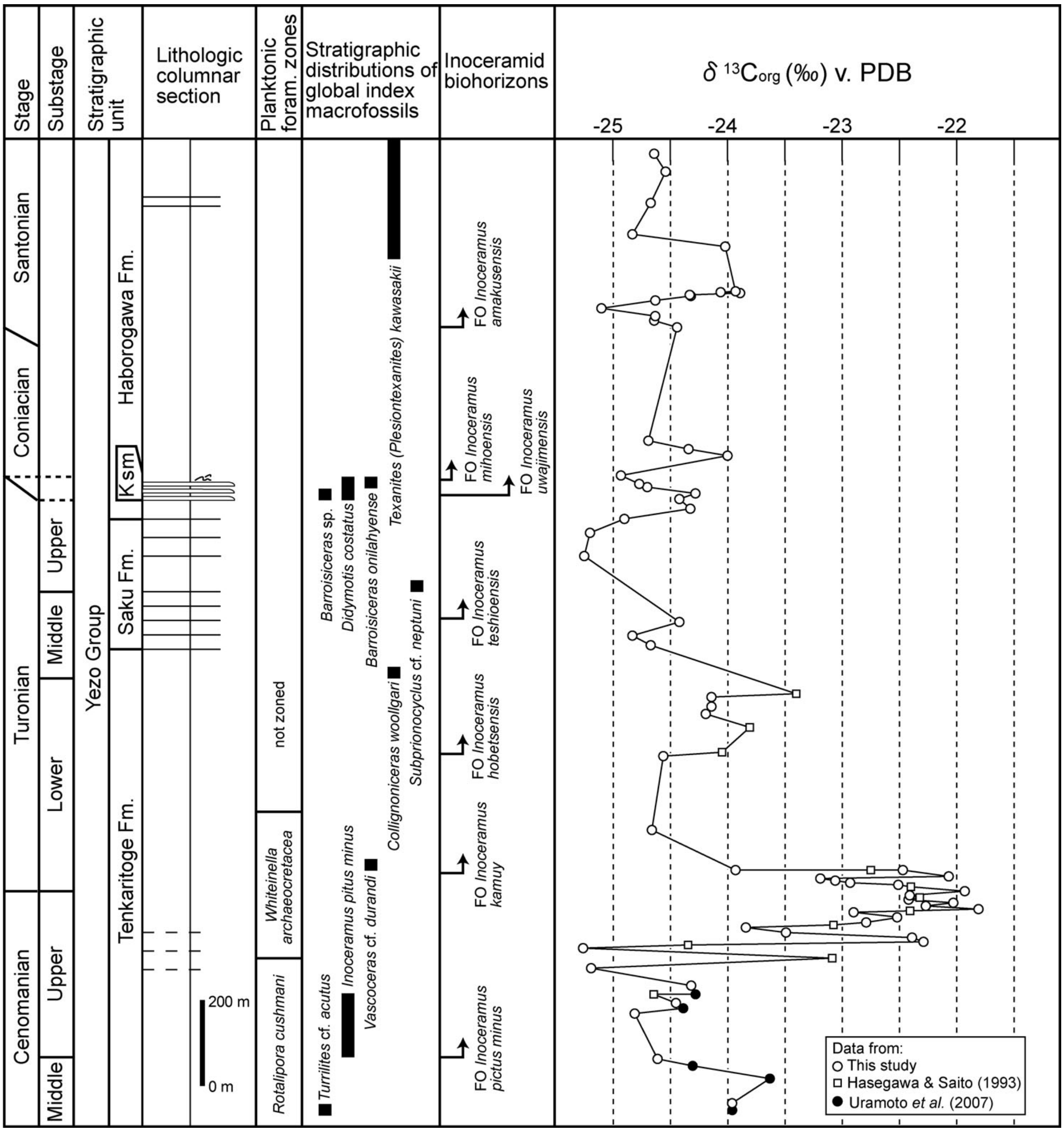

Figure 4. Carbon isotope profile of sedimentary organic matter for the Upper Cretaceous sequence in the Obira area. Macrofossil data are after Tanabe *et al.* (1977), Matsumoto *et al.* (1981), Sekine, Takagi & Hirano (1985), Funaki & Hirano (2004), Oizumi *et al.* (2005), Nishimura, Maeda & Shigeta (2006) and Uramoto *et al.* (2007). Planktonic foraminiferal zones are after Hasegawa & Saito (1993) and Nishi *et al.* (2003). Ksm – Kamikinenbetsu Sandstone Member. For legend see Figure 1.

isotope values of terrestrial plants in the hinterland of the NE Asian region.

6. Discussion

6.a. Correlation of δ^{13}C profiles of terrestrial organic matter within the Yezo Group

We compare significant carbon isotope fluctuations of terrestrial organic matter from the Upper Cretaceous sequence in the Obira area with previously reported δ^{13}C profiles for terrestrial organic matter from Japan and Russia (Hasegawa, 1997, 2003*a*; Hasegawa & Hatsugai, 2000; Hasegawa *et al.* 2003) (Fig. 5). Correlations of δ^{13}C profiles were validated by the biostratigraphic intrabasinal correlation in the global chronological time scale on the basis of worldwide macro- and microfossils, as well as biohorizons of regional marker inoceramids in the NW Pacific.

The correlation suggests that terrestrial organic δ^{13}C profiles show notable fluctuations at specific stratigraphic horizons and all the fluctuations are closely associated with the occurrences of age-diagnostic fossils (Fig. 5). The value and amplitude of each δ^{13}C fluctuation are similar among sections, so the δ^{13}C fluctuation is interpreted to represent the averaged δ^{13}C fluctuation of terrestrial plants in the hinterland of NE Asian region.

The Cenomanian–Turonian boundary δ^{13}C fluctuation is correlated with the sharp positive δ^{13}C excursion (~ +2.5‰) in the Oyubari area (Hasegawa, 1997), because the maximum δ^{13}C value in this

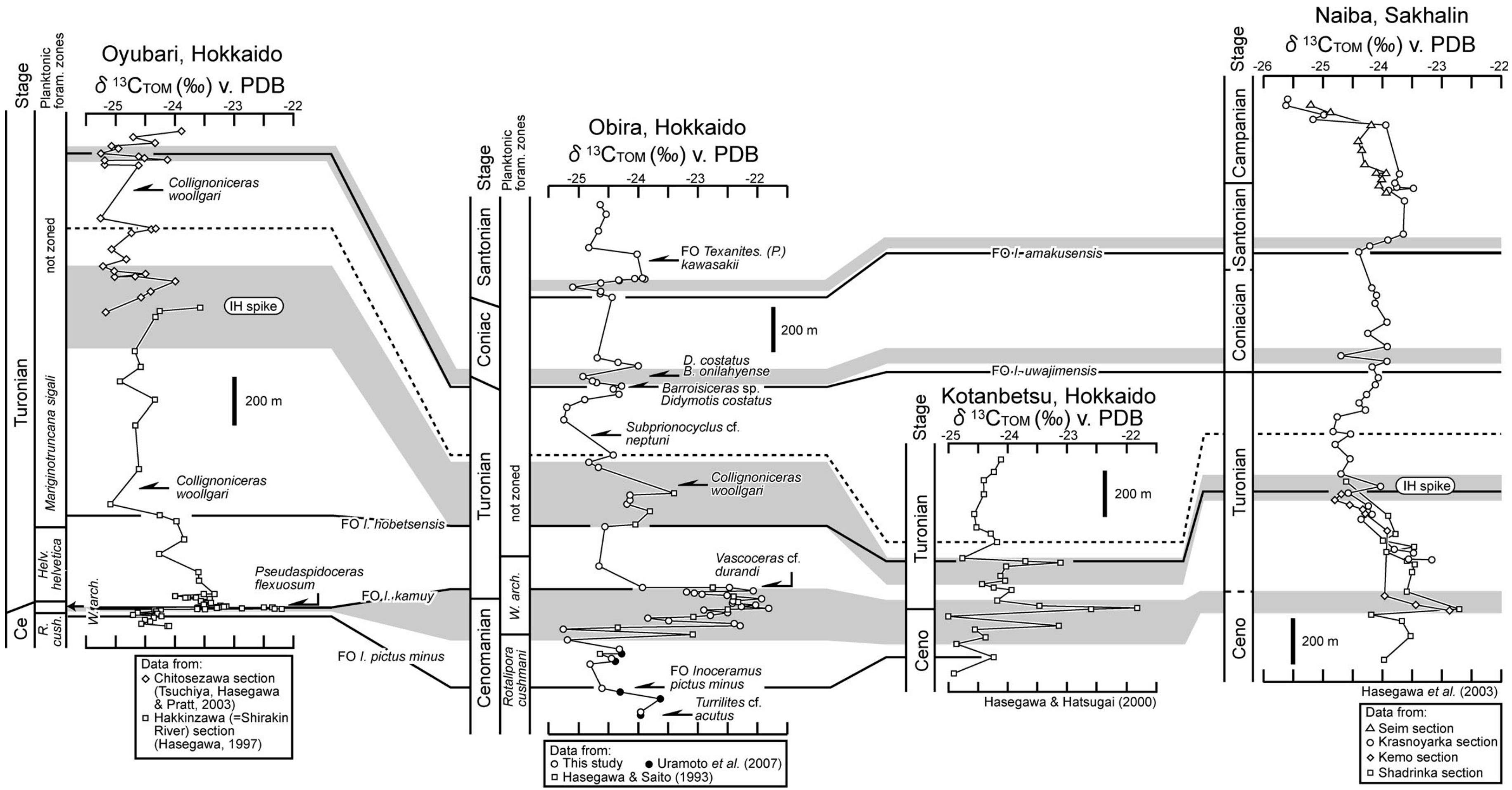

Figure 5. Correlation of the Upper Cretaceous $\delta^{13}C$ curves of terrestrial organic matter ($\delta^{13}C_{TOM}$) of the Yezo Group in Hokkaido, Japan and Sakhalin, Russia (Oyubari area: composite curve of Hasegawa, 1997 and Tsuchiya, Hasegawa & Pratt, 2003, published *in* Hasegawa, 2003*a*; Obira area: this study, Hasegawa & Saito, 1993, and Uramoto *et al*. 2007; Kotanbetsu area: Hasegawa & Hatsugai, 2000; Naiba area: Hasegawa *et al*. 2003). Correlative carbon isotope fluctuations are connected with grey bands and a dashed line. Solid lines are the first occurrence of the regional marker inoceramid of the Yezo Group. The 'IH spike' in the Oyubari and Naiba areas is a Middle Turonian positive $\delta^{13}C$ event reported by Hasegawa (2003*a*). Macrofossil data for the Obira area are after Tanabe *et al*. (1977), Matsumoto *et al*. (1981), Sekine, Takagi & Hirano (1985), Funaki & Hirano (2004), Oizumi *et al*. (2005), Nishimura, Maeda & Shigeta (2006) and Uramoto *et al*. (2007). Planktonic foraminifera zones for the Obira area are after Hasegawa & Saito (1993) and Nishi *et al*. (2003).

positive event is recorded within the planktonic foraminifera *Whiteinella archaeocretacea* Zone. The $\delta^{13}C$ values observed in the Obira area (from −25.3 ‰ to −21.8 ‰) are similar to the $\delta^{13}C$ fluctuations in the Kotanbetsu area (from −25.0 ‰ to −21.8 ‰) (Hasegawa & Hatsugai, 2000), thus suggesting the maximum amplitude of terrestrial organic $\delta^{13}C$ fluctuation during the Cenomanian–Turonian boundary event in the hinterland. The $\delta^{13}C$ fluctuation also corresponds to the positive excursion of ∼ +1.5 ‰ recorded in the lower part of the Naiba area (Hasegawa *et al.* 2003). While previous studies have reported that the Cenomanian–Turonian boundary positive $\delta^{13}C$ fluctuation is characterized by a single or double spike event in the Yezo Group (Hasegawa & Saito, 1993; Hasegawa, 1997; Hasegawa & Hatsugai, 2000; Hasegawa *et al.* 2003), the present study obtained $\delta^{13}C$ data with higher resolution, thereby identifying high-frequency $\delta^{13}C$ fluctuations. The significant short stratigraphic range of $\delta^{13}C$ fluctuation (Hasegawa, 1997) and abrupt change in lithology reported for the Cenomanian–Turnian boundary sequence (Kurihara & Kawabe, 2003; Takashima *et al.* 2004) in the Oyubari area denote the absence of part of the stratigraphic sequence due to hiatus or a winnowing process (Stow, Reading & Collinson, 1996). The different $\delta^{13}C$ patterns in the Kotanbetsu and Naiba areas (Hasegawa & Hatsugai, 2000; Hasegawa *et al.* 2003) are due to the relative scarcity of exposure and the wide sampling interval.

Subsequently, the $\delta^{13}C$ profile of terrestrial organic matter in the Yezo Group shows several positive-and-negative anomalies of ∼ 1 ‰ in the Middle Turonian, upper Turonian–lower Coniacian and Santonian sequences (Fig. 5). The positive $\delta^{13}C$ fluctuation in association with the occurrence of Middle Turonian ammonoid *Collignoniceras woollgari* in the Obira area is correlated with the ‘IH spike’ that is a positive $\delta^{13}C$ event in the Middle Turonian sequence of the Yezo Group documented by Hasegawa (2003*a*) (Fig. 5). Occurrences of *Inoceramus hobetsensis* Nagao and Matsumoto in the Yezo Group support this regional chemostratigraphic correlation. Our correlation of the ‘IH spike’ reinforces the significance of this $\delta^{13}C$ fluctuation for stratigraphic correlation. In addition, biostratigraphic calibration also indicates that another $\delta^{13}C$ positive feature is also present above the ‘IH spike’. The fluctuation is characterized by a small positive peak of ∼ +0.7 ‰ in the Oyubari area (Hasegawa, 2003*a*) and this feature seems to be present in the data from the Obira, Kotanbetsu and Naiba areas (dashed line in Fig. 5). However, this feature is recognized by few data points except for the Oyubari area. Further $\delta^{13}C$ measurements are expected to confirm this correlation.

A positive-and-negative $\delta^{13}C$ fluctuation pattern which is characterized by a negative notch within an overall positive $\delta^{13}C$ fluctuation is recognizable with occurrences of *Didymotis costatus* and *Barroisiceras* spp. in the upper Turonian–lower Coniacian sequence of the Obira area. Correlative counterparts of this fluctuation are present in association with the first occurrence of *Inoceramus uwajimensis* Yehara of the Yezo Group in the Oyubari and Naiba areas (Hasegawa, 2003*a*; Hasegawa *et al.* 2003) (Fig. 5). The positive-and-negative $\delta^{13}C$ pattern is important for correlation. However, because the negative notch is represented by few data, addition of terrestrial $\delta^{13}C$ data is necessary to confirm this correlation.

A Santonian positive $\delta^{13}C$ shift that is closely associated with the first occurrence of *Texanites (Plesiotexanites) kawasakii* in the Obira area is comparable with the positive $\delta^{13}C$ shift just above the first occurrence of *Inoceramus amakusensis* Nagao and Matsumoto in the Naiba area (Hasegawa *et al.* 2003) (Fig. 5). Our correlation of the Santonian terrestrial organic $\delta^{13}C$ fluctuations provides a new chemostratigraphic constraint on the Upper Cretaceous sequence of the Yezo Group.

6.b. $\delta^{13}C$ event correlations in marine carbonates of Europe and terrestrial organic matter of the NW Pacific

The above arguments demonstrate the correlation of terrestrial organic $\delta^{13}C$ events of the Upper Cretaceous sequence at a regional scale. In the following discussion, we demonstrate the global chemostratigraphic correlation of these terrestrial organic $\delta^{13}C$ fluctuations to the carbonate $\delta^{13}C$ record in Europe, based on biostratigraphic data (Figs 6, 7).

6.b.1. *Cenomanian–Turonian boundary $\delta^{13}C$ fluctuation*

The isotopic fluctuations that encompass the Cenomanian–Turonian boundary are the best documented $\delta^{13}C$ event in the Cretaceous sequence (e.g. Pratt & Threlkeld, 1984; Gale *et al.* 1993; Pratt *et al.* 1993; Hasegawa, 1997; Paul *et al.* 1999; Keller *et al.* 2001, 2004; Tsikos *et al.* 2004; Bowman & Bralower, 2005; Jarvis *et al.* 2006; Li *et al.* 2006; Voigt *et al.* 2007). The most detailed carbon isotope records of the Cenomanian–Turonian boundary event have been reported for marine carbonates and marine organic matter from North America and Europe (Paul *et al.* 1999; Keller *et al.* 2004; Tsikos *et al.* 2004; Bowman & Bralower, 2005; Voigt *et al.* 2007). Based on the correlation between North American and European data, Jarvis *et al.* (2006) proposed that the following three positive peaks characterize the $\delta^{13}C$ event of the Cenomanian–Turonian boundary (peak a–c in Jarvis *et al.* (2006); Fig. 7): (1) peak a, the first positive peak; (2) peak b, the positive peak in the middle part of the positive excursion; and (3) peak c, the positive peak at the top of the positive excursion.

Biostratigraphic calibration suggests that the $\delta^{13}C$ patterns of the Cenomanian–Turonian boundary event are highly conformable between marine carbonates and terrestrial organic matter (Fig. 7), so the three peaks are correlated with discrete terrestrial organic $\delta^{13}C$ peaks in Cenomanian–Turonian boundary event of the Obira area, Hokkaido (Fig. 7): (1) the sharp positive $\delta^{13}C$ peak in the uppermost part of the *Rotalipora cushmani*

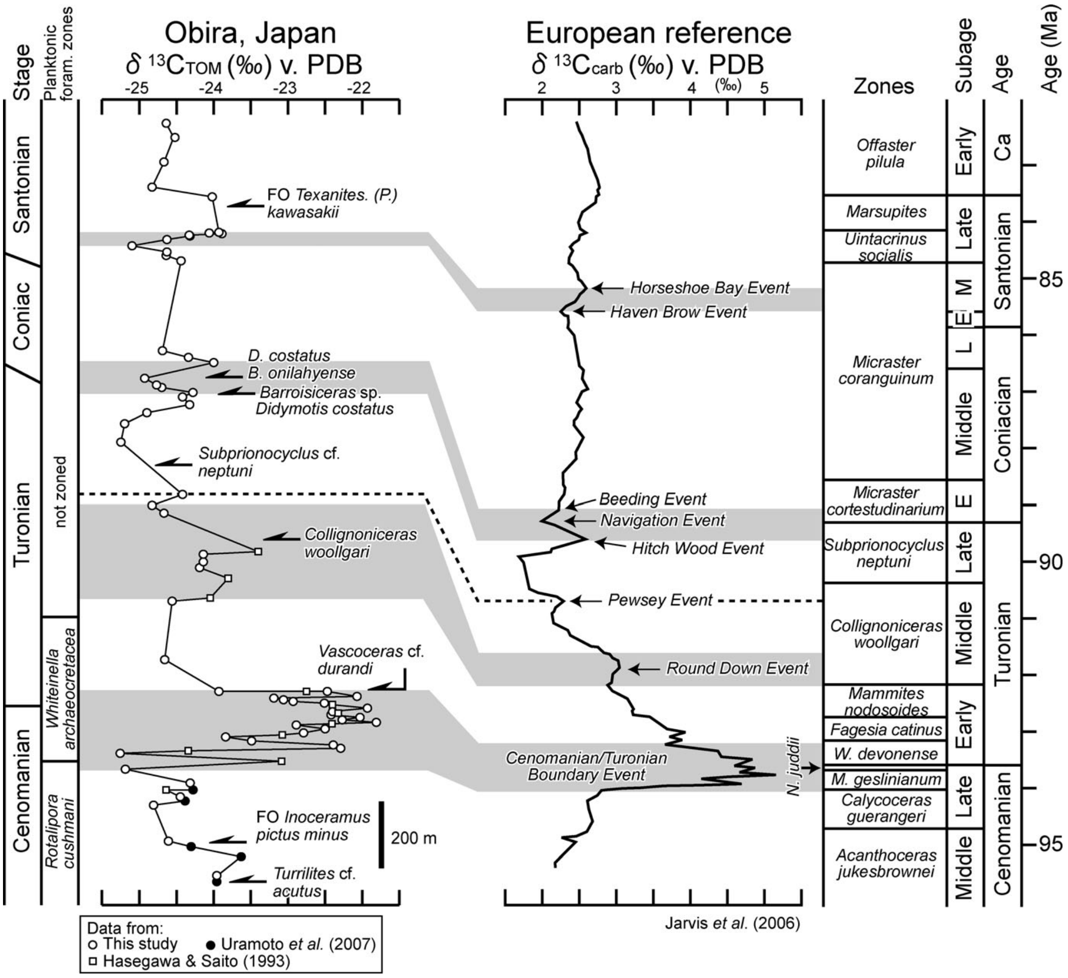

Figure 6. Correlation of the Upper Cretaceous δ^{13}C curves of terrestrial organic matter (δ^{13}C$_{\text{TOM}}$) of the Yezo Group in the Obira area, Hokkaido, Japan (this study; Hasegawa & Saito, 1993; Uramoto *et al.* 2007), and the reference δ^{13}C$_{\text{carbonate}}$ curve in Europe (Jarvis *et al.* 2006). Grey shaded areas and a dashed line mark the correlative carbon isotope fluctuations. Detailed correlation of the Cenomanian–Turonian Boundary carbon isotope event between Europe and Japan is shown in Figure 7. Macrofossil data for the Obira area are after Tanabe *et al.* (1977), Matsumoto *et al.* (1981), Sekine, Takagi & Hirano (1985), Funaki & Hirano (2004), Oizumi *et al.* (2005) and Uramoto *et al.* (2007). Planktonic foraminifera zones for the Obira area are after Hasegawa & Saito (1993) and Nishi *et al.* (2003).

Zone with peak a; (2) the positive δ^{13}C peak in the lower part of the *Whiteinella archaeocretacea* Zone with peak b; and (3) the positive peak that corresponds with the occurrence of Lower Turonian ammonoid *Vascoceras* cf. *durandi* with peak c.

Consistent δ^{13}C patterns allow further correlation of positive-and-negative fluctuations of the Cenomanian–Turonian boundary event: a small positive carbon isotope fluctuation below the first occurrences of Turonian index fossils (peak b' in Fig. 7); minimum carbon isotope value between the each positive peak; and top and bottom of the Cenomanian–Turonian boundary event. Therefore, a total of nine chemostratigraphic horizons are proposed for the key to the high-resolution chemostratigraphy of the Cenomanian–Turonian boundary sequence (Fig. 7).

6.b.2. Middle Turonian δ^{13}C fluctuation

The Middle Turonian δ^{13}C$_{\text{carb}}$ profiles for Europe show a longer-term negative fluctuation. This fluctuation is superimposed by two shorter-term positive isotopic events (Jarvis *et al.* 2006). Previous studies termed the two positive fluctuations as the Round Down Event and the Pewsey Event, in ascending order (Gale, 1996; Jarvis *et al.* 2006).

The Round Down Event and Pewsey Event are tentatively correlated to the 'IH spike' and the above positive fluctuations in the Yezo Group (Fig. 6), based on the occurrences of Middle Turonian ammonoid *Collignoniceras woollgari* and biostratigraphic position of the terrestrial organic δ^{13}C fluctuations. Recently, Hasegawa (2003*a*) correlated the 'IH spike' to the Pewsey Event in Europe. We re-interpret the correlation on the basis of the biostratigraphic calibration, and our new global correlation suggests that the patterns of conspicuous positive δ^{13}C of terrestrial organic matter for the Middle Turonian sequence of the Yezo Group are conformable with the carbonate δ^{13}C curve in Europe. However, this correlation should be confirmed by further δ^{13}C analyses of the Yezo Group, because the correlative counterpart of the Pewsey Event in the Yezo Group is represented by few data points.

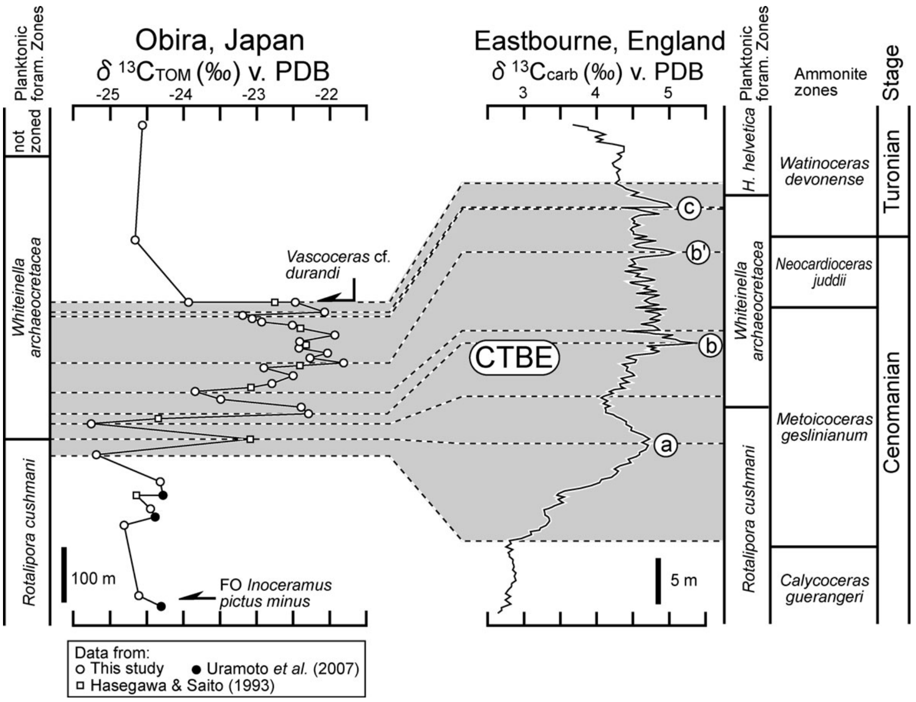

Figure 7. Detailed correlation of the Cenomanian–Turonian boundary δ^{13}C event (CTBE) recorded in marine carbonates ($\delta^{13}C_{carb}$) in Eastbourne, England, and terrestrial organic matter ($\delta^{13}C_{TOM}$) in the Obira area, Hokkaido, Japan. Grey shaded area is the chemostratigraphic range of the CTBE. Correlative δ^{13}C peaks are connected with dashed lines. Positive peaks at a, b and c are defined by global correlation of CTBE between GSSP and England (Jarvis *et al.* 2006). See text for the correlation of positive peak b' and negative carbon isotope fluctuations. Carbon isotope data for Eastbourne are after Paul *et al.* (1999) and biostratigraphic data are after Keller *et al.* (2001) and Gale *et al.* (2005). Carbon isotope data for the Obira area are after this study, Hasegawa & Saito (1993) and Uramoto *et al.* (2007). Macrofossil data for the Obira area are after Sekine, Takagi & Hirano (1985), Funaki & Hirano (2004), and Uramoto *et al.* (2007). Planktonic foraminifera zones for the Obira area after Hasegawa & Saito (1993) and Nishi *et al.* (2003).

6.b.3. Upper Turonian–lower Coniacian δ^{13}C fluctuation

The $\delta^{13}C_{carb}$ fluctuations through the Upper Turonian–Lower Coniacian sequence display a δ^{13}C minimum at the Turonian–Coniacian boundary that is sandwiched by the positive values in the Upper Turonian and Lower Coniacian (Voigt & Hilbrecht, 1997; Voigt, 2000; Jarvis *et al.* 2006). These fluctuations were previously termed the Hitch Wood Event (Upper Turonian), Navigation Event (Turonian–Coniacian boundary) and Beeding Event (Lower Coniacian) (Gale, 1996; Jarvis *et al.* 2006).

The positive-and-negative patterns of $\delta^{13}C_{carb}$ fluctuation are correlated with the upper Turonian–lower Coniacian δ^{13}C fluctuation in the Yezo Group, based on the combined occurrences of *Didymotis costatus* and *Barroisiceras* spp. (Fig. 6); however, this correlation remains tentative, because the negative notch within the overall positive fluctuation in the Yezo Group is represented by few data points. Additional terrestrial organic δ^{13}C analyses are necessary to confirm this correlation.

6.b.4. Santonian δ^{13}C fluctuation

The Santonian $\delta^{13}C_{carb}$ profiles for Europe show an overall increasing trend with several superimposed positive-and-negative isotopic events (Jarvis *et al.* 2006). The most conspicuous positive $\delta^{13}C_{carb}$ shift occurs between the negative $\delta^{13}C_{carb}$ event of the 'Haven Brow Event' and the positive $\delta^{13}C_{carb}$ event of the 'Horseshoe Bay Event'. Association of the first occurrence of ammonoid *Texanites (Plesiotexanites) kawasakii* with the Santonian positive δ^{13}C fluctuation of terrestrial organic matter supports the correlation of the positive shift in the NW Pacific to the European positive shift (Fig. 6), thus underlining the significance of the δ^{13}C fluctuation for the global correlation of the Santonian marine sequences.

6.c. Implications for δ^{13}C fluctuations of terrestrial organic matter

We demonstrate the global correlation of short-term δ^{13}C fluctuation of the Upper Cretaceous sequence between marine and terrestrial records based on the biostratigraphic calibration. The correlation suggests that the patterns of remarkable δ^{13}C fluctuations in marine carbonates are conformable in terrestrial organic δ^{13}C fluctuations. This observation indicates that the terrestrial organic δ^{13}C data mirror the global isotopic patterns in the carbon reservoir of the ocean–atmosphere–terrestrial biosphere system during the global marked δ^{13}C fluctuation period. The factors for fundamental similarity in the δ^{13}C fluctuation between marine and terrestrial records are generally considered to have been induced by the organic carbon burial process changes in association with primary production and sea-level controlled accommodation space changes (e.g. Arthur, Dean & Pratt, 1988; Jarvis *et al.* 2006).

However, it is noteworthy that our correlation also indicates that the amplitude of several terrestrial organic $\delta^{13}C$ fluctuations is larger than that of the coeval carbonate $\delta^{13}C$ fluctuations (Figs 6, 7). Particularly for the Cenomanian–Turonian boundary excursion, the amplitude of the $\delta^{13}C$ event in terrestrial records of the Yezo Group (+3.5 ‰ in the Obira area: this study; +3.2 ‰ in the Kotanbetsu area: Hasegawa & Hatsugai, 2000) is significantly higher than European marine carbonate records (∼ +2.5 ‰: Jarvis *et al.* 2006). This observation suggests that palaeoenvironmental parameters which affect terrestrial organic carbon isotopes on a regional scale may play a role in forming such different fluctuations. Factors such as irradiance, nutrients, age (juvenile v. adult), and seasonal variation have been cited as possible factors in controlling the terrestrial organic $\delta^{13}C$ value (e.g. Arens, Jahren & Amundson, 2000). These factors, however, are homogenized in the case of the Hokkaido sections because terrigenous materials are derived from a sufficiently large hinterland in the NE Asian region (Hasegawa, 2001).

Recently, Hasegawa (2003*b*) inferred that $p\mathrm{CO}_2$ changes, and temperature and humidity-related carbon cycle processes in forests, affected the terrestrial organic $\delta^{13}C$ values of the Yezo Group, based on the discrepancies of $\delta^{13}C$ fluctuation within the middle Cenomanian–early Turonian sequences between Japan and Europe. However, although oxygen isotope and TEX_{86} palaeothermometry have suggested that the climatic optimum during Cretaceous times was in the Late Turonian age (e.g. Bornemann *et al.* 2008), significant $\delta^{13}C$ discrepancies between marine carbonates and terrestrial organic matter are not present in Late Turonian $\delta^{13}C$ records (Fig. 6). Therefore, it is likely that other mechanisms also affected the terrestrial organic $\delta^{13}C$ fluctuations of the Yezo Group.

Additional key factors for terrestrial organic $\delta^{13}C$ fluctuation are the CO_2 emission into the atmosphere on a regional scale by release of terrestrial methane hydrate (Archer, 2007) or biomass burning in the terrestrial realm (Kurtz *et al.* 2003; Finkelstein, Pratt & Brassell, 2006). Because $\delta^{13}C$ values of methane hydrate and terrestrial organic matter (terrestrial vegetation, soil organic matter and peat) are lower than those of atmospheric CO_2, the above CO_2 emission processes generate light $\delta^{13}C$ values of terrestrial organic matter. In the case of the Hokkaido sections, terrestrial organic $\delta^{13}C$ values tend to be negative, especially in Late Cenomanian time (this study; Hasegawa, 2003*b*; Uramoto *et al.* 2007), as opposed to the coeval positive trend of European carbonate $\delta^{13}C$ records (Fig. 6). Such negative and amplified terrestrial organic $\delta^{13}C$ fluctuations of the Yezo Group can be explained by the above CO_2 emission into the atmosphere in the Yezo Group hinterland of the NE Asian region.

7. Conclusions

We obtained a carbon isotope profile of terrestrial organic matter for the Upper Cenomanian–Santonian sequence in the Obira area of Hokkaido, northern Japan. The terrestrial organic $\delta^{13}C$ profile shows remarkable fluctuations for the Cenomanian–Turonian boundary, the Middle Turonian, the Turonian–Coniacian boundary, and the Santonian sequences. Based on the presence of internationally recognizable macro- and microfossils, these short-term terrestrial organic $\delta^{13}C$ events are correlated with the previously reported $\delta^{13}C$ events in Japan and Europe. These correlations reinforce the utility of these isotopic events in terms of global chemostratigraphic correlations. In particular, correlation of the Cenomanian–Turonian boundary event demonstrates that highly conformable $\delta^{13}C$ patterns are recognizable between marine carbonates and terrestrial organic matter. The fundamental similarity of $\delta^{13}C$ fluctuations between marine and terrestrial data indicates that the terrestrial organic $\delta^{13}C$ data from the Cenomanian–Turonian boundary mirror the global isotopic patterns in the carbon reservoir of the ocean–atmosphere–terrestrial biosphere system. In addition, our correlation of short-term $\delta^{13}C$ fluctuations of the Upper Cretaceous sequence between marine and terrestrial records also suggests that the magnitude of several terrestrial organic $\delta^{13}C$ events is greater than that of coeval marine carbonate $\delta^{13}C$ events. Especially for the Cenomanian–Turonian boundary $\delta^{13}C$ excursion, the amplitude of terrestrial organic $\delta^{13}C$ fluctuations of the Yezo Group (+3.5 ‰ in the Obira area: this study; +3.2 ‰ in the Kotanbetsu area: Hasegawa & Hatsugai, 2000) is significantly higher than European carbonate $\delta^{13}C$ fluctuations (+2.5 ‰ in Europe: Jarvis *et al.* 2006). This observation is interpreted to indicate that the regional CO_2 emission into the atmosphere by release of terrestrial methane hydrate or biomass burning of terrestrial organic matter amplified the $\delta^{13}C$ profiles of terrestrial organic matter in the Yezo Group to a magnitude greater than those of the global ocean-atmosphere $\delta^{13}C$ trend.

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