Carbon isotope stratigraphy of terrestrial organic matter for the Turonian (Upper Cretaceous) in northern Japan: Implications for ocean-atmosphere $\delta^{13}C$ trends during the mid-Cretaceous climatic optimum

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ABSTRACT

Carbon isotope data of terrestrial organic matter ($\delta^{13}C_{TOM}$) obtained in Hokkaido, northern Japan, from the marine Cretaceous Yezo Group along the northwestern Pacific margin elucidated a detailed chemostratigraphy for the Turonian Stage in this region of East Asia. Chemostratigraphic intra-basin correlation reveals three positive $\delta^{13}C_{TOM}$ events in the Middle–Upper Turonian of the Yezo Group. $\delta^{13}C_{TOM}$ fluctuations in these events show similar patterns in the Yezo Group, indicating that terrestrial organic matter is mixed sufficiently before deposition in the Yezo Basin. These $\delta^{13}C_{TOM}$ events are correlated with previously documented $\delta^{13}C_{carbonate}$ events in Europe (the Lulworth–Round Down, Glynde–Pewsey, and Late Turonian Events) based on global biostratigraphy. Our chemostratigraphic correlations strengthen the use of these $\delta^{13}C$ events for global correlation of the Turonian marine successions. In addition, global correlation of Turonian marine and terrestrial $\delta^{13}C$ events identifies changes in isotopic difference between $\delta^{13}C_{TOM}$ and $\delta^{13}C_{carbonate}$ ($\Delta_{TOM-carbonate}$), which are interpreted to reflect changes in atmospheric pCO_2 levels, and climate-driven stresses of humidity and soil processes. In earlier stages of Turonian, $\Delta_{TOM-carbonate}$ values are increased. Elevated atmospheric pCO_2, and increased humidity and soil processes in enhanced greenhouse conditions during mid-Turonian, are interpreted to enlarge $\Delta_{TOM-carbonate}$ values. In later stages of Turonian, $\Delta_{TOM-carbonate}$ values are at a constant level, and the lowering of atmospheric pCO_2 or decrease of climate stress related to the diverse paleoclimatic cooling is interpreted to have restored the ocean-atmosphere $\delta^{13}C$ trends.

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INTRODUCTION

The Turonian Stage has been identified as a climatic optimum during the mid-Cretaceous super-greenhouse world with tropical sea-surface temperatures in excess of 35 °C (Wilson et al., 2002; Bice et al., 2003, 2006; Bornemann et al., 2008). In addition to the Turonian climate being characterized by extreme warmth, it was also associated with significant tectonic, geochemical, and biotic perturbations (Jenkyns, 2010), and subsequent rapid climatic cooling indicated by high-frequency sea-level changes and $\delta^{18}O$ fluctuations in marine limestones (Sahagian et al., 1996; Stoll and Schrag, 2000; Voigt et al., 2004; Miller et al., 2005). A clearer picture of Turonian paleoenvironmental conditions is critical for gaining a better understanding of the climatic evolution of the greenhouse Earth. However, available paleoenvironmental studies of this stage have mainly focused on Europe, North America, and the Russian platform. Data from the Pacific region are still limited despite the fact that the paleo-Pacific constituted the largest open ocean in the mid-Cretaceous.

The Cretaceous succession of the Yezo Group that crops out along the northwestern Pacific margin is the sedimentary infill of a forearc basin. The sedimentation rate of the Yezo Group is estimated to be 200–400 m/m.y. (Kaiho and Hasegawa, 1994; Nemoto and Hasegawa, 2011) and is much higher than that of the European Turonian type section. Therefore, detailed paleoenvironmental analysis of the Yezo Group can potentially provide a higher-resolution record of the Turonian paleoenvironmental conditions than studies of European successions. However, the compilation and evaluation of Turonian geological data for the Yezo Group have been limited, partly due to the poorer chronostratigraphic controls on dating of the Yezo Group as compared with the European type section, with the exception of the Cenomanian-Turonian boundary (Hasegawa, 1997; Kurihara and Kawabe, 2003; Moriya et al., 2008; Uramoto et al., 2009; Hasegawa et al., 2010; Nemoto and Hasegawa, 2011; Kurihara et al., 2012). Establishment of higher-resolution chronostratigraphy of the Yezo Group is thus required to clarify the spatial and temporal variations of paleoenvironmental conditions through the Turonian recorded by this group.

Previous studies have documented the stratigraphic $\delta^{13}C$ trends recorded by terrestrial organic matter in the Cretaceous Yezo Group (Hasegawa, 1997; Ando et al., 2002, 2003; Hasegawa et al., 2003; Ando and Kakegawa, 2007; Uramoto et al., 2007, 2009; Moriya et al., 2008; Hasegawa et al., 2010; Nemoto and Hasegawa, 2011). Because of an isotopic linkage between marine and terrestrial carbon reservoirs within Cretaceous ocean-atmosphere-biosphere systems, the time-stratigraphic terrestrial $\delta^{13}C$ fluctuations for Cretaceous time are assumed to reflect $\delta^{13}C$ variations in atmospheric CO_2 (Hasegawa, 1997; Ando et al., 2002; Gröcke et al., 2005; Nunn et al., 2010). Therefore, terrestrial $\delta^{13}C$ records in the Cretaceous Yezo Group (e.g., Hasegawa, 1997; Ando et al., 2002; Uramoto et al., 2009; Nemoto and Hasegawa, 2011) are comparable to reference marine $\delta^{13}C$ records (Jarvis et al., 2006). The carbon

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isotope stratigraphy of terrestrial organic matter might provide a high-resolution correlation tool for the Yezo Group. While Turonian δ^{13}C data have previously been acquired for the Yezo Group (Hasegawa and Saito, 1993; Hasegawa, 1997, 2003a; Hasegawa and Hatsugai, 2000; Hasegawa et al., 2003; Uramoto et al., 2009), the relatively low-resolution sampling employed by previous studies means that available data are still insufficient for global chemostratigraphic correlations and the calibration of δ^{13}C fluctuations in Turonian of the Yezo Group. As such, additional δ^{13}C measurements would facilitate an improved chemostratigraphic framework for the Turonian marine successions of the Yezo Group.

In this study, we present the carbon isotope stratigraphy of sedimentary organic matter with the aim of providing detailed chemostratigraphic constraints on the Turonian Stage of the Yezo Group in Hokkaido, northern Japan. Firstly, we present δ^{13}C data for sedimentary organic matter within the upper Cenomanian–lower Coniacian succession in the Shumarinai and Obira areas of Hokkaido (Fig. 1). The Shumarinai and Obira areas were chosen for this study because previous works reported the Yezo Group was more widely exposed in these areas, with more continuous stratigraphy than in other areas (Figs. 1 and 2). In the Shumarinai area, previous studies have reported the occurrence of age-diagnostic fossils for the upper Cenomanian–lower Coniacian (Nishida et al., 1996, 1998a, 1998b; Sekiya et al., 2009) (Fig. 2). We conducted δ^{13}C measurements for the Shumarinai succession and integrated the chemostratigraphic data with biostratigraphic data. In the Obira area, Hasegawa and Saito (1993) and Uramoto et al. (2009) reported the δ^{13}C data for the Turonian, and biostratigraphic studies have reported the occurrences of age-diagnostic fossils for the upper Cenomanian–lower Coniacian (Tanabe et al., 1977; Matsumoto et al., 1981; Funaki and Hirano, 2004; Kaneko and Hirano, 2005; Oizumi et al., 2005; Nishimura et al., 2006) (Fig. 2). For the Obira section, we conducted additional δ^{13}C measurements, and compiled existing Turonian δ^{13}C data and biostratigraphic data. Subsequently, we compare Turonian δ^{13}C data for the Shumarinai and Obira areas, and other geological traverses across the Yezo Group along northwestern Pacific margin in Japan and Russia. Finally, we correlate the Turonian terrestrial organic δ^{13}C profiles obtained for the Yezo Group with the European reference δ^{13}C profile of marine carbonates (Jarvis et al., 2006) and discuss the ocean-atmosphere δ^{13}C trends in the Turonian Stage based on global chemostratigraphic correlations.

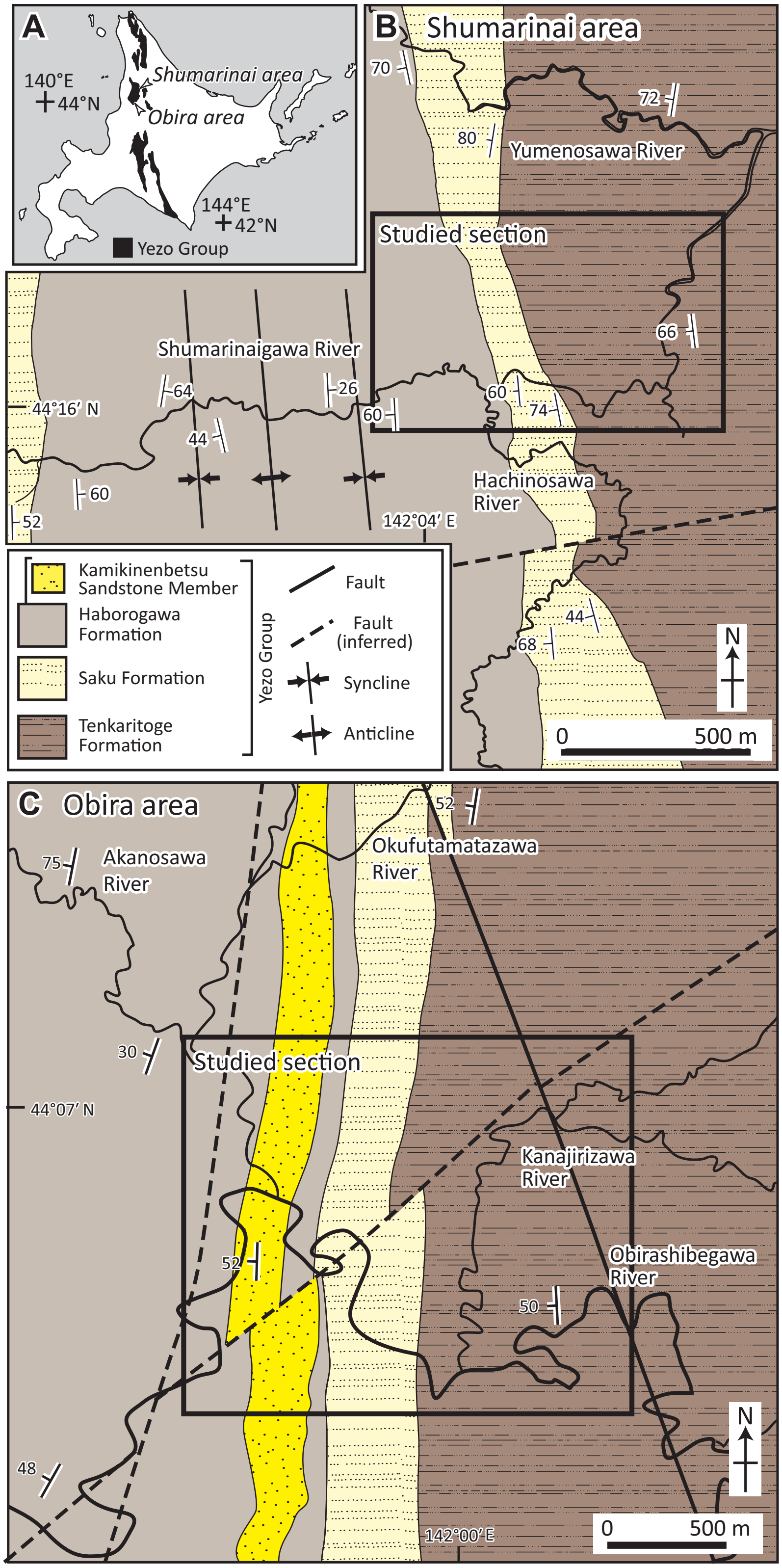


Figure 1. (A) Location map of the studied area and distribution of the Yezo Group in Hokkaido, northern Japan (Takashima et al., 2004). (B) Geological map of the Shumarinai area, Hokkaido, northern Japan. (C) Geological map of the Obira area, Hokkaido, northern Japan (Funaki and Hirano, 2004; Oizumi et al., 2005; Uramoto et al., 2009).

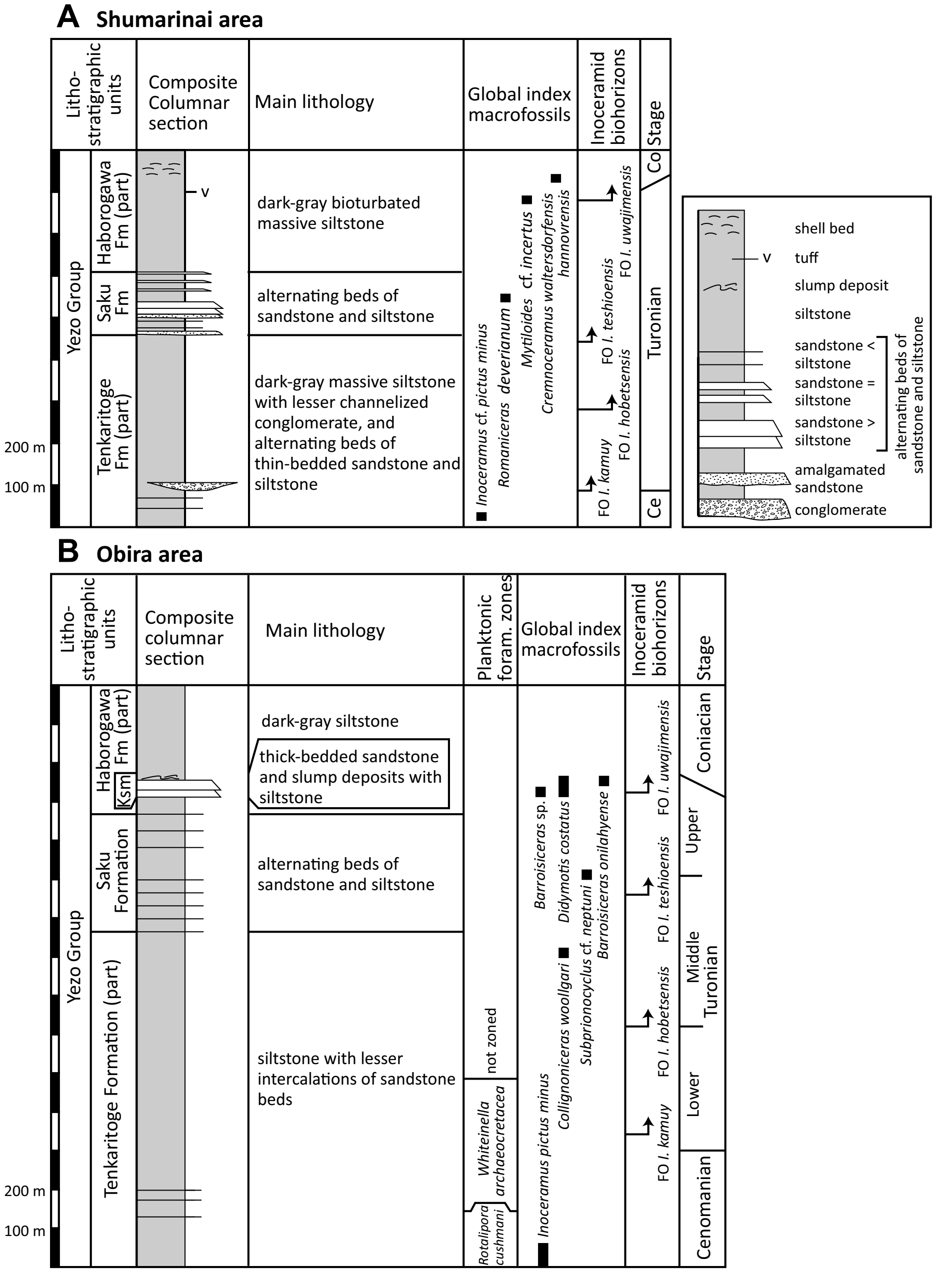


Figure 2. (A) Stratigraphy of the studied sections in the Shumarinai area. Paleontological data are from Nishida et al. (1996, 1998b) and Sekiya et al. (2009). (B) Stratigraphy of the studied sections in the Obira area. Paleontological data are from Tanabe et al. (1977), Matsumoto et al. (1981), Hasegawa and Saito (1993), Nishi et al. (2003), Funaki and Hirano (2004), Kaneko and Hirano (2005), Oizumi et al. (2005), Nishimura et al. (2006), Uramoto et al. (2007), and Kurihara et al. (2012). FO—first occurrence; foram.—foraminifera; KSM—Kamikinenbetsu Sandstone Member.

GEOLOGIC SETTING

The stratigraphy of the Cretaceous Yezo Group has been extensively studied in the Shumarinai and Obira areas, Hokkaido Island, since the pioneering studies by Igi et al. (1958), Tsushima et al. (1958), and Tanaka (1963). The sedimentary succession comprises a series of siliciclastic marine successions. The Yezo Group is widely exposed in the studied sections, with the strata generally striking north-south and younging to the west (Fig. 1). Although some faults are present in the studied sections, the offsets of the Yezo Group along the faults are small, meaning that the complete stratigraphic sequence is preserved (Figs. 1 and 2).

The lithostratigraphy in the studied areas is outlined by Igi et al. (1958), Tsushima et al. (1958), and Tanaka (1963). Takashima et al. (2004) updated and integrated the lithostratigraphic framework of the Yezo Group sections throughout Hokkaido Island. However, as a result of the confined tectonics in the Yezo forearc basin during the Turonian (Tamaki et al., 2009), the Yezo Group shows significant spatial lithological variations, and difficulties remain in reconciling the lithostratigraphy for the Turonian throughout the Yezo Group in Hokkaido Island. Therefore, the lithostratigraphic framework used here is taken from that recently established by Funaki and Hirano (2004) and Oizumi et al. (2005) in the Obira area. The studied succession consists of the following conformable lithostratigraphic units (in ascending stratigraphic order): Tenkaritoge, Saku, and Haborogawa formations (Figs. 1 and 2). The Tenkaritoge Formation is dominated by dark-gray bioturbated siltstone, with lesser intercalations of coarser clastic rocks in the studied areas. The Saku Formation consists of alternating beds of turbiditic sandstone and siltstone. The Haborogawa Formation consists of dark-gray bioturbated siltstone. In the Obira area, thickly bedded sandstone (>1 m) and slump deposits characterize the lower part of the Haborogawa Formation, making it useful as a regional lithostratigraphic marker (Kamikinenbetsu Sandstone Member; Funaki and Hirano, 2004).

The studied sections are of Late Cenomanian to Early Coniacian age, based on occurrences of age-diagnostic ammonoids, bivalves, and planktonic foraminifera. Nishida et al. (1998b) and Funaki and Hirano (2004) reported the occurrence of Late Cenomanian indicator *Inoceramus pictus minus* Matsumoto in the basal part of the studied succession. The Cenomanian-Turonian boundary is recognized in the Tenkaritoge Formation based on integration of planktonic foraminifera and macrofossil biostratigraphy (Shumarinai area: Nishida et al. 1998a, 1998b; Sekiya et al., 2009; Obira area: Hasegawa and Saito, 1993; Nishi et al., 2003; Funaki and Hirano, 2004; Uramoto et al., 2009; Hasegawa et al., 2010; Nemoto and Hasegawa, 2011). Occurrences of global age-diagnostic macrofossils such as *Collignoniceras woollgari* Mantell, *Subprionocyclus* cf. *neptuni* (Geinitz), and *Romaniceras deverianum* (d'Orbigny) characterize the Turonian sequences (Tanabe et al., 1977; Nishida et al., 1998b; Funaki and Hirano, 2004). The Turonian-Coniacian boundary is located within the Haborogawa Formation. The boundary is situated between the stratigraphic levels of the occurrence of inoceramids

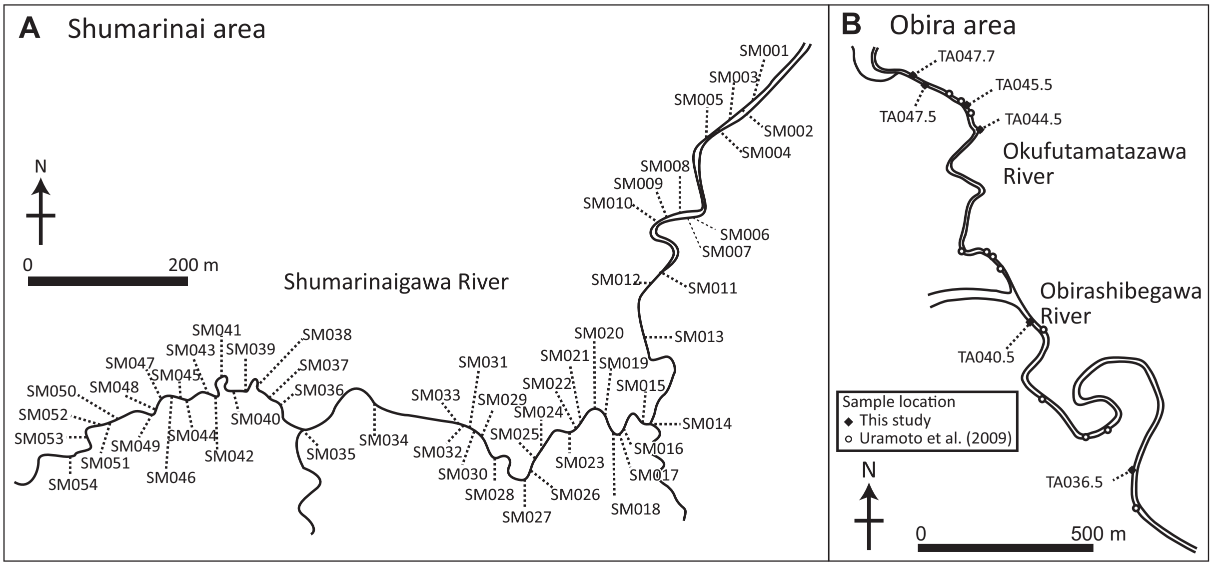


Figure 3. (A) Location sites of mudstone samples collected for analysis in the Shumarinai area. (B) Location sites of mudstone samples collected for analysis in the Obira area.

Mytiloides cf. *incertus* Jimbo and *Cremnoceramus waltersdorfensis hannovrensis* Heinz in the Shumarinai area (Nishida et al., 1998b), and is situated between the stratigraphic levels of the occurrence of *Barroisiceras* spp. and *Didymotis costatus* (Fric) in the Obira area (Tanabe et al., 1977; Matsumoto et al., 1981; Funaki and Hirano, 2004).

MATERIALS AND METHODS

Fifty-four fresh mudstone samples were collected along the Shumarinaigawa River section in the Shumarinai area, and seven samples were collected along the Obirashibegawa River and Okufutamatazawa River sections in the Obira area (Fig. 3). Of these, 10 samples in the Shumarinai area were selected to document the properties of sedimentary organic matter by modal composition analysis of kerogen and Rock-Eval pyrolysis. Previous studies have reported the properties of sedimentary organic matter, including compositional characteristics and thermal diagenesis of mudstone samples from the studied section in the Obira area (Hasegawa and Saito, 1993; Uramoto et al., 2009; Nemoto and Hasegawa, 2011). In order to isolate kerogen, carbonate and silicate minerals were dissolved from coarsely crushed samples using repeated attack with 6 N hydrochloric acid and 40% hydrofluoric acid. The insoluble residue was centrifuged in a $ZnBr_2$ solution (specific gravity of 2.0) to obtain concentrated kerogen. The modal composition of the kerogen was determined by counting 500 points under a transmitted-light microscope, following the visual classification scheme for kerogen proposed by Tyson (1995).

Rock-Eval pyrolysis was performed on powdered bulk samples using a VINCI Technologies model 6 instrument housed at the Japan Petroleum Exploration Company Ltd. (JAPEX) Research Center at Chiba, central Japan. A total of 100 mg of each subsample was pyrolysed from 300 to 650 °C in a nitrogen atmosphere. We measured the temperature (T_{max}) at which the maximum amount of hydrocarbons was released, the ratio of the amount of hydrocarbons released to total organic carbon (TOC) content (hydrogen index), and the ratio of the amount of CO_2 released during heating to 390 °C to TOC content (oxygen index).

For $\delta^{13}C$ analyses of organic matter, powdered bulk subsamples of ~2 g were acidified with a 6 N solution of hydrochloric acid for 24 h to decompose carbonates. The samples were then treated with dichloromethane to eliminate free hydrocarbons. Subsequently, $\delta^{13}C$ ratios were measured using a GV Instruments Isoprime EA mass spectrometer (precision: ±0.10‰) housed at JAPEX Research Center. Carbon isotope ratios ($\delta^{13}C$) are expressed as the per mil (‰) deviation from the Peedee belemnite standard.

RESULTS

Modal Composition of Sedimentary Organic Matter

Visual characterization of the kerogen under transmitted light revealed that terrestrial woody material (opaque, translucent, and biostructured phytoclast) dominates the kerogen modal composition in all of the analyzed samples (Table 1). Spore/pollen grains and dinoflagellate cysts only constitute a minor component of the kerogen modal composition. No amorphous organic matter was detected.

TABLE 1. RESULTS OF MODAL COMPOSITION ANALYSIS OF KEROGEN AND ROCK-EVAL PYROLYSIS OF SELECTED SAMPLES

Sample	Modal composition of kerogen (%)					Rock-Eval pyrolysis		
	Phytoclast			Spore/pollen grains	Dinocysts	T_{max} (°C)	HI	OI
	Opaque	Translucent	Biostructured					
SM01	25.2	70.6	2.2	2.0	–	434	27	66
SM05	15.8	79.4	3.2	1.6	–	432	23	68
SM11	17.6	77.4	3.0	2.0	–	435	34	52
SM16	19.6	73.0	4.2	3.0	0.2	435	28	61
SM22	17.2	76.8	4.4	1.6	–	430	31	83
SM30	14.6	79.6	3.8	2.0	–	431	22	95
SM36	16.6	74.4	4.8	4.0	0.2	427	26	71
SM42	15.6	77.4	3.6	3.4	–	426	25	71
SM45	25.4	67.8	4.2	2.6	–	431	25	87
SM50	26.4	64.6	6.4	2.6	–	434	22	97

Note: HI—hydrogen index (mg HC/g TOC); OI—oxygen index (mg CO_2/g TOC).

Rock-Eval Pyrolysis

The results of Rock-Eval pyrolysis are listed in Table 1. Figure 4 shows a plot of hydrogen index versus T_{max} in the Shumarinai area (this study) and the Obira area (Uramoto et al., 2009). The hydrogen index varies from 22 to 34 mg HC/g TOC and T_{max} data range from 426 to 438 °C in the studied areas. These data correspond to kerogen type III/IV and vitrinite reflectance values for kerogen of less than 0.9 %Ro (Fig. 4).

Stratigraphic Carbon Isotope Fluctuations

The range of carbon isotope ratios of sedimentary organic matter in the studied sections varies from –25.0‰ to –22.2‰ in the Shumarinai area and from –25.2‰ to –21.8‰ in the Obira area (Table 2 and Fig. 5). The $\delta^{13}C$ stratigraphic profiles of sections from the Shumarinai and Obira areas generally have similar patterns. The reproducible $\delta^{13}C$ fluctuations are confirmed by the regional datum levels of age-diagnostic inoceramids in the Yezo Group, including the first occurrence (FO) of *Inoceramus kamuy* Matsumoto and Asai, FO of *I. hobetsensis* Nagao and Matsumoto, FO of *I. teshioensis* Nagao and Matsumoto, and FO of *I. uwajimensis* Yehara. The $\delta^{13}C$ profiles show remarkable variations at specific horizons (Fig. 5). The $\delta^{13}C$ profiles show prominent positive $\delta^{13}C$ values around the Cenomanian-Turonian boundary. Several small fluctuations of +0.5–1.0‰ are present in the overlying Turonian succession.

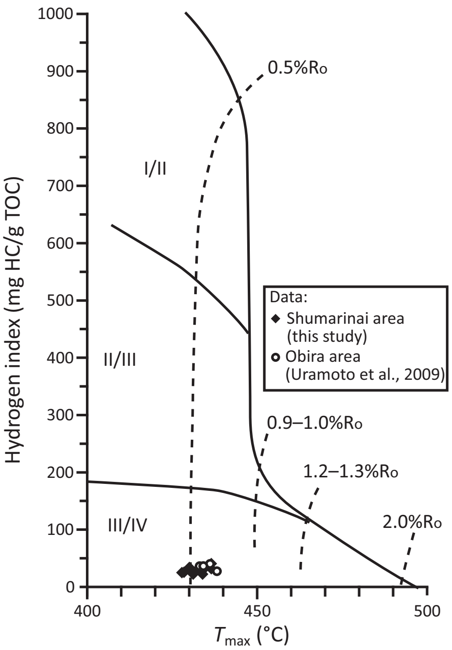


Figure 4. Plot of hydrogen index versus T_{max} for selected mudstone samples from the Shumarinai area (this study) and the Obira area (Uramoto et al., 2009). Dashed lines represent vitrinite reflectance; solid lines represent kerogen zones.

DISCUSSION

Properties of Kerogen

The modal composition of the analyzed kerogen is dominated by terrestrial organic matter (Table 1). A terrigenous origin is also supported by the finding of type III/IV kerogen in the samples (Fig. 4) (Hunt, 1996). The kerogen is therefore derived exclusively from terrestrial organic matter. This result is consistent with previous analyses of bulk sedimentary organic matter in the Yezo Group sections, and Cretaceous marine silicilastic successions in Russia along the northwestern Pacific margin (Hasegawa 1997, 2001; Ando et al., 2003; Hasegawa et al., 2003; Ando and Kakegawa, 2007; Uramoto et al., 2007, 2009; Nemoto and Hasegawa, 2011). As an occasionally higher concentration of marine algal matter has not been visually identified in bulk samples, bulk analysis of organic matter is suitable for studying the $\delta^{13}C$ values of terrestrial organic matter ($\delta^{13}C_{TOM}$) in samples from the sections of the Yezo Group.

Effect of Organic Maturity on $\delta^{13}C$ Values

The analyzed kerogen samples yield vitrinite reflectance values less than 0.9 %Ro (Fig. 4) indicating that the degree of maturity corresponds to the diagenesis to catagenesis stage (Mukhopadhyay, 1994; Hunt, 1996).

It is known that metamorphism can significantly alter the original $\delta^{13}C$ values of organic matter via an isotopic exchange reaction with carbonates (Dunn and Valley, 1992), and that these changes do not occur at temperatures below the metamorphic stage (Teerman and Hwang, 1991; Whiticar, 1996). Our vitrinite reflectance results show that kerogen in the analyzed samples was not subject to metamorphic conditions. Therefore, we can rule out any diagenetic effects on the $\delta^{13}C$ values of kerogen in the sample, and the measured $\delta^{13}C$ ratios represent the original values of terrestrial woody plants in the hinterland of the Yezo Group in East Asia.

Correlation of Turonian $\delta^{13}C_{TOM}$ Profiles within the Yezo Group

We have correlated the $\delta^{13}C_{TOM}$ profiles in the Shumarinai and Obira areas with previously reported $\delta^{13}C_{TOM}$ profiles from the Yezo Group in the Oyubari, Kotanbetsu, and Naiba areas along the northwestern Pacific margin (Hasegawa, 1997; Hasegawa and Hatsugai, 2000; Hasegawa et al., 2003; Tsuchiya et al., 2003) (Fig. 6). The observed $\delta^{13}C_{TOM}$ trends in the Shumarinai and Obira areas are comparable with these published data, except where they differ from the $\delta^{13}C_{TOM}$ patterns in the southern Oyubari area for the stratigraphic level above the FO of *I. hobetsensis* (Hasegawa, 1997) (Fig. 6). In the Oyubari area, the $\delta^{13}C_{TOM}$ profiles for this interval show no marked short-term changes, whereas the FO level of *I. hobetsensis* in other areas shows a +1.0‰ positive $\delta^{13}C_{TOM}$ excursion. This stratigraphic interval is marked by large-scale sedimentary packages of slump deposits in the Oyubari area (Motoyama et al., 1991; Hasegawa, 1997; Kawabe, 2000; Kurihara and Kawabe, 2003) (Fig. 6). Such mass-transport deposits are absent in the lower–middle Turonian sections of the other areas, where the sedimentary successions are dominated by hemipelagic siltstone. The increased occurrence of slump deposits in the Oyubari area is the result of confined subsidence in the Yezo Basin that began in the Turonian (Tamaki et al., 2009). The formation of the confined basin provided sufficient accommodation space that was unique to the Oyubari area. This localized tectonic activity led to tilting of the sea floor, resulting in intense slumping in the Oyubari area.

TABLE 2. WHOLE ROCK CARBON ISOTOPE VALUES (‰) OF STUDIED SAMPLES

Sample	$\delta^{13}C_{wood}$	Sample	$\delta^{13}C_{wood}$
Shumarinai area			
SM001	–23.39	SM028	–24.55
SM002	–23.88	SM029	–24.32
SM003	–22.23	SM030	–24.49
SM004	–22.69	SM031	–24.65
SM005	–22.41	SM032	–24.31
SM006	–23.44	SM033	–24.34
SM007	–24.10	SM034	–25.03
SM008	–24.10	SM035	–24.22
SM009	–24.20	SM036	–24.50
SM010	–24.05	SM037	–24.07
SM011	–24.13	SM038	–24.24
SM012	–24.41	SM039	–24.45
SM013	–24.42	SM040	–24.32
SM014	–24.26	SM041	–24.07
SM015	–24.44	SM042	–23.85
SM016	–24.50	SM043	–24.12
SM017	–24.22	SM044	–23.86
SM018	–24.74	SM045	–23.81
SM019	–24.38	SM046	–23.74
SM020	–24.45	SM047	–23.74
SM021	–24.11	SM048	–23.65
SM022	–24.19	SM049	–23.86
SM023	–24.38	SM050	–24.59
SM024	–23.83	SM051	–24.36
SM025	–23.88	SM052	–24.47
SM026	–24.04	SM053	–24.72
SM027	–24.78	SM054	–24.05
Obira area			
TA036.5	–25.22	TA045.5	–24.40
TA040.5	–24.18	TA047.5	–24.05
TA040.7	–23.80	TA047.7	–24.12
TA044.5	–24.37		

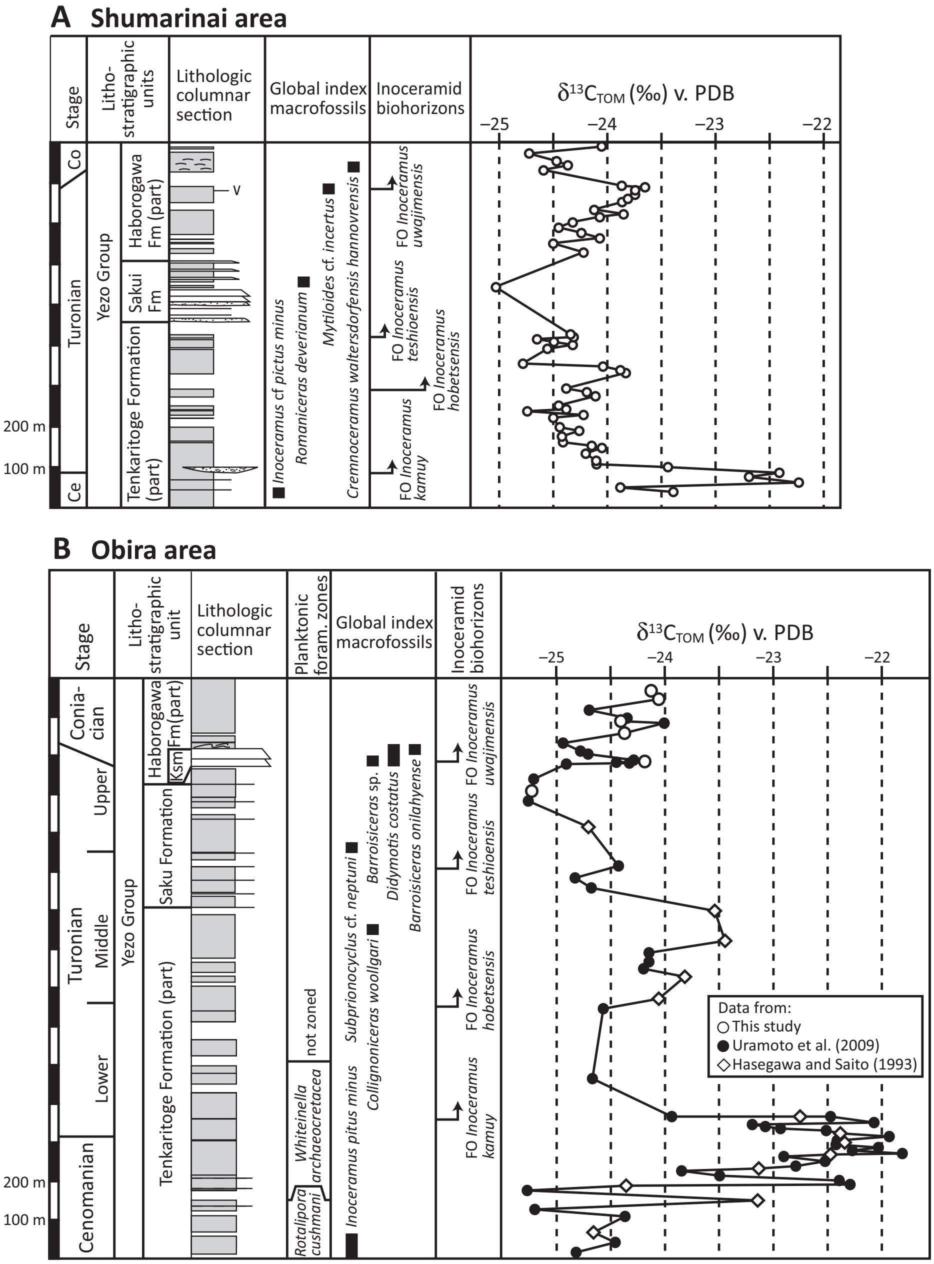


Figure 5. (A) Carbon isotope profile for sedimentary organic matter from the Shumarinai area. Paleontological data are from Nishida et al. (1996, 1998b) and Sekiya et al. (2009). (B) Carbon isotope profile for sedimentary organic matter from the Obira area (Hasegawa and Saito, 1993; Uramoto et al., 2007, 2009; this study). Paleontological data are from Tanabe et al. (1977), Matsumoto et al. (1981), Hasegawa and Saito (1993), Nishi et al. (2003), Funaki and Hirano (2004), Kaneko and Hirano (2005), Oizumi et al. (2005), Nishimura et al. (2006), Uramoto et al. (2007), and Kurihara et al. (2012). FO—first occurrence; foram.—foraminifera; KSM—Kamikinenbetsu Sandstone Member; PDB—Peedee belemnite; TOM—terrestrial organic matter.

The varying pattern of lower–middle Turonian $\delta^{13}C_{TOM}$ fluctuations in the Yezo Group may reflect spatial variations in sediment distribution within the Yezo forearc basin.

With the exception of the slump deposits in the Oyubari area, the $\delta^{13}C_{TOM}$ profiles are fairly similar throughout the Turonian sediments of the Yezo Group (Fig. 6). A notable feature of Turonian $\delta^{13}C_{TOM}$ fluctuations in the Yezo Group is the occurrence of three positive $\delta^{13}C_{TOM}$ anomalies with amplitudes of ~0.5‰–1.0‰ in the middle–upper Turonian (shaded peaks connected with dashed lines in Fig. 6), as revealed by biostratigraphic intra-basin correlations. The three positive fluctuations are labeled YT1, YT2, and YT3 in ascending stratigraphic order (Fig. 6). The maximum $\delta^{13}C_{TOM}$ value (~–23.5‰) of the YT1 positive excursion is recorded above the FO of *I. hobetsensis* (Fig. 6). The presence of middle Turonian *Collignoniceras woollgari* in the Yezo Group (Tsuchiya et al., 2003; Funaki and Hirano, 2004) suggests that YT1 is a Middle Turonian $\delta^{13}C_{TOM}$ event. The YT2 event is found below the FO of *I. teshioensis*. The YT2 event also presents immediately below the Middle/Upper Turonian boundary in the Obira area (Funaki and Hirano, 2004), indicating that YT2 is an upper Middle Turonian $\delta^{13}C_{TOM}$ event (Fig. 6). In the studied areas, however, the YT2 is defined by only a few data points, and more samples within narrower stratigraphic intervals than collected in the present study should be collected and analyzed for more detailed correlation. The YT3 event is characterized by the FO of *I. uwajimensis* (Fig. 6). The YT3 event is also marked by the presence of *Mytiloides incertus* (Nishida et al., 1998b), which occurs worldwide in the upper Upper Turonian successions (Matsumoto and Noda, 1983; Voigt, 1995; Takahashi, 2005). These chemostratigraphic correlations suggest that seven stratigraphic levels (the base, top, and $\delta^{13}C_{TOM}$ maxima of each YT event; shown by dashed lines in Fig. 6) can be used as chemostratigraphic datums in the Turonian Stage of the Yezo Group.

Given that compositional characteristics of bulk sedimentary organic matter in the samples are dominated by terrestrial organic matter (Hasegawa, 1997, 2001; Hasegawa and Hatsugai, 2000; Hasegawa et al., 2003; Uramoto et al. 2009; this study), the regional homogeneity of bulk $\delta^{13}C_{TOM}$ trends in the Yezo Group sections indicates that the terrestrial organic matter was well mixed before its deposition in the Yezo forearc basin. Consequently, we postulate that the bulk $\delta^{13}C_{TOM}$ profiles in the Yezo Group record the average carbon isotopic signature of terrestrial plant material in the hinterland of East Asia during Turonian.

Global Correlation of Turonian $\delta^{13}C$ Profiles

Correlation of Chemostratigraphic Events

Cretaceous marine and terrestrial $\delta^{13}C$ records have been compared in a number of published studies (e.g., Hasegawa, 1997, 2003a; Ando et al., 2002, 2003; Hasegawa et al., 2003; Robinson and Hesselbo, 2004; Gröcke et al., 2005; Ando and Kakegawa, 2007; Uramoto et al., 2007, 2009; Nunn et al., 2010; Nemoto and Hasegawa, 2011). These studies have shown that

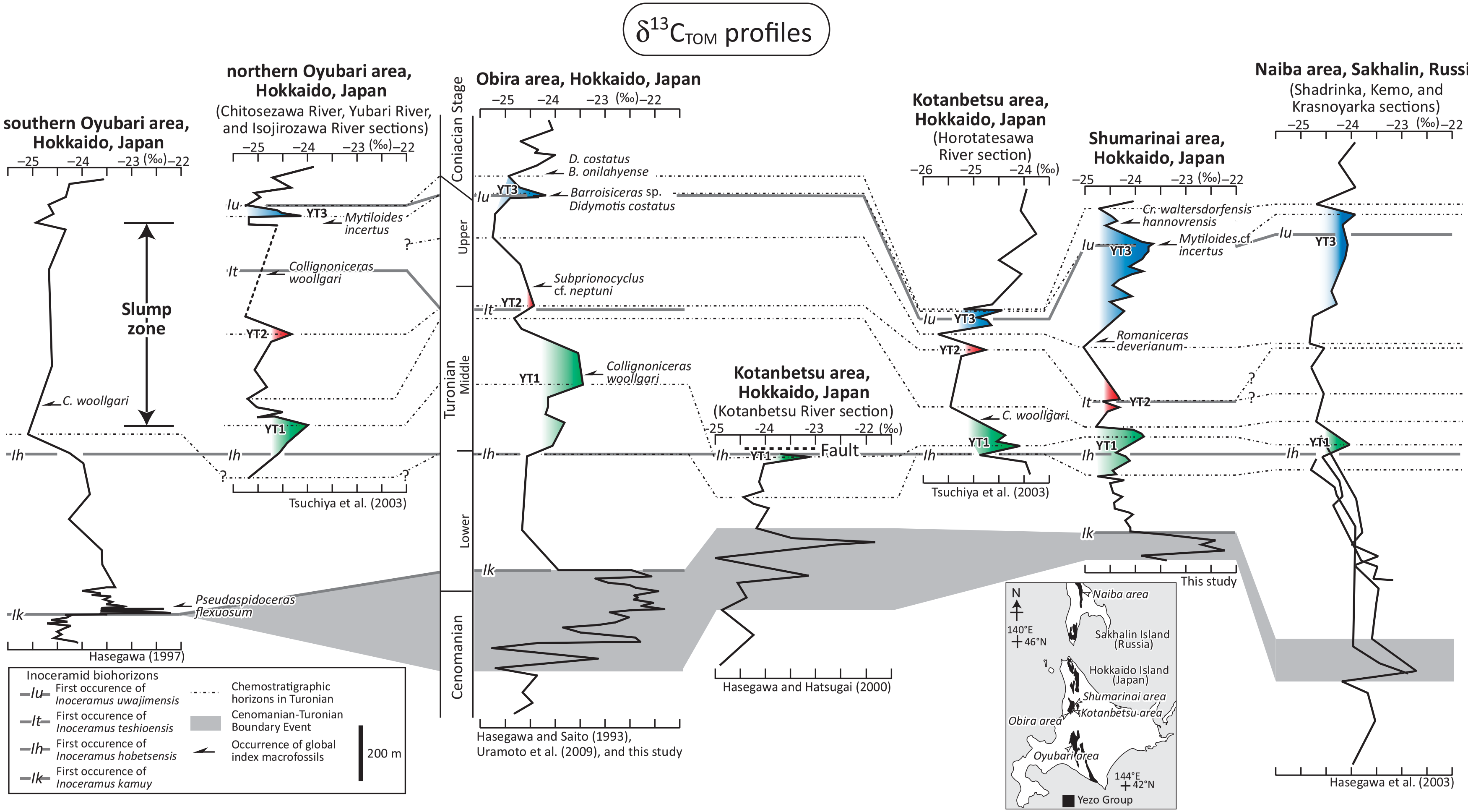


Figure 6. Proposed correlation of terrestrial organic δ^{13}C profiles for the Turonian Stage of the Yezo Group along the northwestern Pacific margin. The inset map shows the locations of the profiles presented in the chemostratigraphic correlation. Dashed lines show the proposed correlation of seven chemostratigraphic horizons. A light gray band highlights the positive δ^{13}C peak of the Cenomanian-Turonian boundary event. Shaded peaks with the YT number notations are short-term positive carbon isotope events in the Turonian Stage of the Yezo Group. Paleontological data were taken from: Shumarinai area (Nishida et al., 1996, 1998b; Sekiya et al., 2009); Obira area (Tanabe et al., 1977; Matsumoto et al., 1981; Funaki and Hirano, 2004; Kaneko and Hirano, 2005; Oizumi et al., 2005; Nishimura et al., 2006; Uramoto et al., 2007; Kurihara et al., 2012); Oyubari area (Matsumoto, 1971; Hirano et al., 1977; Hirano, 1995; Kawabe, 2000; Tsuchiya et al., 2003; Kurihara and Kawabe, 2003); Kotanbetsu area (Hasegawa and Hatsugai, 2000; Tsuchiya et al., 2003); Naiba area (Hasegawa et al., 2003). *Cr*—*Cremnoceramus*; TOM—terrestrial organic matter.

the $\delta^{13}C$ profiles are similar between marine and terrestrial data based on precise biostratigraphic correlation. Recently, Nunn et al. (2010) investigated a geological succession that contains both marine carbonates and terrestrial organic matter, and distinctive $\delta^{13}C$ excursions appear to be synchronous in both the marine and terrestrial data. The synchroneity of the terrestrial and marine $\delta^{13}C$ records provide strong evidence for a $\delta^{13}C$ linkage between marine and terrestrial carbon reservoirs in the ocean-atmosphere system.

The positive $\delta^{13}C_{TOM}$ events within the Turonian Stage in the Yezo Group (YT1–YT3) (Fig. 6) are correlated with the reference $\delta^{13}C_{carbonate}$ trends in Europe (Jarvis et al., 2006). Although the sedimentation rate of the forearc succession of the Yezo Group is much higher (200–400 m/m.y.) (Kaiho and Hasegawa, 1994; Nemoto and Hasegawa, 2011) than that in the European sections, correlations of $\delta^{13}C_{TOM}$ events in Japan with the reference $\delta^{13}C_{carbonate}$ trends in Europe are possible based on a global biostratigraphic calibration (Fig. 7). We correlate the Middle Turonian YT1 event in the Yezo Group with the lower Middle Turonian interval from the Lulworth Event to the Round Down Event (Jarvis et al., 2006), based on the presence of the Middle Turonian ammonoid *C. woollgari* (Fig. 7). The highest-resolution Turonian $\delta^{13}C_{carbonate}$ stratigraphy documented in Europe showed that this stratigraphic interval includes more high-frequency carbon isotope events including Tu6–Tu10 (Voigt et al., 2007). As such, even higher-resolution sampling and carbon isotope study of the Yezo Group would be expected to further refine the detailed global chemostratigraphic correlation of the lower Middle Turonian $\delta^{13}C$ events between Japan and Europe.

Positive $\delta^{13}C_{TOM}$ excursions of the YT2 event in the upper middle Turonian in the Yezo Group are correlated with the Glynde to

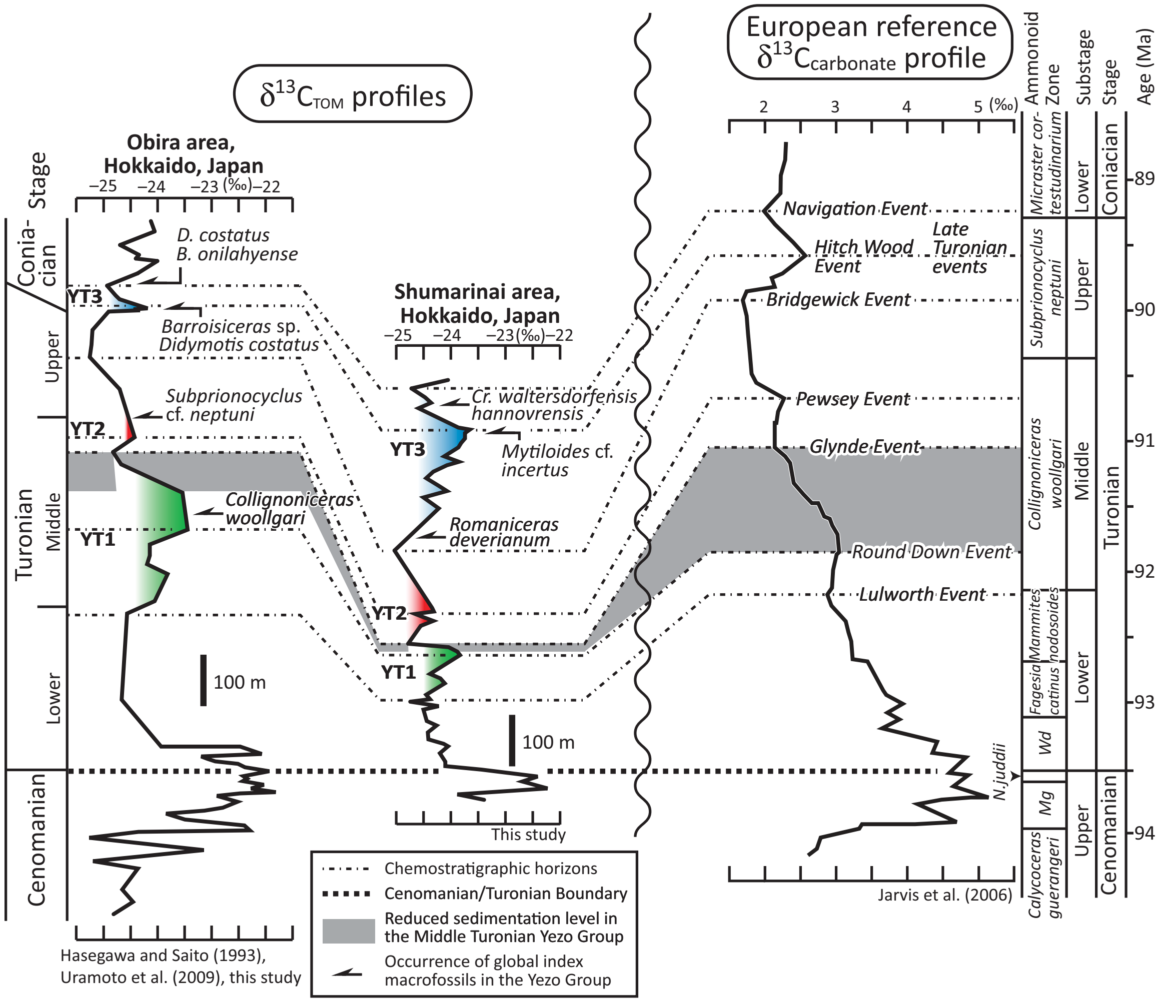


Figure 7. Proposed correlation between the Turonian reference $\delta^{13}C_{carbonate}$ profile in Europe (Jarvis et al., 2006) and $\delta^{13}C_{TOM}$ profiles in northern Japan. Dashed lines show the proposed correlation of seven chemostratigraphic horizons. A light gray band highlights the gradual trend in the middle Turonian to lower $\delta^{13}C_{carbonate}$ that is not observed in the $\delta^{13}C_{TOM}$ profiles in East Asia. Shaded peaks with YT number notations are short-term positive carbon isotope events in the Turonian Stage of the Yezo Group. Paleontological data were taken from: Shumarinai area (Nishida et al., 1996, 1998b; Sekiya et al., 2009); Obira area (Tanabe et al., 1977; Matsumoto et al., 1981; Funaki and Hirano, 2004; Kaneko and Hirano, 2005; Oizumi et al., 2005; Nishimura et al., 2006; Uramoto et al., 2007; Kurihara et al., 2012). *Cr—Cremnoceramus*; *Mg—Metoicoceras geslinianum*; *N.—Neocardioceras*; *Wd—Watinoceras devonense*; TOM—terrestrial organic matter.

Pewsey events (Gale, 1996; Jarvis et al., 2006) in Europe, as the YT2 event in the Yezo Group occurs immediately below the Middle/Upper Turonian boundary (Funaki and Hirano, 2004) (Fig. 7). The high-resolution $\delta^{13}C_{carbonate}$ stratigraphy documented in Europe showed that the Middle/Upper Turonian boundary stratigraphic interval includes more high-frequency carbon isotope events including peaks 1–4 (Jarvis et al., 2006). For confirming the more detailed chemostratigraphic correlation between $\delta^{13}C_{carbonate}$ and $\delta^{13}C_{TOM}$ records, more samples within narrower stratigraphic intervals than in the present $\delta^{13}C_{TOM}$ data should be collected and analyzed in the Yezo Group.

The YT3 event in the Yezo Group can be correlated with the positive $\delta^{13}C_{carbonate}$ excursion from the Bridgewick Event to Navigation Event (late Turonian events) (Gale, 1996; Jarvis et al., 2006) in Europe (Fig. 7). This correlation is supported by the presence of the inoceramid *M. incertus* in the Yezo Group (Nishida et al., 1998b), which occurs worldwide in the uppermost part of the Late Turonian successions (Matsumoto and Noda, 1983; Voigt, 1995; Takahashi, 2005). Global chemostratigraphic correlation of $\delta^{13}C$ trends of the Turonian successions demonstrates that the $\delta^{13}C_{carbonate}$ events are mirrored by $\delta^{13}C_{TOM}$ events. Therefore, our observations strengthen the potential use of these prominent positive $\delta^{13}C_{TOM}$ excursions of the Turonian Stage as a global correlation tool for marine successions based on biochronology.

Correlation of Long-Term $\delta^{13}C$ Trends

The global chemostratigraphic correlations identified in the above discussion suggest that the marked $\delta^{13}C$ variations occurred near simultaneously. However, the long-term decreasing trend in $\delta^{13}C$ that characterizes the stratigraphic interval between the Round Down Event and the Glynde Event in the Middle Turonian $\delta^{13}C_{carbonate}$ profile in Europe (shown by a gray shading in Fig. 7) is absent in the $\delta^{13}C_{TOM}$ profiles from northern Japan (Fig. 7). The presented chemostratigraphic correlations suggest that the Yezo Group does not record a gradual decrease in $\delta^{13}C_{TOM}$ values during the Middle Turonian, and that the $\delta^{13}C_{TOM}$ fluctuations of the upper Middle Turonian Glynde–Pewsey events directly follow the $\delta^{13}C_{TOM}$ fluctuations of the Lulworth–Round Down events (Fig. 7). According to Jarvis et al. (2006), the stratigraphic interval of the gradual decrease in Middle Turonian $\delta^{13}C_{carbonate}$ values is characterized by a significant time interval of more than 0.5 m.y. In general, sedimentation rates of the offshore successions of the Yezo Group are estimated to have been 200–400 m/m.y. (Kaiho and Hasegawa, 1994; Nemoto and Hasegawa, 2011). The absence of a gradual decrease in $\delta^{13}C_{TOM}$ profiles during the Middle Turonian in the Yezo Group might reflect a reduced sedimentation due to prolonged sedimentary hiatus or significant winnowing of sediments at this time on the northwestern Pacific margin. Lithologically, the offshore successions of the Yezo Group during this time interval do not show any significant changes and are mainly characterized by siltstone with lesser sandstone. It is noteworthy that this time interval coincides with an extensive eustatic sea-level fall during the Turonian (Haq et al., 1987). In this respect, paleo-depth indicators such as benthic foraminiferal assemblages in the Yezo Group might be expected to provide better constraints on sedimentary patterns in the northwestern Pacific margin.

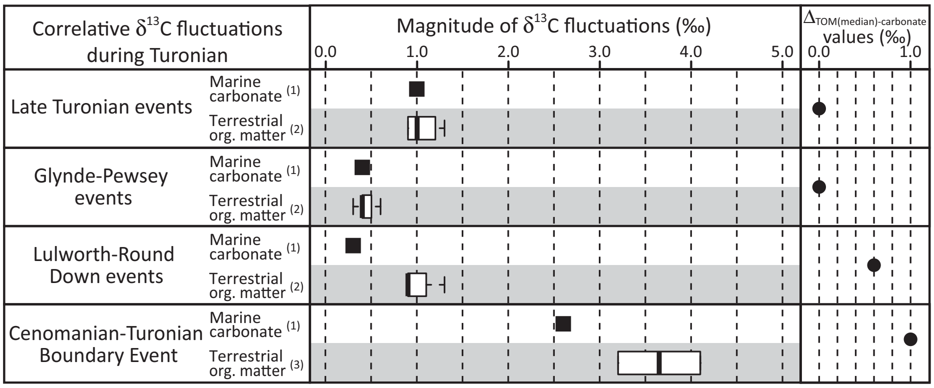
Implications for Mid-Cretaceous Ocean-Atmosphere $\delta^{13}C_{TOM}$ Trend

The global chemostratigraphic correlation suggests that the significant $\delta^{13}C$ excursions occurred simultaneously in the global ocean-atmosphere system (Fig. 7). This indicates that the bulk $\delta^{13}C_{TOM}$ data record isotopic changes in the carbon reservoir of the ocean-atmosphere system during the Turonian. The factors responsible for the similarity in the $\delta^{13}C$ fluctuation between the marine and terrestrial records are generally considered to have been controlled by changes in the organic carbon burial process, in association with changes in primary production and sea level–controlled accommodation space changes (e.g., Arthur et al., 1988; Jarvis et al., 2006), because the organic carbon burial rate directly relates to the atmospheric CO_2 level. Published global correlations of mid-Cretaceous $\delta^{13}C_{TOM}$ and $\delta^{13}C_{carbonate}$ profiles have almost confirmed this, because an isotopic difference between $\delta^{13}C_{TOM}$ and $\delta^{13}C_{carbonate}$ ($\Delta_{TOM-carbonate}$) appears to be constant (Ando et al., 2002, 2003; Ando and Kakegawa, 2007; Uramoto et al., 2007).

Hasegawa (2003b) and Uramoto et al. (2009) noted an increase in $\Delta_{TOM-carbonate}$ values in $\delta^{13}C$ profiles around the Cenomanian-Turonian boundary event (CTBE). This increase is characterized by a significant negative excursion of $\delta^{13}C_{TOM}$ profiles in late Cenomanian from East Asia (Hasegawa, 1997; Hasegawa and Hatsugai, 2000; Uramoto et al., 2007). More recently, Nemoto and Hasegawa (2011) found a negative $\delta^{13}C_{TOM}$ excursion during the CTBE, which has been recognized in $\delta^{13}C_{org}$ data (e.g., Sageman et al., 2006; Gale et al., 2005; Kolonic et al., 2005; Voigt et al., 2006; Kuroda et al., 2007; Nemoto and Hasegawa, 2011) but not in any carbonate sections. This observation suggests that paleoenvironmental parameters that affect $\delta^{13}C_{TOM}$ values on a regional scale may play a role in causing such different fluctuations. Factors such as irradiance, nutrients, organism age (juvenile versus adult), and seasonal variation have been cited as possible factors in controlling the $\delta^{13}C_{TOM}$ value (e.g., Arens et al., 2000; Nunn et al., 2010). These factors, however, are homogenized in the case of the Yezo Group sections because terrigenous materials are derived from a sufficiently large hinterland in the northeast Asian region (Hasegawa, 2003b). Nemoto and Hasegawa (2011) interpreted the $\delta^{13}C_{TOM}$ fluctuations as reflecting enhanced greenhouse conditions and an associated increase in humidity, which were related to volcanically induced elevation of atmospheric pCO_2 at the Cenomanian-Turonian boundary (Kuroda et al., 2007). Also, terrestrial $\delta^{13}C$ decreases were possible in response to climate stress changes in extensive greenhouse conditions that are characterized by increased humidity and related soil processes (Bowen et al., 2004). One or a couple of these factors could ultimately cause lowering of $\delta^{13}C_{plant}$ values by recycling of soil-plant respired CO_2 in the Yezo Group hinterland of East Asia.

Our chemostratigraphic correlations also suggest that increased $\Delta_{TOM-carbonate}$ values appear to occur during mid-Turonian (Fig. 8). The magnitude of the $\delta^{13}C_{TOM}$ excursion of the early Middle Turonian Lulworth–Round Down events is 0.9‰–1.3‰, whereas the reference $\delta^{13}C_{carbonate}$ data for these events (Jarvis et al., 2006) show ~0.3‰ (Fig. 8). It is noteworthy that the timing of the Cretaceous thermal optimum period is of mid-Turonian age (Bornemann et al., 2008) and coincides with the timing of this increase in $\Delta_{TOM-carbonate}$ values in Turonian (Fig. 8). In such paleoenvironmental conditions, the increased $\Delta_{TOM-carbonate}$ values might be explained by the hypotheses of Bowen et al. (2004) and Nemoto and Hasegawa (2011).

One discrepancy of these $\Delta_{TOM-carbonate}$ values is that the $\Delta_{TOM-carbonate}$ values of the CTBE are higher than the $\Delta_{TOM-carbonate}$ values in the Lulworth–Round Down events (Fig. 8). As the timing of the Cretaceous thermal optimum is known to be of mid-Turonian age (Huber et al., 2002; Wilson et al., 2002; Voigt et al., 2004; Bornemann et al., 2008), the higher $\Delta_{TOM-carbonate}$ values of the CTBE are interpreted to reflect not only elevated pCO_2 levels or climatic stress changes, but also other short-term $\delta^{13}C_{TOM}$ disturbances in the Yezo Group hinterland of East Asia. A negative excursion of $\delta^{13}C_{TOM}$ profiles in late Cenomanian characterizes the increased $\Delta_{TOM-carbonate}$ values of the CTBE in the Yezo Group (Hasegawa, 1997; Hasegawa and Hatsugai, 2000; Uramoto et al., 2007). Such negative excursion of bulk $\delta^{13}C_{TOM}$ values can be related to the release of light $\delta^{13}C$ from terrestrial sources (Gröcke et al., 2009; Uramoto



(1) Reference $\delta^{13}C_{carbonate}$ data after Jarvis et al. (2006)
(2) $\delta^{13}C_{TOM}$ data after Hasegawa and Saito (1993), Hasegawa (1997), Hasegawa and Hatsugai (2000), Hasegawa et al. (2003), Tsuchiya et al. (2003), Uramoto et al. (2009), and this study
(3) $\delta^{13}C_{TOM}$ data after Hasegawa and Hatsugai (2000), Moriya et al. (2008), Uramoto et al. (2009), Hasegawa et al. (2010), and Nemoto and Hasegawa (2011)

Figure 8. Plots showing comparison of the magnitude of $\delta^{13}C$ fluctuations in the correlative Turonian $\delta^{13}C$ events between marine carbonates and terrestrial organic (org.) matter, and changes in an isotopic difference between $\delta^{13}C_{TOM}$ and $\delta^{13}C_{carbonate}$ ($\Delta_{TOM-carbonate}$). Plots for marine carbonate data are from reference $\delta^{13}C_{carbonate}$ data (Jarvis et al., 2006). Box and whisker plots for $\delta^{13}C_{TOM}$ data are from the Cretaceous forearc successions along the northwestern Pacific margin. Boundaries of boxes represent the first and third quartiles, and the line within the box represents the second quartile (median); whiskers represent the allowable range of the data (1.5 times the interquartile range). TOM—terrestrial organic matter.

et al., 2009): (1) terrestrial methane hydrate (Archer, 2007), (2) terrestrial biomass burning (Kurtz et al., 2003; Finkelstein et al., 2006), and/or (3) increased weathering of organic-rich sediments and soils (Higgins and Schrag, 2006). A combination of several or all of the above mechanisms in the local carbon cycle of the hinterland of the Yezo Group could be related to the additional factors for negative excursion of bulk $\delta^{13}C_{TOM}$ values in the CTBE.

$\Delta_{TOM-carbonate}$ values decreased significantly in latter stages of the Turonian (Fig. 8); the magnitudes of $\delta^{13}C_{TOM}$ excursions in the late middle Turonian Glynde–Pewsey events and the Late Turonian events are 0.3‰–0.7‰ and 0.9‰–1.3‰, respectively (Fig. 8). The reference $\delta^{13}C_{carbonate}$ data for these events (Jarvis et al., 2006) are ~0.4‰ and ~1.0‰, respectively (Fig. 8). $\Delta_{TOM-carbonate}$ values in latter stages of the Turonian indicate that $\delta^{13}C_{TOM}$ fluctuations in East Asia were consistent with the global $\delta^{13}C$ fluctuations. The factors responsible for the significant decrease in $\Delta_{TOM-carbonate}$ values during late Turonian may be related to lowering of atmospheric pCO_2 and decrease of climate stress in terrestrial ecosystems. Such terrestrial environmental changes may be attributed to the rapid paleoclimatic cooling that occurred in Late Turonian (Voigt and Wiese, 2000; Voigt et al., 2004), which terminated the enlarged $\Delta_{TOM-carbonate}$ values and restored the link of $\delta^{13}C_{TOM}$ values in East Asia to global $\delta^{13}C$ trends. While terrestrial paleoclimate data through this period are still limited in scope, more detailed $\delta^{13}C_{TOM}$ analyses and organic petrographic analyses of the Yezo Group will ultimately lead to a more comprehensive understanding of mid-Cretaceous ocean-atmosphere $\delta^{13}C$ trends.

CONCLUSIONS

We have presented the carbon isotope stratigraphy of sedimentary organic matter for the Turonian (Upper Cretaceous) strata of the Yezo Group in Hokkaido, northern Japan. Characterization of the sedimentary organic matter reveals that the kerogen exclusively derived from terrestrial organic matter and that the degree of organic maturity is immature in the Yezo Group. Therefore, the $\delta^{13}C_{org}$ data represent the values of terrestrial organic material and are largely unaffected by thermal diagenesis.

The Turonian $\delta^{13}C_{TOM}$ profiles in the Yezo Group sections exhibit generally similar patterns. This indicates that the terrestrial plant materials were well mixed after transportation from their original source, which homogenized the $\delta^{13}C_{TOM}$ values. The $\delta^{13}C$ fluctuations presented in this study are interpreted to document the average $\delta^{13}C$ variations of terrestrial higher plants in the eastern Asian hinterland along the northwestern Pacific during Turonian. Three distinct positive excursions of $\delta^{13}C_{TOM}$ (defined here as YT1–YT3 events) were identified. These events can be correlated with the reference $\delta^{13}C_{carbonate}$ records from Europe using biostratigraphic calibration. The chemostratigraphic correlations suggest that three positive chemostratigraphic $\delta^{13}C_{carbonate}$ events are also present in the $\delta^{13}C_{TOM}$ profiles obtained for northern Japan (the Lulworth–Round Down events of the lower Middle Turonian, the Glynde–Pewsey events of the upper Middle Turonian, and the Bridgewick–Navigation events of the Late Turonian). Our chemostratigraphic correlations validate the potential of these $\delta^{13}C$ events to be used for global correlation of Turonian marine sedimentary successions.

Finally, global correlation of the marine and terrestrial $\delta^{13}C$ records suggests that the increased isotopic difference between $\delta^{13}C_{TOM}$ and $\delta^{13}C_{carbonate}$ occurred in early Middle Turonian. This is interpreted to reflect high atmospheric pCO_2 resulting in intensified hydrologic cycles, and increased humidity and soil processes in terrestrial ecosystems in the mid-Turonian enhanced greenhouse conditions (e.g., Bornemann et al., 2008). In the latter stages of the Turonian, $\Delta_{TOM-carbonate}$ values are constant, and the paleoclimatic lowering of atmospheric pCO_2 or decrease of climate stress, and related cooling in late Turonian (e.g., Voigt and Wiese, 2000), were interpreted to have restored global ocean-atmosphere $\delta^{13}C$ trends.

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