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High-resolution sequence stratigraphy in an incised-valley system on the basis of sedimentary organic matter, sulfur content and fossil diatom: An example from Miocene to Pliocene Tatsunokuchi Formation, Iwate Prefecture, Northeast Japan

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This paper discusses relationship among depositional environments, origin of the sedimentary organic matter and diatom fossil assemblage, combined with sequence stratigraphy in an incised-valley system. Interpretation of the environments and their changes was based on sedimentary facies, total sulfur (TS) content, and diatom assemblages. The quantity and composition of organic matter were estimated for the environmental interpretation on the basis of the total organic carbon (TOC) content, reflected-light fluorescent microscopy, and stable carbon isotope ratios of the organic matter ($\delta^{13}C_{org}$). The Miocene to Pliocene Ishibane, Tatsunokuchi and Motohata formations in northeast Japan consist of estuarine and fluvial deposits that were formed during a rise and fall of sea level. The proportions of vitrinite and cutinite that are coarse-grained and terrestrial in origin are relatively high in fluvial deposits of the lowstand systems tract. The proportion of marine alginite, TOC content, and $\delta^{13}C_{org}$ values increase upward in estuary deposits of the transgressive systems tract. This implies the increase in the influence of the sea and the estuarine bottom conditions becoming anoxic. The proportions of vitrinite and cutinite increase upward in the highstand systems tract, whereas the $\delta^{13}C_{org}$ values and TOC content decrease upward. These reflect the strong influence of river discharge. Variations of diatom fossil assemblage of fresh water, brackish water, intertidal, coastal marine and open marine species indicate the same repetitions of transgression and regression in the Tatsunokuchi Formation. Therefore, the transgressive systems tract in the Tatsunokuchi Formation includes four cycles of transgression and regression, while highstand systems tract shows one cycle of transgression and regression.

Key words: depositional environment, diatom assemblage, incised valley, Pliocene, organic matter, sea level, stable carbon isotope, Tatsunokuchi Formation

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Introduction

Variation of organic matter and assemblage of diatom fossils in incised valley-fill sediments are strongly influenced by river discharge and inflow of seawater related to relative sea level changes. Sediments in incised-valley contain rich organic matter, including both land and sea origins. Organic matter that is insoluble in organic solvent is called kerogen or particulate organic matter. The microscopic analysis of kerogen has been widely used for assessing the petroleum source potential in sedimentary basins (e.g., Staplin, 1969; Hunt, 1979; Tissot and Welte, 1984; Senftle et al., 1987) and for interpreting depositional environments (e.g., Tyson, 1987, 1993, 1995; Batten 1996).

Since the 1990s, many papers have discussed the relationship between stacking patterns of depositional systems related to sea-level changes and organic-matter type and preservation in the systems. These studies were mainly conducted on siliciclastics (e.g., Hart et al., 1994; Tyson, 1996; Wood and Gorin, 1998; Omura and Hoyanagi, 2004; Pellaton and Gorin, 2005) and carbonates (e. g., Gorin and Steffen, 1991; Steffen and Gorin, 1993; Bombardiere and Gorin, 1998) deposited in shelf to deep-sea environments. On the other hand, several studies about organic-matter type and preservation have been conducted on estuary to delta deposits (e.g., Hart 1994; Pittet and Gorin, 1997; Carvalho et al., 2006; Omura et al., 2000, 2006; Yoshida et al., 2006, 2007, 2009). However, the relationship between organic-matter preservation in estuary to fluvial deposits and sea-level changes has rarely been studied. In particular, estuarine environments have important factors (e. g., river discharge, inflow of seawater, and redox conditions of the bottom environment) that affect the transportation and preservation of organic matter.

This study addresses the relationship among environmental changes, sequence stratigraphy, and organic-matter preservation of an incised-valley system in a wave-dominated and micro-tidal setting, through a study of the Pliocene to Miocene Tatsunokuchi Formation in Iwate Prefecture, northeast Japan. Environmental change and sequence stratigraphy were explained on the basis of detailed sedimentary facies analysis, measurement of grain size and total sulfur (TS) content. In addition, assemblages of diatom fossils were used mainly to interpret the estuarine conditions. The quantity and composition of organic matter were estimated on the basis of the total organic carbon (TOC) content, reflected-light fluorescent microscopy, and stable carbon isotope ratios of the organic matter ($\delta^{13}C_{org}$).

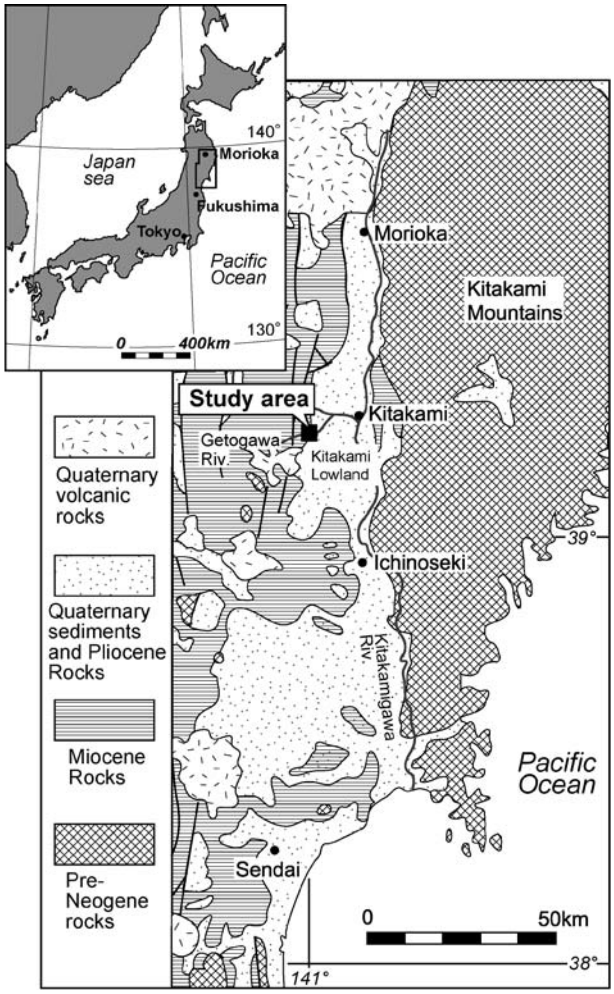

Fig. 1 Location map of the study area and geologic setting around the study area (modified from Oishi et al., 1996).

Geologic and stratigraphic framework

The Tatsunokuchi Formation is distributed from the Fukushima to Iwate prefectures, which are situated on the fore-arc side of the Japanese archipelago (Fig. 1). The coastal area facing the Pacific Ocean is a wave-dominated and microtidal region. The study area is in the Semi-Onsen section that is along the Geto River bluff on western margin of the Kitakami Lowland (Fig. 1). The Tatsunokuchi Formation was deposited in the incised valley (Yagishita, 1998) that was formed along the Kitakami Lowland. The well-exposed section is suitable for high resolution analysis.

Miocene and Pliocene deposits in the study area are divided into the Ishibane, Tatsunokuchi, and Motohata formations, in ascending order (Oishi et al., 1996, 1998) (Figs. 2 and 3). The Tatsunokuchi Formation conformably overlies the Ishibane Formation and is unconformably overlain by the Motohata Formation (Oishi et al., 1996, 1998). The Ishibane Formation unconformably overlies basement rocks of the late Miocene Hishinai Formation and Maetsukami-yama acidic volcanic rock (Oishi et al., 1996, 1998) (Fig. 3). The Ishibane and Motohata formations consist of nonmarine deposits that are composed of conglomerate, sandstone, mudstone, and lignite, whereas the Tatsunokuchi Formation is composd of marine deposits that are composed of bluish gray mudstone with thin sandstone beds (Oishi et al., 1996).

The fission track (FT) age of the Sotomasuzawa tuff in the upper part of the Ishibane Formation is 5.6±0.5 Ma (Oishi and Yoshida, 1998), whereas the age of the Semi-Onsen tuff in the middle part of the Motohata Formation is 3.9±0.4 Ma (Oishi et al., 1996) (Figs. 2 and 3). The Tatsunokuchi Formation is also correlated with the uppermost Miocene to lowermost Pliocene (*Thalassiosira oestrupii* Subzone) on the basis of diatom biostratigraphy (Yanagisawa, 1998).

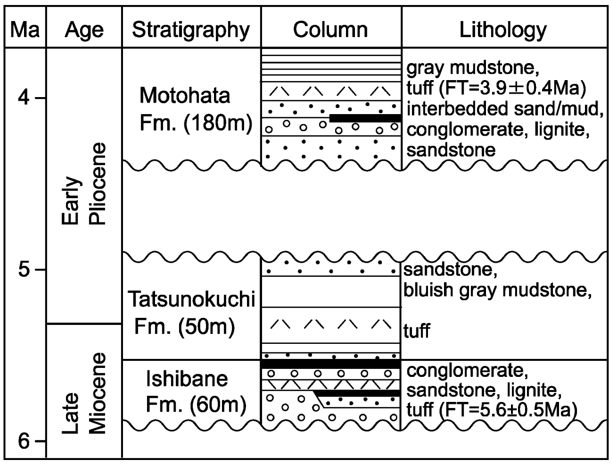

Fig. 2 Stratigraphy and age of the Ishibane, Tatsunokuchi and Motohata formations (modified from Oishi et al., 1998). Fm: Formation, FT: Fission-track age. Lithology of the column refer to legend in Fig. 3.

Material and methods

Samples of clay, silt, and sandy silt in the Ishibane, Tatsunokuchi, and Motohata formations were obtained for grain size, TOC, TS, fluorescent visual kerogen, and stable carbon isotope analyses at about 0.5 to 2.0 m intervals. Samples for diatom analysis were obtained from the Tatsunokuchi Formation.

Samples for grain-size analysis were added to a 10% H_2O_2 solution to eliminate organic matter, and grain size was then measured by a Coulter LS230 (Beckman Coulter Co., Ltd.) laser-diffraction grain-size analyzer, with size range of 0.4 μm to 2 mm.

Samples for TOC and TS analyses were dried at 70℃ for 24-hour and pulverized to about 200 mesh in an agate vessel, and the dried samples were divided in two. One sample was added to 6N HCl to eliminate carbonates. The TOC content was measured by the combustion method at 950℃ in a CHN Yanaco MT-5 Analyzer. Antipyrine (C, 70.19%; N, 14.88 %) was used for the standard, and the analytical precision

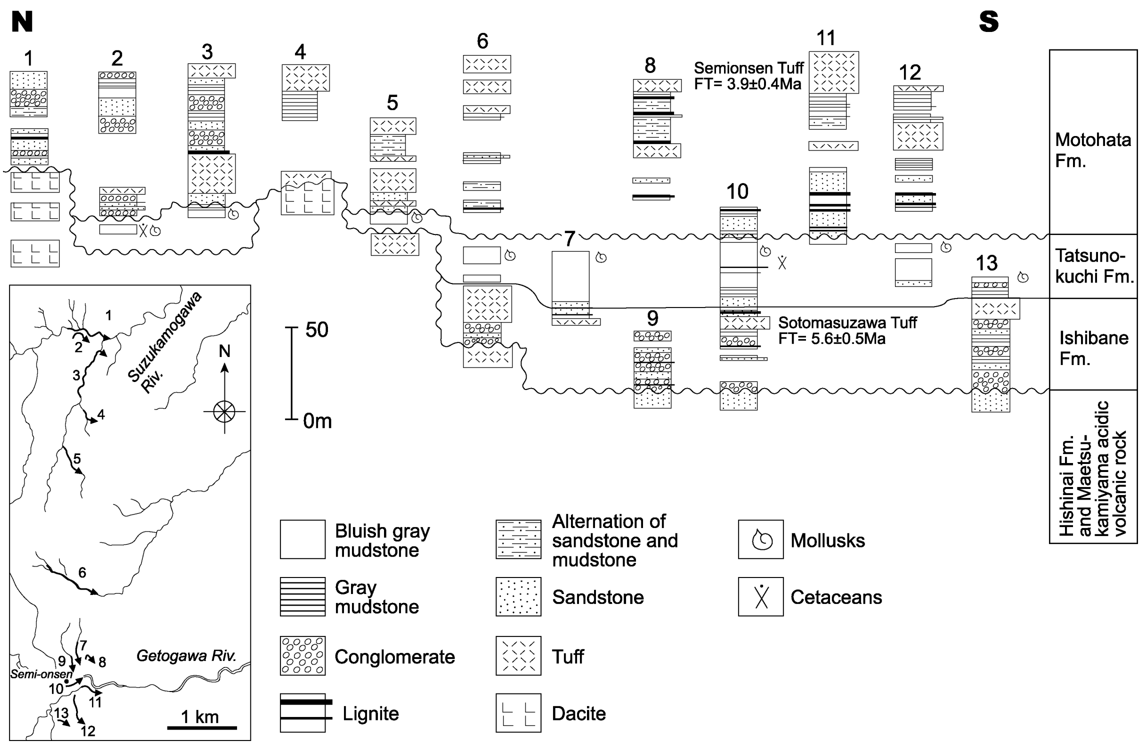

Fig. 3 Geological cross section of the Miocene to Pliocene deposits in western margin of the Kitakami Lowland area (modified from Oishi et al., 1998).

Table 1 Classification of sedimentary organic matter using fluorescent visual kerogen analysis (modified from Omura and Hoyanagi, 2004)

Group	Constituent	Characteristics		Origin
		transmitted light	fluorescent light	
woody-coaly organic matter	vitrinite	translucent, brown semiopaque square- and blade-shaped	no fluorescence	higher plant debris (woody phytoclasts)
	fusinite	black opaque, square- and blade-shaped	no fluorescence	higher plant debris (woody phytoclasts)
herbaceous organic matter with pollen and spores	sporinite	translucent, brown, pollen and spore-shaped	strong to weak fluorescence	pollen and spores
	cutinite	translucent, brown, plant tissue structures	moderate fluorescence yellow to light gray	cuticle and leaves
	resinite		strong orange fluorescence	resin
amorphous organic matter	FA (fluorescent amorphous organic matter)	structureless	strong fluorescence yellow to white	resin, sporinite and cutinite
	NFA (non-fluorescent amorphous organic matter)	structureless	no fluorescence	vitrinite and other terrigenous organic matter
	WFA (weakly fluorescent amorphous organic matter)	structureless	weak fluorescence	marine plankton
aquatic organic microfossils	alginite	translucent, *Pediastrum*-shaped	strong fluorescence yellow green	freshwater algae
		translucent, dinoflagellates- and acritarchs-shaped	strong to weak fluorescence orange to white	marine algae and marine plankton

was approximately 0.17%. The TS content was measured from the other sample by the combustion method at 1, 450℃ in an EMIA-120 (Horiba Co., Ltd.) sulfur analyzer. Coal (S, 2.37%) was used for the standard, and the analytical precision was approximately 0.02%.

Muddy deposits of approximately 1 g for dry samples were used for diatom analysis. The sample was placed in a 200 ml beaker and 30−50 ml of 15% hydrogen peroxide (H_2O_2) was added and then boiled for 10 to 20 minutes until all effervescence stopped. 20 to 30 ml of 10% hydrochloric acid solution was then added to the residue. The sample was boiled for about 20 minutes and then flooded with distilled water. The acid was removed by successive 3- to 5-hour decantations (at least 4 times) with distilled water. Clay minerals were removed by successive 3- to 5-hour decantations (at least 5 times) with 0.01 N sodium pyrophosphate ($Na_2P_2O_7$). Sodium pyrophosphate was then removed by successive 3- to 5-hour decantations (at least 4 times) with distilled water. Strewn slides were prepared on 18×18 mm cover glasses and mounted in Pleurax on glass slides. One slide was prepared for each sample. Two hundred diatom valves were counted for each sample at 1000x magnification. We referred mainly to Krammer and Lange-Bartalot (1986, 1988, 1991a, b) for identification of fresh and brackish water diatoms and to Barron (1985) and Akiba (1986) for marine extinct diatoms.

Organic matter seen under a microscope is generally called kerogen or visual kerogen, the many classifications of which were summarized by Tyson (1995). The present study follows the classification of kerogen and the procedures of Omura and Hoyanagi (2004) for the preparation and observation of organic matter (Table 1). Kerogen is classified into vitrinite (derived from woody phytoclasts), cutinite (cuticle phytoclasts), sporinite (pollen or spores), and alginite (algae or marine plankton). AOM (amorphous organic matter) is subdivided into NFA (non-fluorescent amorphous kerogen), FA (fluorescent amorphous kerogen), and WFA (weakly fluorescent amorphous kerogen) on the basis of their fluorescence (Sawada and Akiyama, 1994; Omura and Hoyanagi 2004). The origins of NFA and WFA in Pleistocene to Miocene shelf and deep-sea deposits of Niigata and Akita prefectures, which are on the back-arc side of the Japanese archipelago are estimated to be terrestrial higher plants and marine plankton, respectively (Sawada and Akiyama, 1994; Omura and Hoyanagi, 2004). While, Omura et al. (2006) analyzed stable carbon isotope of Pleistocene shelf sediments in a fore-arc setting and pointed out that NFA in this area is derived from both terrigenous and marine plankton. The composition of organic matter in each sample was determined by counting 300 points at intervals of 100 μm under transmitted and reflected fluorescent-light microscopy (working magnification 400x, 330 to 385 nm excitation filter, 420 nm absorption filter).

Our stable carbon isotope analytical procedures are as follows: (1) After the procedures mentioned above, a portion of the separated kerogen was treated with a saturated ammonium carbonate solution in order to remove any fluoride, (2) $NaBH_4$ was used to eliminate sulfide, and (3) a few milligrams of the dried sample were heated to 950℃ in an elemental analyzer (FlashEA 1122, ThermoQuest, Ltd.), and the resulting purified CO_2 gas was fed directly into a mass spectrometer (Delta Plus, ThermoQuest, Ltd.) using pure helium carrier gas. Our results are expressed as per mil (‰) relative to the V-PDB standard. We measured a working standard (Atropine; $\delta^{13}C = -23.2$‰) once every six samples. The analytical precision was 0.17‰ in carbon for C.

Results

1. Facies description and depositional environments

The late Miocene to early Pliocene deposits in the study area are classified into nine depositional facies (A to I) on the basis of lithology, grain size, sedimentary structure, basal contacts, and fossils. Depositional facies are shown in Figure 4, and photographs of characteristic depositional facies are shown in Figure 5.

Facies A: Braided River Channel

Description: Facies A (3 m thick) consists of poorly sorted, clast-supported, and pebble-size conglomerate. Conglomerate clasts (max. 8 cm) are rounded to subrounded. The matrix consists of coarse and very coarse-grained sandstone. This facies exhibits normal grading and has channelized bases. Bioturbation and trace fossils are absent. Facies A erosionally overlies Facies C.

Depositional environment: Facies A is interpreted as fluvial channel deposits because it exhibits channelized bases, conglomerate facies, and lack of evidence for biological activity. Furthermore, this facies is characterized by the aggradation of thin channel-fill deposits and lack of mudstone beds. Braided river is characterized by abandoned channels and its rapid migration limits the accumulation of finer material (Walker and Cant, 1984). Therefore, Facies A is interpreted as fluvial channel deposits of a braided river system.

Facies B: Meandering River Channel

Description: Facies B (0.6 to 6.6 m thick) consists of poorly sorted and fine to very coarse-grained sandstone and matrix-supported pebble-size conglomerate. It exhibits an upward-fining facies succession from conglomerate to cross-bedded sandstone. Conglomerate clasts (max. 6 cm) are subrounded to subangular. Conglomerate beds exhibit channelized bases and normal grading. Sandstone beds have normal grading and trough-stratification and include silt rip-up clasts (max. 17 cm). Furthermore, small-scale trough-typed cross-laminae are present within large cross-stratification (epsilon cross-stratification). Wood fragments are scattered throughout Facies B, and bioturbation and trace fossils are absent. Facies B erosionally overlies Facies H and Facies E.

Depositional environment: Facies B is interpreted as fluvial channel deposits because it exhibits a unidirectional flow structure, a channelized base, and a lack of evidence for biological activity. This facies also exhibits fining-upward and is overlain by muddy deposits of Facies C. Meandering river deposits are characterized by an upward-fining facies succession, the dominance of muddy deposits, and the presence of epsilon cross-stratifications formed by the lateral accretion of a point bar (Walker and Cant, 1984). Therefore, Facies B is considered to be fluvial channel deposits of a meandering river system.

Facies C: Flood Plain

Description: Facies C (1 to 5 m thick) consists of claystone, siltstone, silty, and very fine-grained sandstone and lignite. The mudstone is light- to dark-gray, and it contains obscure parallel laminae and water-escape structures. Claystone is present near lignite beds. Sandstone beds contain intercalated lenticular gravel beds with channelized bases and ripple cross-stratification. Many root traces and wood fragments are present. Bioturbation is absent or rare, but *Teredolites* (1 to 6 cm in diameter) is present. Facies C gradually overlies Facies B.

Depositional environment: Facies C is inferred to represent the flood plain of a meandering river system, because it is muddy, includes many root traces, and overlies the meandering river-channel deposits (Facies B). The lenticular gravel beds with channelized bases represent chute-channel deposits (Walker and Cant, 1984). Thick lignite beds and claystone are also interpreted to have been deposited in the oxbow lake environment (Walker and Cant 1984), the sedimentation in which would be restricted to fines (mud) during overbank flooding from the main stream.

Facies D: Lake

Description: Facies D (60 cm thick) consists of greenish-gray siltstone. This facies is generally structureless, except for the local presence of indistinct parallel laminae and water-escape structures. Many leaf fossils and wood fragments are present. Bioturbation is absent. Facies D is distinguished from Facies C by the former's lack of root traces and lignite beds. Facies D gradually overlies Facies B.

Depositional environment: Facies D is interpreted to represent deposition from suspension in stagnant waters, because it contains massive silt with many leaf fossils. An overall lack of bioturbation suggests a non-marine environment with fresh water conditions. Facies D is inferred to have been deposited in a freshwater lake that was too deep for land plants to live in.

Facies E: Tidal Flat

Description: Facies E (4.4 to 5.0 m thick) mainly consists of fine- to medium-grained massive sandstone and interbedded very fine to fine-grained sandstone and siltstone, and it contains medium-grained cross-bedded sandstone. The

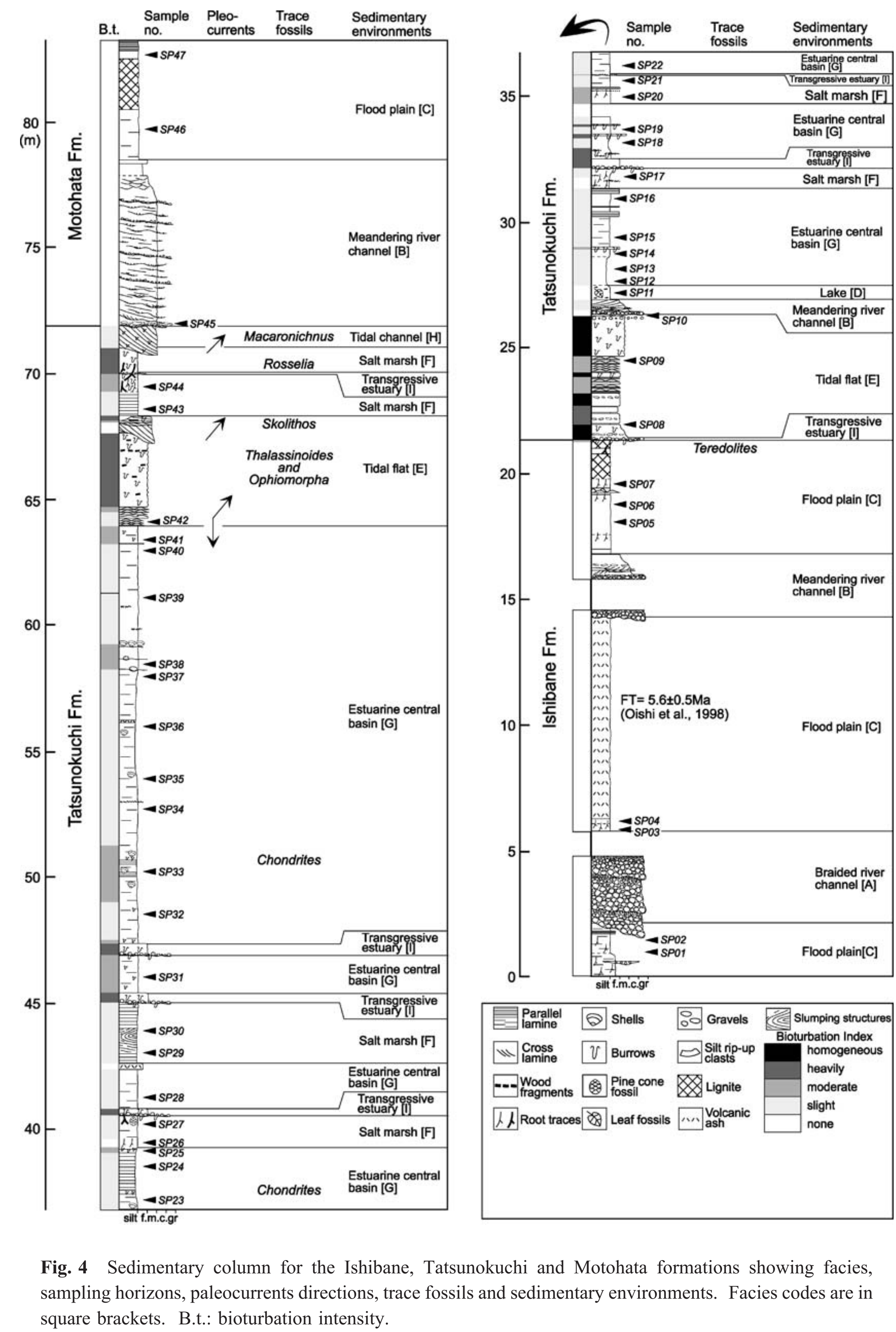

Fig. 4 Sedimentary column for the Ishibane, Tatsunokuchi and Motohata formations showing facies, sampling horizons, paleocurrents directions, trace fossils and sedimentary environments. Facies codes are in square brackets. B.t.: bioturbation intensity.

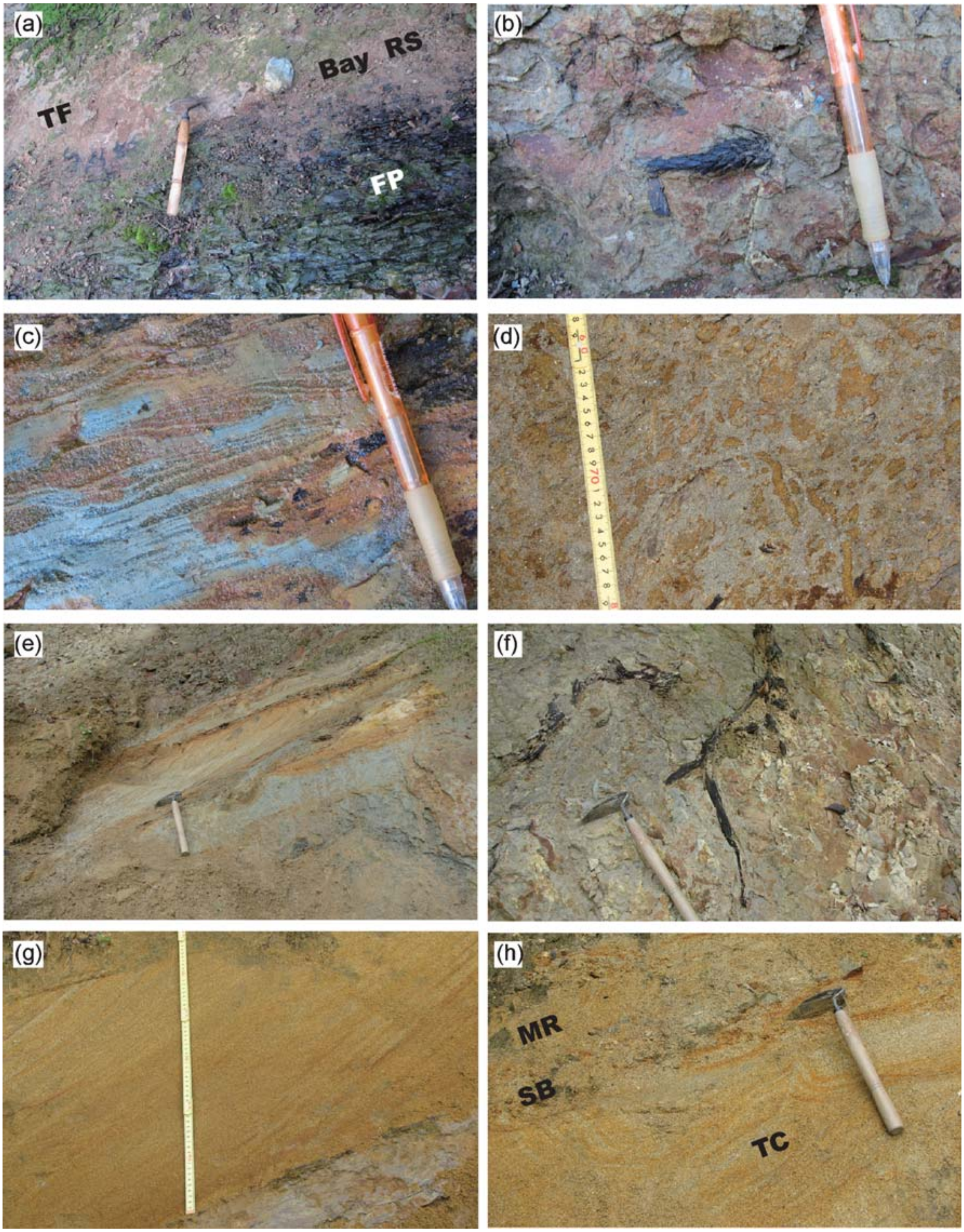

Fig. 5 Facies of the Ishibane, Tatsunokuchi and Motohata formations. (a) bay ravinement surface (Bay RS) between flood plain (FP) and tidal flat (TF) deposits, (b) pine cone fossils in salt marsh deposits, (c) ripple cross laminae with mud drape in tidal flat deposits, (d) intense bioturbation in tidal flat deposits, (e) tidal creek deposits, (f) root traces in salt marsh deposits, (g) large cross-bedded sandstone with *Macaronichnus*-burrows in tidal channel deposits, (h) sequence boundary (SB) between tidal channel (TC) deposits in the Tatsunokuchi Formation and meandering river channel (MR) deposits in the Motohata Formation.

massive sandstone is poorly sorted and includes pebble (1 cm in diameter) and thin siltstone beds. The intervals are characterized by intense bioturbation and large trace fossils such as *Ophiomorpha* and *Thalassinoides* (1 to 6 cm in diameter). Many wood fragments are included. The interbedded sandstone and siltstone show flaser and wavy bedding with short wavelength (several centimeters). Current ripples and wave-ripple cross laminae with mud drapes are present. Paleocurrent directions of current ripples show bidirectional flow (toward the south or northeast). Bioturbation is weak. The large cross-bedded sandstone is moderately well sorted and exhibits a channelized base and foreset laminae. The maximum thickness of the cross-set is about 70 cm. Paleocurrent directions are toward the northeast. Bioturbation is rare, but *Skolithos* is present. Facies E gradually overlies Facies G and Facies I.

Depositional environment: The occurrence of mud drapes on ripples, bidirectional paleocurrent and flaser to wavy bedding characterizes tide-influenced settings (Dalrymple et al. 1992). Large trace fossils and intense bioturbation are evidence for an oxic marine habitat in which benthic organisms were active. Therefore, Facies D is interpreted as tidal flat deposits. The large cross-bedded sandstone is inferred to represent tidal creek deposits formed by flood tide, on the basis of their channelized bases and landward-directed paleocurrents.

Facies F: Salt Marsh

Description: Facies F (0.8 to 1.7 m thick) consists of light- to dark-gray and greenish-gray siltstone with thin sandstone beds. This facies is mostly structureless and contains water-escape structures. It is characterized by the presence of many root traces, wood fragments, and pine cone fossils. Bioturbation is moderate and rare. *Rosselia* (2 cm in diameter) is present. Facies F gradually overlies Facies G and Facies E and gradually overlies Facies I.

Depositional environment: Facies F is interpreted to have been deposited in a muddy marsh environment on land, because this facies consists of siltstone with many root traces and wood fragments. Bioturbation and trace fossils also reflect the influence of seawater. Therefore, Facies D is considered to be salt marsh deposits (Frey and Basan, 1985).

Facies G: Estuarine Central Basin

Description: Facies G (1.4 to 16.7 m thick) mainly consists of bluish, light- to dark-gray claystone and siltstone and contains intercalated thin very fine to fine-grained sandstone beds (less than 10 cm in thickness). Pebbles (2 to 10 cm in diameter) are present in thin sandstone beds. This facies has obscure parallel laminae with bioturbation (or structureless) and partly has distinct parallel laminae (lower part of Facies G). Weak bioturbation and small-size trace fossils such as *Chondrites* (several millimeters in diameter) are present. Many shell fragments are scattered throughout Facies G, and shell fragments are sometimes concentrated as laminae. Facies G sharply overlies Facies D and gradually overlies Facies I.

Depositional environment: The fine-grained deposits imply that deposition occurred in a low-energy environment. The presence of shell fragments and bioturbation indicate a seawater-influenced environment. These characteristics imply that Facies G was deposited in an estuarine central basin (Dalrymple et al. 1992). The parallel-laminated siltstone beds without bioturbation also indicate the presence of stressed conditions for organic activity (Demaison and Moore, 1980), perhaps because of an anoxic sea floor.

Facies H: Tidal Channel or Flood Tidal Delta

Description: Facies H consists of coarse-grained and well to moderately sorted sandstone. The thickness of this facies is about 1 m and varies laterally. This facies shows large foreset laminae and normal grading. Paleocurrents flowed toward the northeast. *Macaronichnus* (a few millimeters in diameter) occurs throughout Facies H. Facies H erosionally overlies Facies F.

Depositional environment: Facies H is interpreted as tidal channel or flood tidal delta deposits (Boothroyd, 1985; Anthony et al.. 2002). This interpretation is based on landward paleocurrents, well-sorted and large cross-bedded sandstone, and erosional bases. The trace fossil *Macaronichnus* indicates a depositional environment within the upper subtidal to lower intertidal zones (Clifton and Thompson, 1978).

Facies I: Lag Deposits on Transgressive Tidal Flat

Description: Facies I (less than 45 cm thick) mainly consists of very poorly to poorly sorted and fine-grained sandstone and contains subrounded pebble-size conglomerate in its basal part. This facies is structureless and shows intense bioturbation. Facies I is gradually overlain by Facies E or G and erosionally overlies Facies C or F for the most part.

Depositional environment: Facies C and F are inferred to have been deposited in shallower environments than Facies E and G. These facies changes imply transgression. Hence, Facies I is considered to be deposited on a tidal flat setting during a transgression. The facies therefore consists of transgressive lag deposits that were formed by tidal erosion of a tidal flat during a transgression. Furthermore, the erosional surface of the base of this facies is inferred to be a bay ravinement surface (Nummedal and Swift, 1987).

2. Grain-size distribution of muddy sediments

The mean grain size of the muddy deposits ranges from 9.4 to 162.4 μm (Fig. 6). The grain size in the Ishibane and Motohata formations fluctuates widely between 9.4 and 162.4 μm. The size is larger than near fluvial channel deposits, whereas it is smaller (less than 27 μm) near lignite beds. The grain size in the Tatsunokuchi Formation ranges between 15.6 and 107.6 μm. This interval shows a fining-upward trend and then a coarsening-upward trend. The boundary between these trends is present at the horizon about 56 m above the base of the stratigraphic column in the study area (Fig. 6).

3. Diatom assemblages

Diatom assemblages in the Tatsunokuchi Formation are dominated by *Paralia sulcata*, and by *Thalassionema nitzschioides*, with *Aulacoseira granulata*, *Navicula erfuga*, *Koizuma tatsunokuchiensis*, *Trochosira concava*, *Neodenticula kamtschatica*, *Thalassiosira antique*, *Thalassiosira oestrupii*, and *Thalassiosira temperei*.

Fresh to brackish water species such as *Aulacoseira granulata*, *Navicula erfuga*, *Achnanthes* spp., and *Pinnularia* spp. constitute 0.5 to 47.5% of the total assemblages and have the fifth highest content spikes (~30% or over ~30%) occurring at salt marsh, tidal flat, and lake deposits in intervals of 21 to 45 m (Fig. 6). A diatom identified as *Trochosira concava* is morphologically very similar to *Pseudopodosira kosugii*, a small centric diatom that is known to occur only in the intertidal zone (Tanimura and Sato, 1997). *T. concava* is found in tidal flat deposits. The marine coastal diatom *Paralia sulcata* occurs throughout the studied stratigraphic section with a maximum of 63% of the total abundance. The species has abundant occurrences in the lowest and uppermost parts (estuarine central basin, salt marsh, and tidal flat deposits) of the Tatsunokuchi Formation. The content of *Thalassionema nitzschioides* exceeds 20% at four horizons of thin central basin deposits in intervals of 21 to 45 m. *Thalassionema nitzschioides* and other open marine species such as *Neodenticula kamtschatica*, *Thalassiosira antique*, *Thalassiosira oestrupii*, and *Thalassiosira temperei* dominantly and are continuously yielded in the upper part (estuarine central basin deposits above 45 m) of the formation.

4. Total sulfur content (TS)

The TS content ranges from 0.01 to 2.56%. The values in fluvial flood plain deposits of the Ishibane and Motohata formations range from 0.01 to 0.09% (Fig. 6). The values in estuary deposits of the Tatsunokuchi Formation fluctuate widely between 0.07 to 2.56%. The values increase upward from 0.2 to 2.0% with facies changes from tidal flat to estuarine central basin deposits. They then decrease upward in the interval of 58 to 72 m above the base of the stratigraphic column.

5. Total organic carbon content (TOC)

The TOC content ranges between 0.12 and 3.69% (Fig. 6). The TOC contents in fluvial flood plain deposits of the Ishibane and Motohata formations are mainly less than 1%, except for one sample. The TOC contents in estuary deposits of the Tatsunokuchi Formation range between 0.23 and 1.84%. The TOC content is high (about 1.5%) in the estuary deposits in the interval of 21 to 40 m above the base of the stratigraphic column, but it is low only in tidal flat deposits. The contents decrease to about 1.0% in the interval of 40 to 58 m and then become lower (0.3%) above 58 m.

6. Organic matter composition

The composition and photographs of observed organic matter are shown in Figures 6 and 7, respectively. Organic matter in the fluvial flood plain deposits of the Ishibane and Motohata formations consists of vitrinite (3.33 to 52.33%), fusinite (0.67 to 56.67%), cutinite (6.67 to 27.00%), sporinite (less than 2.00%), and NFA (30.67 to 63.00%). The NFA is non-fluorescent or has very weak fluorescence. A few broken *Pediastrum* in the freshwater alginite are present on some slides, but they were not encountered in 300 counted points. The organic matter composition is characterized by high proportions of vitrinite, fusinite, cutinite, and sporinite. However, the proportion of NFA increases near lignite beds.

Organic matter in the estuary deposits of the Tatsunokuchi Formation consists of vitrinite (11.00 to 38.33%), fusinite (0.33 to 6.00%), cutinite (7.33 to 29.00%), sporinite (less than 2.00%), alginite (less than 0.67%), and NFA (39.67 to 75.00%). A few broken *Pediastrum* in the freshwater alginite are present on some slides, but they were not encountered in 300 counted points. Well-preserved dinoflagellate cysts and acritarchs are present on each slide. In particular, several tens of dinoflagellate cysts are found in estuarine central basin deposits between 46 and 56 m from the bottom of the column. The proportions of vitrinite and cutinite are relatively high in estuary deposits in the interval of 21 to 46 m. These proportions decrease in the interval of 46 to 56 m, whereas the proportions of NFA and marine alginite are highest. The proportions of vitrinite and cutinite increase upward in the interval of 56 to 72 m.

7. $\delta^{13}C$ values of organic matter

The $\delta^{13}C$ values range from −28.70 to −22.77‰ (Fig. 6). These values in fluvial flood plain deposits of the Ishibane and Motohata formations range from −28.70 to −26.24‰. The values in estuary deposits of the Tatsunokuchi Formation fluctuate widely between −28.39 and −22.77‰. The values are relatively low (about −26‰) in the interval of 21 to 46 m above the base of the stratigraphic column. The values increase to about −23‰ in the interval of 46 to 56 m and decrease upward to about −27‰ above 56 m.

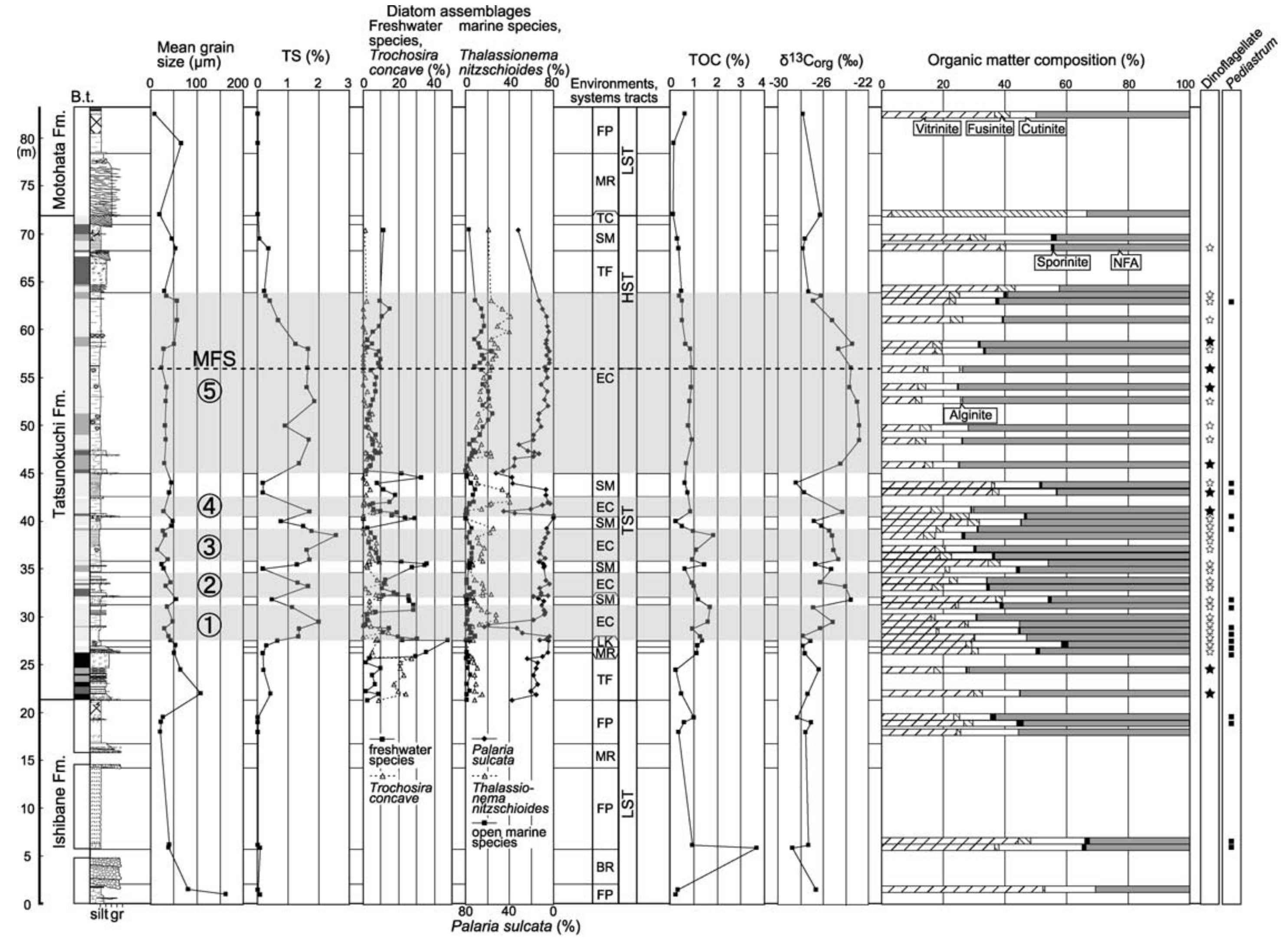

Fig. 6 Summarized column for the Ishibane, Tatsunokuchi and Motohata formations showing facies, sedimentary environments, systems tracts, grain size, total organic carbon (TOC) and total sulfur (TS) contents, stable carbon isotope ratios of organic matter ($\delta^{13}C_{org}$), and organic-matter composition. Square marks show the presence of *Pediastrum*. Black and white stars show more than and less than 20 specimens of dinoflagellate cysts, respectively. LST: lowstand systems tract, TST: transgressive systems tract, HST: highstand systems tract, BR: braided river channel, MR: meandering river channel, FP: flood plain, LK: lake; TF: tidal flat, SM: salt march, EC: estuarine central basin (gray colored), TC: tidal channel, MFS (dashed line): maxmum flooding surface, ① to ⑤: transgression cycles.

Discussion

1. Facies succession and environmental changes

The Ishibane Formation and the lower part (21 to 56 m) of the Tatsunokuchi Formation show a fining-upward facies succession that is composed of braided river or meandering river (Facies A, B, and C), alternation of tidal flat (Facies E) or salt marsh (Facies F) and estuarine central basin deposits (Facies G), and thick estuarine central basin deposits, in ascending order (Figs. 4 and 6). The facies succession shows the expansion of the estuary during transgression. The erosional boundary between meandering river deposits of the Ishibane Formation and tidal flat deposits of the Tatsunokuchi Formation is regarded as a bay ravinement surface (Nummedal and Swift 1987) that is formed by tidal erosion during transgression (Fig. 4). Pebbly deposits (Facies I) located above the surface are interpreted as transgressive lag.

The TS content is generally below 0.3% for nonmarine sediments and is between 0.3 and 3.0% for marine sediments (Keith and Degens, 1959; Berner, 1970, 1984; Nakai et al., 1982; Berner and Raiswell, 1984; Koma, 1987, 1992; Koma et al., 1983, 1989; Sampei et al., 1997). The values are also higher on an anoxic bottom. The TS content in the Tatsunokuchi Formation in interval 21 to 56 m increases from 0.1 to 0.5% with the facies change from meandering river flood plain to tidal flat deposits (Fig. 6). This implies an increase in marine influence. The TS content increases to about 1.5 % in estuarine central basin deposits (Fig. 6). Additionally, the size of trace fossils becomes smaller in estuarine central basin deposits. These results show that the estuarine central basin was subjected to anoxic bottom conditions.

Diatom assemblages in the tidal flat and salt marsh deposits are characterized by high proportions of fresh to brackish water species and an intertidal species *Trochosira concava* (Fig. 6). The marine coastal species, *Palaria sulcata*, increases in estuarine central basin deposits. Alternating predominance of marine and fresh water species corresponds to four repetitions of transgression and regression in an interval of 21 to 45 m. Furthermore, the proportion of

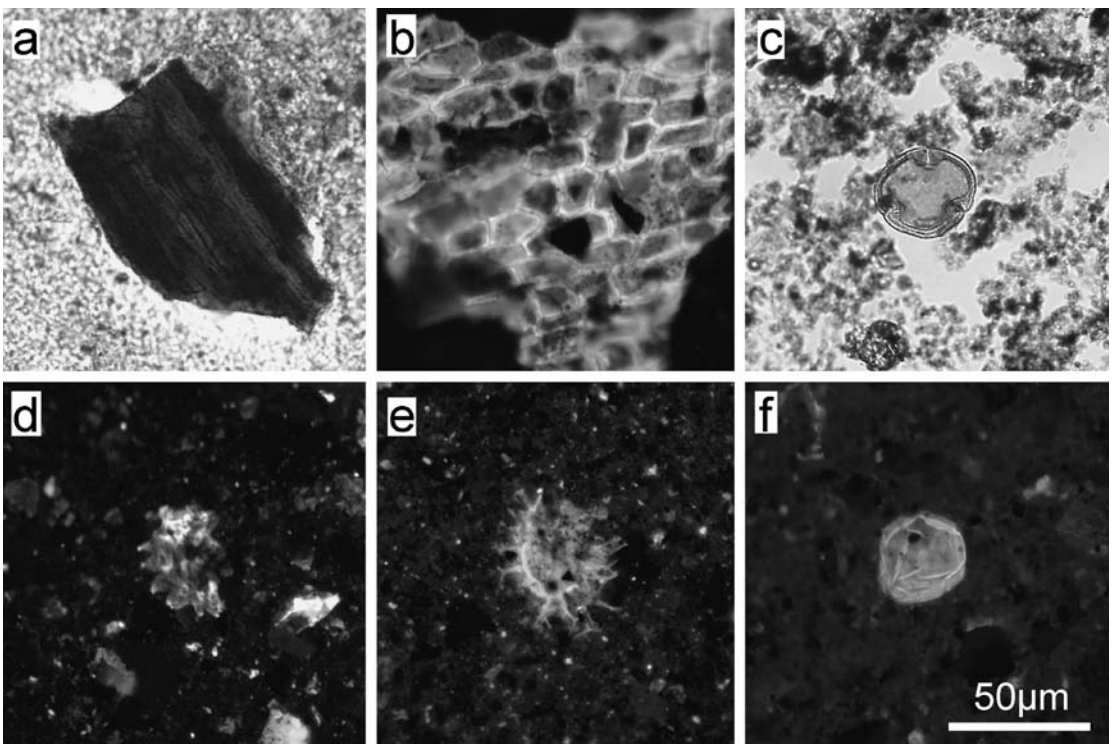

Fig. 7 Organic matter under reflected-light fluorescence microscopy (Olympus BX50) with tungsten light (a and c) and ultraviolet light (b, d, e and f). Scale bar is 50 μm, a: vitrinite in flood plain deposits, b: cutinite in estuarine central basin deposits, c: sporinite in estuarine central basin deposits, d: freshwater alginite (*Pediastrum*) in salt marsh deposits, e: marine alginite (dinoflagellate cysts) in tidal flat deposits, f: marine alginite (acritarchs) in estuarine central basin deposits.

Thalassionema nitzschioides and open marine species increases upward in the estuarine central basin deposits of 45 to 56 m, whereas that of coastal species decreases (Fig. 6). These results suggest that the Tatsunokuchi Foramation includes four transgressive and regressive cycles in the lower part and one cycle of transgression and regression in upper part. According to reconstructions by Chinzei and Iwasaki (1967) and Ogasawara (1994) the bay may have expanded from Fukushima to Iwate prefectures (Fig. 1).

The upper part (56 to 72 m) of the Tatsunokuchi Formation shows a coarsening-upward facies succession that is composed of estuarine central basin (Facies G), tidal flat (Facies E), and salt marsh deposits (Facies F), in ascending order (Figs. 4 and 6). The TS content decreases to 0.3% in this interval, whereas intense bioturbation and large-size trace fossils *Ophiomorpha* and *Thalassinoides* are present. The proportions of freshwater and coastal diatoms also increase upward in this interval, whereas that of open marine diatoms decreases (Fig. 6). These results imply the increased influence of river discharge during regression on the estuarine central basin and the change in the bottom conditions to oxic conditions.

Estuary deposits of the Tatsunokuchi Formation are unconformably overlain by meandering river deposits (Facies B and C) of the Motohata Formation (Figs. 4 and 6). An overall lack of bioturbation and low TS content (less than 0.1%) in this interval shows that the activity of benthic organisms was restricted because of the stress caused by fresh water.

2. Sequence stratigraphy

Fluvial deposits of the Ishibane Formation unconformably overlie basement rocks of the Hishinai Formation (Oishi et al., 1998) (Fig. 3). The boundary between these formations is thought to be a sequence boundary (e.g., Posamentier et al. 1988; Van Wagoner et al. 1988) that was formed by fluvial erosion during a sea-level fall (Fig. 8). In embayment settings, the bay ravinement surface (Nummedal and Swift, 1987), which lies between fluvial deposits of the Ishibane Formation and estuary deposits of the Tatsunokuchi Formation, corresponds to a transgressive surface (Figs. 6 and 8). Therefore, fluvial deposits above the sequence boundary and below the transgressive surface are inferred to represent a lowstand systems tract (e.g., Posamentier et al., 1988; Van Wagoner et al., 1988). A laterally traceable marine bed with molluscs is found in the geological cross section (Oishi et al., 1998) of the Kitakami Lowland (Figs. 3 and 8). This horizon is interpreted as a maximum flooding surface, in which the coastline moved to its most inland site (Fig. 8). The maximum flooding surface (MFS) correlates with the horizon about 56 m above the base of the stratigraphic column in the study area (Fig. 6). Therefore, estuary deposits of the Tatsunokuchi Formation below and above the maximum flooding surface are regarded as a transgressive systems tract and a highstand systems tract (TST and HST) (e.g., Posamentier et al., 1988; Van Wagoner et al., 1988), respectively (Figs. 6 and 8). The Tatsunokuchi Formation is unconformably overlain by fluvial deposits of the Motohata Formation (Oishi et al., 1998). This unconformity is inferred to be a sequence boundary and the fluvial deposits are regarded as a lowstand systems tract (Figs. 6 and 8).

The depositional sequence of the Ishibane and Tatsunokuchi formations is thought to have been formed during about 1 m.y. on the basis of FT ages (Oishi et al., 1996; Oishi and Yoshida, 1998) and diatom biostratigraphy (Yanagisawa, 1998). A cycle of 0.5 to 3 m.y. is defined as a third-order sequence in sequence stratigraphy (e.g., Vail et al., 1991). The Haq curve (Haq et al., 1988) shows that the eustatic sea level began to rise from about 5.5 Ma and reached a maximum transgression at about 5 Ma, subsequently falling from about 4 Ma. The eustatic sea level changes coincide with the conclusions of the present study. Therefore, it is interpreted that the third-order depositional sequence recognized in the Ishibane and Tatsunokuchi formations reflects the eustatic sea level changes. Furthermore, the transgressive systems tract in the Tatsunokuchi Formation includes four cycles of transgression and regression, while highstand systems tract shows one cycle of transgression and regression (Fig.6). These cyles might be caused by higher-order sea-level fluctuations with the duration of 200 to 400 k.y.

3. Relationship among depositional environments, systems tracts, and organic matter preservation

Lowstand systems tract (LST) (Ishibane and Motohata formations)

The TOC content of the LST fluvial deposits is relatively low (about 0.5%), except in one sample including many wood fragments (Fig. 6). In particular, the mean grain size increases to more than 50 μm where the TOC content is low (0.2 to 0.3%). This reflects the fact that the supply of coarse-grained silisiclastics diluted the TOC content of sediments. Organic-matter composition of fluvial deposits is characterized by high proportions of the vitrinite, fusinite, cutinite, and sporinite, which are coarse-grained and terrestrial in origin, and by the occurrence of the freshwater alga *Pediastrum* (Fig. 6). Terrestrial plants (C3 plants), except for C4 and CAM plants, and marine plankton in the low- and mid-latitude ocean have $\delta^{13}C_{org}$ values between −23 and −31‰ (average −27‰) and between −18 and −24‰ (average −22‰), respectively (Deines, 1980; Anderson and Auther, 1983; Boutton, 1991; Tyson, 1995). $\delta^{13}C_{org}$ values in fluvial deposits of the Ishibane and Motohata formations

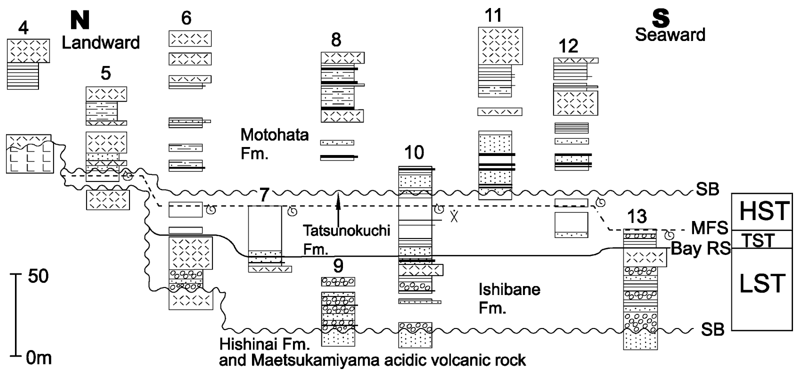

Fig. 8 Schematic cross section of sequence stratigraphy in the Kitakami lowland area. See figure 3 for each columnar section. SB: sequence boundary, Bay RS: bay-ravinement surface, MFS: maximum flooding surface.

range between −26.2 and −28.7‰ (Fig. 6). These phenomena indicate that terrestrial plants were supplied and preserved in sediments, and they support the conclusions based on facies analysis.

However, some samples in fluvial flood plain deposits show a relatively high proportion of NFA, though $\delta^{13}C_{org}$ values are the same (Fig. 6). In general, the proportion of AOM is low in freshwater deposits (e.g., Ujiie, 1992; Hart 1994; Omura et al., 2000; Omura and Hoyanagi, 2004). The present study shows that the proportion of NFA is high (50 to 63%) in intervals that are fine-grained and with intercalating thick lignite beds, whereas NFA is low (about 30%) in intervals that are sandy and near river channels deposits (Figs. 6 and 9a). Fluvial flood plain deposits of the former are interpreted to have been deposited in the oxbow with an anoxic bottom. Therefore, organic matter has possibly been preserved as NFA under low-energy and anoxic bottom conditions in freshwater environments.

Transgressive and highstand systems tracts (Tatsunokuchi Formation)

Lower part of transgressive systems tract: The TOC content in the lower part of the TST increases from 0.3 to 1.5% with facies changes from tidal flat to estuarine central basin deposits (Fig. 6). The TOC trend reflects the change of estuarine bottom conditions to anoxic, which is suitable for preserving organic matter. The estuarine central basin deposits are also characterized by relatively higher TOC content (about 1.5%) and lower $\delta^{13}C_{org}$ values (about −26‰) than those in the upper part of the TST (Figs. 6 and 9b). Furthermore, the proportions of vitrinite and cutinite in the lower part of the TST are higher than those in the upper part (Fig. 9b). These phenomena indicate that the estuary in the early transgressive stage was under a relatively strong influence of rivers and that large amounts of terrigenous organic matter supplied into the estuary were effectively preserved under anoxic bottom conditions (Fig. 9b).

The lower part of the TST is also characterized by repetitions of a facies succession that consists of erosional-based lag, estuarine central basin, and salt marsh deposits, in ascending order (Figs. 4 and 6). The succession is at intervals of 5 m and is definitely present only in the lower part of the TST. The proportions of freshwater diatoms and vitrinite and cutinite increase upward in the lower part of estuarine central basin deposits and decrease in the upper part of estuarine central basin deposits and salt marsh deposits (Fig. 6). In contrast, the proportion of marine diatoms, TS content, and $\delta^{13}C_{org}$ values show opposite patterns (Fig. 6). These results indicate that river discharge and seawater-intrusion were alternatively stronger and weaker in the early transgressive stage. Numerous river floods may have occurred in the transgressive stage because of high precipitation during the warming stage, and they may have supplied large amounts of freshwater and muddy deposits into the estuary. In addition, it was possibly easy to record small variations of river discharge in the lower part of the TST, because of increasing sedimentary space. Therefore, small fluctuations of competition between river discharge and seawater-intrusion are interpreted to reflect subtle variations of the balance between river floods and sea-level rise during the early transgressive stage. The occurrences of river floods also should have affected the occurrence of the high TOC content in the lower part of the TST in terms of supply of terrigenous organic matter.

Upper part of transgressive systems tract: The TOC content decreases to about 0.8% in the upper part of the TST, although the TS content stays high (about 1.5%), as it also does in the lower part of the TST (Figs. 6 and 9b). The proportions of vitrinite and cutinite in this interval are lowest, whereas $\delta^{13}C_{org}$ values are highest (about −23‰) (Fig. 6). In addition, marine plankton dinoflagellate cysts and acritarchs, and marine diatoms are well preserved in this interval, and their proportions increase toward the maximum flooding surface (Fig. 6). These imply the increase in the marine influence during transgression and environmental

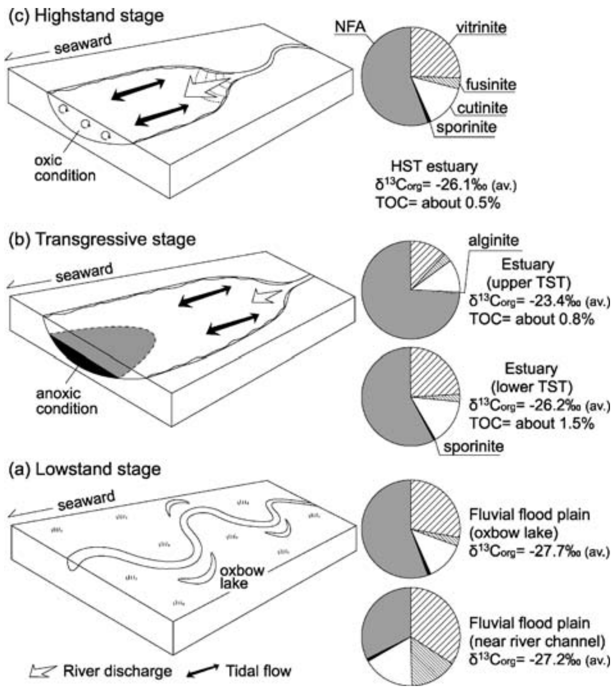

Fig. 9 Sequence stratigraphic interpretation of estuarine environments and organic matter preservation. (a) fluvial environment in the lowstand stage, (b) estuary environment in the transgressive stage, (c) estuary environment in the highstand stage.

changes to a large estuary such as a bay. Therefore, relatively low TOC content in the upper part of the TST is interpreted to reflect the decrease in terrigenous organic matter supplied into the estuarine central basin. The organic-matter composition in the upper part of the TST is also characterized by the high proportion of NFA (Fig. 6). Tyson (1993, 1995) stated that a part of AOM was produced by benthic bacteria under anoxic marine conditions. The TS content is high in the upper part of the TST, as mentioned above. These show that organic matter in the Tatsunokuchi Formation has been preserved as NFA in sediments under anoxic marine conditions.

Highstand systems tract: The TOC and TS contents in the HST gradually decrease upward to 0.5 and 0.3%, respectively (Figs. 6 and 9). The proportions of vitrinite, cutinite, and freshwater diatoms gradually increase upward in this interval, whereas $\delta^{13}C_{org}$ values decrease to about −27‰ (Fig. 6). These phenomena indicate that vertical mixing in the estuary was caused by the increased influence of rivers during regression, and preservation of organic matter under oxic bottom conditions was difficult (Fig. 9c). Alternatively, the supply of coarse-grained silisiclastics may have diluted the TOC content of sediments, because mean grain size sharply increases to more than 50 μm in this interval (Fig. 6).

Conclusions

This study documents depositional environmental changes and their relation to the origin of sedimentary organic matter, combined with sequence stratigraphy of an incised-valley system in the Miocene to Pliocene Ishibane, Tatsunokuchi, and Motohata formations. The present approach has enabled a discussion about the relationship between sedimentary organic matter and other paleoenvironmental proxies such as sedimentary facies, diatom assemblages, total organic carbon (TOC) contents, and total sulfur (TS) contents with more precise chronological resolution than similar previous studies. The depositional pattern and origin of organic matter are summarized as follows.

1. Lowstand systems tract is generally characterized by relatively high proportions of vitrinite and cutinite and low $\delta^{13}C_{org}$ values. The transgressive systems tract is represented by the upward decrease in vitrinite and cutinite with the increase in marine alginite, NFA, and $\delta^{13}C_{org}$ values. The highstand systems tract shows an upward increase in vitrinite and cutinite as well as the decrease in marine alginite, NFA, and $\delta^{13}C_{org}$ values.
2. The TOC content is highest in the lower part of the transgressive systems tract. The interval shows relatively high TS content and low $\delta^{13}C_{org}$ values. In additions, competition between river discharge and seawater intrusion are recorded well in the compositional variations of diatom assemblage, and these fluctuations may reflect subtle variations of a balance between river floods and sea-level rise during the early transgressive stage.

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堆積有機物，イオウ，珪藻化石をもちいた解析谷埋積システムにおける高分解シーケンス層序解析：東北日本岩手県に分布する中新統・鮮新統竜の口層の例

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岩手県南部の北上市に分布する中新統～鮮新統の石羽根，竜の口，本畑層は，エスチュアリーと河川環境での堆積を示している．また，1 回の海水準上昇と引き続く低下で堆積した地層で，低海水準期堆積体，海進期堆積体，高海水準期堆積体に区分できる．その周期は約 100 万年と推定できる．河川成堆積物からなる低海水準期堆積体では陸源有機物片を多く含み，有機物は低い安定炭素同位体比（$\delta^{13}C_{org}$）を示す．引き続くエスチュアリー環境を示す海進期堆積体では，上方に向かって，海棲有機物が増え，$\delta^{13}C_{org}$の値が大きくなり，さらに全有機炭素量も増加する傾向にある．海進初期ではこれらは，小さな増減を 4 回繰り返しているが，やがて最大海進面に向かって安定して増加するようになる．最大海進面より上位の高海水準期堆積体では陸源有機物量が増大し，$\delta^{13}C_{org}$と全有機炭素量は減少し，陸域からの物質供給が増加したことが示される．珪藻化石の淡水種，汽水種，潮間帯種，沿岸性種，外洋種の比率変化も同一の海進海退の繰り返しを竜の口層中に記録している．したがって，竜の口層の海進期堆積体は短周期の 4 回の海進海退と引き続く海進から構成され，一方，高海水準期堆積体は 1 回の海退を示す．これらは数 10 万年周期の海水準変動に対応するかもしれない．