

1 **High-resolution upper Maastrichtian carbon isotope stratigraphy of terrestrial**
2 **organic matter from northern Japan**

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17 **ABSTRACT**

18 High-resolution stable carbon isotope stratigraphy of terrestrial organic matter was
19 established for the upper Maastrichtian Senpohshi Formation of the Nemuro Group in
20 eastern Hokkaido, northern Japan. The Senpohshi Formation, approximately 1,300 m
21 thick, is dominated by hemipelagic mudstone deposited along an active margin in the
22 North Pacific region. Microscopic observations of extracted kerogen samples from the
23 formation revealed the presence of sedimentary organic matter (SOM), predominantly
24 phytoclasts and a minor amount of non-fluorescent amorphous organic matter, indicating
25 material of a terrestrial higher plant origin. The atomic hydrogen/carbon ratios of the
26 kerogen samples indicated a coalification rank at the anthracite stage or below. Therefore,
27 the stable carbon isotope values of the bulk SOM obtained for the Senpohshi Formation
28 represent the unmodified, original values of terrestrial organic matter. The stable carbon
29 isotope profile reconstructed for the Formation provides the first high-resolution
30 terrestrial record of the Mid-Maastrichtian Event (MME), which is comparable to high-
31 resolution marine carbon isotope data from other sections. The carbon isotopic signatures
32 defined by the marine records are recognized in the terrestrial data from the Formation,
33 especially in middle to upper part of the event. However, the terrestrial record showed a
34 discrepancy from the marine data in the lower part of the MME, suggesting local variation

35 of the hinterland environment in the North Pacific region. This study provides new insight
36 into environmental changes during the late Maastrichtian by establishing a detailed
37 carbon isotope record of terrestrial materials.

38

39 *Keywords:* Carbon isotope stratigraphy; terrestrial organic matter; Mid-Maastrichtian
40 Event; hinterland environment; Nemuro Group; North Pacific region

41 **1. Introduction**

42 The Maastrichtian Age is a time marked by extensive environmental changes. Third-
43 order cycle, high-amplitude falls in sea level are recognized in the Maastrichtian, which
44 can be superimposed over the long-term sea-level drop from the latest Cretaceous through
45 the Paleogene (Haq et al., 1987; Crampton et al., 2006; Simmons et al., 2007; Kominz et
46 al., 2008). The Maastrichtian is also characterized by short-term climatic cooling and
47 warming events accompanied by the long-term climatic cooling in the latest Cretaceous
48 (Barrera and Huber, 1990; Barrera and Savin, 1999; Frank and Arthur, 1999; Li and
49 Keller, 1999; Huber et al., 2002; Frank et al., 2005; MacLeod et al., 2005; Isaza-Londoño
50 et al., 2006; Friedrich et al., 2009; Thibault and Gardin, 2010). Furthermore, changes in
51 thermohaline circulation during the Maastrichtian have been suggested, although the
52 mechanism and characteristics remain controversial (MacLeod and Huber, 1996; Barrera
53 et al., 1997; Barrera and Savin, 1999; Frank and Arthur, 1999; MacLeod et al., 2000;
54 Frank et al., 2005; Pucéat et al., 2005; Donnadieu et al., 2006; Hunter et al., 2008;
55 MacLeod et al., 2011; Murphy and Thomas, 2012; Robinson and Vance, 2012; Robinson
56 et al., 2012; Jung et al., 2013; Voigt et al., 2013; Moiroud et al., 2016; Farnsworth et al.,
57 2019; Haynes et al., 2020; Ladant et al., 2020).

58 Environmental changes during the Maastrichtian likely resulted in significant biotic
59 events. For example, evidence from this stage suggests widespread inoceramid bivalve
60 extinction in the mid-Maastrichtian (Ward et al., 1991; MacLeod et al., 1996), rudist reef
61 collapse in tropical regions (Johnson and Kauffman, 1990, 1996; Johnson et al., 1996),
62 and an increase in provinciality in plankton microfossil assemblages (Shafik, 1990; Huber,
63 1992; Huber and Watkins, 1992; Burnett, 1998; Lees, 2002; Thibault et al., 2012a, b).
64 Moreover, there is evidence of latitudinal migration of calcareous plankton (e.g., the
65 calcareous nannofossils *Nephrolithus frequens* and *Micula murus* and the planktonic
66 foraminifer *Abathomphalus mayaroensis*) during the Maastrichtian (Pospichal and Wise,
67 1990; Huber, 1992; Huber and Watkins, 1992; Nifuku et al., 2009; Thibault et al., 2010;
68 Voigt et al., 2012). The present consensus is that sea-level fall, climatic cooling, and
69 changes in thermohaline circulation played important roles in these biotic events
70 (MacLeod and Huber, 1996; Barrera et al., 1997; Barrera and Savin, 1999; Frank and
71 Arthur, 1999; MacLeod et al., 2000; Frank et al., 2005; Pucéat et al., 2005; Donnadiou et
72 al., 2006; Hunter et al., 2008; MacLeod et al., 2011; Murphy and Thomas, 2012; Robinson
73 and Vance, 2012; Robinson et al., 2012; Jung et al., 2013; Voigt et al., 2013; Moiroud et
74 al., 2016; Farnsworth et al., 2019; Haynes et al., 2020; Ladant et al., 2020).

75 Records of stratigraphic trends in stable carbon isotopes are useful tools for
76 investigating paleoenvironments. A variety of marine and terrestrial-origin materials has
77 been measured for this purpose (e.g., bulk carbonate rock, calcareous fossil, bulk
78 sedimentary organic matter (SOM), and fossil wood fragments). Global
79 paleoenvironmental events are recorded in carbon isotope profiles, where measured
80 values are representative of the isotopic composition of ocean-atmosphere reservoirs.
81 Perturbation of global carbon isotope composition may be caused by the environmental
82 events, such as extensive organic carbon burial (Scholle and Arthur, 1980; Jenkyns, 1988),
83 CO₂ emission by massive volcanic events (Hesselbo et al., 2002), or large-scale
84 dissociation of methane hydrate (Dickens et al., 1995). The local environment can also
85 control the stable carbon isotope profile. For instance, carbon isotope values of marine
86 carbonate may be influenced by the local factors such as fluvial runoff, primary
87 productivity, or bottom water circulation (Immenhauser et al., 2008; Wendler, 2013).
88 Hinterland climate can affect the isotopic composition in terrestrial organic matter
89 (Farquhar et al., 1980; Ramesh et al., 1986; Körner et al., 1988; Leavitt, 1993; Feng and
90 Epstein, 1995; Lipp et al., 1996; Pendall et al., 1999; Schleser et al., 1999; Edwards et al.,
91 2000). Therefore, stable carbon isotope composition can record global to local scale
92 changes of the paleoenvironment.

93 Stable carbon isotope stratigraphy has been widely established in the Maastrichtian
94 sections (e.g. Li and Keller, 1998a, b; Barrera and Savin, 1999; Hasegawa et al., 2003;
95 Nordt et al., 2003; Friedrich et al., 2009; Voigt et al., 2010; Batenburg et al., 2012;
96 Thibault et al., 2012a, b; Voigt et al., 2012; Jung et al., 2012; Falzoni et al., 2016; Salazar-
97 Jaramillo et al. 2016; Dameron et al., 2017; Batenburg et al., 2018). Several prominent
98 carbon isotope events have been recognized in previous research. The Campanian–
99 Maastrichtian Boundary Event (CMBE) is characterized by a relatively prolonged
100 negative excursion from the latest Campanian to the early Maastrichtian (Barrera and
101 Savin, 1999). The Mid-Maastrichtian Event (MME) is characterized by two positive
102 peaks interrupted by a negative trough with multiple short-term negative excursions
103 (Voigt et al., 2012). Carbon isotope values dropped abruptly at the end of the Cretaceous,
104 which is marked by the Cretaceous–Paleogene Boundary Event (KPgE) (Hsü et al., 1982;
105 Perch-Nielsen et al., 1982; Zachos and Arthur, 1986; Keller and Lindinger, 1989). These
106 events are global phenomena indicated by carbon isotope events simultaneously recorded
107 in diverse materials from various regions (Hasegawa et al., 2003; Wendler, 2013; Salazar-
108 Jaramillo et al., 2016). In addition to these events, recent high-resolution carbon isotope
109 stratigraphy reveals the presence of further carbon isotope events in the Maastrichtian,

110 which are correlated globally (Jung et al., 2012; Thibault et al., 2012a, b; Voigt et al.,
111 2012; Wendler, 2013; Batenburg et al., 2018).

112 Most of the carbon isotope profiles from the Maastrichtian sections are derived from
113 marine materials, which are bulk carbonate rocks (Zachos et al., 1985; Corfield et al.,
114 1991; Stoll and Schrag, 2000; Voigt et al., 2010; Wendler et al., 2011; Jung et al., 2012;
115 Thibault et al., 2012a, b; Voigt et al., 2012; Batenburg et al., 2018) and single-species of
116 calcareous microfossils (Zachos et al., 1985, 1989; Barrera and Huber, 1990; D'Hondt
117 and Lindinger, 1994; Corfield and Norris, 1996; Li and Keller, 1998a, b; Barrera and
118 Savin, 1999; MacLeod and Huber, 2001; Friedrich et al., 2009).

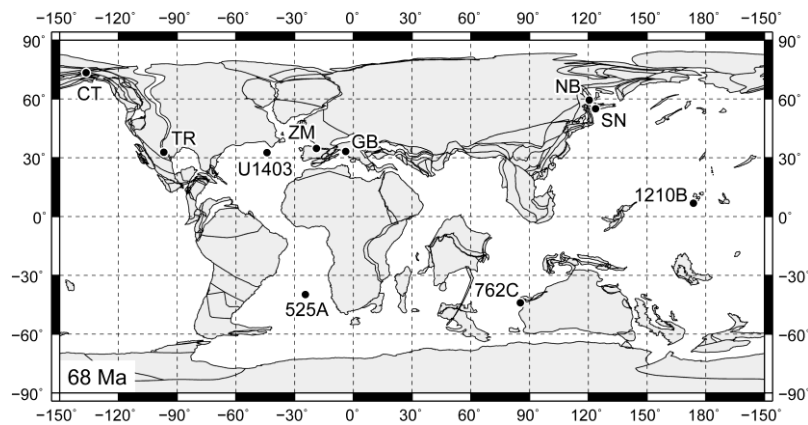
119 In contrast, the carbon isotope profile is rarely obtained from terrestrial materials.
120 Hasegawa et al. (2003) established stable carbon isotope stratigraphy from terrestrial
121 organic matter for the upper Cenomanian through upper Maastrichtian (up to polarity
122 chron C30n) from Naiba section in Sakhalin Island, Russia. Salazar-Jaramillo et al.
123 (2016) presented the profile for terrestrial organic matter in the lower Cantwell Formation,
124 Alaska, which covered the carbon isotope events CMBE and MME. Nordt et al. (2003)
125 obtained stable carbon isotope stratigraphy from pedogenic carbonate in Tornillo Basin,
126 Texas, which ranged from the upper Campanian through the Danian. However, the
127 temporal resolution of the isotope stratigraphies obtained from terrestrial material is much

128 lower than that of the recently established high-resolution carbon isotope stratigraphy
129 from marine carbonates (Batenburg et al., 2012; Jung et al., 2012; Thibault et al., 2012a,
130 b; Voigt et al., 2012; Batenburg et al., 2018). Moreover, limitation of the age control in
131 the lower Cantwell Formation and Tornillo Basin makes it difficult to correlate the
132 terrestrial carbon isotope profiles to the high-resolution marine records. Therefore,
133 acquisition of a high-resolution carbon isotope data from terrestrial material would
134 provide a comprehensive view on isotopic composition of ocean-atmosphere reservoirs
135 during the Maastrichtian. In addition, a detailed terrestrial carbon isotope record would
136 give an insight into terrestrial environment during the stage, which is rarely investigated
137 compared with the paleoenvironment in marine realm. The terrestrial data should be
138 obtained from the sections where sufficient age control has been established because it
139 enables accurate stratigraphic correlation between the terrestrial and marine records.

140 Here, we established a Maastrichtian carbon isotope stratigraphy of terrestrial
141 material at a higher resolution than the previous study on terrestrial materials (Hasegawa
142 et al., 2003; Nordt et al., 2003; Salazar-Jaramillo et al., 2016). We researched the upper
143 Maastrichtian Senpohshi Formation in eastern Hokkaido, northern Japan (Figs. 1 and 2).
144 The rapid sedimentation rate of the formation enabled us to establish a high-resolution
145 carbon isotope stratigraphy of terrestrial-origin SOM. Furthermore, the

146 magnetostratigraphy established in the section provides reliable stratigraphic markers for
147 global correlations (Nifuku et al., 2009). In this paper, we discuss the carbon isotope
148 stratigraphy derived from the Senpohshi Formation and its comparison with the carbon
149 isotope profiles from other sections.

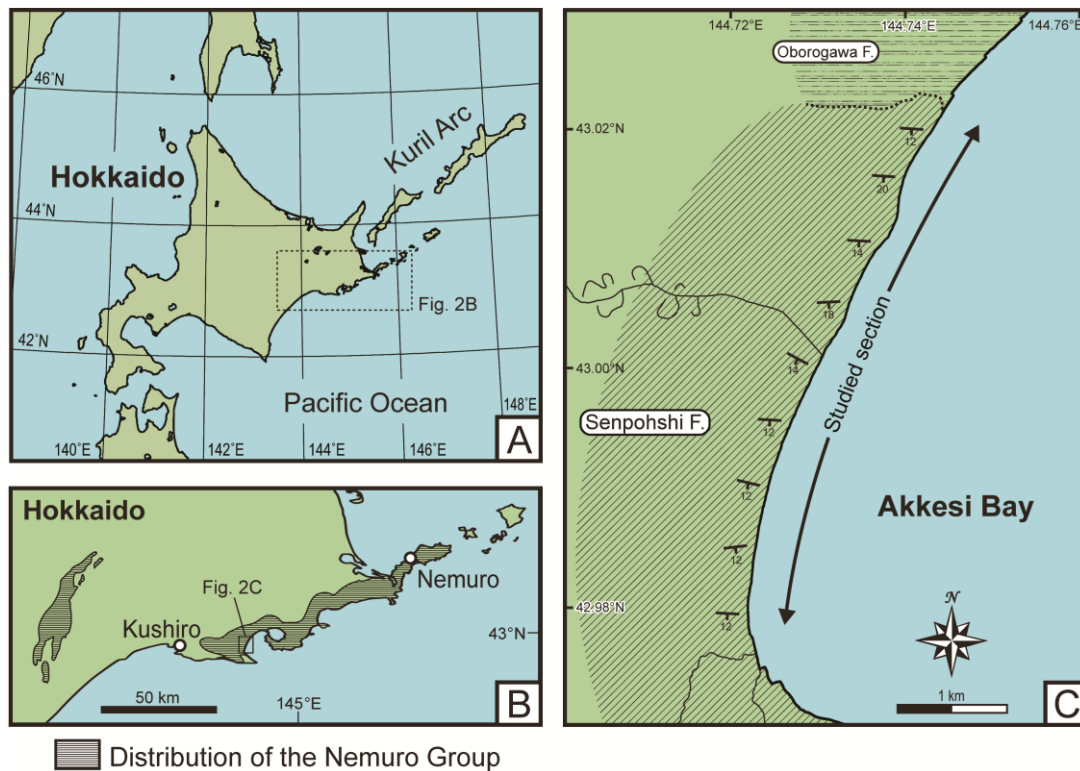
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152 **Fig. 1.** Paleogeographic map of the late Maastrichtian (68 Ma; modified after Hay et al., 1999,
153 https://www.odsn.de/odsn/services/paleomap/adv_map.html) showing the location of the sections
154 used in this study. SN: Senpohshi Formation of the Nemuro Group, northern Japan; NB: Naiba section,
155 Sakhalin, eastern Russia; GB: Bottaccione Gorge and the Contessa Highway sections at Gubbio, Italy;
156 ZM: Zumaia section, Basque country, Northern Spain; TR: Aguja, Javelina, and Black Peaks
157 Formations in the Tornillo Basin, Texas, US; CT: lower Cantwell Formation, Alaska, US. Numbers
158 indicate DSDP, ODP, and IODP sites (DSDP Site 525A, South Atlantic Ocean; ODP Site 762C, Indian
159 Ocean; ODP Site 1210B, equatorial Pacific Ocean; IODP Site U1403, Newfoundland Margin).

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■ Distribution of the Nemuro Group

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Fig. 2. Index maps showing (A) the location of Hokkaido and (B) the distribution of the Nemuro

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Group, and (C) location of the studied section.

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165 2. Geological setting and chronology

166 2.1. Geological setting

167

The Senpohshi Formation is a part of the Cretaceous–Paleogene Nemuro Group,

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which is exposed in eastern Hokkaido, northern Japan (Fig. 2). The Nemuro Group is

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mainly composed of hemipelagic mudstones and sediment gravity flow deposits, such as

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turbidites and submarine slumps deposits (Kiminami, 1978; Naruse, 2003). The Group is

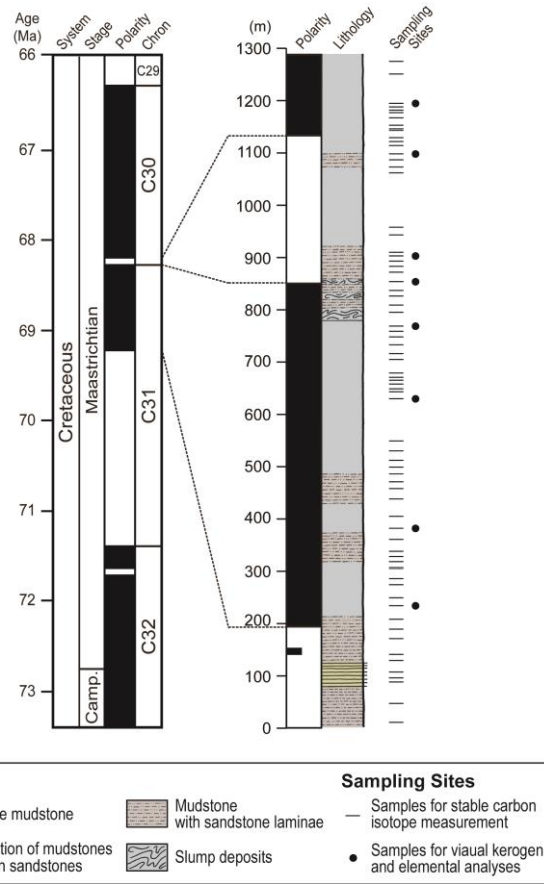
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interpreted to represent deposits of the forearc basin located adjacent to the Paleo-Kuril

172 arc (Kiminami, 1983; Kimura, 1994). Plate tectonic reconstruction suggested the basin
173 located in the North Pacific region, though the paleolatitude during the Late Cretaceous
174 is still controversial (Kimura, 1994; Bazhenov and Burtman, 1994; Fujiwara et al., 1995;
175 Bazhenov et al., 2001; Nifuku et al., 2009; Katagiri et al., 2020).

176 The Senpohshi Formation is widely exposed along the western coast of Akkeshi Bay,
177 eastern Hokkaido (Fig. 2C). It conformably overlies the Oborogawa Formation and is
178 unconformably overlain by the Shiomi Formation (Asano, 1962; Okada et al., 1987).
179 Bedding within the formation is homoclinal, dipping 10°–20° to the south. The thickness
180 of the formation exceeds 1,270 m (the upper boundary is not exposed). No large-scale
181 faults or tectonic folding are recognized. The formation mainly consists of weakly
182 bioturbated, dark grey mudstone layers occasionally intercalated with sandstone laminae
183 or thin beds of sandstones (~1.0 cm), which are interpreted as sediment gravity flow
184 deposits (Fig. 3). Thin slump deposits occur locally. These sedimentary features suggest
185 that the formation was deposited at the base of a submarine slope (Naruse, 2003).

186



187

188 **Fig. 3.** Lithology and magnetostratigraphy (after Nifuku et al., 2009) of the Senpohshi Formation.

189 Correlation with the astronomically calibrated magnetostratigraphic time scale (Option 2 of Husson et

190 al., 2011) is shown. Stratigraphic position of the sampling sites is also indicated in the figure.

191

192 2.2. Chronology

193 The Senpohshi Formation is assigned to the Maastrichtian based on biostratigraphy

194 and magnetostratigraphy. Maastrichtian index fossils have been found in this formation.

195 Naruse et al. (2000) and Nifuku et al. (2009) reported the ammonite *Pachydiscus*

196 *flexuosus* from the lower part of the formation. This species is commonly found in

197 Maastrichtian successions in southern Sakhalin and Hokkaido (Maeda and Shigeta, 2005;
198 Maeda et al., 2005). Shigeta et al. (2015) reported an ammonite fossil assemblage from
199 the same section including *Gaudryceras makarovense*, *Gaudryceras* cf. *seymouriense*,
200 *Anagaudryceras matsumotoi*, and *Diplomoceras* cf. *notabile* fossils, which are indicative
201 of the mid to upper Maastrichtian. Moreover, Nifuku et al. (2009) reported the occurrence
202 of “*Inoceramus*” *awajiensis*, a known Maastrichtian index fossil of the northwest Pacific
203 (Toshimitsu et al., 1995), in the middle part of the formation. Okada et al. (1987) reported
204 the occurrence of the calcareous nannofossil *Nephrolithus frequens*, a known global index
205 fossil of the upper Maastrichtian (Burnett, 1998), in the upper part of the formation.
206 Moreover, the planktonic foraminifera *Globotruncanella petaloidea*, also an index fossil
207 of the Maastrichtian (Caron, 1985), has been reported from the upper part of the formation
208 (Yamada, 1984).

209 Nifuku et al. (2009) established the magnetostratigraphy of the formation, identifying
210 four magnetozones. Considering the relationship between the biostratigraphy and
211 magnetostratigraphy of the formation, the magnetozones were correlated with polarity
212 chrons C31r–C30n, corresponding to the upper part of the lower Maastrichtian to the
213 upper Maastrichtian (Fig. 3, Table 1).

214 **Table 1.** Stratigraphic positions of polarity chron boundaries in the Senpohshi Formation (Nifuku et
215 al., 2009).

Polarity Chron Boundary	Stratigraphic height (m)
C30n/C30r	1118
C30r/C31n	841
C31n/C31r	190

216

217 **3. Materials and methods**

218 Samples were collected from 72 horizons within the Senpohshi Formation at
219 stratigraphic intervals of 2–104 m (average 18 m), depending on exposure (Fig. 3). All
220 samples were taken from hemipelagic mudstone. Turbidites and slump deposits were
221 excluded from sampling and analysis.

222 To investigate the content of the sedimentary organic matter (SOM) and to evaluate
223 the degree of thermal maturation, kerogen samples were extracted from eight mudstone
224 samples. First, moderately crushed samples were immersed repeatedly in HCl and HF
225 and then treated with a surfactant. Processed samples were then centrifuged using ZnCl₂
226 solution (specific gravity, 2.00) to extract the kerogen. To obtain estimates of the SOM
227 content, the extracted kerogen samples were observed under transmitted and fluorescent
228 (blue UV) light following a modified version of the classification scheme for organic
229 particles proposed by Tyson (1995); in our study, amorphous organic matter was divided

230 into nonfluorescent amorphous organic matter (NFA), weakly fluorescent amorphous
231 organic matter (WFA), and fluorescent amorphous organic matter (FA), based on
232 fluorescence properties. The relative abundance of each type was determined by counting
233 500 points at intervals of 100 μm under transmitted and fluorescent light. To evaluate the
234 degree of thermal diagenesis, we determined the atomic hydrogen/carbon ratio (H/C ratio)
235 of the bulk extracted kerogen samples using a Finnigan FlashEA 1112 elemental analyzer.

236 Stable carbon isotopic ratios of the bulk SOM and the total organic carbon (TOC)
237 content were analyzed from the mudstone samples. Powdered mudstone samples were
238 treated with HCl to remove carbonate minerals before analysis. The stable carbon isotope
239 ratios and TOC contents of the treated bulk mudstone samples were measured using a
240 mass spectrometer coupled with an elemental analyzer (Finnigan FlashEA 1112,
241 ConFloIII, and DELTA Plus Advantage). Stable carbon isotopic ratios were expressed in
242 δ -notation relative to the PDB standard:

$$243 \quad \delta^{13}\text{C} (\text{‰}) = (\text{R}_{\text{sample}}/\text{R}_{\text{standard}} - 1) \times 1000 \quad (1)$$

244 where $\text{R} = {}^{13}\text{C}/{}^{12}\text{C}$. L-Histidine (SHOKO Science Co., Ltd.) was used as an internal
245 laboratory standard and the stable carbon isotopic composition was -10.19‰ . Repeated
246 analysis of the laboratory standard indicated an overall uncertainty within $\pm 0.25\text{‰}$ (1σ).

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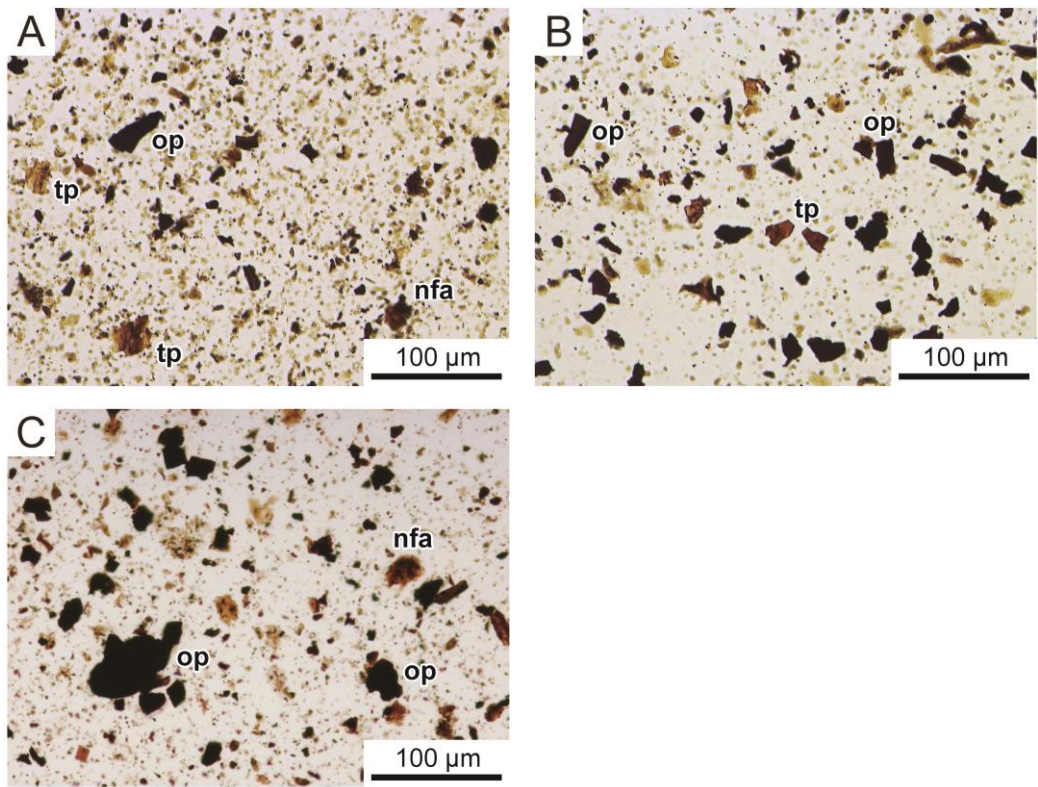
250 **4. Results**

251 *4.1. Constituents and H/C ratio of SOM*

252 Microscopic observations revealed that the SOM within the Senpohshi Formation is
253 dominated by phytoclasts with a minor amount of amorphous organic matter (Figs. 4 and
254 5, Table 2). These two elements made up more than 99% of the SOM in each sample.
255 Phytoclasts constituted 78.4%–98.6% of the SOM and were classified predominantly as
256 translucent phytoclasts without a definitive biostructure (44.4%–62.4%) and opaque
257 phytoclasts (31.2%–54.2%). Translucent phytoclasts with a definitive biostructure are
258 rare, and they are dominantly represented by cuticles. Amorphous organic matter
259 constituted 1.4%–21.4% of the SOM. The NFA was the dominant type of amorphous
260 organic matter, whereas there were very small amounts of WFA and FA. Palynomorphs
261 were rare in the SOM and accounted for less than 0.6% of the SOM.

262 The average atomic H/C value of the extracted kerogen samples was 0.64 and ranged
263 from 0.47 to 0.81 (Table 3).

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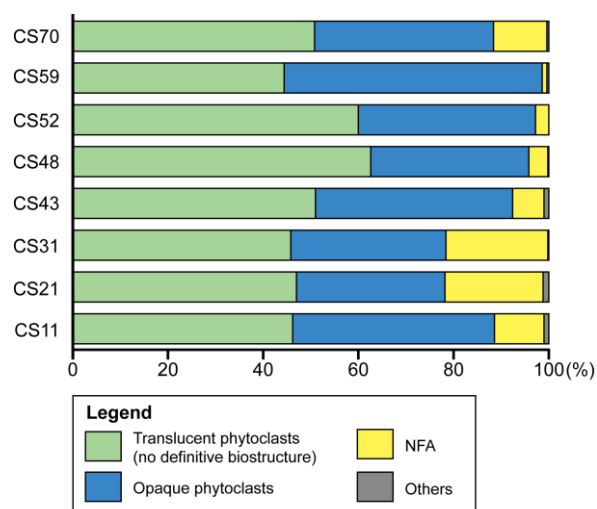
Fig. 4. Photomicrographs of kerogen extracted from selected samples: (A) CS21, (B) CS52, and (C)

267

CS59. tp: translucent phytoclasts; op: opaque phytoclasts; nfa: non-fluorescent amorphous organic

268

matter. Note that these three components dominate the SOM in each sample.



269

270

Fig. 5. Relative abundance (%) of various constituents of SOM extracted from eight selected samples.

271

272 **Table 2.** Relative abundance (%) of various constituents of SOM extracted from eight selected

273 samples.

Sample	Stratigraphic height (m)	Phytoclast			Amorphous organic matter			Palytomorph	
		Opaque	Translucent		NFA	WFA	FA	Pollen/Spore Grains	Phytoplankton
			No definitive biostructure	Definitive biostructure					
CS70	1195	37.6	50.8	-	11.2	-	-	0.4	-
CS59	1098	54.2	44.4	-	1.0	-	0.4	-	-
CS52	903	37.2	60.0	-	2.8	-	-	-	-
CS48	854	33.2	62.6	-	4.0	-	-	0.2	-
CS43	769	41.4	51.0	0.4	6.6	-	-	0.6	-
CS31	630	32.6	45.8	-	21.4	-	-	0.2	-
CS21	382	31.2	47.0	0.6	20.6	0.4	-	-	0.2
CS11	234	42.4	46.2	0.2	10.4	-	0.6	0.2	-

275 **Table 3.** Elemental compositions of kerogen extracted from eight selected samples.

Sample	Stratigraphic height (m)	TOC (wt%)	TH (wt%)	H/C (atomic ratio)
CS70	1195	73.26	3.74	0.61
CS59	1098	72.80	3.93	0.65
CS52	903	72.67	3.81	0.63
CS48	854	72.69	3.79	0.63
CS43	769	71.94	2.80	0.47
CS31	630	65.58	3.93	0.72
CS21	382	63.51	3.02	0.57
CS11	234	71.45	4.84	0.81

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277 *4.2. TOC content and stable carbon isotope ratios*

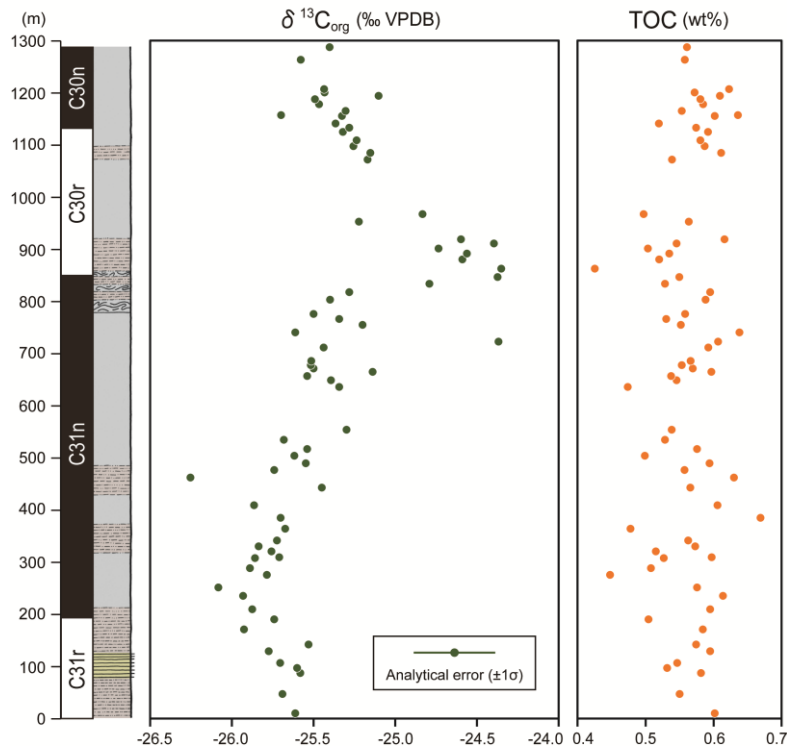
278 The average TOC content was 0.52 wt% and ranged from 0.43 to 0.67 wt% (Table

279 4). Stratigraphic variations in the TOC content were relatively minor, and most values

280 were 0.5–0.6 wt% for the Senpohshi Formation (Fig. 6).

Table 4. TOC content and $\delta^{13}\text{C}_{\text{org}}$ values obtained for the Senpohshi Formation.

Sample	Stratigraphic height (m)	Age (Ma)	TOC (wt%)	$\delta^{13}\text{C}_{\text{org}}$ (‰)	Sample	Stratigraphic height (m)	Age (Ma)	TOC (wt%)	$\delta^{13}\text{C}_{\text{org}}$ (‰)
CS01	11	69.467	0.60	-25.6	CS37	679	68.544	0.57	-25.5
CS02	47	69.417	0.55	-25.7	CS38	705	68.508	0.59	-25.4
CS03	88	69.360	0.58	-25.6	CS39	716	68.493	0.61	-24.4
CS04	96	69.349	0.53	-25.6	CS40	733	68.469	0.64	-25.6
CS05	107	69.334	0.55	-25.7	CS41	748	68.448	0.55	-25.2
CS06	129	69.304	0.59	-25.8	CS42	759	68.433	0.53	-25.3
CS07	141	69.287	0.57	-25.5	CS43	769	68.419	0.56	-25.5
CS08	171	69.246	0.58	-25.9	CS44	795	68.384	0.59	-25.4
CS09	189	69.221	0.50	-25.7	CS45	809	68.364	0.59	-25.3
CS10	208	69.194	0.60	-25.9	CS46	826	68.341	0.53	-24.8
CS11	234	69.159	0.61	-25.9	CS47	837	68.326	0.55	-24.4
CS12	249	69.138	0.58	-26.1	CS48	854	68.314	0.43	-24.4
CS13	274	69.103	0.45	-25.8	CS49	872	68.307	0.52	-24.6
CS14	286	69.087	0.51	-25.9	CS50	883	68.302	0.53	-24.6
CS15	305	69.060	0.53	-25.9	CS51	893	68.297	0.50	-24.7
CS16	307	69.058	0.60	-25.7	CS52	903	68.293	0.55	-24.4
CS17	318	69.042	0.52	-25.8	CS53	910	68.290	0.62	-24.6
CS18	328	69.029	0.57	-25.8	CS54	943	68.276	0.56	-25.2
CS19	338	69.015	0.56	-25.7	CS55	958	68.269	0.50	-24.8
CS20	361	68.983	0.48	-25.7	CS56	1062	68.224	0.54	-25.2
CS21	382	68.954	0.67	-25.7	CS57	1073	68.219	0.61	-25.2
CS22	405	68.922	0.61	-25.9	CS58	1087	68.213	0.59	-25.3
CS23	438	68.877	0.57	-25.5	CS59	1098	68.208	0.58	-25.2
CS24	458	68.849	0.63	-26.3	CS60	1114	68.202	0.59	-25.3
CS25	471	68.831	0.56	-25.7	CS61	1122	68.198	0.57	-25.3
CS26	486	68.810	0.59	-25.5	CS62	1129	68.195	0.52	-25.4
CS27	499	68.792	0.50	-25.6	CS63	1143	68.189	0.60	-25.3
CS28	512	68.774	0.58	-25.5	CS64	1146	68.188	0.64	-25.7
CS29	530	68.750	0.53	-25.7	CS65	1153	68.185	0.55	-25.3
CS30	549	68.723	0.54	-25.3	CS66	1167	68.179	0.58	-25.5
CS31	630	68.611	0.47	-25.3	CS67	1177	68.174	0.58	-25.5
CS32	643	68.594	0.55	-25.4	CS68	1182	68.172	0.61	-25.1
CS33	649	68.585	0.54	-25.5	CS69	1188	68.169	0.57	-25.4
CS34	658	68.573	0.60	-25.1	CS70	1195	68.166	0.62	-25.4
CS35	665	68.563	0.57	-25.5	CS71	1251	68.142	0.56	-25.6
CS36	671	68.555	0.55	-25.5	CS72	1275	68.132	0.56	-25.4



283

284 **Fig. 6.** Stratigraphic variation of stable carbon isotope values of bulk SOM ($\delta^{13}\text{C}_{\text{org}}$) and TOC content
 285 within the Senpohshi Formation. Error bar shows an overall uncertainty ($\pm 0.25\%$; 1σ) indicated by
 286 repeated analysis of the laboratory standard.

287

288 The average stable carbon isotope value of the bulk SOM obtained from the
 289 Senpohshi Formation was -25.4% and ranged from -26.3 to -24.4% (Table 4). The
 290 stratigraphic profile compiled for the measured $\delta^{13}\text{C}_{\text{org}}$ is shown in Fig. 6. From the top
 291 of C31r to the uppermost part of C31n, $\delta^{13}\text{C}_{\text{org}}$ values are almost constant ($\sim -25.5\%$). A
 292 small negative trough appeared at the bottom of C31n ($\sim 0.5\%$ in amplitude), but the
 293 amplitude of this feature was within measurement uncertainty. There is an apparent

294 negative excursion in the middle of C31n (~0.8‰ in amplitude), which is represented by
295 a single measurement point. Following this interval of approximately constant values, the
296 $\delta^{13}\text{C}_{\text{org}}$ profile showed a distinct positive excursion at the C30r/C31n boundary (from ~-
297 25.5 to ~-24.5‰). The stable carbon isotope values declined across C30r before
298 recovering to background values (~-25.5‰) at the bottom of C30n.

299

300 **5. Discussion**

301 *5.1. Origin and diagenesis of SOM*

302 The SOM within the Senpohshi Formation is dominated by phytoclasts with a minor
303 amount of NFA, which together account for more than 99% of the SOM (Fig. 5, Table 2).
304 Previous studies have proposed that phytoclasts originated from terrestrial higher plants
305 (Tyson, 1995), as did NFA (Sawada and Akiyama, 1994; Watanabe and Akiyama, 1998;
306 Omura and Hoyanagi, 2004). Sawada and Akiyama (1994) reached this conclusion based
307 on an analysis of a high NFA content sample from the Toarcian Shale, Yorkshire, England.
308 The authors reported that the $\delta^{13}\text{C}$ value of the sample was equivalent to the $\delta^{13}\text{C}$ values
309 of terrestrial higher plants, and the density of the SOM was similar to that of vitrinite.
310 Watanabe and Akiyama (1998) and Omura and Hoyanagi (2004) reached the same
311 conclusions based on analyses of carbon isotope values of high NFA content samples

312 from the Niigata sedimentary basin, central Japan. The authors showed that $\delta^{13}\text{C}$ values
313 of the SOM were equivalent to those of terrestrial higher plants, and they suggested that
314 the NFA originated from terrestrial higher plants. In the case of the Senpohshi Formation,
315 irrespective of the NFA content, stable carbon isotope values of the bulk SOM range from
316 -26.3 to -24.4‰ (Table 4), which coincides with the $\delta^{13}\text{C}$ values of terrestrial higher C3
317 plants (Deines, 1980; Jasper and Gagosian, 1990; Whelan and Thompson-Rizer, 1993).
318 These results indicate that the SOM from the Senpohshi Formation originated from
319 terrestrial higher plants.

320 The atomic H/C ratios obtained from the extracted kerogen samples from the
321 Senpohshi Formation were used to estimate the degree of thermal diagenesis. The $\delta^{13}\text{C}$
322 composition of woody material is modified from its original value via an isotopic
323 exchange reaction with carbonate during the graphitization stage of metamorphism
324 ($>400\text{ °C}$) (Dunn and Valley, 1992). In contrast, the $\delta^{13}\text{C}$ composition of woody material
325 is not significantly altered by hydrocarbon generation that occurs before metamorphism
326 (Deines, 1980; Teerman and Hwang, 1991; Whiticar, 1996). The atomic H/C ratios of the
327 kerogen samples from the Senpohshi Formation, which range from 0.47 to 0.81, indicate
328 that the coalification rank of the kerogen is largely below the anthracite stage, although
329 some kerogen samples could be classified to be of early anthracite stage (Table 3)

330 (Teichmüller and Teichmüller, 1979). Five of the eight extracted kerogen samples yielded
331 small numbers of pollen/spore grains, supporting our interpretation that the thermal
332 diagenesis of the kerogen resulted in the material below the anthracite stage (Table 2).
333 These observations indicate that the thermal diagenesis of the Senpohshi Formation was
334 less than that corresponding to the graphitization stage of metamorphism. Therefore, it is
335 suggested that the $\delta^{13}\text{C}$ values the SOM were not modified by thermal diagenesis.

336 To summarize, our analysis indicates that the $\delta^{13}\text{C}_{\text{org}}$ values obtained for the
337 Senpohshi Formation represent the original values of the carbon isotope composition of
338 terrestrial higher plants.

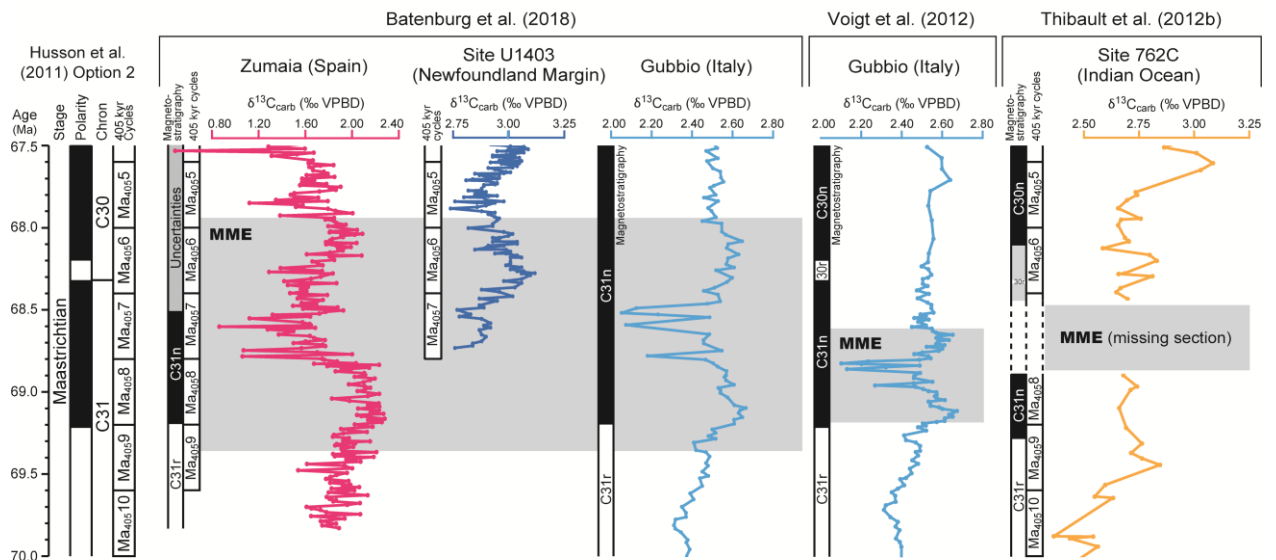
339

340 *5.2. Comparison of the carbon isotope stratigraphy with other sections*

341 The carbon isotope stratigraphy of the middle of the Maastrichtian is characterized
342 by the MME, which consists of two positive peaks separated by a negative trough with
343 short-term negative excursions (Batenburg et al., 2018; Jung et al., 2012; Voigt et al.,
344 2012). The three segments of the event are labelled MME1 to MME3 in ascending order
345 in this study, after the zonation by Voigt et al., (2012).

346 Previous research suggested two possibilities for age correlation of the MME (Fig.
347 7). Batenburg et al. (2018) correlated the event based on the cyclostratigraphy of the

348 Zumaia section in northern Spain and the IODP Site U1403 in the Newfoundland Margin.
349 They identified the event in the Maastrichtian 405 kyr cycles from the upper Ma₄₀₅₉ to
350 lower Ma₄₀₅₅, corresponding to 69.38 Ma through 67.87 Ma (~1,510 kyr in duration).
351 Another interpretation was proposed by Voigt et al. (2012) from the magnetostratigraphy
352 of the Gubbio section, Italy. They distinguished the event in the lower part of the polarity
353 chron C31n. It is correlated to 69.21 Ma through 68.63 Ma (~580 kyr in duration)
354 according to the astronomically calibrated Maastrichtian magnetostratigraphy by Husson
355 et al. (2011) (Option 2). Thibault et al. (2012b) suggested a similar view based on the
356 magnetostratigraphy and cyclostratigraphy of the ODP Site 762C in Indian Ocean. They
357 implied the event in the lower C31n (upper Ma₄₀₅₈ through upper Ma₄₀₅₇); however, it
358 was not confirmed by carbon isotope data because they interpreted the event within a
359 missing section.
360



361

362 **Fig. 7.** Comparison of the age model for the MME after Thibault et al. (2012b), Voigt et al. (2012),
 363 and Batenburg et al. (2018). Ages were adjusted to the astronomically calibrated Maastrichtian time
 364 scale by Husson et al. (2011) (Option 2).

365

366 Comparing the two hypothesis described above, the age model by Batenburg et al.
 367 (2018) provides better agreement among the carbon isotope profiles from multiple
 368 sections. Reliable stratigraphic correlation has been established between the Zumaia
 369 section and Site U1403 based on cyclostratigraphy, and the carbon isotope profiles during
 370 the MME are consistent each other. Moreover, the positive peaks recognized in the
 371 Ma₄₀₅₆ at the Site 762C can be correlated to the upper positive peak of the event in the
 372 Zumaia section and Site U1403, although Thibault et al. (2012b) originally placed the
 373 peaks above the MME. Batenburg et al. (2018) implied uncertainty in the

374 magnetostratigraphy of the Gubbio section, and they made a stratigraphic correlation by
375 using tie-points in the carbon isotope curve. The results showed a good agreement of the
376 carbon isotope profile from the Gubbio section with the other sections. The above
377 observations suggest the age correlation by the Batenburg et al. (2018) is more
378 appropriate because of better consistency of carbon isotope curves among multiple
379 sections.

380 Therefore, this study follows the age model for the MME proposed by Batenburg et
381 al. (2018). It is indicated that the carbon isotope stratigraphy of the Senpohshi Formation
382 covers the stratigraphic level right below the MME through the uppermost part of the
383 event (Figs. 6 and 7). Based on this age model, the carbon isotope profile from the
384 Senpohshi Formation was correlated to other sections (Fig. 1). Six sections representing
385 marine carbon isotope records were selected, namely the Gubbio section in Italy (Voigt
386 et al., 2012), the Zumaia section in northern Spain (Batenburg et al., 2012), Equatorial
387 Pacific ODP Site 1210B (Jung et al., 2012), Indian Ocean ODP Site 762C (Thibault et al.,
388 2012b), South Atlantic DSDP Site 525A (Li and Keller, 1998), and Newfoundland
389 Margin IODP Site U1403 (Batenburg et al., 2018). Bulk carbonate samples were analyzed
390 at these sections except for Site 525A, where they examined single species foraminifera
391 samples. The terrestrial carbon isotope records had been obtained from the Naiba section,

392 Sakhalin, Eastern Russia (Hasegawa et al., 2003), the lower Cantwell Formation, Alaska,
 393 US (Salazar-Jaramillo et al., 2016), and the Aguja, Javelina, and Black Peaks Formations
 394 in the Tornillo Basin, Texas, US (Nordt et al., 2003). The carbon isotope profiles were
 395 derived from terrestrial organic matter in the Naiba section and Cantwell Formation.
 396 Pedogenic carbonate was investigated in the Tornillo Basin. Ages of all the selected
 397 sections were adjusted to the astronomically calibrated Maastrichtian time scale by
 398 Husson et al. (2011) (Option 2). Stratigraphic correlations were made after
 399 cyclostratigraphy (Zumaia, Site U1403, and Site 762C), magnetostratigraphy (Senpohshi
 400 Formation, Site 525A, and Naiba) (Table 5), or tie-points in the carbon isotope profile
 401 (Gubbio, Site 1210B, Cantwell Formation, and Tornillo Basin). The tie-points were after
 402 Voigt et al. (2012) and Batenburg et al. (2018) (Table 6), and the carbon isotope curves
 403 from the four sections were correlated to the Zumaia section. The results of the
 404 comparison of the carbon isotope stratigraphy of the Senpohshi Formation with other
 405 sections are shown in Fig. 8.

406

407 **Table 5.** Ages of the K-Pg boundary and the Maastrichtian polarity chron boundaries after Option 2
 408 proposed by Husson et al. (2011).

Event	Age (Ma)
K/PgB	66 ± 0.07

C29r/C30n	66.3 ± 0.08
C30n/C30r	68.2 ± 0.08
C30r/C31n	68.32 ± 0.07
C31n/C31r	69.22 ± 0.07
C31r/C32n1n	71.4 ± 0.08

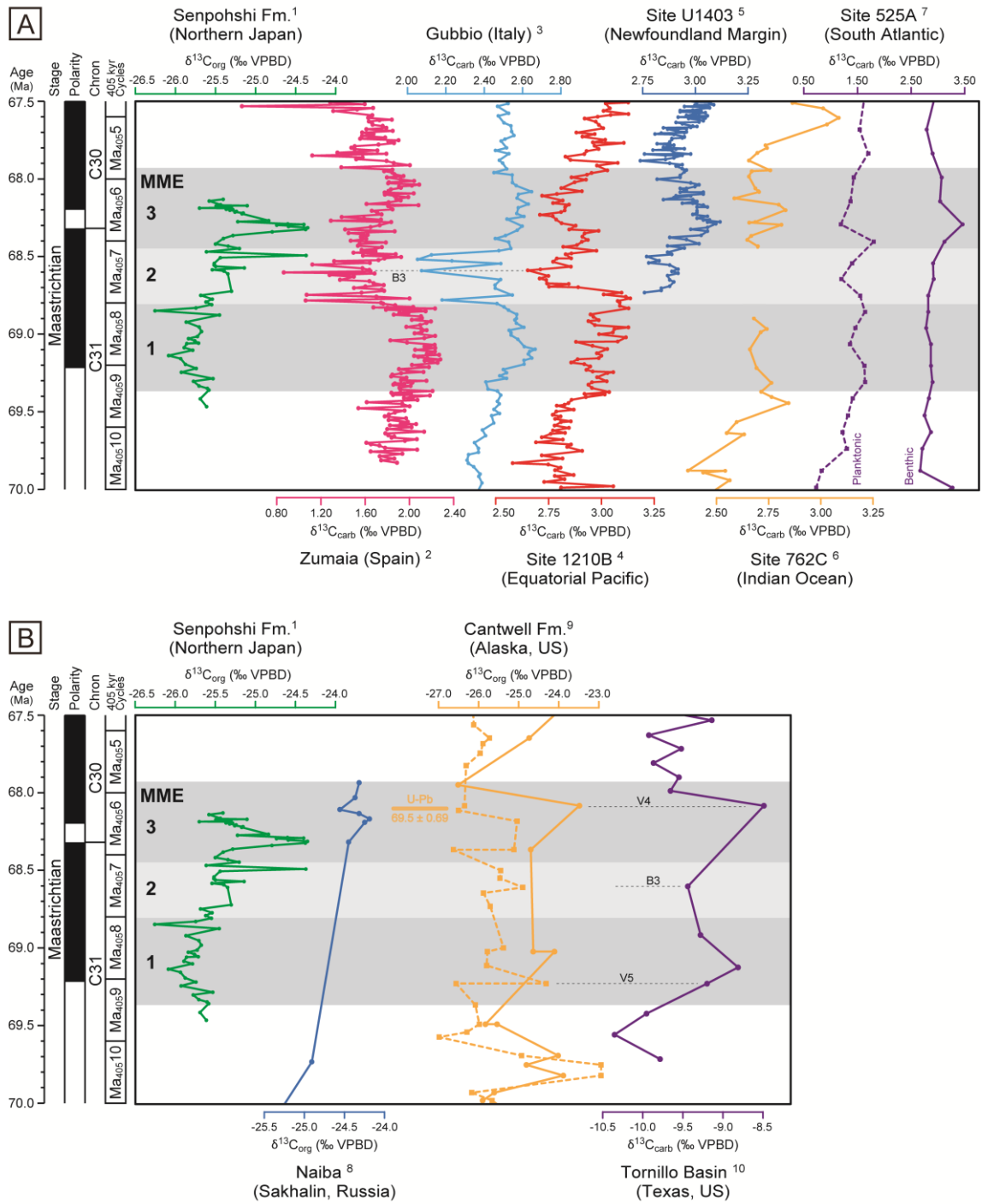
409

410 **Table 6.** Tie-points for correlation of stable carbon isotope profiles, which are defined by Voigt et al.

411 (2012) and Batenburg et al. (2018).

Tie-point	Description	Reference
V2	$\delta^{13}\text{C}$ minimum a short distance below the C29r/C30n boundary	Voigt et al. (2012)
V3a	$\delta^{13}\text{C}$ maximum within C30n, transition towards long-term $\delta^{13}\text{C}$ decline	Voigt et al. (2012)
V4	Upper $\delta^{13}\text{C}$ max of the mid-Maastrichtian $\delta^{13}\text{C}$ plateau	Voigt et al. (2012)
B3	the $\delta^{13}\text{C}$ minimum within a broad plateau of high $\delta^{13}\text{C}$ values in the mid-Maastrichtian, within C31n	Batenburg et al. (2018)
V5	Inflection towards the lower $\delta^{13}\text{C}$ maximum of the mid-Maastrichtian $\delta^{13}\text{C}$ plateau, a short distance above the C31n/C31r boundary	Voigt et al. (2012)
V6	Inflection towards the long-term Maastrichtian $\delta^{13}\text{C}$ rise in C31r, top of the CMBE, top occurrence of <i>Tranolithus orionatus</i>	Voigt et al. (2012)

412



413

414 **Fig. 8.** Correlation of stable carbon isotope stratigraphy of the Senpohshi Formation with other sections.

415 (A) Correlation with carbon isotope profiles obtained from marine materials. (1) Senpohshi Formation

416 of the Nemuro Group, northern Japan (this study); (2) Zumaia section, Basque country, Northern Spain

417 (Batenburg et al., 2012); (3) Bottaccione Gorge and the Contessa Highway sections at Gubbio, Italy
418 (Voigt et al., 2012); (4) ODP Site 1210B, Equatorial Pacific (Jung et al., 2012); (5) IODP Site U1403,
419 Newfoundland Margin (Batenburg et al., 2018); (6) ODP Site 762C, Indian Ocean (Thibault et al.,
420 2012b); (7) DSDP Site 525A, South Atlantic (Li and Keller, 1998). Bulk carbonate was analyzed at
421 these sites except for the DSDP Site 525A, where they measured single-species planktonic
422 (*Rugoglobigerina rugosa*) and benthic foraminifera (*Anomalinoidea acufa*). (B) Correlation with
423 carbon isotope profiles obtained from terrestrial materials. (8) Naiba section, Sakhalin, eastern Russia
424 (Hasegawa et al., 2003). Bulk sedimentary organic matter was analyzed. (9) lower Cantwell Formation,
425 Alaska, US (Salazar-Jaramillo et al., 2016). Stable carbon isotope compositions were obtained from
426 bulk sedimentary organic matter and fossil wood. (10) Aguja, Javelina, and Black Peaks Formations
427 in the Tornillo Basin, Texas, US (Nordt et al., 2003). They measured pedogenic carbonate.
428 Astronomically calibrated magnetostratigraphy and the 405 kyr eccentricity cycles are after Option 2
429 of Husson et al. (2011). The light and dark gray shaded intervals correspond to the MME and its
430 constituting segments after Voigt et al. (2012).

431

432 The MME is widely recognized in the marine carbon isotope records (Fig. 8A) (e.g.,
433 Voigt et al., 2010; Thibault et al., 2012a; Voigt et al., 2012; Jung et al., 2012; Wendler et
434 al., 2011; Wendler, 2013; Batenburg et al., 2018). Recently acquired high-resolution data

435 from Zumaia, Gubbio, Site 1210B, and Site U1403 provided detailed features of the
436 carbon isotope profile during the event (Batenburg et al., 2012; Jung et al., 2012; Voigt
437 et al., 2012; Batenburg et al., 2018). All the sections conformably show the three segments
438 of the event, which consist of two positive peaks intercalated by a negative trough with
439 short-term negative excursions.

440 In contrast, there remains uncertainty in identification of the MME and its
441 constituting segment in the terrestrial records, because of the limitation of data resolution
442 and age control. The presence of the MME was suggested by the terrestrial records from
443 the lower Cantwell Formation and Tornillo Basin (Fig. 8B) (Nordt et al., 2003; Salazar-
444 Jaramillo et al., 2016). The identification of the event largely relied on the features of the
445 carbon isotope curves. Positive peaks in the two sections showed an apparent similarity
446 to the positive peaks in the segments MME1 and MME3 recognized in the marine records.
447 However, their presence is less confident because each peak is represented by a single
448 measurement point. Moreover, the two sections lack sufficient age control to correlate the
449 carbon isotope profiles to the MME. A bentonite U-Pb zircon age of 69.5 ± 0.69 Ma was
450 indicated near the second positive peak in the lower Cantwell Formation (Salazar-
451 Jaramillo et al., 2016). However, the measured age is not fully consistent with the age of
452 the segment MME3 (68.45 Ma – 67.86 Ma) after the age model by Batenburg et al. (2018).

453 There is no stratigraphic marker in the Maastrichtian interval in the Tornillo Basin, even
454 though the KPg boundary had been indicated by a weak iridium anomaly in the studied
455 section (Lehman, 1990; Nordt et al., 2003).

456 Therefore, higher resolution data from the Senpohshi Formation with age control by
457 magnetostratigraphy has significance in the recognition of the MME in the terrestrial
458 record. The terrestrial record of the MME in the Senpohshi Formation suggests both
459 similarity and difference to the marine carbon isotope records (Fig. 8A). Both records
460 show a positive peak in the segment MME3. The Senpohshi Formation shows a positive
461 excursion at the C30r/C31n boundary which is consistent with the peak at the same
462 stratigraphic level at the Sites 1210B and U1403. The excursion can also be correlated
463 with a small positive peak near the C30r/C31n boundary at the Sites 525A and 762C,
464 though it is represented by a single measurement point (Li and Keller, 1998; Thibault et
465 al., 2012b). In the segment MME2, both Senpohshi Formation and marine records show
466 negative shift of the carbon isotope value from the overlying segment. The marine records
467 also illustrate multiple short-term negative excursions superimposed on the negative
468 trough; however, they are not clear in the Senpohshi Formation. A negative peak at 68.85
469 Ma could be correlated to one of the short-term negative excursions in other sections,
470 though there is uncertainty in the recognition of the peak because it is represented by a

471 single measurement point. Lower resolution of the data in the segment MME2 of the
472 Senpohshi Formation possibly caused the lack of short-term negative excursions in the
473 carbon isotope profile. In contrast, there is a clear discrepancy between the Senposhi
474 Formation and the other sections in the segment MME1. The marine records show a
475 positive peak in this interval; however, the Senpohshi Formation indicates a slight
476 negative shift of the carbon isotope values. The difference might suggest local factors
477 affecting carbon isotope values recorded in the Senpohshi Formation.

478 In summary, this study provides the first high-resolution terrestrial carbon isotope
479 record of the MME which is comparable to the high-resolution data from marine materials.
480 The results indicate the carbon isotopic signatures defined by the marine records are
481 recognized in the terrestrial materials, especially in the middle to upper part of the event
482 (segments MME2 and MME3). However, the terrestrial data of the Formation showed a
483 discrepancy from the marine records in the lower part of the event (segment MME1),
484 suggesting influence of local factors in the study area. We will discuss the factors in the
485 following section.

486

487 *5.3. Factors influencing carbon isotope profile recorded in the Senpohshi Formation*

488 The terrestrial organic matter-derived carbon isotope data of the Senpohshi
489 Formation showed a discrepancy from the marine records in the lower part of the MME
490 (Fig. 8A). The difference is seemingly caused by local factors of the Formation. There
491 are some potential factors that can affect carbon isotope values of the terrestrial organic
492 matter: (1) compositional changes in terrestrial organic matter, (2) reworking of older
493 sediment, and (3) changes in the hinterland environment.

494 Regarding the Factor 1, it is unlikely that compositional changes in terrestrial organic
495 matter preserved in sediment contributed to the carbon isotope profile of the Senpohshi
496 Formation because the composition of terrestrial organic matter did not change
497 significantly. The SOM in the Senpohshi Formation is dominated by opaque and
498 translucent phytoclasts and a minor amount of NFA (Fig. 6 and Table 2). The relative
499 abundance of the three macerals in the samples from the segment MME1 (kerogen
500 samples CS11 and CS21) was within the range of the abundances in the overlaying
501 segments (kerogen samples CS31, CS43, CS48, CS52, CS59, and CS70). These
502 observations indicate that a change in terrestrial organic matter composition is unlikely
503 to cause the deviation of the carbon isotope profile.

504 It is also difficult to explain the discrepancy through reworking of terrestrial organic
505 matter from older sediment (Factor 2). Although the interval contains thin layers of
506 sediment-gravity-flow deposits and thin slump deposits (Fig. 6), sampling sites of
507 specimens were chosen to avoid gravity flow deposits. The upper interval of the
508 Formation also contains gravity flow deposits. However, the data from the upper interval
509 shows a positive carbon isotope excursion which can be globally correlated to the positive
510 peak in other sections (Fig. 8). The data from the upper interval indicates the sampling
511 method avoided the influence of reworking materials.

512 A possible cause for the discrepancy is a changing hinterland environment (Factor
513 3). Stable carbon isotope values of terrestrial plants are affected by abiotic factors of the
514 environment of these plants, such as relative humidity, temperature, precipitation, soil
515 moisture, and atmospheric CO₂ concentration (e.g., Farquhar et al., 1980; Ramesh et al.,
516 1986; Körner et al., 1988; Leavitt, 1993; Feng and Epstein, 1995; Lipp et al., 1996;
517 Pendall et al., 1999; Schleser et al., 1999; Edwards et al., 2000). The carbon isotope
518 profiles also reflect plant species variation (Lloyd and Farquhar, 1994; Chikaraishi and
519 Naraoka, 2003), which can be caused by a changing hinterland environment. In addition,
520 a large-scale shift of the sediment source area may change plant species composition
521 delivered to the depositional sites. However, it is not likely in the study area because the

522 sediments were sourced from restricted area of the adjacent island arc (Kiminami, 1983;
523 Kimura, 1994; Fujiwara et al., 1995). Therefore, it is suggested that changes in the
524 hinterland environment and floral assemblage affected the carbon isotope values recorded
525 in the Senpohshi Formation during the early phase of the MME.

526 The discrepancy of the carbon isotope profiles among the Senpohshi Formation and
527 the other sections suggests spatial variation of paleoenvironment during the early phase
528 of the MME. Additional paleoenvironmental research should illuminate the nature and
529 cause of the local variation of the terrestrial environment in the North Pacific region. This
530 study provides new insight into environmental changes during the late Maastrichtian by
531 establishing a detailed carbon isotope record of terrestrial materials.

532

533 **6. Conclusions**

534 High-resolution stable carbon isotope stratigraphy of terrestrial organic matter was
535 established for the upper Maastrichtian Senpohshi Formation of the Nemuro Group of
536 eastern Hokkaido, northern Japan. Geochemical and petrographic analyses of bulk SOM
537 and extracted kerogen samples led to the following conclusions:

538 (1) The SOM within the Senpohshi Formation is dominated by phytoclasts and a minor
539 amount of NFA. Because both phytoclasts and NFA are interpreted to have originated

540 from terrestrial higher plants, the SOM within the Senpohshi Formation is inferred to
541 have the same origin.

542 (2) The atomic H/C ratios obtained for the extracted kerogen samples range from 0.47 to
543 0.81, indicating a coalification rank at the anthracite stage or below. This finding
544 reveals that $\delta^{13}\text{C}_{\text{org}}$ values obtained from the Senpohshi Formation were not
545 significantly varied by thermal diagenesis. Therefore, the $\delta^{13}\text{C}_{\text{org}}$ values obtained for
546 the Senpohshi Formation represent the original values of the carbon isotope
547 composition of terrestrial higher plants.

548 (3) The stable carbon isotope profile reconstructed for the Formation provides the first
549 high-resolution terrestrial record of the MME, which is comparable to high-resolution
550 marine carbon isotope data. The carbon isotopic signatures defined by the marine
551 records are recognized in the terrestrial data from the Formation, especially in middle
552 to upper part of the event. However, the terrestrial record showed a discrepancy from
553 the marine data in the lower part of the MME, suggesting local variation of the
554 hinterland environment in the North Pacific region. This study provides new insight
555 into environmental changes during the late Maastrichtian by establishing a detailed
556 carbon isotope record of terrestrial materials.

557

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573

574 **8. References**

- 575 Arthur, M.A., Fischer, A.G., 1977. Upper Cretaceous-Paleocene magnetic stratigraphy at
576 Gubbio, Italy. I. Lithostratigraphy and sedimentology. Geological Society of
577 America Bulletin 88, 367–371.
- 578 Asano, K., 1962. Japanese Paleogene from the view-point of foraminifera with
579 descriptions of several new species. Contributions Institute of Geology and
580 Paleontology, Tohoku University 57, 1–32. (in Japanese, with English Abstract)
- 581 Barrera, E., Huber, B.T., 1990. Evolution of Antarctic waters during the Maastrichtian:
582 Foraminifer oxygen and carbon isotope ratios, Leg 113. Proceedings of the Ocean
583 Drilling Program, Scientific Results 113, 813–827.
- 584 Barrera, E., Savin, S.M., 1999. Evolution of late Campanian-Maastrichtian marine
585 climates and oceans. In: Barrera, E., Johnson, C.C. (Eds.), Evolution of the
586 Cretaceous ocean-climate system. Geological Society of America Special Paper 332,
587 p. 245–282.
- 588 Barrera, E., Savin, S.M., Thomas, E., Jones, C.E., 1997. Evidence for thermohaline-
589 circulation reversals controlled by sea-level change in the latest Cretaceous.
590 Geology 25, 715–718.

591 Batenburg, S. J., Sprovieri, M., Gale, A. S., Hilgen, F. J., Hüsing, S., Laskar, J., Liebrand,
592 D., Lirer, F., Orue-Etxebarria, X., Pelosi, N., Smit, J., 2012. Cyclostratigraphy and
593 astronomical tuning of the Late Maastrichtian at Zumaia (Basque country, Northern
594 Spain). *Earth and Planetary Science Letters*, 359, 264-278.

595 Batenburg, S. J., Friedrich, O., Moriya, K., Voigt, S., Cournède, C., Moebius, I., Blum, P.,
596 Bornemann, A., Fiebig, J., Hasegawa, T., Hull, P. M., Norris, R. D., Röhl, U., Sexton,
597 P. F., Westerhold, T. Wilson, P. A., the IODP Expedition 342 Scientists, 2018. Late
598 Maastrichtian carbon isotope stratigraphy and cyclostratigraphy of the
599 Newfoundland Margin (Site U1403, IODP Expedition 342). *Newsletters on*
600 *Stratigraphy*, 51, 245-260.

601 Bazhenov, M.L., Burtman, V.S., 1994. Upper Cretaceous paleomagnetic data from
602 Shikotan Island, Kuril Arc: Implications for plate kinematics. *Earth and Planetary*
603 *Science Letters* 122, 19–28.

604 Bazhenov, M.L., Zharov, A.E., Levashova, N.M., Kodama, K., Bragin, N.Y., Fedorov, P.I.,
605 Bragina, L.G. Lyapunov, S.M., 2001. Paleomagnetism of a Late Cretaceous island
606 arc complex from South Sakhalin, East Asia: Convergent boundaries far away from
607 the Asian continental margin? *Journal of Geophysical Research: Solid Earth*, 106,
608 19193-19205.

- 609 Bowman, V. C., Francis, J. E., Riding, J. B., 2013. Late Cretaceous winter sea ice in
610 Antarctica?. *Geology*, 41, 1227-1230.
- 611 Burnett, J.A., 1998. Upper Cretaceous, In: Bown, P.R. (Ed.), *Calcareous nannofossil*
612 *biostratigraphy*. Campman and Hall, London, p. 132–199.
- 613 Caron, M., 1985. Cretaceous planktic foraminifera. In: Bolli, H.M., Saunders, J.B., Perch-
614 Nielsen, K. (Eds.), *Plankton stratigraphy*. Cambridge University Press, Cambridge,
615 p. 17–86.
- 616 Chave, A.D., 1984. Lower Paleocene-Upper Cretaceous magnetostratigraphy, Sites 525,
617 527, 528, and 529, Deep Sea Drilling Project Leg 74. *Initial Reports of the Deep*
618 *Sea Drilling Project DSDP 74*, 525-531.
- 619 Chikaraishi, Y., Naraoka, H., 2003. Compound-specific δD – $\delta^{13}C$ analyses of n-alkanes
620 extracted from terrestrial and aquatic plants. *Phytochemistry*, 63, 361-371.
- 621 Corfield, R.M., Norris, R.D., 1996. Deep water circulation in the Paleocene ocean.
622 *Geological Society, London, Special Publications*, 101, 443–456.
- 623 Corfield, R.M., Cartlidge, J.E., Premoli-Silva, I., Housley, R.A., 1991. Oxygen and
624 carbon isotope stratigraphy of the Palaeogene and Cretaceous limestones in the
625 Bottaccione Gorge and the Contessa Highway sections, Umbria, Italy. *Terra Nova*,
626 3, 414–422.

- 627 Crampton, J.S., Schiøler, P., Roncaglia, L., 2006. Detection of Late Cretaceous eustatic
628 signatures using quantitative biostratigraphy. *Geological Society of America*
629 *Bulletin*, 118, 975-990.
- 630 Dameron, S.N., Leckie, R.M., Clark, K., MacLeod, K.G., Thomas, D.J., Lees, J.A., 2017.
631 Extinction, dissolution, and possible ocean acidification prior to the
632 Cretaceous/Paleogene (K/Pg) boundary in the tropical Pacific. *Palaeogeography,*
633 *Palaeoclimatology, Palaeoecology*, 485, 433-454.
- 634 Deines, P., 1980. The isotopic composition of reduced organic carbon. In: Frits, P., Fontes,
635 J.C. (Eds.), *Handbook of Environmental Isotope Geochemistry 1*. Elsevier,
636 Amsterdam, p. 329–406.
- 637 D'Hondt, S., Lindinger, M., 1994. A stable isotopic record of the Maastrichtian ocean-
638 climate system: South Atlantic DSDP site 528. *Palaeogeography, Palaeoclimatology,*
639 *Palaeoecology*, 112, 363–378.
- 640 Dickens, G.R., O'Neil, J.R., Rea, D.K. Owen, R.M., 1995. Dissociation of oceanic
641 methane hydrate as a cause of the carbon isotope excursion at the end of the
642 Paleocene. *Paleoceanography and Paleoclimatology*, 10, 965-971.

643 Donnadieu, Y., Pierrehumbert, R., Jacob, R., Fluteau, F., 2006. Modelling the primary
644 control of paleogeography on Cretaceous climate. *Earth and Planetary Science*
645 *Letters*, 248, 426-437.

646 Dunn, S.R., Valley, J.W., 1992. Calcite-graphite isotope thermometry: a test for
647 polymetamorphism in marble, Tudor gabbro aureole, Ontario, Canada. *Journal of*
648 *Metamorphic Geology* 10, 487–501.

649 Edwards, T.W.D., Graf, W., Trimborn, P., Stichler, W., Lipp, J., Payer, H.D., 2000. $\delta^{13}\text{C}$
650 response surface resolves humidity and temperature signals in trees. *Geochimica et*
651 *Cosmochimica Acta* 64, 161–167.

652 Falzoni, F., Petrizzo, M.R., Clarke, L.J., MacLeod, K.G., Jenkyns, H. C., 2016. Long-
653 term Late Cretaceous oxygen-and carbon-isotope trends and planktonic
654 foraminiferal turnover: A new record from the southern midlatitudes. *Bulletin*, 128,
655 1725-1735.

656 Farnsworth, A., Lunt, D.J., O'Brien, C.L., Foster, G.L., Inglis, G.N., Markwick, P.,
657 Pancost, R.D., Robinson, S.A., 2019. Climate sensitivity on geological timescales
658 controlled by nonlinear feedbacks and ocean circulation. *Geophysical Research*
659 *Letters*, 46, 9880-9889.

- 660 Farquhar, G. D., 1980. Carbon isotope discrimination by plants: Effects on carbon dioxide
661 concentration and temperature via the ratio of intercellular and atmospheric CO₂
662 concentrations, In: Pearman G.I. (Ed.), Carbon dioxide and climate: Australian
663 research. Australian Academy of Science, Canberra, p. 105–110.
- 664 Feng, X., Epstein, S., 1995. Carbon isotopes of trees from arid environments and
665 implications for reconstructing atmospheric CO₂ concentration. *Geochimica et*
666 *Cosmochimica Acta*, 59, 2599-2608.
- 667 Frank, T.D., Arthur, M.A., 1999. Tectonic forcings of Maastrichtian ocean-climate
668 evolution. *Paleoceanography* 14, 103–117.
- 669 Frank, T.D., Thomas, D.J., Leckie, R.M., Arthur, M.A., Bown, P.R., Jones, K., Lees, J.A.,
670 2005. The Maastrichtian record from Shatsky Rise (northwest Pacific): A tropical
671 perspective on global ecological and oceanographic changes. *Paleoceanography* 20,
672 PA1008.
- 673 Friedrich, O., Herrle, J.O., Wilson, P.A., Cooper, M.J., Erbacher, J., Hemleben, C., 2009.
674 Early Maastrichtian carbon cycle perturbation and cooling event: Implications from
675 the South Atlantic Ocean. *Paleoceanography*, 24, PA2211.
- 676 Fujiwara, Y., Kanamatsu, T., Nanayama, F., 1995. Tectonic evolution of two paleo arc-
677 trench systems in Hokkaido, northern Japan. *Geofisica Internacional*, 34, 283-291.

678 Gardin, S., Galbrun, B., Thibault, N., Coccioni, R., Premoli-Silva, I., 2012. Bio-
679 magnetochronology for the upper Campanian–Maastrichtian from the Gubbio area,
680 Italy: New results from the Contessa Highway and Bottaccione sections.
681 *Newsletters on Stratigraphy* 45, 75–104.

682 Haq, B.U., Hardenbol, J., Vail, P.R., 1987. Chronology of fluctuating sea levels since the
683 Triassic. *Science* 235, 1156–1167.

684 Hasegawa, T., Pratt, L.M., Maeda, H., Shigeta, Y., Okamoto, T., Kase, T., Uemura, K.,
685 2003. Upper Cretaceous stable carbon-isotope stratigraphy of terrestrial organic
686 matter from Sakhalin, Russian Far East: a proxy for the isotopic composition of
687 paleoatmospheric CO₂. *Palaeogeography, Palaeoclimatology, Palaeoecology* 215,
688 179–182.

689 Hay, W.W., DeConto, R., Wold, C.N., Wilson, K.M., Voigt, S., Schulz, M., Wold-Rossby,
690 A., Dullo, W.-C., Ronov, A.B., Balukhovskiy, A.N., Soeding E., 1999, Alternative
691 global Cretaceous paleogeography, In: Barrera, E. and Johnson, C. (Eds.), *The*
692 *evolution of Cretaceous ocean/climate systems*, Geological Society of America
693 *Special Paper* 332, p. 1–47.

694 Haynes, S. J., MacLeod, K. G., Ladant, J. B., Guchte, A. V., Rostami, M. A., Poulsen, C.
695 J., Martin, E. E., 2020. Constraining sources and relative flow rates of bottom waters
696 in the Late Cretaceous Pacific Ocean. *Geology*, 48, 509-513.

697 Hesselbo, S.P., Robinson, S.A., Surlyk, F. Piasecki, S., 2002. Terrestrial and marine
698 extinction at the Triassic-Jurassic boundary synchronized with major carbon-cycle
699 perturbation: A link to initiation of massive volcanism?. *Geology*, 30, 51–254.

700 Huber, B.T., 1992. Paleobiogeography of Campanian-Maastrichtian foraminifera in the
701 southern high latitudes. *Palaeogeography, Palaeoclimatology, Palaeoecology* 92,
702 325–360.

703 Huber B.T., Watkins, D.K., 1992. Biogeography of Campanian-Maastrichtian calcareous
704 plankton in the region of the Southern Ocean: paleogeographic and paleoclimatic
705 implications, In: Kennett, J.P., Warnke, D.A. (Eds.), *The Antarctic*
706 *paleoenvironment: A perspective on global change*. Antarctic Research Series 56.
707 American Geophysical Union, Washington, D.C., p. 31–60.

708 Huber, B.T., Norris, R.D., MacLeod, K.G., 2002. Deep-sea paleotemperature record of
709 extreme warmth during the Cretaceous. *Geology* 30, 123–126.

710 Hsü, K.J., He, Q., McKenzie, J.A., Weissert, H., Perch-Nielsen, K., Oberhänsli, H., Kelts,
711 K., LaBrecque, J., Tauxe, L., Krähenbühl, U., Percival, S.F., Wright, R., Karpoff,

712 A.M., Petersen, N., Tucker, P., Poore, R.Z., Gombos, A.M., Pisciotta, K., Carman,
713 M.F., Schreiber, E., 1982. Mass mortality and its environmental and evolutionary
714 consequences. *Science*, 216, 249-256.

715 Hunter, S. J., Valdes, P. J., Haywood, A. M., Markwick, P. J., 2008. Modelling
716 Maastrichtian climate: investigating the role of geography, atmospheric CO₂ and
717 vegetation. *Climate of the past discussions*, 4, 981-1019.

718 Husson, D., Galbrun, B., Laskar, J., Hinnov, L.A., Thibault, N., Gardin, S., Locklair, R.E.,
719 2011. Astronomical calibration of the Maastrichtian (Late Cretaceous). *Earth and
720 Planetary Science Letters* 305, 328–340.

721 Immenhauser, A., Holmden, C., Patterson, W.P., 2008. Interpreting the carbon isotope
722 record of ancient epeiric seas: lessons from the Recent. In: Pratt, B.R., Holmden, C.
723 (Eds.), *Dynamic of Epiric Seas*. Geological Association of Canada, Special Paper,
724 p. 135–174

725 Isaza-Londoño, C., MacLeod, K.G., Huber, B.T., 2006. Maastrichtian North Atlantic
726 warming, increasing stratification and foraminiferal paleobiology at three
727 timescales. *Paleoceanography* 21, PA1012.

- 728 Jasper, J.P., Gagosian, R.B., 1990. The sources and deposition of organic matter in the
729 Late Quaternary Pigmy Basin, Gulf of Mexico. *Geochimica et Cosmochimica Acta*
730 54, 1117–1132.
- 731 Jenkyns, H.C., 1998. The Early Toarcian (Jurassic) Anoxic Event: stratigraphic,
732 sedimentary, and geochemical evidence, *American Journal of Science* 288, 101–
733 151.
- 734 Johnson, C.C., Kuffman, E.G., 1990. Originations, radiations and extinctions of
735 Cretaceous rudistid bivalve species in the Caribbean Province, In: Kauffman E.G.,
736 Walliser, O.H. (Eds.), *Extinction events in Earth history*. Springer-Verlag, New York,
737 p. 305–324.
- 738 Johnson, C.C., Kuffman, E.G., 1996. Maastrichtian extinction patterns of Caribbean
739 province rudistids, In: MacLeod N., Keller. G. (Eds.), *Cretaceous-Tertiary boundary*
740 *mass extinction: Biotic and environmental changes*. Norton, New York, p. 231–274.
- 741 Johnson, C.C., Barron, E.J., Kauffman, E.G., Arthur, M.A., Fawcett, P.J., Yasuda, M.K.,
742 1996. Middle Cretaceous reef collapse linked to ocean heat transport. *Geology* 24,
743 376–380.

- 744 Jung, C., Voigt, S., Friedrich, O., 2012. High-resolution carbon-isotope stratigraphy
745 across the Campanian–Maastrichtian boundary at Shatsky Rise (tropical Pacific).
746 *Cretaceous Research* 37, 177–185.
- 747 Jung, C., Voigt, S., Friedrich, O., Koch, M.C., Frank, M., 2013. Campanian-Maastrichtian
748 ocean circulation in the tropical Pacific, *Paleoceanography*, 28, 562–573.
- 749 Katagiri, T., Naruse, H., Ishikawa, N., Hirata, T., 2020. Collisional bending of the western
750 Paleo - Kuril Arc deduced from paleomagnetic analysis and U–Pb age
751 determination. *Island Arc*, 29, e12329.
- 752 Keller, G., Lindinger, M., 1989. Stable isotope, TOC and CaCO₃ record across the
753 Cretaceous/Tertiary boundary at El Kef, Tunisia. *Palaeogeography,*
754 *Palaeoclimatology, Palaeoecology*, 73, 243-265.
- 755 Kiminami, K., 1978. Stratigraphic re-examination of the Nemuro Group. *Chikyu Kagaku*
756 32, 120–132. (in Japanese with English abstract)
- 757 Kiminami, K., 1983. Sedimentary history of the late Cretaceous-Paleocene Nemuro
758 Group, Hokkaido, Japan: a forearc basin of the Paleo-Kuril arc-trench system.
759 *Journal of the Geological Society of Japan* 89, 607–624.

- 760 Kimura, G., 1994. The latest Cretaceous - Early Paleogene rapid growth of accretionary
761 complex and exhumation of high pressure series metamorphic rocks in northwestern
762 Pacific margin. *Journal of Geophysical Research: Solid Earth*, 99, 22147-22164.
- 763 Kominz, M.A., Browning, J.V., Miller, K.G., Sugarman, P.J., Mizintseva, S., Scotese,
764 C.R., 2008. Late Cretaceous to Miocene sea-level estimates from the New Jersey
765 and Delaware coastal plain coreholes: an error analysis. *Basin Research* 20, 211–
766 226.
- 767 Körner, C., Farquhar, G.D., Roksandic, Z., 1988. A global survey of carbon isotope
768 discrimination in plants from high altitude. *Oecologia*, 74, 623–632.
- 769 Ladant, J.B., Poulsen, C.J., Fluteau, F., Tabor, C.R., MacLeod, K.G., Martin, E.E., Haynes,
770 S.J., Rostami, M.A., 2020. Paleogeographic controls on the evolution of Late
771 Cretaceous ocean circulation. *Climate of the Past*, 16, 973-1006.
- 772 Leavitt, S.W., 1993. Environmental information from $^{13}\text{C}/^{12}\text{C}$ ratios of wood, In: Swart,
773 P.K., Lohmann, K.C., Mckenzie, J., Savin, S. (Eds.), *Climate Change in Continental*
774 *Isotopic Records*, Geophysical Monograph 78, 325–331.
- 775 Lehman, T., 1990, Paleosols and the Cretaceous/Tertiary transition in the Big Bend region
776 of Texas: *Geology*, 18, 362–364.

- 777 Lees, J.A., 2002. Calcareous nannofossil biogeography illustrates palaeoclimate change
778 in the Late Cretaceous Indian Ocean. *Cretaceous Research* 23, 537–634.
- 779 Li, L., Keller, G., 1998. Maastrichtian climate, productivity and faunal turnovers in
780 planktic foraminifera in South Atlantic DSDP sites 525A and 21. *Marine*
781 *Micropaleontology* 33, 55–86.
- 782 Li, L., Keller, G., 1999 Variability in Late Cretaceous and deep waters: evidence from
783 stable isotopes. *Marine Geology* 161, 171–190.
- 784 Lipp, J., Trimborn, P., Edwards, T., Waisel, Y. Yakir, D., 1996. Climatic effects on the
785 $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ of cellulose in the desert tree *Tamarix jordanis*. *Geochimica et*
786 *Cosmochimica Acta*, 60, 3305–3309.
- 787 Lloyd, J., Farquhar, G.D., 1994. ^{13}C discrimination during CO_2 assimilation by the
788 terrestrial biosphere. *Oecologia* 99, 201–215.
- 789 Lowrie, W., Alvarez, W., 1977. Upper Cretaceous-Paleocene Magnetic Stratigraphy at
790 Gubbio, Italy. 3. Upper Cretaceous Magnetic Stratigraphy. *Geological Society of*
791 *America Bulletin* 88, 374–377.
- 792 Lowrie, W., Alvarez, W., 1981. 100 Million Years of geomagnetic polarity history.
793 *Geology* 9, 392–397.

- 794 MacLeod, K.G., Huber, B.T., 1996. Reorganization of deep ocean circulation
795 accompanying a Late Cretaceous extinction event. *Nature* 380, 422–425.
- 796 MacLeod, K.G., Huber, B.T., 2001. The Maastrichtian record at Blake Nose (western
797 North Atlantic) and implications for global palaeoceanographic and biotic changes,
798 In: Kroon, D., Norris, R.D., Klaus, A. (Eds.), *Western North Atlantic Palaeogene
799 and Cretaceous Palaeoceanography*. Geological Society of London Special
800 Publications 193, p. 111–130.
- 801 MacLeod, K.G., Huber, B.T., Ward, P.D., 1996. The biostratigraphy and
802 paleobiogeography of Maastrichtian inoceramids, In: Ryder, G., Fastovsky, D.,
803 Garter, S. (Eds.), *The Cretaceous-Tertiary event and other catastrophes in Earth
804 history*. Geological Society of America Special Paper 307, p. 361–373.
- 805 MacLeod, K.G., Huber, B.T., Ducharme, M.L., 2000. Paleontological and geochemical
806 constraints on the deep ocean during the Cretaceous greenhouse interval, In: Huber,
807 B.T., MacLeod, K.G., Wing, S.L., (Eds.), *Warm climates in Earth history*.
808 Cambridge University Press, Cambridge, p. 241–274.
- 809 MacLeod, K.G., Huber, B.T., Isaza-Londoño, C., 2005. North Atlantic warming during
810 global cooling at the end of the Cretaceous. *Geology* 33, 437–440.

811 MacLeod, K.G., Londoño, C.I., Martin, E.E., Berrocoso, Á.J., Basak, C., 2011. Changes
812 in North Atlantic circulation at the end of the Cretaceous greenhouse interval.
813 *Nature Geoscience*, 4, 779-782.

814 Maeda, H., Shigeta, Y., 2005. Maastrichtian ammonoid fauna from the Pugachevo area,
815 southern Sakhalin, Russian Far East, In: Shigeta, Y., Maeda, H. (Eds.), *The*
816 *Cretaceous System in the Makarov area, Southern Sakhalin, Russian Far East.*
817 *National Science Museum Monographs 31*, National Science Museum, Tokyo,
818 Japan, p. 121–136.

819 Maeda, H., Shigeta, Y., Fernando, A.G.S., Okada, H., 2005. Stratigraphy and fossil
820 assemblages of the Upper Cretaceous System in the Makarov area, Southern
821 Sakhalin, Russian Far East, In: Shigeta, Y., Maeda, H. (Eds.), *The Cretaceous*
822 *System in the Makarov area, Southern Sakhalin, Russian Far East.* National Science
823 *Museum Monographs 31*, National Science Museum, Tokyo, Japan, p. 25–120.

824 Moiroud, M., Pucéat, E., Donnadieu, Y., Bayon, G., Guiraud, M., Voigt, S., Deconinck,
825 J.F., Monna, F., 2016. Evolution of neodymium isotopic signature of seawater
826 during the Late Cretaceous: Implications for intermediate and deep circulation.
827 *Gondwana Research*, 36, 503-522.

- 828 Murphy, D.P., Thomas, D.J., 2012. Cretaceous deep-water formation in the Indian sector
829 of the Southern Ocean, *Paleoceanography*, 27, PA1211.
- 830 Naruse, H., 2003. Cretaceous to Paleocene depositional history of North-Pacific
831 subduction zone: reconstruction from the Nemuro Group, eastern Hokkaido,
832 northern Japan. *Cretaceous Research* 24, 55–71.
- 833 Naruse, H., Maeda, H., Shigeta, Y., 2000. Newly discovered Late Cretaceous molluscan
834 fossils and inferred K/T boundary in the Nemuro Group, eastern Hokkaido, northern
835 Japan. *Journal of the Geological Society of Japan* 106, 161–164. (in Japanese with
836 English abstract)
- 837 Nifuku, K., Kodama, K., Shigeta, Y., Naruse, H., 2009. Faunal turnover at the end of the
838 Cretaceous in the North Pacific region: Implications from combined
839 magnetostratigraphy and biostratigraphy of the Maastrichtian Senpohshi Formation
840 in the eastern Hokkaido Island, northern Japan. *Palaeogeography,*
841 *Palaeoclimatology, Palaeoecology* 271, 84–95.
- 842 Nordt, L., Atchley, S., Dworkin, S., 2003. Terrestrial evidence for two greenhouse events
843 in the latest Cretaceous. *GSA Today* 13, 4–9.
- 844 Okada, H., Yamada, M., Matsuoka, H., Murota, T., Isobe, T., 1987. Calcareous
845 nannofossils and biostratigraphy of the Upper Cretaceous and Lower Paleogene

846 Nemuro Group, eastern Hokkaido, Japan. *Journal of the Geological Society of*
847 *Japan* 93, 329–348.

848 Omura, A., Hoyanagi, K., 2004. Relationships between composition of organic matter,
849 depositional environments, and sea-level changes in backarc basins, central Japan.
850 *Journal of Sedimentary Research* 74, 620–630.

851 Perch-Nielsen, K., McKenzie, J., He, Q., 1982. Biostratigraphy and isotope stratigraphy
852 and the ‘catastrophic’ extinction of calcareous nannoplankton at the
853 Cretaceous/Tertiary boundary. In: Silver, L.T., Schultz, P.H. (Eds.), *Geological*
854 *implications of impacts of large asteroids and comets on the Earth*, GSA Special
855 *Paper*, 190, 353-371

856 Pendall, E., Betancourt, J.L., Leavitt, S.W., 1999. Paleoclimatic significance of δD and
857 $\delta^{13}C$ values in pinon pine needles from packrat middens spanning the last 40,000
858 years. *Palaeogeography, Palaeoclimatology, Palaeoecology*, 147, 53-72.

859 Pospichal, J.J., Wise Jr, S.W., 1990. Maestrichtian calcareous nannofossil biostratigraphy
860 of Maud Rise, ODP Leg 113 Sites 689 and 690, Weddell Sea. *Proceedings of Ocean*
861 *Drilling Project, Scientific Results* 113, 465–487.

- 862 Puc at, E., Donnadi eu, Y., Ramstein, G., Fluteau, F., Guillocheau, F., 2005. Numerical
863 evidence for thermohaline circulation reversals during the Maastrichtian, *Geochem.*
864 *Geophys. Geosyst.*, 6, Q11012.
- 865 Ramesh, R., Bhattacharya, S.K. and Gopalan, K., 1986. Climatic correlations in the stable
866 isotope records of silver fir (*Abies pindrow*) trees from Kashmir, India. *Earth and*
867 *Planetary Science Letters*, 79, 66-74.
- 868 Robinson, S.A., Vance, D., 2012. Widespread and synchronous change in deep-ocean
869 circulation in the North and South Atlantic during the Late Cretaceous,
870 *Paleoceanography*, 27, PA1102.
- 871 Robinson, S.A., Murphy, D.P., Vance, D., Thomas, D.J., 2010. Formation of “southern
872 component water” in the Late Cretaceous: evidence from Nd-isotopes. *Geology*, 38,
873 871-874.
- 874 Salazar-Jaramillo, S., Fowell, S.J., McCarthy, P.J., Benowitz, J.A.,  liwiński, M.G.
875 Tomsich, C.S., 2016. Terrestrial isotopic evidence for a Middle-Maastrichtian
876 warming event from the lower Cantwell Formation, Alaska. *Palaeogeography,*
877 *Palaeoclimatology, Palaeoecology*, 441, 360-376.

- 878 Sawada, K., Akiyama, M., 1994. Carbon isotope composition of macerals separated from
879 various kerogens by density separation method. *Journal of the Japanese Association*
880 *for Petroleum Technology* 59, 244–255. (in Japanese with English abstract)
- 881 Schleser, G.H., Helle, G., Lücke, A. Vos, H., 1999. Isotope signals as climate proxies: the
882 role of transfer functions in the study of terrestrial archives. *Quaternary Science*
883 *Reviews*, 18, 927–943.
- 884 Shafik, S., 1990. Late Cretaceous nannofossil biostratigraphy and biogeography of the
885 Australian western margin. Bureau of Mineral Resources, Geology and Geophysics
886 Report 295, 1–164.
- 887 Shigeta, Y., Nishimura, T., Nifuku, K., 2015. Middle and late Maastrichtian (latest
888 Cretaceous) ammonoids from the Akkeshi Bay area, eastern Hokkaido, northern
889 Japan and their biostratigraphic implications. *Paleontological Research* 19, 107–127.
- 890 Scholle, P.A. & Arthur, M.A. 1980. Carbon-isotope fluctuations in Cretaceous pelagic
891 limestones: potential stratigraphic and petroleum exploration tool. *AAPG Bulletin*,
892 64, 67–87
- 893 Simmons, M.D., Sharland, P.R., Casey, D.M., Davies, R.B., Sutcliffe, O.E., 2007. Arabian
894 Plate sequence stratigraphy: Potential implications for global chronostratigraphy.
895 *GeoArabia*, 12, 101-130.

- 896 Stoll, H.M., Schrag, D.P., 2000. High-resolution stable isotope records from the upper
897 cretaceous rocks of Italy and Spain: glacial episodes in a greenhouse planet?
898 Geological Society of America. Bulletin 112, 308–319.
- 899 Teerman, S.C., Hwang, R.J., 1991. Evaluation of the liquid hydrocarbon potential of coal
900 by artificial maturation techniques. *Organic Geochemistry*, 17, 749-764.
- 901 Teichmuller, M., Teichmuller, R., 1979. Diagenesis of coal (coalification), In: Larsen, G.,
902 Chilingar, G.V. (Eds.), *Diagenesis in Sediments and Sedimentary Rocks*. Elsevier,
903 Amsterdam, p. 207–246.
- 904 Thibault, N., Gardin, S., 2006. Maastrichtian calcareous nannofossil biostratigraphy and
905 paleoecology in the Equatorial Atlantic (Demerara Rise, ODP Leg 207 Hole 1258A).
906 *Revue de Micropaléontologie*, 49, 199-214.
- 907 Thibault, N., Gardin, S., 2007. The late Maastrichtian nannofossil record of climate
908 change in the South Atlantic DSDP Hole 525A. *Marine Micropaleontology* 65, 163–
909 184.
- 910 Thibault, N., Gardin, S., 2010. The calcareous nannofossil response to the end-Cretaceous
911 warm event in the Tropical Pacific. *Palaeogeography, Palaeoclimatology,*
912 *Palaeoecology* 291, 239–252.

- 913 Thibault, N., Gardin, S., Galbrun, B., 2010. Latitudinal migration of calcareous
914 nannofossil *Micula murus* in the Maastrichtian: implications for global climate
915 change. *Geology* 38, 203–206.
- 916 Thibault, N., Harlou, R., Schovsbo, N., Schiøler, P., Minoletti, F., Galbrun, B., Lauridsen,
917 B.W., Sheldon, E., Stemmerik, L., Surlyk, F., 2012a. Upper Campanian–
918 Maastrichtian nannofossil biostratigraphy and high-resolution carbon-isotope
919 stratigraphy of the Danish Basin: towards a standard $\delta^{13}\text{C}$ curve for the Boreal
920 Realm. *Cretaceous Research* 33, 72–90.
- 921 Thibault, N., Husson, D., Harlou, R., Gardin, S., Galbrun, B., Huret, E., Minoletti, F.,
922 2012b. Astronomical calibration of upper Campanian–Maastrichtian carbon isotope
923 events and calcareous plankton biostratigraphy in the Indian Ocean (ODP Hole
924 762C): implication for the age of the Campanian–Maastrichtian boundary.
925 *Palaeogeography, Palaeoclimatology, Palaeoecology* 337, 52–71.
- 926 Thibault, N., Harlou, R., Schovsbo, N.H., Stemmerik, L., Surlyk, F., 2016. Late
927 Cretaceous (late Campanian–Maastrichtian) sea-surface temperature record of the
928 Boreal Chalk Sea. *Climate of the Past*, 12, 429–438.
- 929 Tobin, T.S., Ward, P.D., Steig, E.J., Olivero, E.B., Hilburn, I.A., Mitchell, R.N., Diamond,
930 M.R., Raub, T.D., Kirschvink, J.L., 2012. Extinction patterns, $\delta^{18}\text{O}$ trends, and

931 magnetostratigraphy from a southern high-latitude Cretaceous–Paleogene section:
932 Links with Deccan volcanism. *Palaeogeography, Palaeoclimatology, Palaeoecology*,
933 350–352, 180–188.

934 Toshimitsu, S., Matsumoto, T., Noda, M., Nishida, T., Maiya, S., 1995. Towards an
935 integrated mega-, micro- and magneto-stratigraphy of the Upper Cretaceous in
936 Japan. *Journal of the Geological Society of Japan* 101, 19–29. (in Japanese with
937 English abstract)

938 Tyson, R.V., 1995. *Sedimentary Organic Matter: Organic Facies and Palynofacies*.
939 Chapman and Hall, London. 633 p.

940 Uramoto, G. I., Tahara, R., Sekiya, T., Hirano, H., 2013. Carbon isotope stratigraphy of
941 terrestrial organic matter for the Turonian (Upper Cretaceous) in northern Japan:
942 Implications for ocean-atmosphere $\delta^{13}\text{C}$ trends during the mid-Cretaceous climatic
943 optimum. *Geosphere*, 9, 355–366.

944 Voigt, S., Friedrich, O., Norris, R.D., Schoenfeld, J., 2010. Campanian–Maastrichtian
945 carbon isotope stratigraphy: shelf-ocean correlation between the European shelf sea
946 and the tropical Pacific Ocean. *Newsletters on Stratigraphy* 44, 57–72.

947 Voigt, S., Gale, A.S., Jung, C., Jenkyns, H.C., 2012. Global correlation of Upper
948 Campanian–Maastrichtian successions using carbon-isotope stratigraphy:

- 949 development of a new Maastrichtian timescale. *Newsletters on Stratigraphy* 45, 25–
950 53.
- 951 Ward, P.D., Kennedy, W.J., MacLeod, K.G., Mount, J.F., 1991. Ammonite and inoceramid
952 bivalve extinction patterns in Cretaceous/Tertiary boundary sections of the Biscay
953 region (southwestern France, northern Spain). *Geology* 19, 1181–1184.
- 954 Watanabe, H., Akiyama, M., 1998. Characterization of organic matter in the Miocene
955 turbidites and hemipelagic mudstones in the Niigata oil field, central Japan. *Organic*
956 *Geochemistry* 29, 605–611.
- 957 Wendler, I., 2013. A critical evaluation of carbon isotope stratigraphy and biostratigraphic
958 implications for Late Cretaceous global correlation. *Earth-Science Reviews*, 126,
959 116–146.
- 960 Wendler, I., Willems, H., Gräfe, K.-U., Ding, L., Luo, H., 2011. Upper Cretaceous
961 interhemispheric correlation between the Southern Tethys and the Boreal: chemo-
962 and biostratigraphy and paleoclimatic reconstructions from a new section in the
963 Tethys Himalaya, S-Tibet. *Newsletters on Stratigraphy* 44, 137–171.
- 964 Whelan J.K., Thompson-Rizer, C.L., 1993. Chemical methods for assessing kerogen and
965 protokerogen types and maturity, In: Engel, M.H., Macko, S. (Eds.), *Organic*
966 *Geochemistry, Principles and Applications*. Plenum Press, New York, p. 289–353.

- 967 Whiticar, M.J., 1996. Stable isotope geochemistry of coals, humic kerogens and related
968 natural gases. *International Journal of Coal Geology*, 32, 191-215.
- 969 Yamada, M., 1984. Calcareous nannofossils and planktonic foraminifera from the west
970 coast of the Akkeshi Bay, Hokkaido, In: Saito, T., Okada H., Kaiho, K. (Eds.),
971 *Biostratigraphy and international correlation of the Paleogene System in Japan*.
972 Faculty of Science, Yamagata University, Yamagata, Japan, p. 15–18. (in Japanese)
- 973 Zachos, J.C., Arthur, M.A., 1986. Paleooceanography of the Cretaceous/Tertiary boundary
974 event: inferences from stable isotopic and other data. *Paleoceanography* 1, 5–26.
- 975 Zachos, J. C., Arthur, M. A., Thunell, R. C., Williams, D. F., Tappa, E. J., 1985. Stable
976 isotope and trace element geochemistry of carbonate sediments across the
977 Cretaceous/Tertiary boundary at Deep Sea Drilling Project Hole 577, Leg 861.
978 *Initial Reports of the Deep Sea Drilling Project* 86, 513–532.
- 979 Zachos, J. C., Arthur, M. A., Dean, W. E., 1989. Geochemical evidence for suppression
980 of pelagic marine productivity at the Cretaceous/Tertiary boundary. *Nature*, 337,
981 61–64.

982

983 **9. Figure captions**

984

985 **Fig. 1.** Paleogeographic map of the late Maastrichtian (68 Ma; modified after Hay et al.,
986 1999, https://www.odsn.de/odsn/services/paleomap/adv_map.html) showing the location
987 of the sections used in this study. SN: Senphoshi Formation of the Nemuro Group,
988 northern Japan; NB: Naiba section, Sakhalin, eastern Russia; GB: Bottaccione Gorge and
989 the Contessa Highway sections at Gubbio, Italy; Zumaia section, Basque country,
990 Northern Spain; TR: Aguja, Javelina, and Black Peaks Formations in the Tornillo Basin,
991 Texas, US; CT: lower Cantwell Formation, Alaska, US. Numbers indicate DSDP, ODP,
992 and IODP sites (DSDP Site 525A, South Atlantic Ocean; ODP Site 762C, Indian Ocean;
993 ODP Site 1210B, equatorial Pacific Ocean; IODP Site U1403, Newfoundland Margin).

994

995 **Fig. 2.** Index maps showing (A) the location of Hokkaido, (B) the distribution of the
996 Nemuro Group, and (C) location of the studied section.

997

998 **Fig. 3.** Lithology and magnetostratigraphy (after Nifuku et al., 2009) of the Senpohshi
999 Formation. Correlation with the astronomically calibrated magnetostratigraphic time
1000 scale (Option 2 of Husson et al., 2011) is shown. Stratigraphic position of the sampling
1001 sites is also indicated in the figure.

1002

1003 **Fig. 4.** Photomicrographs of kerogen extracted from selected samples: (A) CS21, (B)
1004 CS52, and (C) CS59. tp: translucent phytoclasts; op: opaque phytoclasts; nfa: non-
1005 fluorescent amorphous organic matter. Note that these three components dominate the
1006 SOM in each sample.

1007

1008 **Fig. 5.** Relative abundance (%) of various constituents of SOM extracted from eight
1009 selected samples.

1010

1011 **Fig. 6.** Stratigraphic variation of stable carbon isotope values of bulk SOM ($\delta^{13}\text{C}_{\text{org}}$) and
1012 TOC content within the Senpohshi Formation. Error bar shows an overall uncertainty
1013 ($\pm 0.25\%$: 1σ) indicated by repeated analysis of the laboratory standard.

1014

1015 **Fig. 7.** Comparison of the age model for the MME after Thibault et al. (2012b), Voigt et
1016 al. (2012), and Batenburg et al. (2018). Ages were adjusted to the astronomically
1017 calibrated Maastrichtian time scale by Husson et al. (2011) (Option 2).

1018

1019 **Fig. 8.** Correlation of stable carbon isotope stratigraphy of the Senposhi Formation with
1020 other sections. (A) Correlation with carbon isotope profiles obtained from marine

1021 materials. (1) Senpohshi Formation of the Nemuro Group, northern Japan (this study);
1022 (2) Zumaia section, Basque country, Northern Spain (Batenburg et al., 2012); (3)
1023 Bottaccione Gorge and the Contessa Highway sections at Gubbio, Italy (Voigt et al.,
1024 2012); (4) ODP Site 1210B, Equatorial Pacific (Jung et al., 2012); (5) IODP Site U1403,
1025 Newfoundland Margin (Batenburg et al., 2018); (6) ODP Site 762C, Indian Ocean
1026 (Thibault et al., 2012b); (7) DSDP Site 525A, South Atlantic (Li and Keller, 1998). Bulk
1027 carbonate was analyzed at these sites except for the DSDP Site 525A, where they
1028 measured single-species planktonic (*Rugoglobigerina rugosa*) and benthic foraminifera
1029 (*Anomalinoidea acufa*). (B) Correlation with carbon isotope profiles obtained from
1030 terrestrial materials. (8) Naiba section, Sakhalin, eastern Russia (Hasegawa et al., 2003).
1031 Bulk sedimentary organic matter was analyzed. (9) lower Cantwell Formation, Alaska,
1032 US (Salazar-Jaramillo et al., 2016). Stable carbon isotope compositions were obtained
1033 from bulk sedimentary organic matter and fossil wood. (10) Aguja, Javelina, and Black
1034 Peaks Formations in the Tornillo Basin, Texas, US (Nordt et al., 2003). They measured
1035 pedogenic carbonate. Astronomically calibrated magnetostratigraphic time scale and the
1036 405 kyr eccentricity cycles are after Option 2 of Husson et al. (2011). The light and dark
1037 gray shaded intervals correspond to the MME and its constituting segments after Voigt et
1038 al. (2012).

1039

1040 **10. Table captions**

1041

1042 **Table 1.** Stratigraphic positions of polarity chron boundaries in the Senpohshi Formation

1043 (Nifuku et al., 2009).

1044

1045 **Table 2.** Relative abundance (%) of various constituents of SOM extracted from eight

1046 selected samples.

1047

1048 **Table 3.** Elemental compositions of kerogen extracted from eight selected samples.

1049

1050 **Table 4.** TOC content and $\delta^{13}\text{C}_{\text{org}}$ values obtained for the Senpohshi Formation.

1051

1052 **Table 5.** Ages of the K-Pg boundary and the Maastrichtian polarity chron boundaries

1053 after Option 2 proposed by Husson et al. (2011).

1054

1055 **Table 6.** Tie-points for correlation of stable carbon isotope profiles, which are defined by

1056 Voigt et al. (2012) and Batenburg et al. (2018).