- 1 Carbon isotopes, stratigraphy and environmental change: the Middle/Upper
- 2 Cambrian Positive Excursion (SPICE) in Port-au-Port Group, Western
- 3 Newfoundland, Canada
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32 In many basins, Upper Cambrian carbonate successions display intervals with a positive carbon isotope excursion (CIE) of up to +5%. In North America, this marks the boundary between the 33 Sauk II-III super-sequences. A Steptoean Positive Carbon Isotope Excursion (SPICE) locality 34 previously identified in the Port-au-Port peninsula, western Newfoundland, has been revisited and 35 an additional potential SPICE locality found. In both locations, a CIE is found to be associated with 36 37 a prominent bioherm and sandstone layer within a sequence of carbonate rocks. At March Point, 38 columnar stromatolites occur while at Felix Cove thrombolites can be seen. In the latter, the 39 sandstone immediately overlies the thrombolites coincident with the CIE, while at March Point a 40 dolomitized grainstone occurs above the stromatolites. The sandstone at this locality post-dates the CIE. Although lower than the SPICE in some localities, a positive CIE is present in both sections. 41 March Point (+1.1 ‰) and Felix Cove (+1.8 ‰). Additionally, $\delta^{13}C_{org}$ rises from -30.0 ‰ to -22.0 42 ‰ at March Point, and from -27 ‰ to −24.0 ‰ at Felix Cove and, in accordance with previously 43 published work, we suggest that this could be the SPICE. Comparison of the stratigraphy and 44 45 petrography between the two localities suggest that both depositional and diagenetic factors could have influenced the nature of the interpreted SPICE in Newfoundland. It is also possible that the 46 local carbon isotopic signature may have been influenced by a semi-restricted depositional and 47 48 early diagenetic environment, related to the paleogeographic configuration, rather than the global marine excursion. 49

50 Keyword: SPICE, Port au Port Group, Stable Isotopes

Introduction

- 52 Upper Cambrian (Steptoean Stage) carbonate successions are found worldwide, including
- examples in North America, Siberia, Kazakhstan, China Australia and Oman (Figure 1), with a
- positive carbonate carbon isotope ($\delta^{13}C_{carb}$) excursion (CIE) of up to +5%, lasting nearly 4 Ma
- 55 (Saltzman et al. 2000, 2004, Kouchinsky et al. 2008, Woods et al. 2011, Neilson et al. 2016). This
- excursion has also been observed in the δ^{13} C of organic-rich (δ^{13} C_{org}) mudstones and shales
- 57 (Ahlberg et al. 2009, Saltzman et al. 2011). This CIE was first reported from the John's Wash
- 58 Limestone of Utah by Brasier (1993), followed by sections described in the House Range Utah;
- 59 McGill Nevada; Wind Rover Range and Gros Ventre Range Wyoming (Saltzman et al. 1995).
- The term Steptoean Positive Carbon Isotope Excursion (SPICE) was proposed by Saltzman et al.
- 61 (1998), who suggested a global occurrence comparing data from South China, Kazakhstan,
- Australia and Laurentia (Shingle Pass, Nevada) (Saltzman et al. 2000). This positive carbon isotope
- excursion is coeval with a trilobite mass extinction horizon (Palmer 1965, Sepkoski 1982,
- 64 Yochelson 1984, Saltzman et al. 1995, 1998, 2000, Bond and Grasby 2017). Saltzman et al. (2004,
- 65 2011) and Bond and Grasby (2017) suggested that the positive excursion represents a perturbation
- in the global carbon cycle that occurs on the boundary between the Sauk II and Sauk III second
- order super-sequences from the Sauk megasequence (Sloss 1963).
- 68 Other CIE's of similar magnitude and duration have been described from the Late Ordovician, late
- 69 Silurian, Upper Devonian, early Mississippian, and early late Permian (Gruszczynski et al. 1989;
- 70 Brenchley 1994; Joachimski et al. 2002; Saltzman 2002, Saltzman et al. 2011). Each positive CIE
- may correspond to different causes, but in general they are thought to reflect an increased burial of
- isotopically lighter organic carbon (resulting in higher δ^{13} C in the residual pool) related to
- 73 increased organic productivity, with organic preservation due to high sedimentation rates and/or
- anoxic conditions (Arthur et al. 1987, Derry et al. 1992, Schidlowski 1992, Schrag et al. 2002).
- 75 The SPICE was first described in Western Newfoundland in the Petit Jardin Formation (Felix and
- overlying Man O' War members) at Felix Cove by Saltzman et al. (2004). In this unit, δ^{13} C values
- of carbonates rise from -1% to 0% V-PDB, reaching a peak δ^{13} C of +2.2% just above a quartz
- 78 sandstone layer that marks the Sauk II Sauk III boundary. Although the presence of an excursion
- has been reported in the Port au Port rocks (Saltzman et al. 2004), the magnitude of the SPICE
- 80 excursion in the Petit Jardin Formation is smaller than observed around the world (Glumac and
- 81 Walker 1998).
- This raises questions about whether the presence of the global SPICE excursion in the Port au Port
- Peninsula rocks is real, or whether it is just a local environmental or diagenetic shift. In an attempt
- 84 to address this issue, two Western Newfoundland stratigraphic sections were logged in detail,

followed by petrographic analysis of the carbonates, and δ^{13} C analysis of both carbonate and 85 86 organic carbon samples. 87 **Geologic Setting** 88 The study area is located on the southern coast of the Port au Port Peninsula, western 89 Newfoundland (Figure 2). The SPICE was identified in Felix Cove (48°31'49.2"N / 58°47'12"E), a 90 section previously studied by Saltzman et al. (2004) and another potential SPICE locality identified at March Point (48°30'42.1"N / 59°05'31.1"E), 91 The Port au Port Group forms part of the autochthonous Cambrian-Ordovician sequence of the 92 93 tectono-stratigraphic Humber zone of the northern Appalachian orogeny (Williams 1976, 1979). It 94 is interpreted as shallow-marine deposits from the outer part of a stable carbonate platform that 95 surrounded the Iapetus Ocean (Williams 1976, 1979, Palmer and James 1979). These mixed carbonate-siliciclastic deposits record the gradual transition from early Cambrian siliciclastic 96 97 deposition to predominantly carbonate sedimentation in the Early Ordovician (Chow 1985, Chow and James 1987b, Cowan and James 1993). The Group is composed of three formations, namely 98 99 the March Point, Petit Jardin (Cape Ann, Campbells, Big Cove, Felix and Man O' War Members) 100 and Berry Head Formations. 101 Transgressive-regressive Grand Cycles are identified through the Port au Port Group (Chow 1985, Chow and James 1987a, 1987b) (Figure 3). The marine transgression at the beginning of the Grand 102 103 Cycles resulted in a shallow subtidal-intertidal environment, where relatively muddy carbonates 104 were deposited (shaley packages and parted limestone; Chow and James 1987b). These deposits 105 pass upwards into more open marine oolitic shoals, probably deposited near the platform edge 106 (carbonate packages; Chow and James 1987b). As sea level dropped at the top of the cycles, the 107 environment returned to a more restricted water circulation situation, although according to Chow 108 and James (1987b) extensive evidence of subaerial exposure at the top of cycle B is absent. Carbon 109 isotope data, previously collected in the Port au Port Group at Felix Cove, show a strong positive δ^{13} C_{carb} excursion between the Felix and Man O' War Members (Figure 1), which has been 110 111 interpreted as the SPICE (Saltzman et al. 2004). 112 Methodology The field work was performed along two main outcrops on the Port au Port Peninsula – March 113 114 Point and Felix Cove. Detailed sedimentological logging on a cm scale (1:50) and systematic sampling was carried out at 1m resolution, from the base of accessible units in the Petit Jardin 115 Formation to the top of each exposed Petit Jardin or Berry Head Formation section. The 116 117 stratigraphic section logged in the March Point locality (MP) was 270m thick, and in the Felix

118	Cove locality (FC), 40m thick. A third outcrop was found, in which the diagnostic
119	sedimentological characteristics of the SPICE layer in the Port-au-Port peninsula are exhibited
120	(West Felix Cove $48^{\circ}31'51.6"N / 58^{\circ}47'49.1"E$), but logging was not carried out at this location.
121	Representative layers were selected for thin sections (March Point: 45; Felix Cove: 8), carbonate
122	δ^{13} C and δ^{18} O analyses (March Point: 41; Felix Cove: 16), and organic carbon (δ^{13} Corg) (March
123	Point: 14; Felix Cove: 4). Polished thin sections were prepared at the Mineral and Rock Analyses
124	Laboratory (LAMIR) at the Universidade Federal do Paraná (UFPR) and at Petrografia BR, after
125	impregnation with blue resin for porosity identification. Petrographic analysis was performed at the
126	Universidade Federal do Rio Grande do Sul (UFRGS). For carbonate identification, the thin
127	sections were stained with a solution of alizarin red.
128	Carbonate stable isotope ($\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$) analysis was carried out at LAMIR/UFPR in Brazil
129	and at the BGS in the UK. For isotope analyses, carbonate samples were collected using a
130	microdrill (0.5mm diamond drill), in order to avoid mixing of primary and diagenetic constituents
131	whenever possible. At LAMIR, the amount of material drilled was around 0.3 mg, while a larger
132	amount of powder was required by the BGS for the analysis (10mg). At LAMIR, the isotope ratios
133	were determined using a GasBench II attached to a Thermo Scientific Delta Advantage continuous
134	flow isotope ratio mass spectrometer, following the Spötl and Vennemann (2003) and Paul and
135	Skrzypek (2007) routine with a reproducibility <0.1% (1 σ). At BGS, isotopic analysis was carried
136	out using a vacuum extraction technique and a VG Optima mass spectrometer with overall
137	analytical reproducibility better than <0.1% (1 σ) for $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$. The main difference in
138	the carbonate isotope methodology of the two laboratories is the time of carbonate-acid reaction. At
139	the BGS a quick reaction at low temperature (16°C) was performed over two hours in order to
140	release CO ₂ from calcite with minimal contribution from any CO ₂ derived from secondary
141	dolomite. However, at LAMIR, the time taken for reaction was between one to two hours at a
142	temperature of 72°C. The isotope composition of samples is reported in the standard δ notation,
143	defined as the relative difference, in parts per thousand (‰) without any further conversion. Each
144	laboratory used their own in house standards previously calibrated to Vienna Pee Dee Belemnite
145	(V-PDB).
146	For organic carbon isotope ($\delta^{13}C_{org}$) analysis at the BGS, the samples were reacted with 5% HCl
147	overnight to remove carbonate, and dried down at 40°C. After being cleaned, homogenized, and
148	dried, c. 15–25 mg of decarbonized sediment were used for determination of carbon isotope ratios
149	using a carbon isotopic ratio Carlo Erba 1500 on-line to a VG TripleTrap (with a secondary
150	cryogenic trap for very low carbon content samples) and Optima dual-inlet mass spectrometer.
151	$\delta^{13}C_{org}$ values were calculated to the V-PDB scale using a within-run laboratory standard (BROC1)

153 precision of $\pm < 0.1\%$ (1 σ). 154 Results The stratigraphic interval that comprises the purported SPICE in Felix Cove (as identified by 155 Saltzman, 2004, at 22.5 m, Figure 4a) comprises several sets of fining-upward oolitic grainstones 156 (20-40 cm thickness), followed by a layer of pillow-shaped thrombolite, around 25 cm thick, with a 157 clotted center tending to a more regular shape at the edges (Figure 4b). The sedimentary layers that 158 underly the thrombolite layer contain trilobites of the Crepicephalus Zone (Terranovella dorsalis 159 and Crepicephalus iowensis in the FC1 and FC2 collections of Westrop, 1992). Westrop (1992) 160 161 also records the occurrence of the trilobite Dytremacephalus strictus in the thrombolite layer itself (R. Levesque, Geological Survey of Canada, location 96635), indicating that it belongs to the 162 Aphelaspis Zone (Figure 3). These bioherms are overlain by a thin glauconitic sandstone layer (~25 163 164 cm thickness) before returning upwards to grainstones and parted limestone sets. These sets show 165 different degrees of dolomitization, but it is still possible to identify primary constituents. 166 Saltzman et al. (2004) note that Elvinia Zone fauna (A.R. Palmer, Institute for Cambrian Studies 167 collection ICS-1406) were found approximately 8 m above the quartz sandstone. The trilobite data 168 confirms the CIE as being the SPICE. A similar lithological succession is observed in West Felix Cove (Figure 4c). 169 170 In the March Point section, the interval that comprises the positive carbon excursion, between 222m and 226 m, consists of a layer of columnar stromatolites (Figure 4d), around 20 cm thick, 171 either forming as separated columns or laterally continuous mats. These are overlain by a thin 172 173 dolomitized glauconitic grainstone, 20 cm thick (Figure 4e). A meter-thick glauconitic, fine-174 grained sandstone (Figure 4f) occurs 3-4 m above. Below the stromatolite layer, the oolitic 175 grainstone sets are extensively dolomitized. No well constrained faunal information is available for 176 this layer. The uncompacted bioclastic sandstone that overlies the bioherms in Felix Cove is cemented by 177 calcite with very minor replacive dolomite (Figure 5a) although in West Felix Cove the dolomite 178 179 content is considerably higher, showing small relict calcite areas (pinkish stain) (Figure 5b). Quartz 180 grains are well rounded and well sorted, medium coarse - coarse sand, with 465 µm diameter average (Table 1). The glauconitic grainstone that overlies the stromatolites at March Point is 181 182 extensively dolomitized (Figure 5c) including the glauconite peloids (Figure 5d). The glauconitic 183 sandstone layer, 3-4 m above it, contains very poorly sorted, sub-rounded to angular silt to medium 184 sand grains (89 µm diameter average) which is also partially cemented by dolomite (Figure 5e and

f) and is very compacted. The sets of oolitic grainstones that underlie the potential SPICE layer in

calibrated against NBS-19 and NBS-22. Replicate analysis of well-mixed samples embraces a

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186	March Point are extensively dolomitized although the ooids outlines may be partially preserved
187	(Figure 6a-c). For several meters above the potential SPICE layer, carbonate-clastic sandstones
188	(composed of quartz grains and carbonate particles) occur (Figure 6d), replaced by dolomite.
189	The isotope data ($\delta^{13}C_{carb}$, $\delta^{18}O_{carb}$, $\delta^{13}C_{org}$ and %C) analysis for the March Point section are
190	presented in Table 2 and Figure 7. The $\delta^{13}C_{org}$ data are relatively stable below the base of the Felix
191	Member, with values between -29.5% and -24.8%. Above this point, however, the values become
192	more variable, between -30% and -21.3%, showing a general enrichment in ¹³ C organic carbon up
193	section, with a maximum within the Felix Member. The total organic carbon level (%C), however,
194	is generally very low, ranging between 0.04% and 0.3%, except for layers at 170 and 180m, with
195	1.9% and 2.1% respectively. The $\delta^{13}C_{carb}$ data show significant depletion at the top of the
196	Campbells Member (-4.1‰), and a maximum (+1.1‰) at the boundary between the Felix and Man
197	O' War Members. Above this, the $\delta^{13}C_{carb}$ general trend is towards lower values between -2 and $-$
198	1‰. In the March point section the best defined carbonate carbon excursion occurs over a thickness
199	of approximately 5 m (Figure 7). $\delta^{18}O_{carb}$ values are not directly related to the $\delta^{13}C_{carb}$ values ($r^2 =$
200	0.21, n = 41), although they show similar trends. However, $\delta^{18}O_{carb}$ becomes less variable in the
201	Felix Member, showing a more positive peak, around -4.5%, which coincides with the positive
202	shift in $\delta^{13}C_{carb}$ at the boundary between the Felix Cove and Man O' War Members.
203	In the Felix Cove section, only part of the Petit Jardin Formation is exposed, starting within the
204	Felix Member. The $\delta^{13}C_{carb}$, $\delta^{18}O_{carb}$, $\delta^{13}C_{org}$ and %C data are given in Table 2 and Figure 8. Near
205	the lowermost part of the outcrop exposed in the Felix Member, the $\delta^{13}C_{carb}$ value is around -2.0% ,
206	reaching a high of +1.8% near to the transition between the Felix and Man O' War Members
207	(Figure 8). This section corresponds to the same interval studied by Saltzman et al. (2004). The
208	data present a general upward increase in $\delta^{13}C_{carb}$ values (Figure 8). Compared to $\delta^{13}C_{carb}$, the
209	$\delta^{18}O_{carb}$ data shows a relatively stable trend, as observed for the Felix Member in the March Point
210	section, with a negative trough (though only of one data point) of -9.0% just before an interval of
211	higher values that reach -5.4% . A weak correlation between $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ occurs for the
212	Felix Cove section (r ² = 0.57, n= 16). As in the March Point section, %C is low, ranging between
213	0.04% and 0.17%. The stratigraphic thickness of the excursion upwards from 22.25 m, is about 15
214	m (Figure 8).
215	Discussion
216	In the Port au Port Peninsula, particular lithologies were found to be related to the peak $\delta^{13}C$ values
217	in outcrop. This positive $\delta^{13}C$ excursion was observed and identified by Saltzman et al. (2004) at
218	Felix Cove and, based on trilobite data presented by Westrop (1992), identified as the SPICE. The
219	positive $\delta^{13} C$ excursion occurs at a time when there was an influx of glauconitic quartzose sand

believed to be as a result of a decrease in water depth (regression). This was followed by a rise in 220 221 sea level and deepening (Saltzman et al. 2004), reflected in the resumption of carbonate deposition. 222 Detailed logging of the SPICE section at Felix Cove has shown that the positive peak is 223 characterized by a lithological sequence of bioherms, overlain by glauconitic sand rich layers, also 224 observed by Saltzman et al. (2004). A very similar lithological sequence has also been identified during this study approximately 0.8 km west of Felix Cove (West Felix Cove) although limited 225 226 data is available. 227 In the March Point section (approximately 25 km further west, Figure 2), a similar lithological 228 sequence of bioherms and glauconitic sandstone occur in the interval and is accompanied by a positive carbon isotope excursion (Figure 7). No biostratigraphic data are available for the upper 229 230 part of the Felix Member. Westrop (1992) provides data for the underlying Big Cove Member but 231 this lies well below where the SPICE would be expected in that section. 232 Although there are similarities in the lithological sequence between these localities, there are also 233 differences. Folk (1951) suggested that textural and compositional maturity of the sediment are 234 products of the degree of reworking, energy and distance of transport. The fine, poorly sorted, 235 slightly immature sandstones at March Point (Figure 5e and f, Table 1) contrast with the "clean", 236 coarse and mature sandstone observed at Felix Cove (Figure 6a and b). Also, the position of the 237 distinctive glauconitic sandstone is different. At Felix Cove it directly overlies the thombolites 238 while at March Point it occurs 3-4m m above it. At March Point, the stromatolite is overlain by a 239 dolomitized glauconitic grainstone prior to deposition of the glauconitic sandstone. Moreover, even 240 though it is unlikely that the morphology of specific microbial buildups was related to particular 241 environments, lateral differences do exist. The bioherm morphology at March Point comprise 242 columnar stromatolites, (Figure 4e) while thrombolites are observed in this layer at Felix Cove 243 (Figure 4a-c). The interval at March Point can therefore only be identified as a potential SPICE 244 locality. The Felix Cove section indicates that the lead up to the SPICE occurred in a shallow marine 245 246 environment, with bioclastic grainstone shoals (e.g. echinoderms) and bioherms. The influx of 247 coarse, well rounded, quartz sandstone marks the base of the positive carbon excursion and start of 248 a regression. The sandstone layer is followed by several sets of fining upward oolitic bioclastic graisntones, up to the Felix/Man O' War member transition, where parted limestone and shale 249 250 become abundant. These deposits have been interpreted by Chow and James (1987b) as subtidal to 251 intertidal. Comparatively, in the March Point section the positive carbon excursion (potential SPICE) occurs within a set of a glauconitic grainstones. The fine, angular, carbonate-clastic 252 sandstone occurs a few metres above the peak of the positive $\delta^{13}C_{carb}$ excursion rather than at its 253 beginning, near to the transition between Felix and Man O' War members. This suggests that the 254

sandstones at the two locations are diachronous and may have been deposited in different 255 environments with the start of the regression being slightly later at March Point than at Felix Cove. 256 257 While the SPICE interval is 200 m thick in Nevada (Saltzman et al. 1998) and 80 m in the Southern Appalachians (Glumac and Walker 1998), it is only 15 m thick in the Felix Cove section and the 258 259 potential SPICE only 5 m thick in the March Point section (Figure 9). This has already been noted by James and Stevens (1986), who found a thickness of less than 20 m, similar to our observations 260 at Felix Cove. However we observed a much thinner interval in the March Point section. 261 Additionally, the $\delta^{13}C_{carb}$ of the SPICE has a relatively lower magnitude than observed in other 262 suggested localities around the world. Globally, the SPICE is described as a $\delta^{13}C_{carb}$ positive shift 263 of around +5% (Saltzman et al. 2000, 2004, Kouchinsky et al. 2008, Woods et al. 2011) in 264 carbonates. The results presented here reach a $\delta^{13}C_{carb}$ peak of about +1.13 % for the March Point 265 section and +2.2 ‰ (Saltzman et al. 2004), and +1.8 ‰ (this work) for the Felix Cove section, with 266 267 an average shift of about +2 ‰, half of the typical global value. There are other locations which 268 have lower values, for example in the Southern Appalachians and South China (~+3%; Fan et al. 269 2011). At March Point, dolomitisation is significant in the immediate lead up to and around the potential 270 271 SPICE interval and this may have affected the isotopic results. To minimize the effect of this, the 272 data around the potential SPICE interval itself were collected at the BGS (Figure 7) using a quick 273 acid reaction time. This was performed over two hours at low temperature (16°C) to release CO₂ 274 mainly from calcite and minimize the contribution from dolomite. At March Point, the higher $\delta^{13}C_{carb}$ values are accompanied by an increase in $\delta^{18}O_{carb}$ (up to c. -4‰, Figure 7). A similar 275 pattern in $\delta^{18}O_{carb}$ values is recorded from early dolomites in the Lower-Middle Triassic Moenkopi 276 Formation (Marenco et al. 2008). The early, relatively heavy Moenkopi Formation dolomites were 277 contemporaneous with inner-middle shelf limestones which showed much lighter $\delta^{18}O_{carb}$ values of 278 -9 to -6%. The $\delta^{18}O_{carb}$ data observed here may therefore reflect a contribution from early 279 dolomitisation which may be more likely to have retained the original carbon isotopic signature due 280 281 to a lack of subsequent recrystallisation. Considering the wider effects of diagenesis, the more negative $\delta^{18}O_{carb}$ data observed in both 282 283 sections could be the result of burial and interaction with hot or meteoric diagenetic fluids (Scholle 1977, Moore 2002). Thus, the large depletion in $\delta^{18}O_{carb}$ values (down to -9%, Figures 7 and 8) 284 could be interpreted as a diagenetic effect, marked by resetting of the original isotope ratios (Brand 285 and Veizer 1980). Despite different diagenetic patterns around the SPICE and potential SPICE 286 intervals, both sections show similar δ^{18} O values at that level (between –4.5 and –5.75 ‰, Figures 287 7 and 8). These values are heavier than most of the samples that lie stratigraphically below or 288

above the potential SPICE at March Point. This may suggest that the original signature has been 289 290 preserved better around the SPICE and potential SPICE interval. A positive excursion was observed in $\delta^{13}C_{org}$ data from Avalonia (Woods et al. 2011), Australia, 291 China and North America (Iowa) (Saltzman et al. 2011). In Newfoundland this is also observed 292 although an offset between $\delta^{13}C_{org}$ and $\delta^{13}C_{carb}$ occurs in the March Point section and, to a lesser 293 degree, in the Felix Cove section (Figure 10). The isotope offset between $\delta^{13}C_{org}$ and $\delta^{13}C_{carb}$ curves 294 has been noted previously in Queensland, Australia and Iowa, USA by Saltzman et al. (2011). 295 There, $\delta^{13}C_{org}$ was found to track the $\delta^{13}C_{carb}$ curve on the rising limb, with $\delta^{13}C_{org}$ shifting back to 296 lighter values before peak SPICE in the inorganic data. Although the pattern is clear at March 297 298 Point, the reduced thickness of the Felix Member exposed at Felix Cove makes it more difficult to identify there. The highly variable trend in $\delta^{13}C_{org}$ below the potential SPICE at March Point is 299 300 most likely due to the low %C values in most of the samples. The SPICE is a global phenomenon and represents the characteristics of open ocean seawater at 301 302 that time. However, as shown earlier, the Petit Jardin Formation was deposited in a more marginal marine environment (Chow and James, 1987b) rather than open marine. Gilleaudeau and Kah, 303 (2013) have shown that carbonates deposited in epicratonic seas are typically 2-4 % lighter in δ^{13} C 304 than those deposited in open-marine pericratonic environments. Gilleaudeau and Kah (2013) 305 suggest that general mechanisms for this include ¹³C-depletion due to marine upwelling, ¹³C-306 307 depletion from terrigenous plant material (even in pre-Silurian environments from early bryophytes) or ¹³C-depletion from in situ organic remineralisation. 308 309 Local tectonics may also have played a part in reducing the extent of the CIE. It has been suggested 310 that local tectonics influenced the sedimentation patterns observed in the Port-au-Port peninsula in 311 the inner flexural wedge of the Laurentian cratonic margin (Cowan and James, 1993; Lavoie et al, 312 2012). Even for relatively adjacent locations such as Felix Cove and March Point (c. 25 km apart), 313 local tectonics could have accounted for the variations in input and timing of the clastics observed related to the SPICE and potential SPICE (Figure 9) and could have generated local, more 314 315 restricted environments. These environments would have had differences in energy (Knight and Boyce 2009) which again could have affected the geochemical signature (Gilleaudeau and Kah, 316 317 2013). 318 As the platforms evolved due to closure of Iapetus, Western Newfoundland and its surroundings, 319 located in southern Laurentia, became progressively more restricted whereas SPICE outcrops 320 situated on the west/northwest of Laurentia maintained open circulation (Figure 11). Likewise, 321 deposition in the southern Appalachians also occurred in a similar progressively restricted environment. The CIE in the southern Appalachians, like the SPICE and potential SPICE in 322 323 Newfoundland, is thinner with a reduced magnitude.

The isotopic signal observed in the Petit Jardin Formation therefore, could reflect superposition of 324 325 a regional and local signature onto ocean chemistry as a result of semi-restricted/restricted water 326 circulation in the Newfoundland area due to SL changes and closure of the Iapetus Ocean (Cowan 327 and James 1993, Patterson and Walter 1994, Saltzman et al. 2004). Hence, despite recording a positive excursion plausibly related to the SPICE, the Petit Jardin Formation may have imparted its 328 own isotopic signature onto the SPICE, driven by local tectonic, sedimentary and diagenetic 329 330 processes. 331 **Conclusions** 332 Two upper Cambrian sections in the Port-au-Port peninsula, Western Newfoundland have been studied and compared in detail (March Point and Felix Cove) These are predominantly carbonate in 333 334 lithology but contain specific sandstone horizons that occur in close proximity to bioherms. At March Point, this bioherm is of stromatolitic character and a pooly sorted glauconitic sandstone 335 336 layer occurs 3-4 m above it. At Felix Cove, the bioherm is thromboltic and directly overlain by a coarse glauconitic sandstone. The sandstones are therefore diachronous. 337 Carbon isotope data (from both carbonate carbon and organic carbon) show a positive excursion (at 338 339 the level which marks the boundary between Sauk II and III. Saltzman et al. (2004) presented data 340 for the section at Felix Cove which has been dated by trilobite fauna and identified as the SPICE. 341 New data from a potential SPICE horizon at March Point is presented in this work but no 342 biostratigraphic data are available. The $\delta^{13}C_{carb}$ excursion presented here and by Saltzman et al. (2004) is reduced in magnitude 343 344 compared to other global occurrences of the SPICE. This may be driven by local tectonic, 345 sedimentary and/or diagenetic processes. A restricted environment would result in a less well developed excursion rather than the expected global CIE due to ocean chemistry. The stratigraphic 346 347 and isotope variations observed in the Port-au-Port peninsula associated with the SPICE suggest 348 that local as well as global conditions were important in determining its character. 349 Acknowledgments 350 This work is a part of the PhD of the first author, developed at Universidade Federal do Rio Grande 351 352 do Sul (UFRGS) and University of Aberdeen (UoA). The authors gratefully acknowledge support from Shell Brasil through the "BG05: UoA-UFRGS-SWB Sedimentary Systems" project at 353 354 UFRGS and the strategic importance of the support given by ANP through the R&D levy 355 regulation. BGS and LAMIR staffs are also acknowledged for assisting with stable isotope

analysis. We also would like to tank Dr. Duncan McIlroy for all his help during the field work, and

- 357 Dr. Marc Laflamme, Dr. Pedro Jose Marenco and an anonymous reviewer for their help in
- 358 improving the manuscript.
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Table 1. Quartz grain size for March Point and Felix Cove sections.

	March	Point		Felix Cove	
Hight	222.3 m	222.5m	23 m	23.4 m	24 m
Average	88	89	473	466	456
Maximum	338	224	734	815	858
Minimum	37	36	116	227	138
Median	75	78	504	439	409
n	31	31	31	31	31

Table 2. March point section isotopic results.

Sample description	Sample height (m)	δ ¹³ C _{org} (% ₀)**	%C	δ ¹³ C _{carb} (‰)*	δ ¹⁸ O _{carb} (‰)*	Laboratory
Glauconitic grainstone	9.25	-		-0.96	-8.09	LAMIR
Grainstone	56.75	-		-1.79	-5.51	LAMIR
Grainstone	59.00	-		-1.32	-5.70	LAMIR
Grainstone	67.00	-		-0.41	-7.63	LAMIR
Grainstone	83.50	-		-1.80	-7.37	LAMIR
Grainstone	86.50	-		-1.26	-7.28	LAMIR
Grainstone	106.00	-		-2.46	-6.74	LAMIR
Grainstone	109.50	-		-3.84	-7.62	BGS
Grainstone interbedded with mudstone	115.25	-		-1.77	-6.55	LAMIR
Glauconitic grainstone	122.5	-29.2	0.2	-	-	BGS
Reddish grainstone interbedded with shale	126.00	-		-2.16	-8.01	LAMIR
Shale	132.50	-28.41	0.4	-	-	BGS
Shale	133	-27.9	0.3	-	-	BGS
Shale	133.50	-29.47		-	-	BGS
Grainstone interbedded with mudstone	136.25	-		-3.18	-7.38	BGS
Shale	136.5	-30.8		-	-	BGS
Grainstone interbedded with mudstone	137.00	-		-4.16	-7.57	LAMIR
Grainstone	140.25	-		-2.54	-8.19	LAMIR
Shale	145.00	-27.48	0.1			BGS

Glauconitic bioclastic grainstone	152.25	-		-1.29	-8.00	LAMIR
Glauconitic bioclastic grainstone	152.50	-		-0.82	-7.84	BGS
Shale	153.25	-28.71	0.1	-	-	BGS
Shale	154.25	-26.61	0.1	-	-	BGS
Shale	155.25	-27.50	0.1	-	-	BGS
Shale	157.25	-28.40	0.1	-	-	BGS
Shale	157.25	-28.70	0.1	-	-	BGS
Mudstone interbedded with shale	160.25	-26.68	0.3	-	-	BGS
Mudstone interbedded with shale	162.25	-27.07	0.1	-	-	BGS
Mudstone interbedded with shale	166.75	-26.79	0.04	-	-	BGS
Glauconitic grainstone	167.00	-		-0.90	-7.94	LAMIR
Glauconitic grainstone	167.50	-27.88	0.3	-	-	BGS
Glauconitic grainstone	167.75	-27.35	0.1	-	-	BGS
Mudstone	168.75	-27.82	0.3	-	-	BGS
Glauconitic grainstone	170.75	-28.01	1.9	-	-	BGS
Glauconitic grainstone interbedded with mudstone	174.00	-27.70	0.5	-	-	BGS
Glauconitic grainstone interbedded with mudstone	175.50	-		-1.16	-5.90	BGS
Mudstone	175.50	-27.97	0.2	-	-	BGS
Glauconitic grainstone interbedded with mudstone	176.50	-		-1.49	-6.72	LAMIR
Glauconitic grainstone interbedded with mudstone	176.50	-24.77	0.3	-	-	BGS
Parted limestone dolomitized	179.00	-		-0.84	-7.19	LAMIR
Dolomitized parted limestone	179.00	-		-0.82	-6.50	LAMIR
Dolomitized parted limestone	179.75	-25.67	0.4	-	-	BGS
Glauconitic grainstone	180.50	-27.36	2.1	-	-	BGS
Dolomitized oolitic grainstone	183.75	-30.02	0.2	-	-	BGS
Dolomitized oolitic grainstone	184.50	-		-1.72	-6.87	LAMIR
Dolomitized oolitic grainstone	189.50	-		-1.53	-6.56	LAMIR

Mudstone	194.00	-22.47	0.3	-	-	BGS
Dolomitized oolitic grainstone	194.25	-27.21	0.5	-	-	BGS
Dolomitized oolitic grainstone	194.75	-26.68	0.5	-	-	BGS
Dolomitized oolitic grainstone interbedded with mudstone	198.50	-28.24	0.1	-	-	BGS
Parted limestone	199.50	-28.55	0.04	-	-	BGS
Dolomitized oolitic grainstone	200.25	-		-1.61	-6.76	LAMIR
Dolomitized oolitic grainstone	201.50	-28.17	0.1	-	-	BGS
Dolomitized oolitic grainstone	206.00	-21.31	0.8	-	-	BGS
Dolomitized oolitic grainstone	208.00	-		-1.29	-6.66	LAMIR
Dolomitized oolitic grainstone	215.50	-		-0.68	-6.93	LAMIR
Dolomitized oolitic grainstone	218.50	-		-0.46	-5.27	BGS
Parted limestone	218.50	-22.55	0.5	-	-	BGS
Dolomitized oolitic grainstone	222.75	-		+1.13	-5.08	BGS
Dolomitized oolitic grainstone	223.60	-		+0.52	-4.58	BGS
Dolomitized oolitic grainstone	223.75	-		+0.79	-5.76	BGS
Parted limestone	224.75	-25.79	0.1	-	-	BGS
Parted limestone	225.50	-		-0.31	-4.75	BGS
Parted limestone	226.25	-		-0.18	-4.04	BGS
Parted limestone	226.25	-25.55	0.1	-	-	BGS
Parted limestone	226.75	-26.3	1.1	-	-	BGS
Glauconitic grainstone	230.00			-1.74	-7.73	BGS
Glauconitic grainstone	230.00	-26.24		-	-	BGS
Mudstone	230.75	-		-1.35	-8.18	BGS
Mudstone	231.50	-		-1.83	-8.47	BGS
Mudstone	233.50	-		-1.05	-7.95	BGS
Mudstone	235.00	-		-1.19	-8.08	BGS
Glauconitic grainstone	236.00	-		-1.73	-8.32	BGS

Glauconitic grainstone	236.00	-	-1.67	-8.62	BGS
Glauconitic grainstone	236.75	-	-2.01	-7.86	BGS
Grainstone	239.25	-	-1.07	-6.57	LAMIR
Grainstone	239.25	-23.05	-	-	BGS
Dolomitized oolitic grainstone	244.50	-	-1.38	-3.32	LAMIR

^{*}Overall analytical reproducibility for ^{13}C and ^{18}O (1 σ): BGS - 0.1%; LAMIR: not determined **Overall analytical reproducibility for $^{13}C_{org}$ (1SD): BGS - $\pm < 0.1\%$

Table 3. Felix Cove section isotopic results.

Sample description	Sample height (m)	δ ¹³ C _{org} (‰)**	%C	δ ¹³ C _{carb} (‰)*	δ ¹⁸ O _{carb} (‰)*	Laboratory
Grainstone	1.00	-		-2.0	-7.3	BGS
Grainstone	6.50	-		-1.3	-5.9	BGS
Shale	9.00	-26.6	1.0	-	-	BGS
Grainstone	13.00	-		-1.2	-7.0	BGS
Grainstone	16.80	-		-1.2	-7.5	BGS
Shale	17.25	-26.4	0.1	-	-	BGS
Shale	17.50	-27.5	0.17	-	-	BGS
Dolomitized oolitic grainstone	19.00	-		-0.5	-6.5	BGS
Dolomitized oolitic grainstone	22.25	-		-1.1	-9.0	BGS
Dolomitized oolitic grainstone	23.40	-		+0.4	-5.4	BGS
Dolomitized oolitic grainstone	24.00	-		+0.9	-5.4	BGS
Dolomitized oolitic grainstone	24.00	-		+0.87	-5.55	BGS
Dolomitized oolitic grainstone	25.50	-		+0.8	-5.4	BGS
Dolomitized oolitic grainstone	26.00	-		+1.02	-5.57	BGS
Shale	26.60	-23.5	0.01	-	-	BGS
Dolomitized oolitic grainstone	27.25	-		+0.9	-5.7	BGS
Dolomitized oolitic grainstone	27.50	-		+1.59	-5.76	BGS
Dolomitized oolitic grainstone	29.75	-		+1.83	-5.34	BGS

Shale	30.75	-25.7	0.04	-	-	BGS	
Shale	33	-25.0	0.04	-	-	BGS	
Dolomitized oolitic grainstone	35.25	-		+1.70	-5.46	BGS	
Shale	36.00	-26.1	0.19	-	-	BGS	
Mudstone	36.50	-		+1.10	-5.30	BGS	

^{*}Overall analytical reproducibility for $^{13}C_{carb}$ and ^{18}O (1 σ): BGS - 0.1% **Overall analytical reproducibility for $^{13}C_{org}$ (1 σ): BGS - \pm <0.1%

- 490 Figure 1. Record of the SPICE excursion and global chronostratigraphic framework (after
- 491 Saltzman et al. 2004).
- 492 Figure 2. Study area in the Port au Port Peninsula. Location of the logged sections are shown
- 493 by black rectangles: 1) March Point and 2) Felix Cove.
- 494 Figure 3. Schematic stratigraphic log of the Port au Port Group (after Chow, 1986). Each of
- 495 the Grand Cycles (A, B and C) consists of a lower shaley half-cycle mainly composed of
- 496 parted limestone (interbedded shale and mudstone) and an upper carbonate half-cycle of
- oolitic grainstone / bioherms. Estimated Sauk II-III sequence limit is indicated (Chow and
- 498 James 1987b, Saltzman et al. 2004).
- 499 Figure 4. Outcrop photos of SPICE-bearing layers: (a) Felix Cove outcrop at 22.5 m showing
- 500 the massive sandstone (S) that overlies the thrombolite layer at 22.25 m. Some intervals in the
- thrombolite contains vugs parallel to bedding that may also occur in the sandstone (arrow);
- 502 (b) Detail from Felix Cove of thrombolite at 22.25 m; (c) West Felix Cove outcrop with pillow
- thrombolites (T) and the overlying sandstone (S); (d) Detail from March Point of columnar
- stromatolite at 222.5 m; e) Detail of the dolomitized glauconitic grainstone (DG) above the
- stromatolite (STR); f) View of the outcrop with the dolomitized glauconitic grainstone (DG)
- overlying the stromatolite.
- Figure 5. Felix cove section: (a) Photomicrograph of the sandstone at 22.5m (peak SPICE),
- with quartz (Qz), crinoid bioclasts (Cr) and a few glauconite grains (G) and peloids (P),
- cemented by calcite (pinkish stain), with minor dolomite replacement (uncrossed polarizers)
- 510 (b) Photomicrograph of the equivalent sandstone layer in West Felix Cove cemented by
- 511 dolomite replacing calcite (pinkish stain), with minor dolomite replacement (uncrossed
- polarizers); March Point section: (c and d) Photomicrographs of dolomitized (D) glauconitic
- 513 (G) bioclastic (Cr) peloidal grainstone at 222.6m (peak SPICE). (d) Detail of the glauconite
- 514 (G) glauconite peloid partially replaced by dolomite (D) in (c); (e and f) Photomicrographs of
- 515 glauconitic (G) sandstone partially cemented by euhedral dolomite rhombs (D) at 226.5m.
- 516 Figure 6. Photomicrographs of March Point dolomitized layers. (a) Dolomitized grainstone at
- 517 189.5 m; (b) Dolomitized grainstone at 208 m with ghost of ooids (black arrows); (c)
- Dolomitized grainstone at 215.5m; (d) Quartzose (Q) arenite cemented by dolomite (D) at
- 519 **239.5 m.**
- Figure 7. March Point isotope results, showing $\delta^{13}C_{org}$, $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ isotopic data.
- 521 Dolomitized samples indicated in red.
- Figure 8. Felix Cove isotope results, showing $\delta^{13}C_{org}$, $\delta^{13}C_{carb}$ and $\delta^{18}O_{carb}$ isotopic data.
- 523 Dolomitized samples indicated in red.

524	$ Figure \ 9. \ Comparison \ between \ the \ expression \ of \ the \ SPICE \ in \ the \ March \ Point \ (1) \ and \ Felix $
525	Cove (2) sections. The grey shaded area indicates the correlation based on the carbonate
526	carbon signature.
527	Figure 10. $\delta^{13}C_{org}$ and $\delta^{13}C_{carb}$ data for A) March Point Sections and B) Felix Cove Section.
528	The comparison between the curves shows a peak of organic excursion a few meters below
529	(but nearly coincident with) the peak of the carbonate carbon excursion in the March Point
530	section, while in Felix Cove both peaks occurs at same level.
531	Figure 11. Laurentia paleogeographic map (Blakey 2016), showing its evolution between 510
532	Ma and 485 Ma. The red square indicates the position of western Newfoundland, yellow
533	square indicates the position of southern Appalachians (Glumac and Walker 1998), and black
534	squares indicates the positions of other SPICE localities in the United States (Nevada, Utah,
535	Iowa and Minnesota) (Saltzman 2004). Detail of Newfoundland (NF) paleogeography and
536	paleolatitudes are indicated on the map.





















