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SUPPLEMENTARY INFORMATION

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Limited contribution of permafrost carbon to methane release from thawing peatlands

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Description of vegetation communities and peat types

The presence of permafrost in these peatlands results in the peat surface being raised above the water table, allowing non-wetland plant communities to develop, at least on top of hummocks[1]. The peat plateaus in both Teslin and Yellowknife were forested, albeit relatively sparsely with small stature black spruce (*Picea mariana*) trees. In Teslin, the understorey vegetation on hummocks was dominated by dwarf shrubs, including *Ledum* spp, *Vaccinium vitis-idaea*, lichens and feather mosses including *Pleurozium schreberi*, and inter-hummock areas were dominated by relatively dry-adapted *Sphagnum* moss species including *S. capillifolium* and *S. fuscum*. Reflecting the difference in the hummock and inter-hummock plant communities, the underlying plateau peat was a mixture of relatively-humified, woody (sylvic) peat and poorly- or moderately-decomposed *Sphagnum* peat. This was the case for both the active layer and permafrost but with levels of humification generally increasing with depth. In the collapse wetland, peat that accumulated after thaw was composed of poorly-decomposed sedge-derived material to a depth of approximately 60 cm.

In Yellowknife, the plateau vegetation was similar, with the understorey vegetation on hummocks again dominated by lichen and dwarf shrubs including *Ledum spp*, and the inter-hummocks were dominated by *Sphagnum* mosses as well as *Rubus chamaemorus*. The plateau peat was predominantly moderately-decomposed *Sphagnum* peat with some woody material. In the collapse wetlands, post-thaw peat was dominated by green undecomposed moss with or without sedge shoots, with a clear transition to darker plateau peat at depths of 20 to 25 cm.

Experimental Design

Full-profile collars were inserted 40 cm into the wetland profile (Fig S1). Near-surface collars had sealed bases and were filled with 40 cm intact cores and then re-inserted into the wetland, thus excluding any contribution of CH_4 produced at depth. Probes were established at depths of 100 cm at Teslin and 65 cm at Yellowknife due to differences in the thickness of the sedge/moss peat layer that had developed since collapse at the two locations. This ensured that the probes projected approximately 40 cm into the underlying plateau peat and thus sampled from a depth that was roughly equivalent to the top of the permafrost before thaw took place (active-layer thickness on the plateaus was ~50 cm and plateaus peat will have been compacted slightly following collapse).

Intercomparison between DP-IR and Gas Chromatography

During the DP-IR measurements, fifteen ml samples of chamber headspaces were collected and injected into 12 ml pre-evacuated Exetainers® (Labco Ltd., Ceredigion, UK) prior to shipping back to the UK. These were subsequently analysed by gas chromatography (GC) at the Centre of Ecology and Hydrology (CEH) in Edinburgh, UK. Using a total of 75 samples with concentrations ranging from ~60 to ~1200 ppm, we carried out a comparison between the DP-IR and GC concentrations. Overall, there was strong agreement between the concentrations measured by the two approaches (Fig S2; y = 0.8068x + 8.673; R^2 = 0.975). The CH_4 concentrations were slightly lower when measured by the DP-IR (on average 18%). However, this same difference was also observed for exetainers which were filled with certified CH_4 standards in Canada and shipped back to the UK, with the GC measured concentrations being greater than the standards. Thus, it appears the over-pressurising of the vials and/or the shipping from Canada to the UK caused this constant small disagreement between methodologies. However, overall the DP-IR approach was demonstrated to be suitable for measuring fluxes in these ecosystems.

Environmental Monitoring

In Teslin in 2013, the mean air temperature was -0.6°C, whilst the precipitation was 267 mm (23% below 1981 to 2010 average). Despite this, the water table was persistently at or above the collapse wetland soil surface throughout the growing season, with an average depth of 2 cm (Fig. S3). In contrast, in Yellowknife in 2014, precipitation between June and September was 76.4 mm, of which 23.4 mm occurred in a single event at the end of August. This was 48% lower than the long-term average for this period (Environment Canada 2015). On average the water-table level was 16 cm below the surface, reaching a maximum of nearly 30 cm below surface, due to the dry weather conditions (Fig. S3).

Sensitivity analysis and calculation of maximum potential contribution of the previously-frozen C

The CH_4 released from near-surface collars was more enriched in bomb ^{14}C than the full-profile collars. This demonstrates that CH_4 derived from older sources below 40 cm was diluting the ^{14}C signature of CH_4 collected from the full-profile collars. The differences in ^{14}C content between full-profile and near-surface collars were used to calculate the contribution of CH_4 produced from previously-frozen C. Probe samples had highly variable ^{14}C contents, and modern signatures were observed in Yellowknife perhaps reflecting the fact that it was difficult to reliably sample water from the 65 cm probes given the water table was at 30 cm (Table S1). For these reasons, we used a sensitivity analysis approach to calculate the contribution of CH_4 derived from previously-frozen layers (Equation S1).

$$\mathrm{PF\ CH_4\ (\%)} = \left(\frac{\mathrm{FP^{14}CH_4 - NS^{14}CH_4}}{\mathrm{P}\mathit{age} - \mathrm{NS^{14}CH_4}} \right) *100 \qquad \text{(Equation S1)}$$

Where PF CH_4 (%) is the % contribution of previously-frozen C to the total CH_4 efflux, $FP^{14}CH_4$ is the ^{14}C content of the CH_4 collected from the Full-profile collars, $NS^{14}CH_4$ is the ^{14}C content of the CH_4 collected from the Near-surface collars, and P*age* is the ^{14}C content of previously-frozen C. P*age* was varied between ^{14}C contents of 92.80 %modern and 79.93 %modern, representing conventional radiocarbon ages of 600 and 1800 y BP.

The White River Ash tephra layer (1250 y BP[2]) was observed close to the base of the active layer, indicating that the minimum age of the organic matter at the top of the permafrost within the peat plateau was ~1200 BP. In addition, although variable, the average age of the CH_4 collected from the 1 m probes in Teslin was 1216 ± 213 y BP (Table S1; mean ± 1SE, N=9). Using 1200 y BP as *P*age (Equation S1), we calculated the contribution of previously-frozen C to the surface flux to be 1.7 g CH_4 m^{-2} during the growing season for Teslin and 1.0 g CH_4 m^{-2} during the growing season for Yellowknife. Although, the proportional contribution from old C was greater in Yellowknife, the total release of previously-frozen C was lower due to the smaller total CH_4 fluxes.

It is important to emphasise that our calculations represent maximum possible contributions of previously-frozen C. The sensitivity analysis presented in Fig. 4 demonstrates the effects uncertainty in *P*age has on the calculations. By using an age of 1200 y BP we use an age indicative of the top of the previously-frozen layer. Increasing the estimated age of the CH_4 released from previously-frozen organic matter reduces its proportional contribution to the flux (Fig. 4). If a proportion of the CH_4 being released was derived from deeper in the profile, as indicated by ages of up to 2800 y BP in the probe samples from Teslin, then the use of 1200 y BP would overestimate the total contribution of previously-frozen C. Furthermore, in Yellowknife, CH_4 was collected at a time when fluxes were low due to surface production being inhibited by the dry conditions. Therefore, multiplying the whole growing-season flux by the proportional contribution of previously-frozen C measured in late summer, likely overestimates previously-frozen C release; the proportional contribution of previously-frozen C would likely have been lower early in the year when near-surface layers were wetter and also contributing to methane release. Finally, seasonal ice in early summer may act as a barrier to CH_4 diffusion from depth. For this reason, we limited our measurements to late summer when all seasonal ice had melted, but again this could have resulted in an overestimation of the contribution of previously-frozen C early in the growing season.

In summary, our values therefore represent a maximum possible contribution of previously-frozen C, adding further confidence to our conclusion that rates of CH_4 release from the decomposition of previously-frozen C are low.

Testing the secondary hypotheses

Further to the main research aim of determining whether permafrost thaw promotes anaerobic decomposition of previously-frozen organic matter, site specific secondary-hypothesises were also tested:

Hypotheses 1: The rate of methane release from old, previously-frozen soil organic matter will be greater in more recently thawed peatland areas. In order to test the hypothesis, three sampling locations were established along a time since thaw gradient in the collapse wetland at Teslin.

Sampling locations:

- Margin: located in the immediate vicinity to collapsing plateau edge: The plots were established in the wetland as close as possible to the collapsing edge of the plateau, and in the first area in which sedge growth had been established.
- 5 m: located 5 m from the collapsing plateau margin. This area still contained standing dead trees and thus was part of the permafrost plateau in recent decades.

- Wetland centre: located within the centre of the unfrozen wetland: this area had no standing dead trees and ^{210}PB and radiocarbon dating of the sedge/plateau peat transition indicates that collapse occurred ~60 years ago.

CH_4 fluxes did not vary with time since the collapse. However, the ^{14}C content of CH_4 released at the margin sampling location (Fig. 3a) sampling location was significantly lower than at both 5 m into the wetland (LSD post-hoc test, following repeated measures two-way ANOVA: $P = 0.012$) and in the wetland centre ($P = 0.030$). This may have been due to the fact that collapse here occurred after the 1960s peak in atmospheric ^{14}C caused by nuclear weapons testing, and thus the accumulating sedge peat is less enriched in ^{14}C than further into the centre of the wetland. By replacing *P*age in equation 1 with the average age of the three probe-collected CH_4 samples in each area (Table S1), we calculated contributions of previously-frozen C of 8.9% at the margin, 6.9% at 5 m, and 10.1% in the wetland centre. These differences were not statistically significant (one-way ANOVA: $P = 0.875$) and were caused by differences in the probe sample ^{14}C contents with the 2800 y BP sample being collected from the 5 m area (Table S1). As Fig. 4 demonstrates, greater *P*age results in lower calculated contributions of previously-frozen C. If the average probe age for the whole wetland was used then the contributions of previously-frozen C were 8.1 %, 8.4 % and 8.7 % for the margin, 5 m and wetland centre, respectively ($P = 0.998$). Therefore, despite the lower absolute ^{14}C contents of the CH_4 released at the margin, we did not observe a significant difference in the contribution of previously-frozen C to the CH_4 flux, and, thus contrary to our hypothesis, there was no significant effect of time since collapse on the amount of previously-frozen C being released as CH_4.

Hypothesis 2: We predicted that methane release from old, previously-frozen soil organic matter will be greater in areas dominated by sedges, due to the potential for rapid transport through aerenchyma (air-filled channels in the sedge roots), thus reducing the potential for oxidation in the surface layers of the wetland. We investigated this in Yellowknife, by establishing sampling locations in collapse wetlands dominated by either sedge (*Carex rostrata*) or moss (*Sphagnum spp.*). However, we observed no significant differences in the ^{14}C content of the CH_4 being released from the two different vegetation communities (two-way repeated-measures ANOVA: $P = 0.982$). Therefore, we did not find a significant difference in the proportional contribution of previously-frozen C to the surface fluxes. The greater total flux from the sedge-dominated collapse wetlands may suggest that the amount of previously-frozen C being released was greater in these wetlands but overall the data did not generate strong support for this hypothesis.

References

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Table S1 | Summary of the ^{14}C contents, $\delta^{13}C$ ratios and radiocarbon ages for all CH_4 samples collected from a. Teslin and b. Yellowknife. NERC Radiocarbon Facility laboratory analysis codes are included.

a.

Study site	Collar Type	Sampling Location Code	Replicate Number	^{14}C Enrichment (% Modern)	Conventional Radiocarbon Age (years BP)	$\delta^{13}C$ VPDB‰	Lab Code
Teslin	Full-profile	Margin	1	102.48	*modern*	-57.7	SUERC-49663
Teslin	Full-profile	Margin	2	102.74	*modern*	-57.9	SUERC-49667
Teslin	Full-profile	Margin	3	102.06	*modern*	-57.6	SUERC-49673
Teslin	Near-surface	Margin	1	104.1	*modern*	-54.8	SUERC-49664
Teslin	Near-surface	Margin	2	103.22	*modern*	-57.4	SUERC-49670
Teslin	Near-surface	Margin	3	104.25	*modern*	-54.2	SUERC-49674
Teslin	Probes	Margin	1	87.34	1087	-54.5	SUERC-49667
Teslin	Probes	Margin	2	84.61	1343	-55.1	SUERC-49673
Teslin	Probes	Margin	3	91.33	728	-54.2	SUERC-49676
Teslin	Full-profile	5 m	1	103.81	*modern*	-55.4	SUERC-49677
Teslin	Full-profile	5 m	2	104.36	*modern*	-61.1	SUERC-49683
Teslin	Full-profile	5 m	3	103.55	*modern*	-54.8	SUERC-49687
Teslin	Near-surface	5 m	1	105.4	*modern*	-46.6	SUERC-49680
Teslin	Near-surface	5 m	2	105.26	*modern*	-51.5	SUERC-49684
Teslin	Near-surface	5 m	3	105.92	*modern*	-53.7	SUERC-49690
Teslin	Probes	5 m	1	85.65	1244	-54.1	SUERC-49682
Teslin	Probes	5 m	2	89.84	861	-53.4	SUERC-49686
Teslin	Probes	5 m	3	70.51	2806	-57.4	SUERC-49692
Teslin	Full-profile	Wetland Centre	1	103.73	*modern*	-52.6	SUERC-49693
Teslin	Full-profile	Wetland Centre	2	104.67	*modern*	-55.3	SUERC-49697
Teslin	Full-profile	Wetland Centre	3	103.27	*modern*	-52.6	SUERC-49703
Teslin	Near-surface	Wetland Centre	1	107.3	*modern*	-45.6	SUERC-49694
Teslin	Near-surface	Wetland Centre	2	103.62	*modern*	-49.7	SUERC-49700
Teslin	Near-surface	Wetland Centre	3	105.82	*modern*	-45.9	SUERC-49704
Teslin	Probes	Wetland Centre	1	89.48	893	-54.2	SUERC-49696
Teslin	Probes	Wetland Centre	2	85.49	1260	-54.5	SUERC-49702
Teslin	Probes	Wetland Centre	3	91.4	722	-52.5	SUERC-49706

b.

Study site	Collar Type	Sampling Location Code	Replicate Number	^{14}C Enrichment (% Modern)	Conventional Radiocarbon Age (years BP)	$\delta^{13}C$ VPDB‰	Lab Code
Yellowknife	Full-profile	Sedge	1	90.85	771	-58.4	SUERC-56844
Yellowknife	Full-profile	Sedge	2	101.66	*modern*	-54.7	SUERC-56847
Yellowknife	Full-profile	Sedge	3	90.57	796	-40.7	SUERC-56850
Yellowknife	Near-surface	Sedge	1	97.85	174	-47.6	SUERC-56845
Yellowknife	Near-surface	Sedge	2	93.2	565	-46.7	SUERC-56848
Yellowknife	Near-surface	Sedge	3	101.54	*modern*	-47.5	SUERC-56853
Yellowknife	Probes	Sedge	1	93.87	508	-56.5	SUERC-56846
Yellowknife	Probes	Sedge	2	98.13	152	-55.2	SUERC-56849
Yellowknife	Probes	Sedge	3	90.49	803	-58.9	SUERC-56855
Yellowknife	Full-profile	Moss	1	92.97	586	-61.5	SUERC-56833
Yellowknife	Full-profile	Moss	2	103.41	*modern*	-54.5	SUERC-56836
Yellowknife	Full-profile	Moss	3	91.43	719	-32.6	SUERC-56839
Yellowknife	Near-surface	Moss	1	97.98	164	-62.9	SUERC-56834
Yellowknife	Near-surface	Moss	2	101.46	*modern*	-54.1	SUERC-56837
Yellowknife	Near-surface	Moss	3	98.35	133	-43.7	SUERC-56840
Yellowknife	Probes	Moss	1	98	163	-74.6	SUERC-56835
Yellowknife	Probes	Moss	2	97.12	235	-65.8	SUERC-56838
Yellowknife	Probes	Moss	3	97.4	212	-57	SUERC-56843

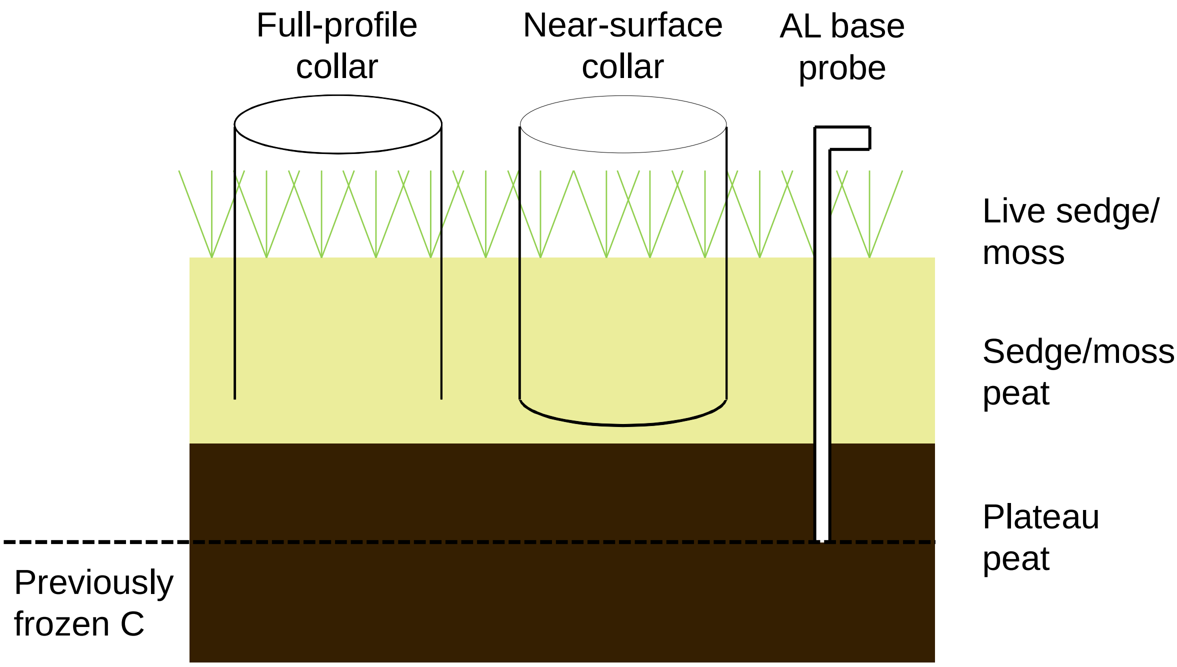

Figure S1 | Schematic of experimental set up for sampling CH_4 for radiocarbon analysis to calculate the maximal contribution of previously frozen C. The full-profile collars were inserted 40 cm while the near-surface collars contained intact cores that were extracted and reinserted into sealed cylinders within the peat profile. The probes were inserted through the sedge/moss peat, which had formed since the peat plateau collapsed, and then 40 cm into the underlying plateau peat. This represented approximately the depth at which the peat was previously frozen (base of the active layer).

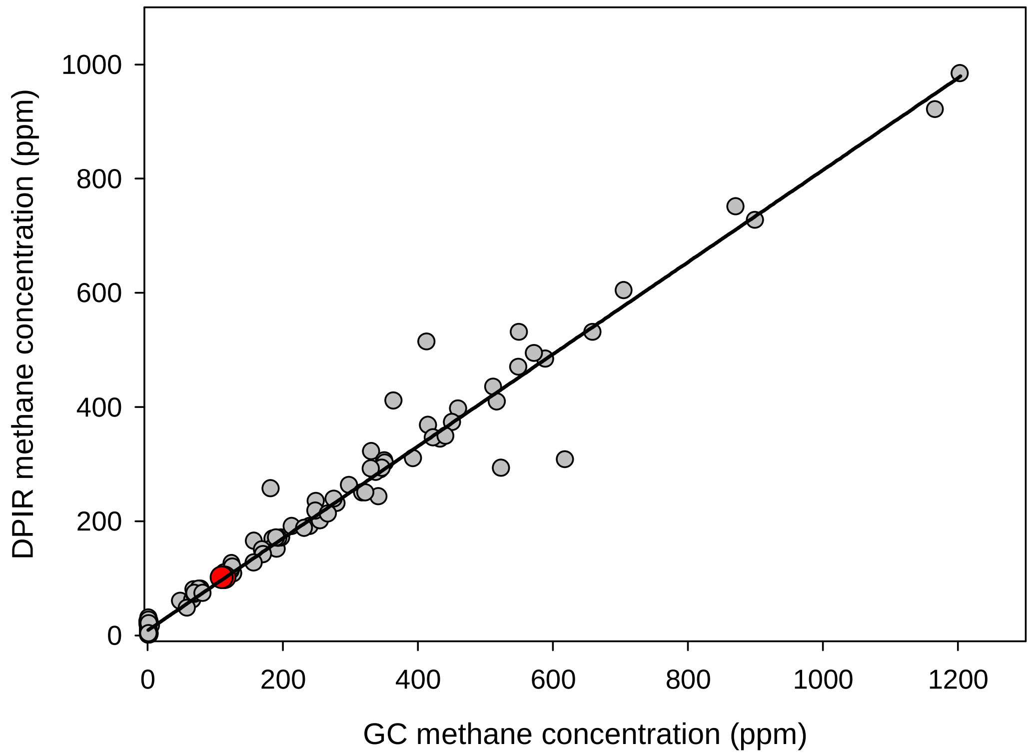

Figure S2 | Correlation between methane concentrations measured in chamber headspaces by the DP-IR and methane concentrations measured by GC analyses on samples collected from the headspaces at the time of the DP-IR measurements. The red circles indicate certified standard gas samples (100 ppm) which were also injected into exetainers in Canada and shipped to the UK.

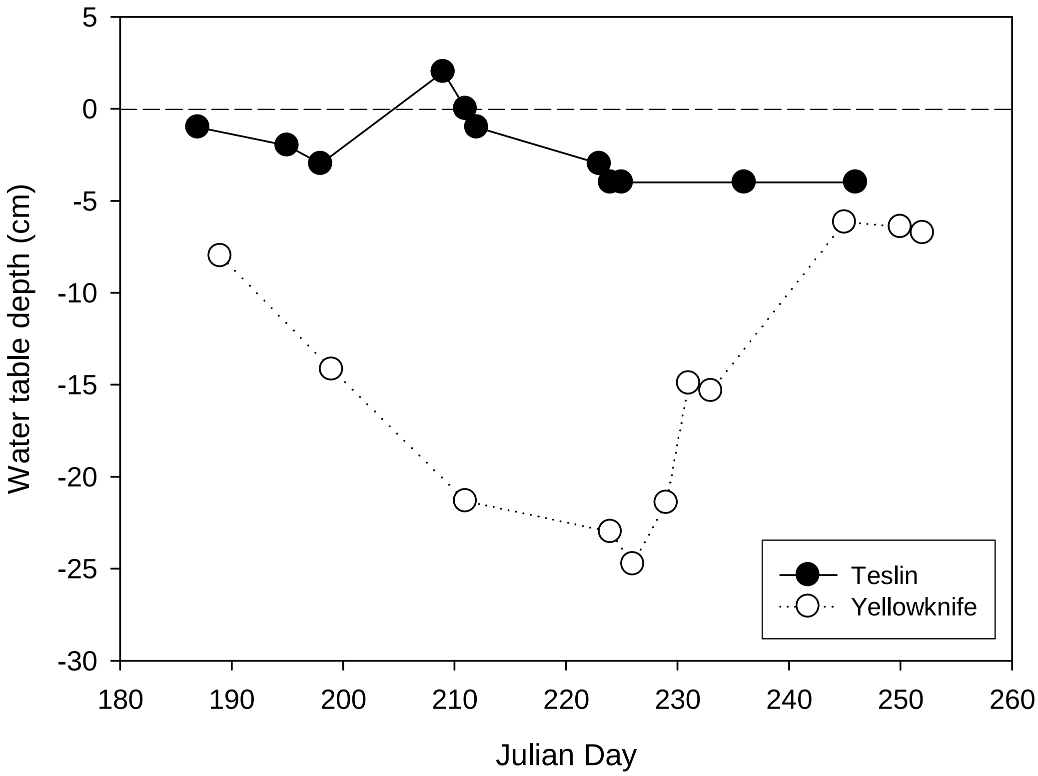

Figure S3 | Seasonal changes in mean water table depth at Teslin in 2013 and Yellowknife in 2014. For Teslin there was only one measurement location because a single collapse wetland with a uniform water table was being measured. In Yellowknife, the mean of three sedge and three moss collapse wetlands is presented (n = 6; standard errors were on average 1.40 cm and thus too small to be seen clearly on the graph).