Microbial iron(III) reduction during palsa collapse promotes greenhouse gas emissions before complete permafrost thaw

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The data contained within this data set is arranged according to the main text and the supplementary information of this publication. Details of the files are outlined below:

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Background information

Field site: Stordalen mire, Abisko, Sweden (68 22′ N, 19 03′ E)

Along palsa hillslopes: Palsa to front

Thaw stages: Palsa, bog and fen

Type of samples: Gas samples, porewater samples, soil core samples

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Main text

1) Gas analysis

This file contains the gas analysis obtained by the gas chambers along the palsa hillslope (68°21'18.70"N, 19° 2'38.00"E), from palsa with an active layer and permafrost underneath, to the collapsing front into bog, water-saturated soil with permafrost below. The file consists of one sheet:

(A) Carbon dioxide and methane

Two columns per each stage along the palsa hillslope (palsa, front, bog) contain

For carbon dioxide:

- Carbon dioxide [$\mu mol/m^2/sec$] (average of gas measurements collected on three days in July 2019 at three separate time points)
- Error bars (standard deviation of gas measurements collected on three days in Uly 2019 at three separate time points)

For methane:

- Methane [µmol/m^2/sec] (average of gas measurements collected on three days in July 2019 at three separate time points)
- Error bars (standard deviation of gas measurements collected on three days in July 2019 at three separate time points)

Method used: used plastic chambers with metal frames sealed with a rubber stopper; gas samples from the field and standard gases used for calibration measured with a gas chromatograph (Hewlett Packard, 5890 Series II) equipped with an electron capture detector (^{63}Ni-ECD)

2) Porewater analysis

This file contains the porewater analysis from 30 cm soil depth obtained by lysimeters along the palsa hillslope (68°21'18.70"N, 19° 2'38.00"E) in July 2019, from palsa with an active layer and permafrost underneath, to the collapsing front into bog, water-saturated soil with permafrost below. The file consists of one sheet:

(A) DOC and Fe

Three columns per each stage along the palsa hillslope (palsa (3x), front (1x), bog (3x)) contain

For dissolved organic carbon (DOC):

- Distance [m]
- Dissolved organic carbon (DOC) [mg/L] (average of eight palsa to bog hillslopes samples in June/July 2019)
- Error bars (standard deviation of eight palsa to bog hillslopes sampled in June/July 2019)

For aqueous iron(II) (Fe^{2+}):

- Distance [m]
- Aqueous Fe^{2+} [mg/L] (average of eight palsa to bog hillslopes samples in June/July 2019)

- Error bars (standard deviation of eight palsa to bog hillslopes sampled in June/July 2019)

Method used for iron in the porewater: by ferrozine assay

Method used for DOC in the porewater: total organic carbon analyzer (High TOC II, Elementar, Elementar Analysensysteme GmbH, Germany); inorganic carbon was removed by acidifying the samples with 2M HCl

3) Relative 16S rRNA gene sequence abundance

This file contains the relative 16S rRNA gene amplicon sequencing abundance (DNA-based) of iron- and methane-cycling microorganisms per each soil horizon (organic horizon, transition zone, mineral horizon) along the palsa hillslope (68°21'27.33"N, 19° 3'1.91"E), from intact palsa (Palsa A) to more collapsed palsa (Palsa B) to collapsing front (Front). The file consists of one sheet:

A) Iron- and methane-cycling microorganisms

Eight columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

- Fe(II)-oxidizing microorganisms (FeOx) [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Fe(III)-reducing microorganisms (FeRed) [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Methanogens [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Methanotrophs [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

Method used: A Naive Bayes classifier fitted with 16S rRNA (gene) sequences extracted with the PCR primer sequences from the QIIME compatible, 99%-identity clustered SILVA v132 database; amplicon sequencing variants (ASVs) classified by taxon using the fitted classifier; 225 ASVs that classified as chloroplasts or mitochondria removed, totalling to < 7% (average 0.6%) relative abundance per sample, and the remaining 9,351 ASVs had their abundances extracted by feature-table (https://github.com/qiime2/q2-feature-table)

4) Relative 16S rRNA (gene) sequence abundance

This file contains the relative 16S rRNA (gene) amplicon sequencing abundance (DNA- and RNA-based) of specific iron- and methane-cycling microorganisms per each soil horizon (organic horizon, transition zone, mineral horizon) along the palsa hillslope (68°21'27.33"N, 19° 3'1.91"E), from intact palsa (Palsa A) to more collapsed palsa (Palsa B) to collapsing front (Front). The file consists of four sheets:

    A) DNA-based % of iron-cycling microorganisms

Eight columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For iron(III)-reducing microorganisms (DNA-based):
- *Geobacter* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Clostridium* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Rhodoferax* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Myxococcales* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

Six columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For iron(II)-oxidizing microorganisms (DNA-based):
- *Gallionella* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Sideroxydans* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Other Gallionellaceae* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

B) RNA-based % of iron-cycling microorganisms

Eight columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For iron(III)-reducing microorganisms (RNA-based):

- *Geobacter* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Clostridium* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Rhodoferax* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Myxococcales* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

Six columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For iron(II)-oxidizing microorganisms (RNA-based):

- *Gallionella* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Sideroxydans* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Other Gallionellaceae* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

C) DNA-based % of methane-cycling microorganisms

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For methanogenic microorganisms (DNA-based):
- *Methanobacterium* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Bathyarchaeia* average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

For methanotrophic microorganisms (DNA-based):
- *Roseiarcus* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Other Beijerinckiacaeae* average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

D) RNA-based % of methane-cycling microorganisms

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For methanogenic microorganisms (RNA-based):

- *Methanobacterium* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- *Bathyarchaeia* average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

For methanotrophic microorganisms (RNA-based):

- *Roseiarcus* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- *Other Beijerinckiacaeae* average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

Method used: See also 3) A) method used; list of iron- and methane-cycling microorganisms manually compiled by cross-checking known iron- and methane-cycling microorganisms

5) Solid phase analysis

This file contains the solid phase analysis (reactive iron (Fe) and reactive Fe-associated organic carbon (OC)) per each soil horizon (organic horizon, transition zone, mineral horizon) along the palsa hillslope (68°21'18.70"N, 19° 2'38.00"E), from intact palsa (Palsa A) to more collapsed palsa (Palsa B) to collapsing front (Front). The file consists of two sheets:

A) Reactive Fe-associated OC

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

- Reactive iron (Fe) (control corrected) [mg/g soil] (duplicate analysis)
- Error bars (combined standard deviation of sodium chloride bicarbonate extractable Fe (control) and sodium dithionite citrate extractable Fe (not control corrected))
- Associated organic carbon (OC) (control corrected) [mg/g soil] (duplicate analysis)
- Error bars (combined standard deviation of the citrate blank, sodium chloride bicarbonate extractable OC (control) and sodium dithionite citrate extractable OC (not control corrected))

B) Control (sodium chloride)

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

- Leachable iron (Fe) = sodium chloride bicarbonate extractable Fe (control, same ionic strength and pH as the dithionite/citrate extraction) [mg/g soil] (duplicate analysis)
- Error bars (range of duplicate analyses of each soil horizon in each thaw stage)
- Leachable organic carbon (OC) = sodium chloride bicarbonate extractable OC (control, same ionic strength and pH as the dithionite/citrate extraction) [mg/g soil] (duplicate analysis)
- Error bars (range of duplicate analyses of each soil horizon in each thaw stage)

Method used for iron in the extracts: by ferrozine assay
Method used for DOC in the extracts: total organic carbon analyzer (High TOC II, Elementar, Elementar Analysensysteme GmbH, Germany); inorganic carbon was removed by acidifying the samples with 2M HCl

6) Fine fraction analysis

Raw data of nanoSIMS analysis of the fine fraction of Palsa A and Front (68°21'18.70"N, 19°2'38.00"E)

(A) Palsa A (transition zone) (4 fine particles) (depth_p3_2.im)

(B) Palsa A (mineral horizon) (1 fine particle) (S19144_MinA_2_4.im)

(C) Front (transition zone) (4 fine particles) (S19147_TransC_5_2.im)

(D) Front (mineral horizon) (1 fine particle) (S19148_MinC_6_3.im)

Method used: performed at the Cameca nanoSIMS 50L of the Chair of Soil Science (TU München, Germany); primary beam (~1.2 pA) was focused at a lateral resolution ~100 nm and scanned over the sample with 12C-, 16O-, 12C14N-, 31P-, 32S-, 27Al16O- and 56Fe16O- secondary ions collected using electron multipliers; the nanoSIMS images were analyzed using the Open MIMS Image plugin available within ImageJ

7) Redox properties and molecular weight of dissolved organic carbon

This file contains the redox properties and molecular weight analysis of dissolved organic carbon (OC) along the palsa hillslope (68°21'18.70"N, 19° 2'38.00"E), from palsa with an active layer and permafrost underneath, to the collapsing front into bog, water-saturated soil with permafrost below. The file consists of one sheet:

(A) Redox properties and molecular weight (MW)

Three columns per each stage along the palsa hillslope (palsa (3x), front (1x), bog (3x)) contain

For the nominal oxidation state of carbon = NOSC:
- Distance [m]
- Average (duplicate analysis)
- Error bars (range of duplicate analysis)

For the double bound equivalence = DBE:
- Distance [m]
- Average (duplicate analysis)
- Error bars (range of duplicate analysis)

For the molecular weight = MW:

- Distance [m]
- MW Average (duplicate analysis)
- Error bars (range of duplicate analysis)

Method used: modified aromaticity index (ModAI) was calculated, and nominal oxidation state of carbon (NOSC) was calculated according to La Rowe&Van Cappellen. Data processing post formula assignment was performed with RStudio utilizing R software (V4.0.3)

8) Acetate analysis

This file contains the acetate analysis from porewater of 30 cm soil depth obtained by lysimeters along the palsa hillslope (68°21'18.70"N, 19° 2'38.00"E), from palsa with an active layer and permafrost underneath, to the collapsing front into bog, water-saturated soil with permafrost below. The file consists of one sheet:

(A) Acetate

Three columns per each stage along the palsa hillslope (palsa (3x), front (1x), bog (3x)) contain
- Distance [m]
- Acetate [mg C/L] (average of eight palsa to bog hillslopes samples in June/July 2019)
- Error bars (standard deviation of eight palsa to bog hillslopes sampled in June/July 2019)

Method used: See also 2) A) method used. High performance liquid chromatography (HPLC; class VP with refractive index detector [RID] 10A and photo-diode array detector SPD-M10A VP detectors; Shimadzu, Japan) was used to determine the fatty acid concentrations

Supplementary Information

1) Gas analysis

This file contains the gas analysis obtained by the Eosense gas chambers along the palsa hillslope (68°21'18.70"N, 19° 2'38.00"E), from palsa with an active layer and permafrost underneath, to the collapsing front into bog, water-saturated soil with permafrost below. The file consists of two sheets:

A) Carbon dioxide emissions from palsa

Five columns per stage palsa:

- Month
- Day
- Year
- Time [hour:min:sec]
- Flux [$\mu mol/m^2/sec$]

B) Carbon dioxide emissions from bog

Five columns per stage bog:

- Month
- Day
- Year
- Time [hour:min:sec]
- Flux [$\mu mol/m^2/sec$]

Method used: performed with two eosense instruments (eosFD Forced Diffusion chamber in conjunction with the eosLink-FD software, EOSENSE INC, Dartmouth, Canada)

2) Porewater analysis (transition zone)

This file contains the porewater analysis of additional elements (phosphorous, sulphur, ammonium) from 30 cm soil depth obtained by lysimeters along the palsa hillslope (68°21'18.70"N, 19° 2'38.00"E), from palsa with an active layer and permafrost underneath, to

the collapsing front into bog, water-saturated soil with permafrost below. The file consists of one sheet:

A) Total phosphorous, total sulphur and ammonium

Three columns per each stage along the palsa hillslope (palsa (3x), front (1x), bog (3x)) contain

For dissolved total phosphorous:
- Distance [m]
- Total phosphorous [mg/L] (average of eight palsa to bog hillslopes samples in June/July 2019)
- Error bars (standard deviation of eight palsa to bog hillslopes sampled in June/July 2019)

For total sulphur:
- Distance [m]
- Total sulphur [mg/L] (average of eight palsa to bog hillslopes samples in June/July 2019)
- Error bars (standard deviation of eight palsa to bog hillslopes sampled in June/July 2019)

For ammonium:
- Distance [m]
- Ammonium [mg/L] (average of eight palsa to bog hillslopes samples in June/July 2019)
- Error bars (standard deviation of eight palsa to bog hillslopes sampled in June/July 2019)

Method used: Phosphorous (P) and sulphur (S) concentrations were measured with ICP-MS (inductively coupled plasma mass spectrometry); a flow injection analysis (FIA) instrument equipped with a dialysis membrane for removal of Fe to prevent side reactions during measurement (Seal Analytical, Germany) was applied for quantification of NH_4^+, NO_3^- and NO_2^- concentrations

3) Porewater analysis (mineral horizon)

This file contains the porewater analysis of from deeper soil horizons (60 cm soil depth) obtained by lysimeters along the palsa hillslope (68°21'18.70"N, 19° 2'38.00"E), from palsa with an active layer and permafrost underneath, to the collapsing front into bog, water-saturated soil with permafrost below. The file consists of two sheet:

A) DOC and Fe

Three columns per each stage along the palsa hillslope (palsa (2x), front (1x), bog (3x)) contain

For dissolved organic carbon (DOC):
- Distance [m]
- Dissolved organic carbon (DOC) [mg/L] (average of eight palsa to bog hillslopes samples in June/July 2019)
- Error bars (standard deviation of eight palsa to bog hillslopes sampled in June/July 2019)

For aqueous iron(II) (Fe^{2+}):
- Distance [m]
- Aqueous Fe^{2+} [mg/L] (average of eight palsa to bog hillslopes samples in June/July 2019)
- Error bars (standard deviation of eight palsa to bog hillslopes sampled in June/July 2019)

Method used for iron in the porewater: by ferrozine assay
Method used for DOC in the porewater: total organic carbon analyzer (High TOC II, Elementar, Elementar Analysensysteme GmbH, Germany); inorganic carbon was removed by acidifying the samples with 2M HCl

B) Total phosphorous, total sulphur and ammonium

Three columns per each stage along the palsa hillslope (palsa (2x), front (1x), bog (3x)) contain

For dissolved total phosphorous:

- Distance [m]
- Total phosphorous [mg/L] (average of eight palsa to bog hillslopes samples in June/July 2019)
- Error bars (standard deviation of eight palsa to bog hillslopes sampled in June/July 2019)

For total sulphur:

- Distance [m]
- Total sulphur [mg/L] (average of eight palsa to bog hillslopes samples in June/July 2019)
- Error bars (standard deviation of eight palsa to bog hillslopes sampled in June/July 2019)

For ammonium:

- Distance [m]
- Ammonium [mg/L] (average of eight palsa to bog hillslopes samples in June/July 2019)
- Error bars (standard deviation of eight palsa to bog hillslopes sampled in June/July 2019)

Method used: Phosphorous (P) and sulphur (S) concentrations were measured with ICP-MS (inductively coupled plasma mass spectrometry); a flow injection analysis (FIA) instrument equipped with a dialysis membrane for removal of Fe to prevent side reactions during measurement (Seal Analytical, Germany) was applied for quantification of NH_4^+, NO_3^- and NO_2^- concentrations

4) Copy numbers

This file contains the gene copy numbers of bacteria and archaea based on qPCR analysis specific for 16S rRNA genes of each soil horizon (organic horizon, transition zone, mineral horizon along the palsa hillslope (palsa to front) (68°21'27.33"N, 19° 3'1.91"E). The file consists of two sheet:

A) Gene copy numbers of bacteria and archaea – DNA-based

Three columns per each stage along the palsa hillslope (palsa (2x), front (1x)) contain the copy numbers of bacteria and archaea for each soil horizon (organic horizon, transition zone, mineral horizon):

- Average [Copy numbers per g soil] (average of triplicate analysis per each stage and soil horizon)
- Error bars (standard deviation of triplicate analysis per each stage and soil horizon)

B) Gene copy numbers of bacteria and archaea – RNA-based

Three columns per each stage along the palsa hillslope (palsa (2x), front (1x)) contain the copy numbers of bacteria and archaea for each soil horizon (organic horizon, transition zone, mineral horizon):

- Average [Copy numbers per g soil] (average of triplicate analysis per each stage and soil horizon)
- Error bars (standard deviation of triplicate analysis per each stage and soil horizon)

Method used: Copy numbers based on qPCR analysis specific for 16S rRNA genes (DNA- and RNA-based)

5) Taxonomic identification of the microbial communities along the palsa hillslope based on 16S rRNA gene amplicon analysis (DNA-based)

This file contains the taxonomic identification of the microbial communities along the palsa hillslope (68°21'27.33"N, 19° 3'1.91"E) based on 16S rRNA gene amplicon analysis (DNA-based). The file consists of one sheet:

A) Microbial communities

Sixteen columns per each stage along the palsa hillslope (palsa (2x), front (1x)) and each soil horizon (organic horizon, transition zone, mineral horizon) contain

- Crenarchaeota
- Euryarchaeota
- Acidobacteria
- Actinobacteria

- Armatimonadetes

- Bacteroidetes

- Chloroflexi

- Cyanobacteria

- Firmicutes

- Gemmatimonadetes

- Nitrospirae

- Patescibacteria

- Planctomycetes

- Proteobacteria

- Verrucomicrobia

- Others

Method used: Based on 16S rRNA gene amplicon analysis (DNA-based), data were averaged among triplicate analysis of each soil horizon

6) C/N weight ratios

This file contains the C/N weight ratios of the three soil horizons along the palsa hillslope (68°21'18.70"N, 19° 2'38.00"E) and thaw gradient and the C/N weight ratios of living plant samples of dominant Stordalen species (values obtained by Hodgkins et al., 2014: Hodgkins, S.B., Tfaily, M.M., McCalley, C.K., Logan, T.A., Crill, P.M., Saleska, S.R., Rich, V.I., Chanton, J.P. Changes in peat chemistry associated with permafrost thaw increase greenhouse gas production. *Proc Natl Acad Sci U S A* **111**(16), 5819-5824 (2014)). The file consists of two sheets:

A) C/N weight ratios of soil samples of three soil horizons along the palsa hillslope and thaw stage

One column per each stage along the palsa hillslope (palsa (2x), front (1x)) and thaw stage (bog (1x) and fen (1x)) contains per each soil horizon (organic horizon, transition zone, mineral horizon):

- C/N weight ratio (average of triplicate analysis of each soil horizon for cores Palsa A, Palsa B, Front (transect 1), one bog (Bog C) and one fen core (Fen E) (see also Patzner et al., 2020: Patzner, M.S., Mueller, C.W., Malusova, M. *et al.* Iron mineral dissolution releases iron and associated organic carbon during permafrost thaw. *Nat Commun* **11,** 6329 (2020)).

- Error bars (standard deviation of triplicate analysis of each soil horizon for cores Palsa A, Palsa B, Front (transect 1), one bog (Bog C) and one fen core (Fen E) (see also Patzner et al., 2020: Patzner, M.S., Mueller, C.W., Malusova, M. *et al.* Iron mineral dissolution releases iron and associated organic carbon during permafrost thaw. *Nat Commun* **11,** 6329 (2020)).

B) C/N weight ratios of living plants of dominant Stordalen species (values obtained from Hodgkins et al., 2014)

Three columns per each stage along the palsa hillslope and thaw stage contain

- Plant species (*Eriophorum vaginatum, Spaghnum* spp., *Eriophorum angustifolium, Carex* spp.)
- C/N weight ratio
- Error bars
-

Method used: quantified in triplicate with a total organic carbon and nitrogen analyzer (High TOC II, Elementar, Elementar Analysensysteme GmbH, Germany); inorganic carbon was removed by acidifying the samples with 2M HCl

7) Replicate analysis of relative 16S rRNA (gene) sequence abundance

This file contains the replicate analysis of relative 16S rRNA (gene) amplicon sequencing abundance (DNA- and RNA-based) of specific iron- and methane-cycling microorganisms per each soil horizon (organic horizon, transition zone, mineral horizon) along the palsa hillslope (68°21'27.33"N, 19° 3'1.91"E), from intact palsa (Palsa A) to more collapsed palsa (Palsa B) to collapsing front (Front). The file consists of four sheets:

A) DNA-based % of iron-cycling microorganisms

Eight columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For iron(III)-reducing microorganisms (DNA-based):

- *Geobacter* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Clostridium* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Rhodoferax* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Myxococcales* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

Six columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For iron(II)-oxidizing microorganisms (DNA-based):

- *Gallionella* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Sideroxydans* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- *Other Gallionellaceae* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

B) RNA-based % of iron-cycling microorganisms

Eight columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For iron(III)-reducing microorganisms (RNA-based):

- *Geobacter* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- *Clostridium* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- *Rhodoferax* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- *Myxococcales* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

Six columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For iron(II)-oxidizing microorganisms (RNA-based):

- *Gallionella* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Sideroxydans* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Other Gallionellaceae* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

C) DNA-based % of methane-cycling microorganisms

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For methanogenic microorganisms (DNA-based):
- *Methanobacterium* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Bathyarchaeia* average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

For methanotrophic microorganisms (DNA-based):
- *Roseiarcus* spp. average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

- *Other Beijerinckiacaeae* average [DNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

D) RNA-based % of methane-cycling microorganisms

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

For methanogenic microorganisms (RNA-based):
- *Methanobacterium* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Bathyarchaeia* average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

For methanotrophic microorganisms (RNA-based):
- *Roseiarcus* spp. average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- *Other Beijerinckiacaeae* average [RNA-based %] (average of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

Method used: See also Main text 3) A) method used; list of iron- and methane-cycling microorganisms manually compiled by cross-checking known iron- and methane-cycling microorganisms

8) Metabolic pathways along the palsa hillslope (analyzed with MetaCyc)

This file contains the metabolic pathways along the palsa hillslope (68°21'27.33"N, 19°3'1.91"E), analyzed with MetaCyc. The file consists of four sheets:

A) Cores presented in the main text (DNA-based)

Six columns per each stage along the palsa hillslope (palsa (2x), front (1x)) and each soil horizon (organic horizon, transition zone, mineral horizon) contain

- Average (triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

    For the following pathways and descriptions:
    1) Pathway: PWY-5100, Description: Pyruvate fermentation to acetate and lactate II
    2) Pathway: METH-ACETATE-PWY, Description: Methanogenesis from acetate
    3) Pathway: METHANOGENESIS-PWY, Description: Methanogenesis from H_2 and CO_2
    4) Pathway: PWY-7616, Description: Methanol oxidation to carbon dioxide

B) Cores presented in the main text (RNA-based)

Six columns per each stage along the palsa hillslope (palsa (2x), front (1x)) and each soil horizon (organic horizon, transition zone, mineral horizon) contain

- Average (triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

    For the following pathways and descriptions:
    5) Pathway: PWY-5100, Description: Pyruvate fermentation to acetate and lactate II
    6) Pathway: METH-ACETATE-PWY, Description: Methanogenesis from acetate

7) Pathway: METHANOGENESIS-PWY, Description: Methanogenesis from H_2 and CO_2

8) Pathway: PWY-7616, Description: Methanol oxidation to carbon dioxide

C) Replicate cores (DNA-based)

Six columns per each stage along the palsa hillslope (palsa (2x), front (1x)) and each soil horizon (organic horizon, transition zone, mineral horizon) contain

- Average (triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

For the following pathways and descriptions:

9) Pathway: PWY-5100, Description: Pyruvate fermentation to acetate and lactate II

10) Pathway: METH-ACETATE-PWY, Description: Methanogenesis from acetate

11) Pathway: METHANOGENESIS-PWY, Description: Methanogenesis from H_2 and CO_2

12) Pathway: PWY-7616, Description: Methanol oxidation to carbon dioxide

D) Replicate cores (RNA-based)

Six columns per each stage along the palsa hillslope (palsa (2x), front (1x)) and each soil horizon (organic horizon, transition zone, mineral horizon) contain

- Average (triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)
- Error bars (standard deviation of triplicate analysis of each soil horizon per thaw stage along the palsa hillslope)

For the following pathways and descriptions:

13) Pathway: PWY-5100, Description: Pyruvate fermentation to acetate and lactate II

14) Pathway: METH-ACETATE-PWY, Description: Methanogenesis from acetate

15) Pathway: METHANOGENESIS-PWY, Description: Methanogenesis from H_2 and CO_2

16) Pathway: PWY-7616, Description: Methanol oxidation to carbon dioxide

Method used: Pathways, i.e. MetaCyc ontology predictions, were inferred with PICRUSt2 version 2.2.0-b (Phylogenetic Investigation of Communities by Reconstruction of Unobserved States) (Langille, M. G. I. *et al.* Predictive functional profiling of microbial communities using 16S rRNA marker gene sequences. *Nat Biotechnol* **31**, 814-821 (2013)) and MinPath (Minimal set of Pathways) (Ye, Y. & Doak, T.G. A Parsimony Approach to Biological Pathway Reconstruction/Inference for Genomes and Metagenomes. *PLoS Comput. Biol.* **5**, e1000465 (2009)) using ASVs and their abundance counts

9) Replicate solid phase analysis

This file contains the replicate solid phase analysis (reactive iron (Fe) and reactive Fe-associated organic carbon (OC)) per each soil horizon (organic horizon, transition zone, mineral horizon) along the palsa hillslope, from intact palsa (Palsa A) to more collapsed palsa (Palsa B) to collapsing front (Front). The file consists of six sheets:

A) Replicate 1 of reactive Fe and associated OC (68°21'27.33"N, 19° 3'1.91"E)

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

- Reactive iron (Fe) (control corrected) [mg/g soil] (duplicate analysis)
- Error bars (combined standard deviation of sodium chloride bicarbonate extractable Fe (control) and sodium dithionite citrate extractable Fe (not control corrected))
- Associated organic carbon (OC) (control corrected) [mg/g soil] (duplicate analysis)
- Error bars (combined standard deviation of the citrate blank, sodium chloride bicarbonate extractable OC (control) and sodium dithionite citrate extractable OC (not control corrected))

B) Replicate 2 of reactive Fe and associated OC (68°21'27.33"N, 19° 3'1.91"E)

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

- Reactive iron (Fe) (control corrected) [mg/g soil] (duplicate analysis)
- Error bars (combined standard deviation of sodium chloride bicarbonate extractable Fe (control) and sodium dithionite citrate extractable Fe (not control corrected))
- Associated organic carbon (OC) (control corrected) [mg/g soil] (duplicate analysis)
- Error bars (combined standard deviation of the citrate blank, sodium chloride bicarbonate extractable OC (control) and sodium dithionite citrate extractable OC (not control corrected))

C) Replicate 3 of reactive Fe and associated OC (68°21'17.95"N, 19° 2'35.33"E)

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

- Reactive iron (Fe) (control corrected) [mg/g soil] (duplicate analysis)
- Error bars (combined standard deviation of sodium chloride bicarbonate extractable Fe (control) and sodium dithionite citrate extractable Fe (not control corrected))
- Associated organic carbon (OC) (control corrected) [mg/g soil] (duplicate analysis)
- Error bars (combined standard deviation of the citrate blank, sodium chloride bicarbonate extractable OC (control) and sodium dithionite citrate extractable OC (not control corrected))

D) Replicate 1 of control (sodium chloride) (68°21'27.33"N, 19° 3'1.91"E)

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

- Leachable iron (Fe) = sodium chloride bicarbonate extractable Fe (control, same ionic strength and pH as the dithionite/citrate extraction) [mg/g soil]
- Error bars (range of duplicate analyses of each soil horizon in each thaw stage)
- Leachable organic carbon (OC) = sodium chloride bicarbonate extractable OC (control, same ionic strength and pH as the dithionite/citrate extraction) [mg/g soil]
- Error bars (range of duplicate analyses of each soil horizon in each thaw stage)

E) Replicate 2 of control (sodium chloride) (68°21'27.33"N, 19° 3'1.91"E)

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

- Leachable iron (Fe) = sodium chloride bicarbonate extractable Fe (control, same ionic strength and pH as the dithionite/citrate extraction) [mg/g soil]

- Error bars (range of duplicate analyses of each soil horizon in each thaw stage)

- Leachable organic carbon (OC) = sodium chloride bicarbonate extractable OC (control, same ionic strength and pH as the dithionite/citrate extraction) [mg/g soil]

- Error bars (range of duplicate analyses of each soil horizon in each thaw stage)

F) Replicate 3 of control (sodium chloride) (68°21'17.95"N, 19° 2'35.33"E)

Four columns per stage (Palsa A, Palsa B, Front) and soil horizon (organic horizon, transition zone, mineral horizon) contain

- Leachable iron (Fe) = sodium chloride bicarbonate extractable Fe (control, same ionic strength and pH as the dithionite/citrate extraction) [mg/g soil]

- Error bars (range of duplicate analyses of each soil horizon in each thaw stage)

- Leachable organic carbon (OC) = sodium chloride bicarbonate extractable OC (control, same ionic strength and pH as the dithionite/citrate extraction) [mg/g soil]

- Error bars (range of duplicate analyses of each soil horizon in each thaw stage)

Method used for iron in the extracts: by ferrozine assay

Method used for DOC in the extracts: total organic carbon analyzer (High TOC II, Elementar, Elementar Analysensysteme GmbH, Germany); inorganic carbon was removed by acidifying the samples with 2M HCl

10) Correlative SEM and nanoSIMS

Images of scanning electron microscopy (SEM) images of fine particles of palsa soil horizons (transition zone, mineral horizon) along the palsa hillslope (palsa to front) (68°21'18.70"N, 19° 2'38.00"E).

A) Palsa A (transition zone) (1 fine particle) (A-9.tif)
B) Palsa A (mineral horizon) (1 fine particle) (A-61.tif)
C) Front (transition zone) (1 fine particle) (MP_5_03.tif)
D) Front (mineral horizon) (1 fine particle) (MP_6_03.tif)

Method used: performed at a a field emission scanning electron microscope (FE-SEM; Jeol JSM-6500F), equipped with secondary electron detector; the acceleration voltage was set to 5 kV, with a working distance of 10; prior analysis: samples were sputter-coated with ~30 nm Au/Pd conductive layer using a Bal-Tec SCD005 sputter coater (Baltec GmbH, Germany)

11) Fine fraction analysis

Raw data of replicate nanoSIMS analysis of the fine fraction of Palsa A and Front (68°21'18.70"N, 19° 2'38.00"E)

   (A) Palsa A (transition zone) (3 fine particles):
       Replicate 1 (depth_p5_3.im)
       Replicate 2 (depth_p7_3.im)
       Replicate 3 (depth_p9_3.im)
   (B) Palsa A (mineral horizon) (3 fine particle)
       Replicate 1 (S19144_MinA_2_1.im)
       Replicate 2 (S20019_3rd_4_2.im)
       Replicate 3 (S19144_MinA_2_3.im)
   (C) Front (transition zone) (3 fine particles)
       Replicate 1 (S19147_TransC_5_1.im)
       Replicate 2 (S19147_TransC_5_2.im)
       Replicate 3 (S19147_TransC_5_4.im)
   (D) Front (mineral horizon) (3 fine particle)
       Replicate 1 (S19148_MinC_6_2.im)
       Replicate 2 (S19148_MinC_6_4.im)
       Replicate 3 (S19148_MinC_6_1.im)

Method used: performed at the Cameca nanoSIMS 50L of the Chair of Soil Science (TU München, Germany); primary beam (~1.2 pA) was focused at a lateral resolution ~100 nm and scanned over the sample with $^{12}C^-$, $^{16}O^-$, $^{12}C^{14}N^-$, $^{31}P^-$, $^{32}S^-$, $^{27}Al^{16}O^-$ and $^{56}Fe^{16}O^-$ secondary ions collected using electron multipliers; the nanoSIMS images were analyzed using the Open MIMS Image plugin available within ImageJ

12) Mössbauer spectroscopy

This file contains Mössbauer spectroscopy fits (measured at 77 and 6 K) using the Voigt Based Fitting (VBF) routine (Rancourt, D. G., Ping, J.Y . Voigt-based methods for arbitrary-shape static hyperfine parameter distributions in Mössbauer spectroscopy. Nucl Instrum Methods Phys Res Sect B 58, 85-97 (1991)) in the Recoil software (University of Ottawa, Canada). Fits are obtained for the two soil horizons (transition zone and mineral horizon) of each of the three thaw stages (palsa (1x), bog (1x) and fen (1x)):

Palsa A (68°21'18.70"N, 19° 2'38.00"E)
Bog C (68°21'18.60"N, 19° 2'39.20"E)
Fen E (68°21'16.80"N, 19° 2'40.30"E)

The file consists of one sheet:

A) Raw data of VBF fits

Seven columns per horizon (transition zone, mineral horizon) per thaw stage (palsa, bog or fen) and per temperature (77K or 6K) contain

- Velocity [mm/s]
- Data
- Fit
- Fe(II)
- Fe(III)
- Fe-S
- Unknown

Method used: performed in a standard transmission setup (Wissel, Wissenschaftliche Elektronik GmbH), and absorption spectra were collected at 77 and 6 K controlling with a closed-cycle cryostat (SHI-850-I, Janis Research Co)