Supporting Online Material

S1. Mass Balance Calculations

a. Triple mass balance model derivation

Using the $\delta^{13}CH_4\uparrow$, $\delta D\text{-}CH_4\uparrow$, and $^{14}CH_4\uparrow$ values, separate mass balances can be constructed for each isotope constraint (Eqns 1 – 3 in the main text):

$$\delta^{13}C\uparrow_C \cdot\Delta Q_C = \delta^{13}C_1\cdot\Delta Q_1 + \delta^{13}C_2\cdot\Delta Q_2 + \delta^{13}C_3\cdot\Delta Q_3 \tag{S1}$$

$$\delta D\uparrow_C \cdot\Delta Q_C = \delta D_1\cdot\Delta Q_1 + \delta D_2\cdot\Delta Q_2 + \delta D_3\cdot\Delta Q_3 \tag{S2}$$

$$\Delta^{14}C\uparrow_C \cdot\Delta Q_C = \delta^{14}C_1\cdot\Delta Q_1 + \delta^{14}C_2\cdot\Delta Q_2 + \delta^{14}C_3\cdot\Delta Q_3 \tag{S3}$$

where ΔQ_n and $\delta^{13}C_n$, δD_n , $\Delta^{14}C_n$ are the fractional mass flux and isotope values ($\delta^{13}CH_4$, $\delta D\text{-}CH_4$, $\Delta^{14}CH_4$) of the n-th source term. ΔQ_C is total mass flux (taken as 1) and $\delta^{13}C\uparrow$, $\delta D\uparrow$, $\Delta^{14}C\uparrow$ are isotope values of the aggregated source. For simplicity, Eqns S1 – S3 can be represented, in order, as:

$$a\cdot\Delta Q_C = b\cdot\Delta Q_1 + c\cdot\Delta Q_2 + d\cdot\Delta Q_3 \tag{S4}$$

$$e\cdot\Delta Q_C = f\cdot\Delta Q_1 + g\cdot\Delta Q_2 + h\cdot\Delta Q_3 \tag{S5}$$

$$i\cdot\Delta Q_C = j\cdot\Delta Q_1 + k\cdot\Delta Q_2 + l\cdot\Delta Q_3 \tag{S6}$$

This series of equations can be solved simultaneously as (derived using Mathematica®):

$$\Delta Q_1 = -\frac{dgi - chi - dek + ahk + cel - agl}{-dgj + chj + dfk - bhk - cfl + bgl} \tag{S7}$$

$$\Delta Q_2 = -\frac{-dfi + bhi + dej - ahj - bel + afl}{-dgj + chj + dfk - bhk - cfl + bgl} \tag{S8}$$

$$\Delta Q_3 = -\frac{-cfi + bgi + cej - agj - bek + afk}{dgj - chj - dfk + bhk + cfl - bgl} \tag{S9}$$

The triple mass balance source mass fractional fluxes are validated by recalculating equations S1 – S3 with the model output of ΔQ_1, ΔQ_2, and ΔQ_3. If the mass balance constraints can be satisfied by only two sources then the remaining ΔQ term is zero. Acceptable scenarios must meet the limits described in the main text (±0.3‰ of $\delta^{13}CH_4\uparrow_T$; ±4‰ of $\delta D\text{-}CH_4\uparrow_T$; ±10‰ of $^{14}CH_4\uparrow_T$, and ΔQ_1, ΔQ_2, and ΔQ_3 summed to 1.0 ± 0.1).

The results of the standard scenario, which is described in the main text, are presented in Tables 1 and S6. Table S6 lists the triple mass balance results for all 29 possible scenarios that could theoretically satisfy the ice record isotope constraints, while Table 1 contains only the subset that have non-negative fractional source contributions. The scenarios that passed the acceptance criteria determine the range of possible source ΔQ values based upon the range of values for $\delta^{13}CH_4\uparrow$, $\delta D\text{-}CH_4\uparrow$, and $^{14}CH_4\uparrow$ (Table S5).

Table S1: Påkitsoq IRMS raw measurement values. [CH_4] is derived from IRMS *m/z* 44 peak height. Sampling profile distance is relative to an arbitrary reference location that is invariant across sampling seasons (Petrenko et al., 2006). The contemporaneous GISP2 methane concentration for each Påkitsoq sample is linearly interpolated from Brook et al. (2000). Values excluded due to anomalous [CH_4] are in parentheses.

Sampling season	Sampling profile distance (m)	Gas age (ka BP)	GISP2 [CH_4] (ppb)	Sample mass (g)	[CH_4] (ppb)	$\delta^{13}CH_4$ (‰ vs. VPDB)
2003	1.23	12.238	500	137.7	(1802)	(-42.18)
	1.29	12.191	503	202.8	517	-46.17
				76.4	485	-44.34
	1.44	11.752	508	115.7	428	-44.95
	2.65	11.430	713	79.7	840	-45.73
				96.5	899	-45.41
	2.95	11.350	748	106.5	795	-45.25
				75.5	(910)	(-45.48)
	3.14	11.332	743	211.6	(856)	(-46.14)
				195.4	(873)	(-46.95)
	3.52	11.278	694	135.1	(920)	(-43.85)
	3.57	11.270	692	66.4	(871)	(-44.73)
	4.08	11.198	680	196.9	(832)	(-46.53)
	4.58	11.091	742	155.6	830	-45.07
				170.9	845	-46.37
	4.99	11.004	739	48.4	682	(-44.59)
				88.1	763	-45.32
	5.03	10.996	738	96.1	864	-46.13
				104.8	866	-45.92
2004	2.07	11.586	517	165.0	449	-46.24
				114.0	475	-45.47
	2.12	11.572	554	150.0	466	-46.13
				188.0	527	-46.53
				113.0§	--	--
	2.17	11.559	590	135.0	429	-46.70
				172.8	(2321)	(-45.88)

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Sampling season	Sampling profile distance (m)	Gas age (ka BP)	GISP2 [CH_4] (ppb)	Sample mass (g)	[CH_4] (ppb)	$\delta^{13}CH_4$ (‰ vs. VPDB)
				179.0	548	-47.27
	2.21	11.527	605	116.0	530	-46.37
				174.4	(709)	(-47.44)
				169.0	543	-46.93
	2.30	11.483	639	169.0	636	-46.66
				201.0	669	-45.84
	2.40	11.475	662	145.0	693	-45.55
				139.0	648	-46.38
				179.0	623	-46.76
				127.5	(918)	(-45.99)
	2.46	11.465	690	170.0	691	-46.31
				137.0	(286)*	(-45.52)
				124.0	609	-46.43
	2.52	11.447	731	142.5	690	-45.52
				156.8	660	-46.10
				128.0	632	-46.26
				143.0	726	-46.49
	2.57	11.433	692	134.0	778	-46.59
				179.8	620	-46.40
				121.0	674	-46.27
2005	1.29	12.191	503	118.5	(654)	(-47.69)
				132.3	(744)	(-46.88)
				160.2	(651)	(-46.83)
				169.1	(713)	(-46.76)
				211.0	(742)	(-47.23)
	1.36	12.139	506	116.8	(569)	(-46.62)
				162.9	(704)	(-45.27)
				144.7	499	-45.88
				140.0	415	-45.80
				133.8	456	-45.08
				115.7	428	-44.48

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Sampling season	Sampling profile distance (m)	Gas age (ka BP)	GISP2 $[CH_4]$ (ppb)	Sample mass (g)	$[CH_4]$ (ppb)	$\delta^{13}CH_4$ (‰ vs. VPDB)
	1.41	12.099	508	259.0	452	-45.72
				111.0	542	-44.83
				117.3	(830)	(-44.33)
				255.6	(657)	(-46.05)

*Sample signal was below shot noise threshold of 290 mV (0.97 nA)

§Sample lost due to capillary breakage

Table S2: Final corrected values for Påkitsoq ice measurements of $\delta^{13}CH_4$.

Gas Age (ka BP)	Sampling Season	Median measured $\delta^{13}CH_4$ (‰ VPDB)	$\delta^{15}N$ correction (‰)	Firn diffusion correction (‰)	Corrected $\delta^{13}CH_4$ (‰VPDB)	Standard uncertainty (‰)	Isotopic disequi-librium correction (‰)	Final equilibrated atmospheric value (‰ vs. VPDB)
10.996	2003	-46.03	0.42	0.00	-46.44	0.15	0.00	-46.4
11.004	2003	-45.32	0.42	0.00	-45.74	0.29	0.00	-45.7
11.091	2003	-45.72	0.45	0.00	-46.17	0.92	0.00	-46.2
11.350	2003	-45.25	0.52	0.00	-45.77	0.29	0.00	-45.8
11.430	2003	-45.57	0.52	0.57	-45.52	0.23	0.07	-45.5
11.433	2004	-46.40	0.54	0.60	-45.82	0.28	0.07	-45.8
11.447	2004	-46.18	0.54	0.61	-46.12	0.42	0.07	-46.1
11.465	2004	-46.22	0.53	0.68	-46.22	0.09	0.08	-46.1
11.475	2004	-46.38	0.52	0.70	-46.20	0.62	0.08	-46.1
11.483	2004	-46.25	0.53	0.74	-46.04	0.58	0.08	-46.0
11.527	2004	-46.65	0.50	0.69	-46.46	0.40	0.09	-46.4
11.559	2004	-46.99	0.46	0.41	-47.03	0.40	0.09	-47.0
11.572	2004	-46.33	0.40	0.00	-46.73	0.28	0.00	-46.7
11.586	2004	-45.85	0.38	0.00	-46.23	0.55	0.00	-46.2
11.752	2003	-44.95	0.37	0.00	-45.32	0.29	0.00	-45.3
12.099	2005	-45.27	0.37	0.00	-45.65	0.63	0.00	-45.7
12.139	2005	-45.44	0.37	0.00	-45.81	0.66	0.00	-45.8
12.191	2003	-45.26	0.35	0.00	-45.61	0.29	0.00	-45.6

Table S3: Age tie points for the Påkitsoq ice sampling profile. Some sample profile locations have multiple possible age assignments for a particular climate proxy due to non-unique matches in the reference geochemical records. For each age tie point based on Påkitsoq $\delta^{18}O_{atm}$ with relatively poor precision the ± 1 SD possible range in age is listed. Three additional age tie points used for the 2003 sampling season are also included below.

Påkitsoq sampling profile distance (m)	Climate proxy for age determination	Possible ages of tie point (ka BP)	Final age of tie point (ka BP)
0.69	[CH_4]	12.838	12.838
0.97	$\delta^{18}O_{ice}$	12.426	12.426
1.62	$\delta^{15}N$	11.928 12.783	11.928
	$\delta^{18}O_{atm}$	11.710(-SD) 12.710(+SD)	
1.74	$\delta^{18}O_{ice}$	11.938	11.938
1.92	[CH_4]	11.602	11.602
2.15	$\delta^{15}N$	11.570	11.570
	$\delta^{18}O_{atm}$	11.515(-SD) 12.510(+SD)	
2.18	[CH_4]	11.587	11.587
2.42	$\delta^{15}N$	11.468	11.468
	$\delta^{18}O_{atm}$	11.260(-SD) 11.450(+SD)	
	[CH_4]	11.464 11.490	
3.20	$\delta^{15}N$	11.339	11.339
	$\delta^{18}O_{atm}$	11.300(-SD) 11.500(+SD)	
3.43	$\delta^{15}N$	11.311 11.154	11.311
	$\delta^{18}O_{atm}$	11.400(-SD) 11.690(+SD)	
3.68	$\delta^{15}N$	11.193 11.084	11.193
	$\delta^{18}O_{atm}$	11.295(-SD) 11.490(+SD)	
3.90 (2003)	$\delta^{15}N$	11.211	11.210
4.20 (2003)	$\delta^{15}N$	10.962	10.962
4.33 (2003)	$\delta^{18}O_{ice}$	11.181	11.181

Table S4. Triple isotope mass balance model results for all possible YD-PB source scenarios. Fractional source contributions are calculated from the mean $\delta^{13}CH_4\uparrow$, $\delta D\text{-}CH_4\uparrow$, $^{14}CH_4\uparrow$ values. Scenarios are termed valid if the model output, recalculated in mass balances, gives values within ±0.3‰ of $\delta^{13}CH_4\uparrow$, ±4‰ of $\delta D\text{-}CH_4\uparrow$, and ±10 ‰ of $^{14}CH_4\uparrow$ and the fractional contributions sum to 1.0 ± 0.1. All valid scenarios are highlighted in bold font. Scenarios from this table with non-negative fractional source contributions are also presented in Table 3.

Scenario #	Source Fractional Contribution										
	Biomass burning	GEM	Thermokarst lakes	Biogenic marine gas hydrates	Aerobic plant methane	Ruminants	Tropical wetlands	Boreal wetlands	Termites	ΔQ_T	Satisfy acceptance criteria?
1	**0.54**		**0.43**		**0.08**					**1.05**	**Y**
2	**0.55**		**0.43**			**0.08**				**1.06**	**Y**
3	**0.56**		**0.43**				**0.08**			**1.07**	**Y**
4	**0.56**		**0.43**					**0.07**		**1.06**	**Y**
5	**0.53**		**0.43**						**0.08**	**1.04**	**Y**
6		-3.43	4.33		-1.85					9.62	N
7		-2.50	3.24			-1.26				7.00	N
8		-1.90	2.60				-0.93			5.43	N
9		-2.13	2.90					-1.00		6.03	N
10		-7.14	8.59						-3.91	19.64	N
11		-0.55	1.22	-0.22						1.99	N
12	0.60	0.38			0.29					1.28	N
13	0.64	0.39				0.28				1.31	N
14	0.68	0.38					0.28			1.33	N
15	0.65	0.37						0.26		1.28	N
16	0.56	0.37							0.29	1.22	N
17	0.52	-0.14	0.59							1.26	N
18	1.71			0.55	-0.50					2.76	N
19	1.62			0.53		-0.46				2.60	N
20	1.59			0.55			-0.47			2.62	N
21	1.67			0.58				-0.47		2.72	N
22	0.71		0.37	0.08						1.15	N
23	1.80			0.56					-0.51	2.86	N
24	1.01	0.24		0.20						1.45	N
25		0.58		-0.30	0.73					1.62	N
26		0.64		-0.35		0.76				1.75	N
27		0.65		-0.41			0.83			1.89	N
28		0.61		-0.37				0.73		1.71	N
29		0.54		-0.25					0.65	1.44	N

Table S5: Triple mass balance results to determine range of source values from the valid scenarios of Table 3. All valid scenarios are shown here in bold font.

Scenario description	Source Fractional Contribution							
	BB	TK	APM	RUM	TW	BW	TERM	Total
mean $\delta^{13}\mathrm{CH}_4\uparrow$ (−49.2‰),	**0.54**	**0.43**	**0.08**					**1.05**
mean $\delta\mathrm{D}\text{-}\mathrm{CH}_4\uparrow$ (−314‰),	**0.55**	**0.43**		**0.08**				**1.06**
mean $^{14}\mathrm{CH}_4\uparrow$ (−138‰)	**0.56**	**0.43**			**0.08**			**1.07**
	0.56	**0.43**				**0.07**		**1.06**
	0.53	**0.43**					**0.08**	**1.04**
min $\delta^{13}\mathrm{CH}_4\uparrow$ (−50.44‰),	**0.43**	**0.41**	**0.17**					**1.02**
mean $\delta\mathrm{D}\text{-}\mathrm{CH}_4\uparrow$(−314‰),	**0.45**	**0.42**		**0.17**				**1.03**
mean $^{14}\mathrm{CH}_4\uparrow$(−138‰)	**0.48**	**0.41**			**0.16**			**1.05**
	0.46	**0.40**				**0.16**		**1.02**
	0.41	**0.41**					**0.17**	**0.99**
max $\delta^{13}\mathrm{CH}_4\uparrow$(−47.92‰),	0.65	0.45	-0.01					1.11
mean $\delta\mathrm{D}\text{-}\mathrm{CH}_4\uparrow$(−314‰),	0.65	0.45		-0.01				1.10
mean $^{14}\mathrm{CH}_4\uparrow$(−138‰)	0.65	0.45			-0.01			1.11
	0.65	0.45				-0.01		1.11
	0.65	0.45					-0.01	1.11
mean $\delta^{13}\mathrm{CH}_4\uparrow$(−49.2‰),	0.67	0.46	0.00					1.12
min $\delta\mathrm{D}\text{-}\mathrm{CH}_4\uparrow$(−322‰),	0.66	0.46		0.00				1.12
mean $^{14}\mathrm{CH}_4\uparrow$(−138‰)	0.66	0.46			0.00			1.12
	0.66	0.46				0.00		1.12
	0.67	0.46					0.00	1.12
mean $\delta^{13}\mathrm{CH}_4\uparrow$(−49.2‰),	**0.42**	**0.40**	**0.17**					**0.99**
max $\delta\mathrm{D}\text{-}\mathrm{CH}_4\uparrow$(−306‰),	**0.44**	**0.41**		**0.16**				**1.01**
mean $^{14}\mathrm{CH}_4\uparrow$(−138‰)	**0.46**	**0.40**			**0.16**			**1.02**
	0.45	**0.40**				**0.15**		**1.00**
	0.40	**0.40**					**0.16**	**0.96**
mean $\delta^{13}\mathrm{CH}_4\uparrow$(−49.2‰),	0.61	0.59	-0.13					1.32
mean $\delta\mathrm{D}\text{-}\mathrm{CH}_4\uparrow$(−314‰),	0.59	0.58		-0.12				1.30
min $^{14}\mathrm{CH}_4\uparrow$(−276‰)	0.57	0.59			-0.12			1.28
	0.58	0.59				-0.12		1.29
	0.63	0.59					-0.13	1.34
mean $\delta^{13}\mathrm{CH}_4\uparrow$(−49.2‰),	**0.48**	**0.27**	**0.29**					**1.05**
mean $\delta\mathrm{D}\text{-}\mathrm{CH}_4\uparrow$(−314‰),	**0.51**	**0.28**		**0.28**				**1.07**
max $^{14}\mathrm{CH}_4\uparrow$(2‰)	**0.55**	**0.27**			**0.27**			**1.10**
	0.53	**0.26**				**0.26**		**1.06**
	0.44	**0.27**					**0.29**	**1.00**

Table S6: Triple mass balance sensitivity test results with valid scenarios. Each test is described in Section 3.4. Note that none of the sensitivity test scenarios result in valid source combinations with the majority contributions from sources other than biomass burning and thermokarst lakes.

Scenario Description	Source Fractional Contribution										
	Biomass Burning	GEM	Thermokarst lakes	Biogenic marine gas hydrates	APM	Ruminants	Tropical wetlands	Boreal wetlands	Termites	Total	Passes acceptance criteria?
Modern scenario	0.49		0.42		0.13					1.04	Y
	0.51		0.43			0.11				1.05	Y
	0.52		0.42				0.12			1.06	Y
	0.50		0.41					0.12		1.04	Y
	0.46		0.42						0.13	1.01	Y
Preboreal scenario	0.53		0.43		0.09					1.05	Y
	0.54		0.43			0.09				1.06	Y
	0.55		0.43				0.09			1.07	Y
	0.55		0.42					0.08		1.05	Y
	0.52		0.43						0.09	1.04	Y
No MBL sink	0.57		0.43		0.04					1.05	Y
	0.58		0.43			0.04				1.05	Y
	0.59		0.43				0.04			1.06	Y
	0.58		0.43					0.04		1.05	Y
	0.57		0.43						0.04	1.04	Y
	0.66		0.40	0.04						1.10	Y
Mean MBL sink	0.52		0.43		0.12					1.06	Y
	0.53		0.43			0.11				1.07	Y
	0.54		0.43				0.11			1.08	Y
	0.54		0.42					0.10		1.06	Y
	0.50		0.43						0.11	1.04	Y
Max MBL sink	0.49		0.43		0.15					1.06	Y
	0.51		0.43			0.14				1.08	Y
	0.53		0.42				0.14			1.09	Y
	0.52		0.42					0.13		1.07	Y
	0.47		0.42						0.15	1.04	Y
Fischer et al. (2008)	0.47		0.42			0.16				1.05	Y
	0.43		0.40				0.21			1.03	Y
	0.47		0.41					0.16		1.04	Y
Lassey et al. (2007)	0.53		0.43			0.11				1.07	Y
	0.54		0.42				0.12			1.08	Y
	0.45		0.40					0.18		1.03	Y
	0.26		0.39						0.29	0.94	Y

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