1. Methods

1.1. Stable Carbon and Nitrogen Isotope Analysis – Calibration and Analytical Uncertainty

Carbon and nitrogen isotopic and elemental compositions were determined using an IsoPrime continuous flow isotope-ratio mass spectrometer (CF-IRMS) coupled to a Vario Micro elemental analyzer (Elementar, Hanau, Germany) at the University of British Columbia. Sample measurements were calibrated relative to VPDB (δ^{13}C) and AIR (δ^{15}N) using USGS40 and USGS41 (Qi et al. 2003). The standard deviations and number of calibration (quality control) standards used in all of the analytical sessions are listed in Table S2.

Table S2. Standard deviations for the carbon and nitrogen isotopic compositions of the calibration standards used in all of analytical session associated with the data presented in this paper.

Standard	***n***	**δ^{13}C (±1σ)**	**δ^{15}N (±1σ)**
USGS40	177	0.08	0.14
USGS41	177	0.16	0.24

The following standards were used to monitor accuracy and precision (Table S3). The isotopic compositions used as the accepted values for these internal standards represent long-term averages.

Table S3. Isotopic reference materials used to monitor internal accuracy and precision.

Standard	Material	Mean δ^{13}C (‰, VPDB)	Mean δ^{15}N (‰, AIR)
MET	Methionine[1]	–28.61±0.10	–5.04±0.13
NIST-1577c	Bovine liver[1]	–17.52±0.09	+8.15±0.14
SRM-1	Caribou bone collagen[1]	–19.40±0.08	+1.82±0.11
SRM-2	Walrus bone collagen[1]	–14.72±0.14	+15.59±0.13
USGS42	Human hair	–21.09±0.10	+8.05±0.10
USGS43	Human hair	–21.28±0.10	+8.44±0.10

1. Internal standard with mean isotopic compositions representing long-term values as measured in three different laboratories.

Table S4 summarizes the mean and standard deviation of carbon and nitrogen isotopic compositions for all check (quality assurance) standards analyzed alongside the samples presented in this study.

Table S4. Mean and standard deviations of all the check (QA) standards analyzed in the analytical sessions associated with data presented in this paper.

		δ^{13}C (‰, VPDB)	δ^{15}N (‰, AIR)
Standard	***n***	**Mean ± 1σ**	**Mean ± 1σ**
MET	177	–28.60 ± 0.08	–5.00 ± 0.16
NIST-1577c	97	–17.52 ± 0.09	+8.13 ± 0.12
SRM-1	77	–19.33 ± 0.12	+1.81 ± 0.07
SRM-2	78	–14.70 ± 0.13	+15.59 ± 0.08
USGS42	4	–21.09 ± 0.02	+7.98 ± 0.03
USGS43	3	–21.28 ± 0.02	+8.41 ± 0.06

All of the samples were analyzed in at least duplicate. One internal standard (SUBC-1, seal bone collagen) was in the process of attaining an average long-term value, so we treated this as a sample replicate rather than a QA standard (163 aliquots of this material were analyzed alongside these samples). The pooled standard deviations for the sample replicates were ±0.15 ‰ for δ^{13}C and ±0.15 ‰ for δ^{15}N (*df*=335).

Standard uncertainty for the δ^{13}C and δ^{15}N measurements of the samples was estimated following Szpak et al. (2017b), which largely follows the method presented in Magnusson et al. (2012). Systematic errors ($u_{(bias)}$) were calculated to be ±0.11 ‰ for δ^{13}C and ±0.13 for δ^{15}N based on the known uncertainty in the check standards and the observed standard deviations of those check standards from the known values. Random errors ($uR_{(w)}$) were calculated to be ±0.16 ‰ for δ^{13}C and ±0.20 ‰ for δ^{15}N based on the pooled standard deviations of the check standards and sample replicates. Standard uncertainty, calculated as the root-sum-square of $u_{(bias)}$ and ($uR_{(w)}$) was determined to be ±0.19 for δ^{13}C and ±0.24 for δ^{15}N.

1.2. Stable Sulfur Isotope Analysis – Calibration and Analytical Uncertainty

Sulfur isotopic and elemental compositions were determined using a Europa ANCA-SL/20-20 elemental analyzer/isotope-ratio mass spectrometer at Iso-Analytical. Sulfur isotopic compositions were calibrated relative to VCDT, using IA-R0611 (Ba_2SO_4) and IAEA-SO-5 (Ba_2SO_4). The standard deviations and number of calibration (quality control) standards used in all of the analytical sessions are listed in Table S5.

Table S5. Standard deviations for the carbon and nitrogen isotopic compositions of the calibration standards used in all of analytical session associated with the data presented in this paper.

Standard	***n***	**δ^{34}S (±1σ)**
IA-R061	20	0.14
IA-R062	24	0.15

The following standards were used to monitor accuracy and precision (Table S6). The isotopic compositions used as the accepted values for these internal standards represent long-term averages.

Table S6. Isotopic reference materials used to monitor internal accuracy and precision.

Standard	Material	Mean $\delta^{34}S$ (‰, VCDT)
IA-R068	Soy protein	+5.3±0.2
IAEA-SO-5	Barium sulfate	+0.5±0.2
IA-R069	Tuna protein	+18.9±0.2

Table S1 in Supplement 1 (www.int-res.com/articles/suppl/m653p205_supp1.xlsx) summarizes the mean and standard deviation of sulfur isotopic compositions for all check (quality assurance) standards analyzed alongside the samples presented in this study.

Table S7. Mean and standard deviations of all the check (QA) standards analyzed in the analytical sessions associated with data presented in this paper.

		$\delta^{34}S$ (‰, VCDT)		
Standard	***n***	**Mean**	**±**	**1σ**
IA-R068	21	+5.1	±	0.2
IAEA-SO-5	17	+0.5	±	0.2
IA-R069	17	+18.9	±	0.1

Sixty-two samples were analyzed in duplicate. he pooled standard deviations for the sample replicates were ±0.4 ‰ for $\delta^{34}S$ (*df*=62).

Standard uncertainty for the $\delta^{34}S$ measurements of the samples was estimated following Szpak et al. (2017b), which largely follows the method presented in Magnusson et al. (2012). Systematic errors ($u_{(bias)}$) were calculated to be ±0.2 ‰ based on the known uncertainty in the check standards and the observed standard deviations of those check standards from the known values. Random errors ($uR_{(w)}$) were calculated to be ±0.3 ‰ based on the pooled standard deviations of the check standards and sample replicates. Standard uncertainty, calculated as the root-sum-square of $u_{(bias)}$ and ($uR_{(w)}$) was determined to be ±0.4 for $\delta^{34}S$.

1.3. AMS Radiocarbon Dating

New AMS radiocarbon dates were obtained from the purified bone collagen of terrestrial mammal bones for several of the sites. Bone collagen was extracted and purified for AMS dating following the same procedure detailed for the samples prepared for stable isotope analysis but an Amicon® 30 kDa ultrafilter was used (Millipore, Darmstadt, Germany) and was cleaned following the procedure presented in Beaumont et al. (2010). Samples were AMS dated at the A.E. Lalonde Radiocarbon Laboratory at the University of Ottawa. Prior to dating, stable carbon and nitrogen isotopic compositions were determined as outlined above to confirm the terrestrial origin of the sample so as to eliminate the possibility of a misidentified polar bear (Gorlova et al. 2015) or an arctic fox that scavenged a significant amount of marine prey. All radiocarbon dates were calibrated using IntCal13 (Reimer et al. 2013).

2. Results

2.1. Stable Isotope Analysis

The isotopic and elemental compositions for all the samples analyzed are presented in Table S1 in Supplement 1 (www.int-res.com/articles/suppl/m653p205_supp1.xlsx). The collagen

yields approached those of modern cortical bone (mean = 16.5%) despite the fact that the demineralized collagen was subjected to ultrafiltration, a process that significantly reduces the collagen yield in ancient (Szpak et al. 2017a) and even modern bone (Guiry et al. 2016). The atomic $C:S_{atomic}$ and $N:S_{atomic}$ ratios were all within the ranges identified by Nehlich & Richards (2009) as indicating collagen with unaltered $\delta^{34}S$ values (600±300 for $C:S_{atomic}$ and 200±100 for $N:S_{atomic}$). One walrus sample (2216) had a $C:N_{atomic}$ ratio of 4.17, which falls outside the widely cited acceptable range of 2.90 to 3.60 (DeNiro 1985). We excluded this sample from all subsequent analyses. No other samples failed bone collagen QC criteria of yield, wt% C, or wt% N (Ambrose 1990). Samples that were excluded from the analysis because they failed the collagen quality criteria are indicated with an X in the column "Excluded".

Results of the statistical tests comparing the $\delta^{34}S$ values of walrus and ringed seal are presented in Table S8.

Table S8. Results of statistical comparisons between ringed seal and walrus $\delta^{34}S$ values by region and overall. Statistically significant differences ($p<0.05$) are in boldface.

$\delta^{34}S$	Mann Whitney U test		t-test	
Region	***U***	***p***	***t***	***p***
Kotzebue Sound (A)			2.94	**0.015**
Devon Island (D)			0.28	0.79
Somerset Island (E)			2.04	0.11
NW Hudson Bay (G)			1.25	0.23
Nunavik (I)	2	**0.006**		
Hudson Strait (J)			1.90	0.11
Ellesmere Island (L)			4.52	**0.0004**
NW Greenland (M)			3.45	**0.002**
All Regions	980	**<0.0001**		

Results of the statistical tests comparing the $\delta^{13}C$ values of walrus and ringed seal are presented in Table S9.

Table S9. Results of statistical comparisons between ringed seal and walrus $\delta^{13}C$ values by region and overall. Statistically significant differences ($p<0.05$) are in boldface.

$\delta^{13}C$	Mann Whitney U test		t-test	
Region	***U***	***p***	***t***	***p***
Kotzebue Sound (A)			0.41	0.69
Devon Island (D)			2.18	**0.048**
Somerset Island (E)			1.12	0.32
NW Hudson Bay (G)			1.45	0.17
Nunavik (I)			1.01	0.33
Hudson Strait (J)	5	0.86		
Ellesmere Island (L)			6.28	**<0.0001**
NW Greenland (M)			1.64	0.11
All Regions	2079	0.32		

Results of the statistical tests comparing the $\delta^{15}N$ values of walrus and ringed seal are presented in Table S10.

Table S10. Results of statistical comparisons between ringed seal and walrus δ^{15}N values by region and overall. Statistically significant differences (p<0.05) are in boldface.

δ^{15}N	Mann Whitney U test		t-test	
Region	***U***	***p***	***t***	***p***
Kotzebue Sound (A)			10.85	**<0.0001**
Devon Island (D)			16.468	**<0.0001**
Somerset Island (E)			4.72	**0.009**
NW Hudson Bay (G)			4.52	**0.0007**
Nunavik (I)			9.32	**<0.0001**
Hudson Strait (J)			2.58	**0.05**
Ellesmere Island (L)			12.90	**<0.0001**
NW Greenland (M)			10	**0.0002**
All Regions			22.80	**<0.0001**

2.2. Radiocarbon Dates

New AMS radiocarbon dates are presented in Table S11. References providing additional information on the sites from which samples were taken in this study are provided in Table S12.

Table S11. New AMS radiocarbon dates produced in this study.

Sample ID	Radiocarbon Lab ID	Site	Feature	Context	Species	^{14}C Age	±	Calibrate Age (years BP, 2 σ range)[1]
7415	UOC-5915	SfFk-4	House 21	Floor	Arctic fox	746	41	569–582 (0.02) 650–740 (0.98)
7420	UOC-1798	SfFk-4	House 21		Arctic fox	632	27	553–611 (0.59) 621–663 (0.41)
7410	UOC-1797	SfFk-4	House 20		Arctic fox	628	25	553–611 (0.60) 620–661 (0.40)
8452	UOC-5919	SfFk-4	House 15	Floor	Caribou	620	41	545–662
8615	UOC-5923	JfEl-4	4B-76		Caribou	760	41	656–760
8296	UOC-5918	JfEl-4	74	K-49	Caribou	759	41	655–760
8588	UOC-5921	JfEl-4	73		Caribou	695	41	557–604 (0.31) 627–697 (0.69)
8592	UOC-5922	JfEl-4	73		Caribou	642	41	551–669
8251	UOC-5916	JfEl-4			Caribou	535	41	507–564 (0.65) 589–641 (0.35)
8224	UOC-1808	NaPi-2	S16/W16	Level 1-5	Caribou/musk ox	613	23	551–653
8222	UOC-1806	NaPi-2	S16/W16	Level 1-5	Caribou/musk ox	593	26	540–569 (0.27) 581–650 (0.73)
8223	UOC-1807	NaPi-2	S16/W16	Level 1-5	Caribou/musk ox	580	26	535–567 (0.33) 585–646 (0.68)
8199	UOC-1805	NcPf-1	N30/W0	Level 1C	Caribou/musk ox	611	25	549–653
8181	UOC-1804	NcPf-12	2a		Caribou	632	23	555–608 (0.60) 623–662 (0.40)

[1.] Numbers in parentheses represent relative area under probability distribution for calibrated ages.

Table S12. List of locations and ages for sites from which samples were obtained and references providing additional information about these sites.

Site	Region	Approximate Date (calibrated years BP)	Reference[1]
KTZ-304	NW Alaska (A)	650–850	Darwent et al. (2013)
KTZ-087	NW Alaska (A)	350–550	Darwent et al. (2013)
KTZ-088	NW Alaska (A)	100–150	Darwent et al. (2013)
NkRi-3	Amundsen Gulf (B)	650–750	Moody and Hodgetts (2013)
OjRl-3	Amundsen Gulf (B)	2150-2750	Arnold (1983)
OkRn-1	Amundsen Gulf (B)	300–500	Kotar (2016)
OlRr-1	Amundsen Gulf (B)	650-750	Manning (1956)
NaPi-2	Coronation Gulf (C)	550–650[2]	Morrison (1983)
NcPf-1	Coronation Gulf (C)	550–650[2]	Morrison (1983)
NcPf-12	Coronation Gulf (C)	550–650[2]	Morrison (1983)
RbJr-1	Devon Island (D)	500–650	Park (1983)
PcJq-5	Somerset Island (E)	550–650	Rick (1980)
PeJr-1	Somerset Island (E)	550–650	Rick (1980)
NgFv-9	Foxe Basin (F)	500–600	Savelle and Dyke (2014)
KkJg-1	NW Hudson Bay (G)	325–500	Staab (1979) Dyke et al. (2019)
IbGk-3	Eastern Hudson Bay (H)	0–650	Desrosiers et al. (2010)
KbFk-7	Nunavik (I)	1900–2100	Todisco and Monchot (2008)
KcFs-2	Nunavik (I)	450–1450	Thompson (2011)
JfEl-4	Hudson Strait (J)	550–700[2]	Badgley (1980)
KkDo-1	SW Baffin Island (K)	650–150	Stenton (1987)
SfFk-4	Ellesmere Island (L)	550–650[2]	Howse (2013)
Iita	NW Greenland (M)	850–20	LeMoine and Darwent (2010)
Cape Grinnell	NW Greenland (M)	550–650	LeMoine and Darwent (2010)
Qaqaitsuit	NW Greenland (M)	1300–1600	LeMoine and Darwent (2010)

Collagen Fingerprinting

Sample collagen spectra for bearded seal and walrus analyzed as part of this study are presented in Figure S1.

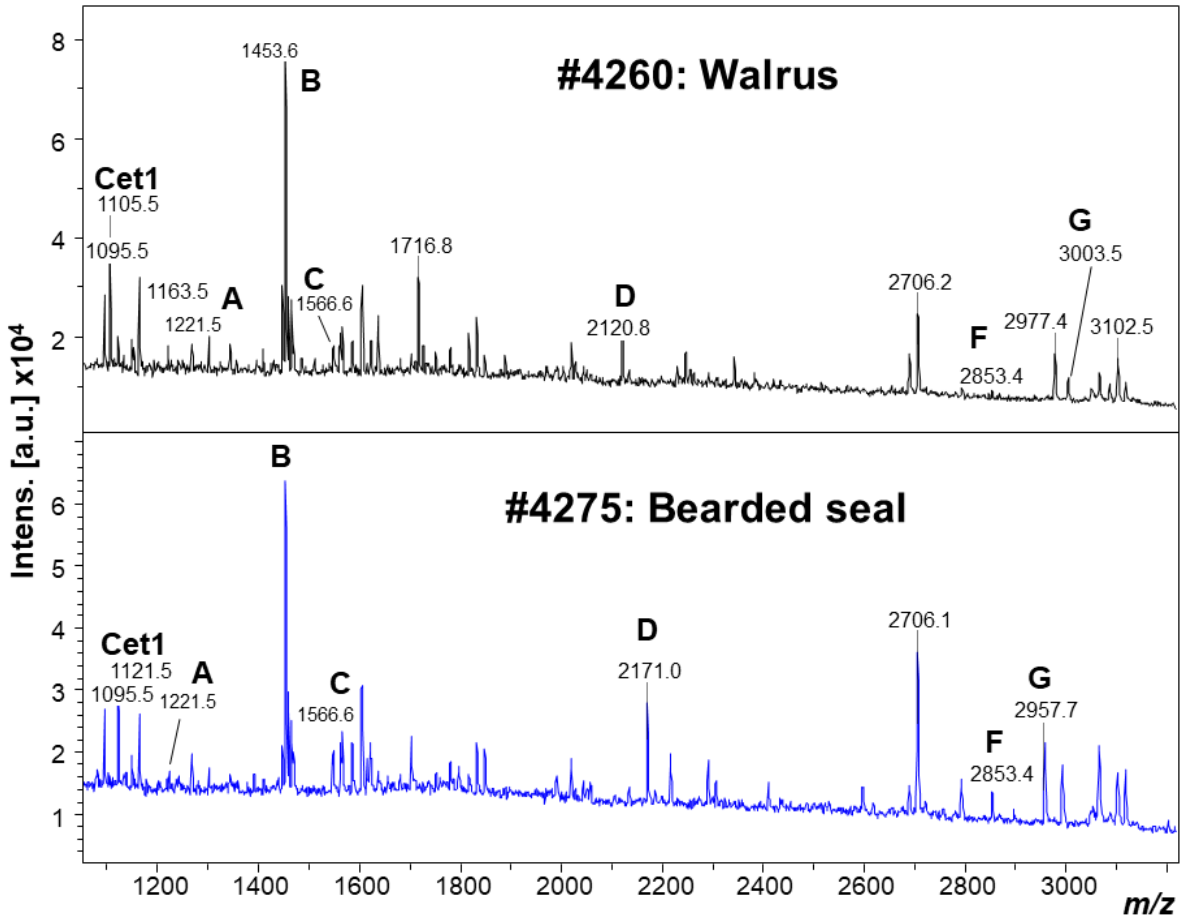

Figure S1. Representative collagen fingerprints for bearded seal and walrus analyzed as part of this study.

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