

Diet and behavior of the Saint-Césaire Neanderthal inferred from biogeochemical data inversion

Vincent Balter*, Laurent Simon

UMR 5125 CNRS-Lyon 1 "PaléoEnvironnement et PaléobioSphère", Université Claude Bernard, Campus de la Doua, Bâtiment Géode, Bd du 11/11/1918, 69622 Villeurbanne Cedex, France

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Abstract

Biogeochemistry is a powerful tool for dietary reconstruction, and mixing equations can be used to quantify the contribution of multiple sources to an individual's diet. The goals of this paper are: 1) to generalize the inverse method to dietary mixtures; and 2) to reconstruct the diet of the Saint-Césaire Neanderthal using Sr/Ca and Ba/Ca data of the mineral fraction of bone (hydroxylapatite), and with published $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ data of the associated organic fraction of bone (collagen). A new method is proposed to calculate the maximum diagenetic contribution of the Sr/Ba ratio, assuming that the soil soluble fraction is the diagenetic end-member and, for a given fraction of diagenesis, allows the restoration of the original Sr/Ba ratio. Considering the Saint-Césaire Châtelperronian mammalian assemblage as the meat source, and on the basis of available Sr, Ba, and Ca contents of plants, the results indicate that the percentage of plants in the Neanderthal's diet must have been close to zero for realistic Sr and Ba impoverishment between diet and hydroxylapatite. Contrary to previous studies, it is shown that fish could constitute a significant proportion (30%) of the diet of the Saint-Césaire Neanderthal. However, this mass balance solution is not supported by the zooarchaeological data. When the entire faunal assemblage is considered as the dietary source, the calculation shows that bovids (except reindeer) represent the greatest percentage of consumed meat (58%), followed by horses/rhinoceros (22%), reindeer (13%), and mammoths (7%). These respective percentages are in close accordance with zooarchaeological records, suggesting that the faunal assemblage associated with the Neanderthal of Saint-Césaire reliably reflects what he ate during the last few years of his life. In behavioral terms, this result supports the hypothesis that this Neanderthal carried the foodstuffs back to the Saint-Césaire shelter before their consumption.

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Introduction

Analysis of carbon ($^{13}\text{C}/^{12}\text{C}$) and nitrogen ($^{15}\text{N}/^{14}\text{N}$) stable isotope ratios of collagen, and of strontium/calcium (Sr/Ca) and barium/calcium (Ba/Ca) trace element ratios of hydroxylapatite, has yielded information on the diet of Neanderthals and anatomically modern humans (Fizet et al., 1995; Richards et al., 2000, 2001a; Balter et al., 2002; Bocherens et al., 2005). One advantage of these biogeochemical approaches is that dietary reconstructions are feasible independent of the taxonomic assessment of the fauna (anthropic and taphonomic

biases) associated with human remains. The biogeochemical method is based on the principle that stable isotopic and elemental ratios of a consumer are a function of its diet. Thus, the $^{13}\text{C}/^{12}\text{C}$ and $^{15}\text{N}/^{14}\text{N}$ values (noted hereafter in δ notation relative to PDB and AIR international standards) of a consumer ($\delta^{13}\text{C}_c$ and $\delta^{15}\text{N}_c$, respectively) are linked to the $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values of the proteins in its diet ($\delta^{13}\text{C}_d$ and $\delta^{15}\text{N}_d$, respectively) by the relationships

$$\delta^{13}\text{C}_c = \delta^{13}\text{C}_d + \Delta^{13}\text{C} \quad (1)$$

and

$$\delta^{15}\text{N}_c = \delta^{15}\text{N}_d + \Delta^{15}\text{N}, \quad (2)$$

* Corresponding author.

E-mail address: vincent.balter@univ-lyon1.fr (V. Balter).

where $\Delta^{13}\text{C}$ and $\Delta^{15}\text{N}$ are the ^{13}C and ^{15}N enrichments between a consumer and the dietary proteins. If the δ notation is not used, equations (1) and (2) are written

$$(^{13}\text{C}/^{12}\text{C})_c = (^{13}\text{C}/^{12}\text{C})_d \alpha_{c-d} \quad (3)$$

and

$$(^{15}\text{N}/^{14}\text{N})_c = (^{15}\text{N}/^{14}\text{N})_d \alpha_{c-d}, \quad (4)$$

where $^{13}\alpha_{c-d}$ and $^{15}\alpha_{c-d}$ are the ^{13}C and ^{15}N fractionation factors between a consumer and the dietary proteins. These isotopic enrichments (or fractionations) of collagen relative to the food were thought to be due to isotopic fractionation that occurs during deamination, transamination, and amino-acid synthesis (e.g., Ambrose and Norr, 1993; Ambrose, 2000). Despite poor understanding of the mechanisms that account for these isotopic enrichments, empirical data show that $\Delta^{13}\text{C}$ and $\Delta^{15}\text{N}$ may vary for large mammals from 3.3‰ to 3.8‰ and 3‰ to 5‰, respectively (e.g., Ambrose and Norr, 1993; Ambrose, 2000). Similar to isotopic systems, the Sr/Ca and Ba/Ca ratios of the mineral fraction of a consumer's bone [(Sr/Ca)_c and (Ba/Ca)_c, respectively] are linked to the Sr/Ca and Ba/Ca ratios of its diet [(Sr/Ca)_d and (Ba/Ca)_d, respectively] by the relationships

$$(\text{Sr}/\text{Ca})_c = (\text{Sr}/\text{Ca})_d \text{OR}_{\text{Sr}} \quad (5)$$

and

$$(\text{Ba}/\text{Ca})_c = (\text{Ba}/\text{Ca})_d \text{OR}_{\text{Ba}}, \quad (6)$$

where OR_{Sr} and OR_{Ba} are the Sr and Ba “Observed Ratios” (i.e., equivalent to the above mentioned α) between diet and bone. The Sr/Ca and Ba/Ca ratios diminish during metabolic processes involving Ca because Sr and Ba are not essential elements and are discriminated relative to Ca. This process, termed Ca biopurification (Comar et al., 1957; Wasserman et al., 1957), implies that the Sr/Ca and Ba/Ca ratios decrease with ascending trophic position in food chains. A review of the available data shows that the values of OR_{Sr} and OR_{Ba} vary from 0.16 to 0.39 and from 0.08 to 0.33, respectively, with a mean of 0.30 ± 0.08 and 0.16 ± 0.08 , respectively (Balter, 2004). Various environmental factors lead to isotopic and elemental variations in trophic chains, including for instance the proportion of C3 vs C4 plants (e.g., van der Merwe, 1989), the speciation of available N in soils (Nadelhoffer et al., 1996; Michelsen et al., 1998), and the composition of the geological substrate (Elias et al., 1982; Burton et al., 1999). As a consequence, the isotopic and elemental patterns are always independently calibrated on the spatial and temporal scale, and the geochemical results of the fossil hominids are presented with those of the associated fauna. The trophic position of fossil hominids is thus qualitatively deduced by comparison with contemporary herbivores and carnivores.

However, the quantitative determination of diet composition can be performed by mass balance equations which are of the form

$$(y/Y)_{\text{mx}} [Y]_{\text{mx}} = (y/Y)_1 [Y]_1 X_1 + (y/Y)_2 [Y]_2 X_2, \quad (7)$$

where the subscripts 1, 2, and mx represent two food sources and the mixture, respectively, (y/Y) represents the isotopic or elemental ratios, [Y] equals the concentration of the most abundant isotope or element, and X is the fractional contribution of each food source. For instance, considering Sr/Ca and Ba/Ca ratios and three end-members, the set of equations is written as follows:

$$(\text{Sr}/\text{Ca})_{\text{mx}} = \left(\frac{\text{Sr}}{\text{Ca}}\right)_1 \frac{[\text{Ca}]_1}{[\text{Ca}]_{\text{mx}}} X_1 + \left(\frac{\text{Sr}}{\text{Ca}}\right)_2 \frac{[\text{Ca}]_2}{[\text{Ca}]_{\text{mx}}} X_2 + \left(\frac{\text{Sr}}{\text{Ca}}\right)_3 \frac{[\text{Ca}]_3}{[\text{Ca}]_{\text{mx}}} X_3, \quad (8)$$

$$(\text{Ba}/\text{Ca})_{\text{mx}} = \left(\frac{\text{Ba}}{\text{Ca}}\right)_1 \frac{[\text{Ca}]_1}{[\text{Ca}]_{\text{mx}}} X_1 + \left(\frac{\text{Ba}}{\text{Ca}}\right)_2 \frac{[\text{Ca}]_2}{[\text{Ca}]_{\text{mx}}} X_2 + \left(\frac{\text{Ba}}{\text{Ca}}\right)_3 \frac{[\text{Ca}]_3}{[\text{Ca}]_{\text{mx}}} X_3, \quad (9)$$

which can be solved with a classical pivoting method. However, as the number of equations increases, the resolution of the set of equations becomes rapidly cumbersome. Another way to write this set of equations is in matrix form:

$$\begin{bmatrix} \left(\frac{\text{Sr}}{\text{Ca}}\right)_{\text{mx}} \\ \left(\frac{\text{Ba}}{\text{Ca}}\right)_{\text{mx}} \end{bmatrix} = \begin{bmatrix} \left(\frac{\text{Sr}}{\text{Ca}}\right)_1 \frac{[\text{Ca}]_1}{[\text{Ca}]_{\text{mx}}} & \left(\frac{\text{Sr}}{\text{Ca}}\right)_2 \frac{[\text{Ca}]_2}{[\text{Ca}]_{\text{mx}}} & \left(\frac{\text{Sr}}{\text{Ca}}\right)_3 \frac{[\text{Ca}]_3}{[\text{Ca}]_{\text{mx}}} \\ \left(\frac{\text{Ba}}{\text{Ca}}\right)_1 \frac{[\text{Ca}]_1}{[\text{Ca}]_{\text{mx}}} & \left(\frac{\text{Ba}}{\text{Ca}}\right)_2 \frac{[\text{Ca}]_2}{[\text{Ca}]_{\text{mx}}} & \left(\frac{\text{Ba}}{\text{Ca}}\right)_3 \frac{[\text{Ca}]_3}{[\text{Ca}]_{\text{mx}}} \end{bmatrix} \times \begin{bmatrix} X_1 \\ X_2 \\ X_3 \end{bmatrix}, \quad (10)$$

which can be noted as

$$m = AX, \quad (11)$$

where m is the matrix of the mixing values (i.e., the measurements), and $A \cdot X$ is the matrix product of A (i.e., the values of the parameters $(\text{Sr}/\text{Ca})_{1 \rightarrow 3} [\text{Ca}]_{1 \rightarrow 3} / [\text{Ca}]_{\text{mx}}$ and $(\text{Ba}/\text{Ca})_{1 \rightarrow 3} [\text{Ca}]_{1 \rightarrow 3} / [\text{Ca}]_{\text{mx}}$) by X (i.e., the values of $X_{1 \rightarrow 3}$). In the matrix inverse method, we solve for the matrix variable X by left-multiplying both sides of the above matrix equation by the inverse matrix of A (A^{-1}). We thus obtain:

$$X = A^{-1}m. \quad (12)$$

The inverse methods, which were developed in the field of earth sciences (e.g., Tarantola, 1987; Albarède, 1995), are easy to compute with matrix calculations (Phillips and Koch, 2002). However, the state of information of the system to resolve is a problem of threshold: when the system is over-determined (i.e., at least $n+1$ proxies for n sources), the solution is non-unique. When the system is under-determined (i.e., at least $n+1$ sources for n proxies), the solution becomes unstable. An over-determined system is potentially useful because the variability of the solution will be reduced if a unique

solution is obtained. One way to obtain a unique solution for an over-determined system is to work with least-square methods (Tarantola, 1987). These calculations involve the determination of \bar{X} (the matrix of the least-square solutions of X), which brings the model parameters and measurement values as close together as possible through the relation

$$\bar{X} = B^{-1}A'm, \quad (13)$$

where A' is the transposed matrix of A , and the matrix B^{-1} equals the inverse matrix of the matrix product $A' \cdot A$. Finally, it should be noted that this calculation is for a system which could be described with linear equations (here the mixing equations).

The purpose of this paper is to apply the least-squares method to biogeochemical data inversion in order to quantitatively reconstruct the diet of the Saint-Césaire I Neanderthal. In each case, we deliberately set the system to an over-determined state using elemental values (Sr/Ca and Ba/Ca) and isotopic values ($\delta^{13}\text{C}$ and $\delta^{15}\text{N}$) which were measured on the Neanderthal and the associated mammalian fauna (Balter et al., 2002; Bocherens et al., 2005). Prior to any dietary reconstruction, the effect of diagenesis is quantified with a new mass balance model.

Materials and methods

The materials consist of the faunal remains which were recovered in the Castelperronian layer EJOP_{sup} of Saint-Césaire in association with the Neanderthal specimen (Lévêque, 1993; Patou-Mathis, 1993; Vandermeersch, 1993). These materials include reindeer (*Rangifer tarandus*), giant deer (*Megaloceros giganteus*), red deer (*Cervus elaphus*), bovids (*Bos sp.*), horses (*Equus caballus*), woolly rhinoceros (*Coelodonta antiquitatis*), and mammoths (*Mammuthus primigenius*).

All the samples were taken from the cortical part of the bone and were ground and etched with an excess of dilute acetic acid (0.1 M) for one hour. The residual material was rinsed and etched with nitric acid (4.5 M) for one hour, and the solution was kept for analysis. Ca, Sr, and Ba concentrations were measured by atomic absorption spectrometry. Fluorine (F) is known to accumulate in the hydroxyl crystallographic site of apatite as a function of the intensity of diagenesis (e.g., Michel et al.,

1996). Here, the F concentrations were measured by potentiometry using a specific electrode. The standard NIST SRM1400 “Bone Ash” was used for quality control. All the results are reported in Table 1.

The problem of diagenesis is quantitatively approached by testing if the measured Sr/Ca and Ba/Ca values of the Saint-Césaire Neanderthal could result from a balance between biological (dietary) and diagenetic end-members. To assess the diagenetic end-member value, we measured the Sr/Ba ratio in the soluble fraction of sediment according to the procedure of Shirahata et al. (1980). This ratio does not take Ca into account, excluding any calcite dissolution and incorrect diagenetic Sr/Ca and Ba/Ca ratios. Thus, the mixing equation takes into account the soil soluble fraction Sr/Ba ratio (i.e., the diagenetic end-member) and the Sr/Ba ratio of the Saint-Césaire I Neanderthal's bone (i.e., the biological end-member), whose value is stated to derive from diet. This mixing model is developed in the Appendix.

The end-member values of Sr, Ba, Ca, $\delta^{13}\text{C}$, $\delta^{15}\text{N}$, ^{12}C , and ^{14}N are listed in Table 2. A rarely mentioned condition of the trophic impoverishment of the Sr/Ca and Ba/Ca ratios is that they must be equal in meat and bones. This condition is implicitly verified because the OR_{Sr} and OR_{Ba} ratios between herbivores and plants on the one hand, and between carnivores and herbivores on the other, are similar (Elias et al., 1982; Gilbert et al., 1994; Burton et al., 1999). Thus, meat Sr and Ba proportions are deduced from measurements carried out on the bones of the ungulates of Saint-Césaire, assuming a constant value of 100 $\mu\text{g/g}$ for the meat Ca content (see Table 2 and references herein). A value of 2.5‰ was set for the ^{13}C enrichment between meat and collagen (e.g., Jim et al., 2004). The elemental values for plants are derived from the extensive survey of Burton et al. (1999).

For calculations, we rearrange equation (7) as follows:

$$0 = ((y/Y)_1 - (y/Y)_{\text{mx}})[Y]_1 X_1 + ((y/Y)_2 - (y/Y)_{\text{mx}})[Y]_2 X_2. \quad (14)$$

Considering the Sr/Ca, Ba/Ca, $\delta^{13}\text{C}$, and $\delta^{15}\text{N}$ proxies, the matrix equation is written for a mixing of two dietary sources A and B and their mixture as follows:

$$\begin{bmatrix} 0 \\ 0 \\ 0 \\ 0 \end{bmatrix} = \begin{bmatrix} X_A \\ X_B \end{bmatrix} \begin{bmatrix} \left(\left(\frac{\text{Sr}}{\text{Ca}} \right)_A - \left(\frac{\text{Sr}}{\text{Ca}} \right)_{\text{mx}} \right) [\text{Ca}]_A & \left(\left(\frac{\text{Sr}}{\text{Ca}} \right)_B - \left(\frac{\text{Sr}}{\text{Ca}} \right)_{\text{mx}} \right) [\text{Ca}]_B \\ \left(\left(\frac{\text{Ba}}{\text{Ca}} \right)_A - \left(\frac{\text{Ba}}{\text{Ca}} \right)_{\text{mx}} \right) [\text{Ca}]_A & \left(\left(\frac{\text{Ba}}{\text{Ca}} \right)_B - \left(\frac{\text{Ba}}{\text{Ca}} \right)_{\text{mx}} \right) [\text{Ca}]_B \\ \left(\left(\frac{^{13}\text{C}}{^{12}\text{C}} \right)_A - \left(\frac{^{13}\text{C}}{^{12}\text{C}} \right)_{\text{mx}} \right) [^{12}\text{C}]_A & \left(\left(\frac{^{13}\text{C}}{^{12}\text{C}} \right)_B - \left(\frac{^{13}\text{C}}{^{12}\text{C}} \right)_{\text{mx}} \right) [^{12}\text{C}]_B \\ \left(\left(\frac{^{15}\text{N}}{^{14}\text{N}} \right)_A - \left(\frac{^{15}\text{N}}{^{14}\text{N}} \right)_{\text{mx}} \right) [^{14}\text{N}]_A & \left(\left(\frac{^{15}\text{N}}{^{14}\text{N}} \right)_B - \left(\frac{^{15}\text{N}}{^{14}\text{N}} \right)_{\text{mx}} \right) [^{14}\text{N}]_B \end{bmatrix}. \quad (15)$$

Table 1
List of log(Sr/Ca), log(Ba/Ca), %F, and %N of the analyzed material from Saint-Césaire

Lab N°	Field N°	Species	Sample [#]	Log (Sr/Ca)	Log (Ba/Ca)	F (%)	N (%)
P706	EJOPsup. E5	<i>Rangifer tarandus</i>	Mandible	-3.32	-3.21	0.74	
P723	EJOPsup. I4(I) 22i	<i>Rangifer tarandus</i>	Metapod	-3.28	-3.20	0.51	
P730	EJOPsup. G5(II) 23s	<i>Rangifer tarandus</i>	Mi	-3.23	-2.93	2.18*	0.1
P746	EJOP E7(III) 32s	<i>Rangifer tarandus</i>	Mandible	-3.52	-3.14	0.70	0.4
P708	EJOPsup. I5 22s	<i>Bos sp.</i>	Metapod	-3.39	-3.15	0.37	
P709	EJOPsup. I5 22s	<i>Bos sp.</i>	Metapod	-3.39	-3.15	0.45	
P718	EJOPsup. F6(II) 26i	<i>Bos sp.</i>	M3i	-3.44	-3.12	0.64	
P724	EJOPsup. H6(I) 25	<i>Bos sp.</i>	Mandible	-3.39	-3.09	0.47	
P725	EJOPsup. I5(IV) 24i	<i>Bos sp.</i>	Metapod	-3.38	-3.04	0.37	
P733	EJOPsup. D6(II) 28i	<i>Bos sp.</i>	Bone (ind.)	-3.32	-3.11	0.56	0.2
P734	EJOPsup. H6(I) 25i	<i>Bos sp.</i>	Bone (ind.)	-3.37	-3.08	1.25*	0.1
P748	EJOP E7(I) 32s	<i>Bos sp.</i>	Ps	-3.38	-3.14	0.57	0.3
P749	EJOP D5(II) 27s	<i>Bos sp.</i>	I	-3.22	-3.12	0.69	0.2
P750	EJOP G2(IV) 32i	<i>Bos sp.</i>	Tooth	-3.36	-3.10	0.65	0.3
P751	EJOP E7(I) 31i	<i>Bos sp.</i>	Tooth	-3.33	-3.15	0.53	0.2
P752	EJOP G5(IV) 24s	<i>Bos sp.</i>	Tooth	-3.36	-3.29	0.42	0.1
P753	EJOP D5(II) 26	<i>Bos sp.</i>	Tooth	-3.09	-3.35	0.56	0.1
P711	EJOPsol E8(I)	<i>Mammuthus primigenius</i>	M	-3.48	-3.32	0.39	
P713	EJOPsol E8(I)	<i>Mammuthus primigenius</i>	M	-3.24	-3.38	0.46	
P729	EJOPsup. H5(III) 24i	<i>Mammuthus primigenius</i>	M	-3.34	-3.37	0.61	0.1
P714	EJOPsup. (III-IV) 29s	<i>Caelodonta antiquitatis</i>	M1-2i	-3.32	-3.17	0.80	
P715	EJOPsup. F5(IV)	<i>Caelodonta antiquitatis</i>	P2s	-3.21	-3.27	0.77	
P731	EJOPsup. G5(III) 23i	<i>Caelodonta antiquitatis</i>	Tooth	-3.33	-3.38	0.49	0.1
P747	EJOP G5(IV) 24s	<i>Caelodonta antiquitatis</i>	Humerus	-3.43	-3.46	0.55	0.1
P732	EJOPsup. I4(IV) 25s-26s	<i>Equus sp.</i>	Tooth	-3.30	-3.40	0.44	0.1
P736	EJOPsup. E5(IV) 27i	<i>Equus sp.</i>	C	-3.36	-3.47	2.03*	0.1
P738	EJOPsup. G4(II) 23	<i>Megaloceros giganteus</i>	Maxillary	-3.53	-3.26	1.40*	0.7
P739	EJOPsup. G4(II) 24	<i>Megaloceros giganteus</i>	M2s	-3.63	-3.29	0.42	0.1
P757	EJOP sup. F6 (II) 25s	<i>Cervus elaphus</i>	M1-2i	-3.27	-3.25	0.64	
P728	EJOPsup. E6(II) 24i-26i	<i>Sus scrofa</i>	Maxillary	-3.34	-3.45	0.63	0.1
P737	EJOPsup. E6(II) 24i-26i	<i>Sus scrofa</i>	Ms	-3.54	-3.42	1.53*	0.1
P755	EJOP sup. H4(IV) 23s	<i>Sus scrofa</i>	M3i	-3.55	-3.51	0.57	
P754	EJOP sup. G7(III) 30s	<i>Vulpes vulpes</i>	M	-3.49	-3.42	0.63	
P740	EJOP F3(IV) 23s	<i>Carnivore ind.</i>	C	-3.66	-3.70	0.56	0.1
P741	EJOP E6(II) 27s	<i>Carnivore ind.</i>	Tibia	-3.31	-3.40	0.58	0.1
P743	EJOPsup.	<i>Homo sapiens neanderthalensis</i>	Fibula	-3.57	-3.65	0.48	0.6
G714	RPB H6-G6	Sediment		-3.61 [§]	-3.63 [§]		
G722	RPB H5(IV) 24	Sediment		-3.59 [§]	-3.66 [§]		

[#] For teeth, only dentine was sampled. No difference was found between dentine and bone from the elemental point of view (Balter et al., 2002).

* These values were rejected due to high fluorine content.

§ These values were rejected due to calcite dissolution.

$(\text{Sr}/\text{Ca})_{\text{mx}}$, $(\text{Ba}/\text{Ca})_{\text{mx}}$, $(^{13}\text{C}/^{12}\text{C})_{\text{mx}}$, and $(^{15}\text{N}/^{14}\text{N})_{\text{mx}}$ are replaced by $(\text{Sr}/\text{Ca})_{\text{Hsn}}/\text{OR}_{\text{Sr}}$, $(\text{Ba}/\text{Ca})_{\text{Hsn}}/\text{OR}_{\text{Ba}}$, $(^{13}\text{C}/^{12}\text{C})_{\text{Hsn}}/^{13}\alpha$, and $(^{15}\text{N}/^{14}\text{N})_{\text{Hsn}}/^{15}\alpha$, respectively, where $10^3\ln(^{13}\alpha) = \Delta^{13}\text{C}$ and $10^3\ln(^{15}\alpha) = \Delta^{15}\text{N}$, and where the subscript Hsn stands for the Neanderthal measured value.

Concerning the matrix computing, we include a closure condition which constrains the least-square solution to sum up the different $X_{1 \rightarrow n}$ to unity (Albarède, 1995). All the steps of the matrix calculation are clearly explained and illustrated in Albarède (1995).

Results and discussion

The Sr/Ca and Ba/Ca values are lower in carnivores than in herbivores, a pattern in agreement with expected natural trophic biopurification of Ca (Table 1). The Ba/Ca values of monogastric ungulates (mammoth, horses, and rhinos) are significantly lower (Student's t test: 5.21; $p = 0.00002$) than

for digastric ungulates (bison, deer, and reindeer). Correlation analysis between Sr/Ca and nitrogen content (%N; $n = 34$; $r = 0.229$; $p = 0.193$) and fluorine content (%F; $n = 48$; $r = -0.014$; $p = 0.925$), and between Ba/Ca and %N ($n = 34$; $r = 0.308$; $p = 0.076$) and %F ($n = 48$; $r = 0.050$; $p = 0.737$) indicates no significant diagenetic effect. Nevertheless, some samples exhibit high F content (i.e., >1%) and were rejected for interpretations (Table 1). However, the absence of an obvious relationship between diagenetic and paleobiological proxies does not necessarily imply that diagenesis has not altered the original trace element composition of bones.

In order to test whether any diagenetic fraction could contribute to the overall elemental signature of the bone, we developed a simple mixing model which takes into account the soil soluble fraction Sr/Ba ratio, $(\text{Sr}/\text{Ba})_{\text{Dg}}$, and the Sr/Ba ratio of the Saint-Césaire Neanderthal's bone $(\text{Sr}/\text{Ba})_{\text{Bg}}$, which is assumed to derive from a dietary component. The measured Sr/Ba value of the Saint-Césaire Neanderthal, $(\text{Sr}/\text{Ba})_{\text{Hsn}}$,

Table 2

Sr, Ba, Ca, $\delta^{13}\text{C}$, $\delta^{15}\text{N}$, ^{12}C , and ^{14}N values for meat, plant matter, fish, and soil end-members and for the Saint-Césaire Neanderthal. Values in brackets refer to reference: [1] <http://www.nal.usda.gov/fnic/foodcomp/search/>; [2] Elias et al. (1982); [3] Randouin et al. (1996); [4] Burton et al. (1999); [5] Allen et al. (2001); [6] Ciesielski et al. (2006); [7] Dufour et al. (1999); [8] Richards et al. (2001b). The apparent elemental and isotopic fractionation between the Neanderthal and a given end-member is obtained with equations (1), (2), (5), and (6)

		Sr ($\mu\text{g/g}$)	Ba ($\mu\text{g/g}$)	Ca ($\mu\text{g/g}$)	$\delta^{13}\text{C}$ (‰ vs PDB)	$\delta^{15}\text{N}$ (‰ vs AIR)	^{12}C (wt %)	^{14}N (wt %)
Meat	Total	0.0461 ± 0.0113	0.0615 ± 0.0154	100 [1,2,3]	-20.3 ± 0.7	6.2 ± 1.3	50 [1]	15 [1]
	Monogastric	0.0475 ± 0.0088	0.0462 ± 0.0096	<i>id.</i>	-20.8 ± 0.7	6.8 ± 1.5	<i>id.</i>	<i>id.</i>
	Digastric	0.0455 ± 0.0122	0.0686 ± 0.0119	<i>id.</i>	-20.0 ± 0.6	6.0 ± 1.1	<i>id.</i>	<i>id.</i>
	Bovinae (except reindeer)	0.0459 ± 0.0131	0.0693 ± 0.0134	<i>id.</i>	-20.4 ± 0.3	6.1 ± 1.0	<i>id.</i>	<i>id.</i>
	Horse + Rhinoceros	0.04875 ± 0.0088	0.0475 ± 0.0132	<i>id.</i>	-20.5 ± 0.6	6.3 ± 1.4	<i>id.</i>	<i>id.</i>
	Reindeer	0.0435 ± 0.0118	0.0657 ± 0.0059	<i>id.</i>	-19.0 ± 0.4	5.5 ± 1.5	<i>id.</i>	<i>id.</i>
	Mammoth	0.0455 ± 0.0122	0.0441 ± 0.0033	<i>id.</i>	-21.6 ± 0.3	8.3 ± 0.4	<i>id.</i>	<i>id.</i>
	Vegetable	28 ± 20 [4]	69 ± 56 [4]	9300 \pm 6600 [4]	—	—	—	—
Fish	1–100 [5,6] [§]	0.1–10 [5,6] [§]	300 [1,3]	-22.0 [7,8]	10.0 [7,8]	55 [1]	12 [1]	
Soil	93	83	<i>n.a.</i>	<i>n.a.</i>	37E^4 #	<i>n.a.</i>	<i>n.a.</i>	
Neanderthal	86	73	3.2E^5	-19.8	11.4	<i>n.a.</i>	<i>n.a.</i>	

id. idem to total concentration.

n.a. not applicable.

[§] In all cases, the Sr/Ba ratio is about 10.

This value was rejected as calcite dissolution occurred.

can thus be calculated as a mixture of $(\text{Sr}/\text{Ba})_{\text{Dg}}$ and $(\text{Sr}/\text{Ba})_{\text{Bg}}$; these fractions (X_{Dg} and X_{Bg} , respectively) sum up to 1 following the equation:

$$(\text{Sr}/\text{Ba})_{\text{Hsn}} = X_{\text{Dg}}(\text{Sr}/\text{Ba})_{\text{Dg}} + X_{\text{Bg}}(\text{Sr}/\text{Ba})_{\text{Bg}} \quad (16)$$

The complete relationship is fully described in the Appendix. The results indicate that when the $\text{OR}_{\text{Sr}}/\text{OR}_{\text{Ba}}$ ratio is set at 1.87 (the mean value calculated from the extant trophic web), the proportion of the diagenetic component increases with decreasing amounts of vegetation in the diet, reaching a maximum value of 27% when Saint-Césaire 1 is assumed to be 100% carnivorous (Fig. 1). This diagenetic fraction

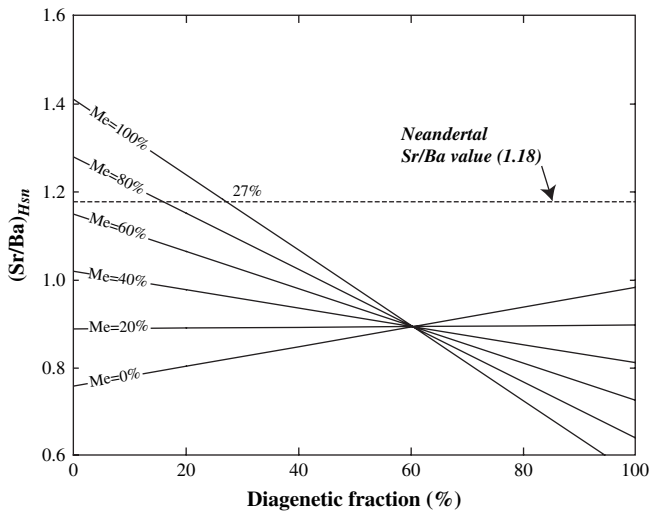


Fig. 1. Variation of the diagenetic fraction of bone as a function of the measured Sr/Ba ratio of the Saint-Césaire Neanderthal. The variation of diagenetic fraction is given as a function of meat percentage in the diet (Me%). Mean values are set for meat and plant Sr/Ba ratios (Table 2).

should be considered only as a possible maximum value because we cannot precisely assess the real value of OR_{Sr} and OR_{Ba} between the Neanderthal and his diet. However, using equation (16), we can calculate for this maximum diagenetic fraction a “reconstructed” $(\text{Sr}/\text{Ba})_{\text{Bg}}$ ratio equal to 1.25, giving a value of 1.18 and 0.98 for $(\text{Sr}/\text{Ba})_{\text{Mes}}$ and $(\text{Sr}/\text{Ba})_{\text{Dg}}$, respectively. If we state that no diagenesis occurred at Saint-Césaire, then $(\text{Sr}/\text{Ba})_{\text{Mes}}$ equals $(\text{Sr}/\text{Ba})_{\text{Bg}}$ and an “apparent” $\text{OR}_{\text{Sr}}/\text{OR}_{\text{Ba}}$ ratio of 1.60 can be calculated. It is likely that the variability of the dietary solutions ranges within limits that are calculated by the “apparent” and “reconstructed” sets of parameters. Thus, the dietary results will be systematically calculated with these two sets of values. This mixing model could be generalized for further trace element-based paleodietary reconstructions to quantify the possible contribution of diagenesis. However, while simpler mixing could be achieved with Sr/Ca and Ba/Ca ratios, extreme care is needed for the determination of Ca concentration in soil soluble fractions to avoid the dissolution of calcite.

The variability of the dietary solutions is investigated for a binary mixing of plant matter and meat using the Sr/Ca and Ba/Ca ratios and varying OR_{Sr} and OR_{Ba} values. When the “reconstructed” set of parameters is used ($\text{OR}_{\text{Sr}}/\text{OR}_{\text{Ba}} = 1.87$; $(\text{Sr}/\text{Ba})_{\text{Hsn}} = 1.25$; case (1), Fig. 2), the fraction of consumed plant matter increases with decreasing values of OR_{Sr} . For an OR_{Sr} of 0.1, a value lower than the minimum OR_{Sr} mean value, the fraction of consumed plant matter is about 1.1%. Using the “apparent” set of parameters ($\text{OR}_{\text{Sr}}/\text{OR}_{\text{Ba}} = 1.60$; $(\text{Sr}/\text{Ba})_{\text{Hsn}} = 1.18$; case (2), Fig. 2), the maximum fraction of consumed plant matter is about 0.9% for an OR_{Sr} of 0.1. Taken together, these results indicate that there is no significant difference in the fraction of consumed plant matter calculated with the “reconstructed” and “apparent” sets of parameters. This implies that the involvement of

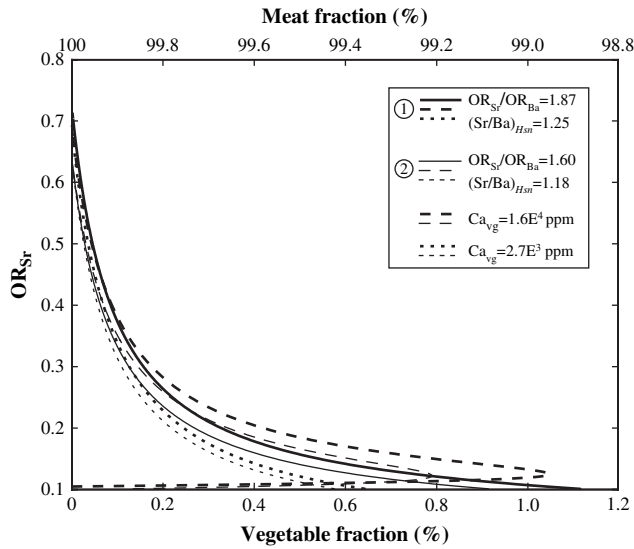


Fig. 2. Variation of the plant matter and meat fractions consumed by the Neanderthal as a function of OR_{Sr} . The calculations involve the “reconstructed” and “apparent” sets of parameters which are figured by case (1) and (2), respectively. The vegetable Ca content (Ca_{veg}) is $9.3E^3$ ppm when not mentioned (see Table 2 for end-member values). The curve obtained for the inverse value of (28/69), i.e., 0.4, and $Ca_{veg} = 9.3E^3$ ppm, is not shown because it is indistinguishable from the solid line of case (2).

a diagenetic fraction as deduced from equation (13) does not alter the overall dietary pattern obtained with a mixture of plant matter and meat. The fact that sizable fractions of plant matter are only computed with very low OR_{Sr} and OR_{Ba} values is due to the tremendous difference between the elemental composition of plant matter and meat, the latter being one order of magnitude lower than the former. This conclusion remains valid even when extreme values are set for plant resources: a similar fraction of consumed plant matter ($<0.8\%$) is calculated when the plant matter Ca content is set at $2.7E^4$ ppm and $15.9E^4$ ppm, and when the plant matter Sr/Ba ratio is inverted relative to its normal value (2.5, i.e., $1/(28/69)$; Table 2).

Let us consider now a mixture of fish and meat (Table 2; Fig. 3). The two curves obtained with the “reconstructed” and “apparent” sets of parameters are indistinguishable, except when the fish Sr/Ca ratio, $(Sr/Ca)_{Fi}$, is set at $3E^{-3}$. In this case, the fraction of consumed fish reaches a maximum of 3.8% and 2.6% when the “apparent” and “reconstructed” sets of parameters are used. However, it should be noted that significant fractions of consumed fish are obtained with realistic OR_{Sr} ($0.2 < OR_{Sr} < 0.4$; Fig. 3). In order to test whether these respective fractions of consumed fish and meat are supported by stable isotopes, we compute a mixing model including Sr/Ca, Ba/Ca, $^{13}C/^{12}C$, and $^{15}N/^{14}N$ ratios, leading to a matrix equation with the form of equation (12). The fraction of consumed fish, obtained with mean end-member $\delta^{13}C$ and $\delta^{15}N$ values (Table 2), mean enrichment factor $\Delta^{13}C$ and $\Delta^{15}N$ values (1‰ and 4‰, respectively), and an $(Sr/Ca)_{Fi}$ of $3E^{-2}$, is 15% whatever the value of OR_{Sr} (Fig. 4A). Moreover, the fraction of consumed fish does not vary with $\Delta^{15}N$ (Fig. 4B). It appears that the most sensitive parameter is

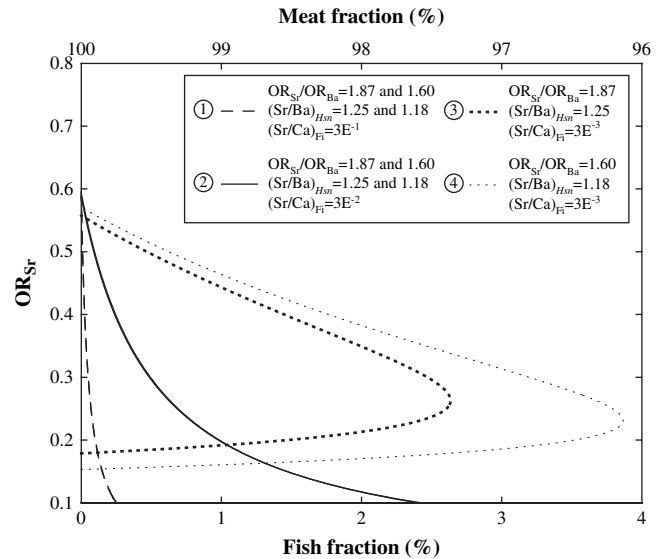


Fig. 3. Variation of the fish and meat fractions consumed by the Neanderthal as a function of OR_{Sr} . The calculations involve the “reconstructed” and “apparent” sets of parameters. In cases (1) and (2), the results are the same with both sets of parameters. Differences in the fish and meat fractions are introduced only when $(Sr/Ca)_{Fi}$ is set at $3E^{-3}$ (see Table 2 for end-member values).

$\Delta^{13}C$: the fraction of consumed fish is calculated to be about 30% and 2% when $\Delta^{13}C$ is set at 1.5‰ and 0.5‰, respectively, and when $(Sr/Ca)_{Fi}$ is set at $3E^{-2}$ (Fig. 4B).

Because of the variability associated with the above results, this hypothesis of fish consumption by the Saint-Césaire Neanderthal does not lead to definitive conclusions, nor can it be definitively rejected. This contrasts with the study of Richards et al. (2001b) which concludes that fish consumption is characteristic of anatomically modern humans. Clearly, more experimental work is needed to better control the variability of $\Delta^{13}C$. This might be achieved through $^{13}C/^{12}C$ controlled feeding studies of mammals (e.g., Ayliffe et al., 2004; Jim et al., 2004), and should result in a better understanding of the metabolic conditions that lead to an isotopic steady-state and of the respective routings that ^{12}C -composed macronutrients experience. However, it is recommended that carbon isotopic results be systematically measured for collagen and apatite in order to provide different and complementary insights into paleodiet (e.g., Ambrose and Norr, 1993). Zooarchaeological data at Saint-Césaire indicate that fish and shellfish were rare (Lavaud-Girard, 1993). Although this may be due to taphonomic processes, we hypothesize that fish consumption did not represent a significant fraction of the Neanderthal’s diet. The fact that the fish fraction is not sizable implies a $\Delta^{13}C$ value lower or equal to 0.5‰.

We further investigate the relative fractions of the different taxa on the basis of the Ba/Ca difference between digastric and monogastric ungulates. The meat fraction of digastric mammals equals 62.6% (Fig. 5A); this result does not vary significantly when Sr/Ca and Ba/Ca end-member variability is introduced into the mixing model. We performed another calculation which introduced end-member C and N stable isotopic variability, and found that the fraction of digastric

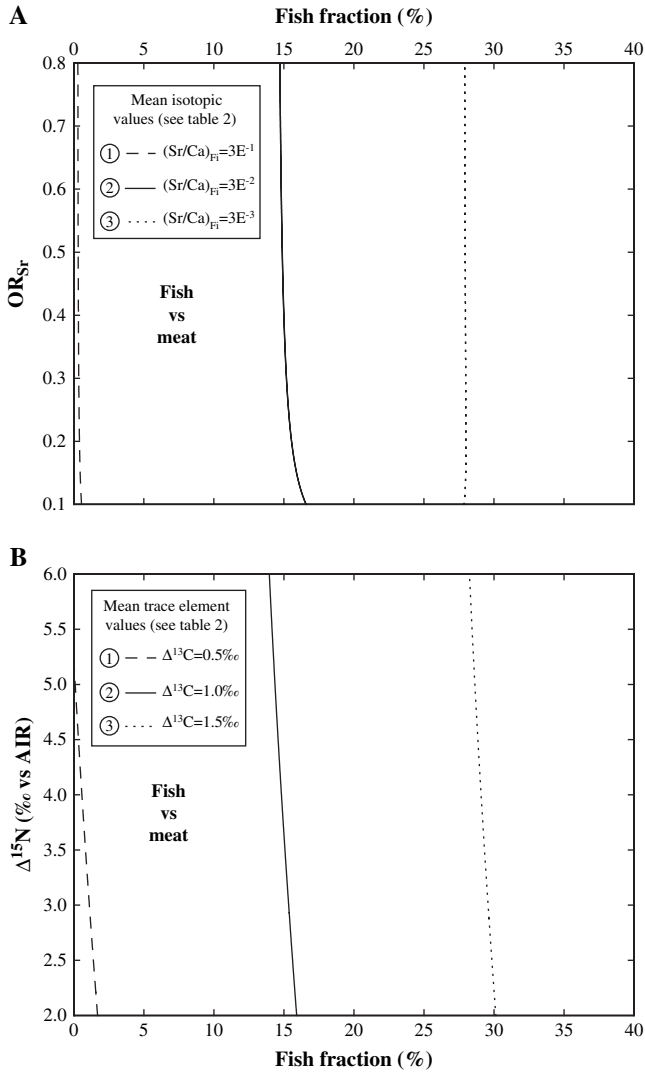


Fig. 4. A, Variation of the fish and meat fractions consumed by the Neanderthal as a function of OR_{Sr} (meat fractions are obtained with 100% – fish%). The “apparent” set of parameters is used for these calculations because it yields the higher proportion of fish (case (4) in Fig. 3). Mean isotopic values are set in each case (see Table 2 for end-member values). B, Variation of the fish and meat fractions consumed by the Neanderthal as a function of Δ¹⁵N (meat fractions are obtained with 100% – fish%). In each case, (Sr/Ca)_{F_i} is set at 3E⁻² (case (2) in Fig. 4A). The resulting fish fraction when (Sr/Ca)_{F_i} is set at 3E⁻³ (case (3) in Fig. 4A) for a Δ¹³C of 1.5‰ is about 45% (not shown). “Apparent” elemental fractionation coefficients are used in each case (see Table 2 for end-member values).

ungulate meat becomes highly variable when the difference between the monogastric and digastric δ¹³C meat values is minimal (case (2); Fig. 5B). However, such a small difference is unlikely to occur because reindeer and mammoth are clearly distinct on the basis of their collagen δ¹³C (Table 2). With the exception of this extreme case, taking into account overall variability of the stable isotopic and elemental parameters leads to a digastric meat fraction of 60 ± 2.5%. The zooarchaeological study conducted at Saint-Césaire, based on the minimal number of individuals and meat weight by taxon, yielded similar results of about 63% digastric meat (Patou-Mathis, 1993).

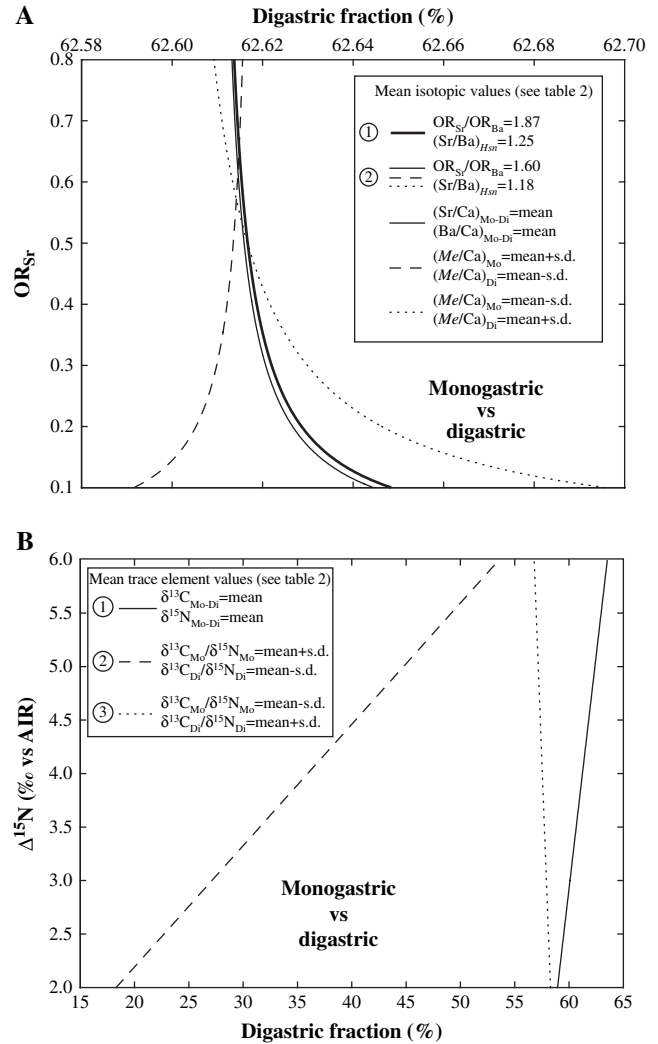


Fig. 5. A, Variation of the monogastric and digastric meat fractions consumed by the Neanderthal as a function of OR_{Sr} (monogastric fractions are obtained with 100% – digastric%). The calculations involve the “reconstructed” (case (1)) and “apparent” (case (2)) sets of parameters. *Me* stands for metals, i.e., Sr and Ba. Mean isotopic values are used in each case, except for Δ¹³C which is set at 0.5‰ (see Table 2 for end-member values). The calculations for Sr/Ca and Ba/Ca mean ± standard deviation (SD) values for monogastric (Mo) and digastric (Di) are derived from Table 2. B, Variation of the monogastric and digastric meat fractions consumed by the Neanderthal as a function of Δ¹⁵N (monogastric fractions are obtained with 100% – digastric%). The “apparent” set of parameters (case (2) in Fig. 5A) and “apparent” elemental fractionation coefficients are used for these calculations (see Table 2 for end-member values). The calculations for δ¹³C and δ¹⁵N mean ± standard deviation (SD) values for monogastric (Mo) and digastric (Di) fractions are derived from Table 2.

A last mixing model was carried out in which ungulates were pooled according to their elemental and isotopic pattern. Mammoths and reindeer were treated as separate end-members due to their contrasting δ¹³C and δ¹⁵N values. Bovids and deer were merged because they are both digastric and isotopically similar. Finally, horses and rhinoceros were associated to form a monogastric end-member with a homogeneous stable isotopic signature (Table 2). Results of this mixing model show that the dietary fractions of mammoth

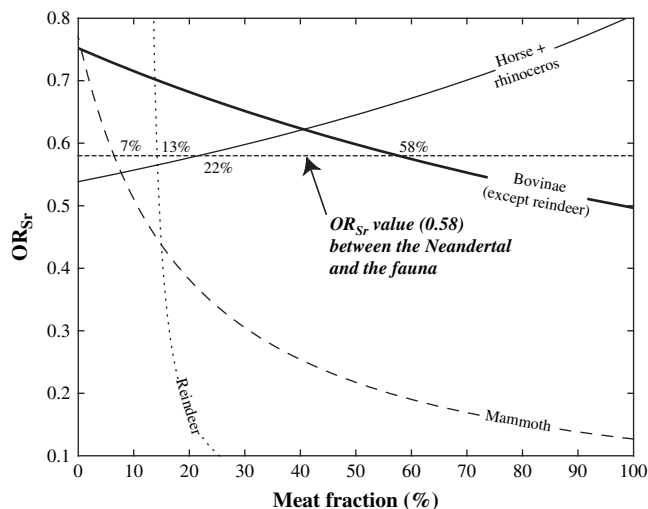


Fig. 6. Variation of the bovine (except reindeer), horse + rhinoceros, mammoth, and reindeer meat fractions consumed by the Neanderthal as a function of OR_{Sr} . Mean elemental and isotopic values are set in each case for end-member values (see Table 2). “Apparent” elemental and isotopic fractionation is used in each case (except for $\Delta^{13}C$ which is set at 0.5‰). Differences in the respective meat fractions with varying values of $\Delta^{15}N$ are indistinguishable from the presented curves.

and reindeer do not vary significantly with OR_{Sr} , and are equal to about 7% and 13%, respectively (Fig. 6). Using the calculated OR_{Sr} ratio between Saint-Césaire I and overall fauna (0.58) leads to a consumed fraction of reindeer and bovines of 22% and 58%, respectively. These results contrast significantly with those from Bocherens et al. (2005), in which the most representative ungulates in the Neanderthal’s diet are monogastric animals. However, in the present study, we found that the fraction of Bovinae (*Bos sp.*, *Megaloceros*, *Cervus*) alone can represent between one-half and two-thirds the total meat supply, a result in accordance with the zooarchaeology. The relative proportion of reindeer and mammoth in the diet are reversed depending on whether the zooarchaeological (3% and 16%, respectively) or biogeochemical (13%, and 7%, respectively) methods are considered, whereas the fraction of meat derived from horses and rhinoceros remains similar with both approaches (18% and 22% for zooarchaeology and biogeochemistry, respectively).

One must keep in mind that the steady-state biogeochemical composition of a mammal’s tissue is only governed by the residence time of the considered chemical proxy. In other words, the respective proportion of ungulates deduced from the present study must reflect an average diet acquired over several years (the residence time of trace elements and stable isotopes in apatite and collagen; e.g., Enlow, 1963). Thus, the similarity of the results obtained by biogeochemistry and zooarchaeology supports the hypothesis that the faunal assemblage at Saint-Césaire fairly reflects the dietary composition of the Neanderthal. This implies he might have brought all the foodstuffs back to the shelter. Likewise, Patou-Mathis (1993) demonstrated that the bone assemblage is anthropogenic, and the results presented here suggest that the game was transported back to the shelter for its final preparation and

consumption, excluding passive scavenging as a predominant subsistence strategy. However, if the Saint-Césaire Neanderthal was a scavenger, then the relative proportions of meat consumed in the shelter and through scavenging were similar, a pattern which cannot be excluded but remains improbable. Although this behavior cannot be attributed to the entire Neanderthal population, which was largely differentiated in time and space, the present study adds a new line of evidence that late Neanderthals were active hunters.

We stress that future paleodietary research should be carried out with a maximum of geochemical tracers including new dietary proxies (e.g., $\delta^{44}Ca$ and $\delta^{34}S$; Richards et al., 2001a; Clementz et al., 2003) in order to better constrain the chemical fluxes between a consumer and its diet. The geochemical results should in turn be compared to the zooarchaeological evidence for interpretations of behavior. However, it remains the case that the Sr/Ca and Ba/Ca ratios are interesting proxies because they can be measured with in situ instrumentation in enamel, providing a unique source of high resolution paleoecological and paleobiological data (Dolphin et al., 2005).

Conclusion

We show in this study that a least-squares-constrained inverse method of biogeochemical data can be applied to the problem of paleodietary reconstruction. Moreover, a mass balance model is proposed to quantify the maximum fraction of diagenesis in bones, and allows the restoration of the original Sr/Ba ratio for a given fraction of diagenesis. In the case of the Middle-Upper Palaeolithic site of Saint-Césaire, it is shown that even if 27% of the Sr/Ba ratio of the Neanderthal could be explained by diagenesis, plant resources did not constitute a sizable proportion of the diet of the Neanderthal. However, from the elemental and isotopic points of view, it is possible that fish may have contributed to the overall biogeochemical signature of the Neanderthal, although this result is not supported by zooarchaeological data. When the ungulates are pooled according to their elemental and isotopic pattern, their respective proportions in the diet of the Neanderthal resemble those deduced by zooarchaeology. This suggests that this Neanderthal brought the foodstuffs back to the site before their consumption, and thus did not practice passive scavenging.

The mass balance model for the determination of maximum diagenesis presented in this paper requires experimental confirmation, but it opens the possibility of the restoration of original trace element signatures in fossil bones, as has been proposed by Zazzo et al. (2004) concerning $\delta^{18}O$ in enamel. We also stress that a more comprehensive knowledge of the $^{13}C/^{12}C$ metabolic routing is needed for an accurate use of $\delta^{13}C$ in paleodietary reconstructions. Finally, it must be kept in mind that robust paleodietary conclusions are obtained only with an over-determined system (i.e., when the number of proxies is higher than the number of end-members). This is particularly true if one considers omnivorous diets (e.g., Plio-Pleistocene hominids and Holocene

humans), highlighting the need to continue the development of new dietary proxies.

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Appendix

If one states that the Sr/Ba ratio, $(\text{Sr/Ba})_{\text{Hsn}}$, of the Saint-Césaire Neanderthal (Hsn) is a balance between a diagenetic Sr/Ba end-member, $(\text{Sr/Ba})_{\text{Dg}}$, and a biological Sr/Ba end-member, $(\text{Sr/Ba})_{\text{Bg}}$, it can be written that $(\text{Sr/Ba})_{\text{Hsn}}$ is equal to

$$(\text{Sr/Ba})_{\text{Hsn}} = X_{\text{Dg}}(\text{Sr/Ba})_{\text{Dg}} + X_{\text{Bg}}(\text{Sr/Ba})_{\text{Bg}}, \quad (\text{A1})$$

where X_{Dg} and X_{Bg} are the diagenetic and biological fractions, respectively, and $X_{\text{Dg}} + X_{\text{Bg}} = 1$. $(\text{Sr/Ba})_{\text{Dg}}$ is the ratio of the Sr and Ba concentrations in the soil soluble fraction, i.e., the concentration in the solution. At steady-state conditions, the chemical composition of a mineral which is forming is a function of that in the solution. The relationship between the Sr/Ba ratio of the bone mineral fraction, i.e., hydroxylapatite $(\text{Sr/Ba})_{\text{ha}}$, and the Sr/Ba ratio of the solution, $(\text{Sr/Ba})_{\text{w}}$, is

$$K_{\text{Sr/Ba}}^{\text{ha-w}} = (\text{Sr/Ba})_{\text{ha}} / (\text{Sr/Ba})_{\text{w}}, \quad (\text{A2})$$

where $K_{\text{Sr/Ba}}^{\text{ha-w}}$ is the Sr/Ba partitioning coefficient between hydroxylapatite and the solution (i.e., water). If $(\text{Sr/Ba})_{\text{ha}}$ is the diagenetic component of the Saint-Césaire Neanderthal, it can be replaced by $(\text{Sr/Ba})_{\text{Dg}}^{\text{Hsn}}$ from equation (A1):

$$(\text{Sr/Ba})_{\text{Dg}}^{\text{Hsn}} = K_{\text{Sr/Ba}}^{\text{a-w}} (\text{Sr/Ba})_{\text{w}}. \quad (\text{A3})$$

The partitioning coefficient is temperature-dependent. In an experimental study, Balter and Lécuyer (2004) determined the values of $K_{\text{Sr/Ba}}^{\text{ha-w}}$ between 5 °C and 60 °C. At ambient temperature (25 °C), the value of $K_{\text{Sr/Ba}}^{\text{ha-w}}$ is 0.882.

If no dietary assumption is made, one can state that the biological component of equation (A1) is derived from an omnivorous diet. Combining equation (A1) with equations (5) and (6) leads to

$$(\text{Sr/Ba})_{\text{Dg}}^{\text{Hsn}} = \left[X^{\text{Vg}} (\text{Sr/Ba})_{\text{Cmp}}^{\text{Vg}} + X^{\text{Mc}} (\text{Sr/Ba})_{\text{Bg}}^{\text{Mc}} \right] (\text{OR}_{\text{Sr}} / \text{OR}_{\text{Ba}}), \quad (\text{A4})$$

where X^{Vg} and X^{Mc} are the fractions of vegetable and meat, respectively. The Sr/Ba value of vegetable, $(\text{Sr/Ba})_{\text{Cmp}}^{\text{Vg}}$, and meat, $(\text{Sr/Ba})_{\text{Bg}}^{\text{Mc}}$, is obtained through a compilation (Cmp)

of extant vegetables (Table 2) and the measurement of the associated fauna, respectively. However, the diagenesis also affects the Sr/Ba value of the fauna and must be taken into account. Using equations (A1) and (A3), the biological Sr/Ba component of the meat, $(\text{Sr/Ba})_{\text{Bg}}^{\text{Mc}}$, is equal to

$$(\text{Sr/Ba})_{\text{Bg}}^{\text{Mc}} = \frac{1}{X_{\text{Bg}}} (\text{Sr/Ba})_{\text{Mes}}^{\text{Mc}} - \frac{X_{\text{Dg}}}{X_{\text{Bg}}} K_{\text{Sr/Ba}}^{\text{a-w}} (\text{Sr/Ba})_{\text{w}}, \quad (\text{A5})$$

where $(\text{Sr/Ba})_{\text{Mes}}^{\text{Mc}}$ is the measured value of the meat.

Introducing equation (A5) into equation (A4), and the resulting equation into equations (A1) and (A3), leads to the final relationship:

$$\begin{aligned} (\text{Sr/Ba})_{\text{Hsn}} = & \left[X^{\text{Vg}} X_{\text{Bg}} (\text{Sr/Ba})_{\text{Cmp}}^{\text{Vg}} + X^{\text{Mc}} (\text{Sr/Ba})_{\text{Mes}}^{\text{Mc}} \right. \\ & \left. - X^{\text{Mc}} X_{\text{Dg}} K_{\text{Sr/Ba}}^{\text{a-w}} (\text{Sr/Ba})_{\text{w}} \right] (\text{OR}_{\text{Sr}} / \text{OR}_{\text{Ba}}) \\ & + X_{\text{Dg}} K_{\text{Sr/Ba}}^{\text{a-w}} (\text{Sr/Ba})_{\text{w}} \end{aligned} \quad (\text{A6})$$

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