Incorporating $\delta^{18}O$ values of past waters in the calibration of radiocarbon dating

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ABSTRACT

All scientists who study the late Quaternary are confronted by the problem of radiocarbon calibration. Since 1969, numerous high-frequency time series have been developed through the use of several stable isotopes (e.g., $\delta^{18}O$). Such time series are inefficient as dating methods because too many age possibilities are obtained for a given unique value. However, the Bayesian framework permits incorporation of these time series as a prior assumption on ages in order to improve calibrated ages obtained from a more precise absolute dating approach such as that using radiocarbon. The method is tested on data obtained from the Gigny cave (French Jura). Highly discontinuous posterior distributions are obtained with narrow highest posterior density regions. Thus, when the radiocarbon method alone gives large uncertainties, the incorporation of $\delta^{18}O$ values of meteoric waters largely diminishes these uncertainties.

Keywords: isotopes, oxygen, radiocarbon calibration, Bayesian statistic.

INTRODUCTION

Radiocarbon content of the biosphere varies with time depending on the geomagnetic field strength, solar fluctuations, and rearrangements in equilibrium between reservoirs. In order to obtain the true calendar date, radiocarbon dating needs to be calibrated in relation to a reference curve. Radiocarbon calibration is fundamental in natural sciences working on the late Quaternary. In these scientific areas, a precise temporal framework is required in order to estimate the precise time span of events, such as the onset or the duration of an extinction (Holdaway et al., 2002), or to derive an accurate depth-based age model which, for example, may permit a highly resolved paleoclimatic record to be obtained (Cannariato and Kennett, 1999; Kennett et al., 2000).

After an initial phase in which scientists applied simple calibration methods, Bayesian statistics are now widely used to calibrate radiocarbon dating by incorporating additional available information from stratigraphy or dendrochronology as prior assumptions following different models using Markov chain Monte Carlo (MCMC) simulation (Buck et al., 1996). In the Bayesian calibration method, improvements have been developed that include alternative prior models for stratigraphic information (Nicholls and Jones, 2001), the model-choice framework (Sahu, 2004), and the modeling of the calibration curve (Gomez Portugal Aguilar et al., 2002). Moreover, improvements in calibration data sets for glacial ages and their combination in the CALPAL2000 data set (Weninger et al., 2002) permit extending these latter methodologies to ages older than the Holocene. Thus, Quaternary time possesses a potential dating framework for highly resolved analysis.

Since Dansgaard et al. (1969), Quaternary time possesses some highly resolved records (proxies) of the terrestrial climate such as the Greenland Ice Sheet Project 2 (GISP2) ice core (Grootes et al., 1993); such records may have, at least, a regional importance. One can expect that isotopic fluctuations recorded in the GISP2 core will be recorded in other areas of the Northern Hemisphere. However, $\delta^{18}O$ of biogenic apatites from mammal teeth and bones coupled with a present-day model have been widely used as proxies of the $\delta^{18}O$ of meteoric waters since Longinelli's (1984) work. However, this isotope record proved to be an inefficient dating tool because of the flickering nature of Quaternary climate superimposed on glacial-interglacial cycles (Dansgaard et al., 1993; Grootes et al., 1993). In spite of this problem, time information is contained in measured oxygen isotope compositions when they are compared to the ice-core records, and the Bayesian framework permits one to envisage the utilization of this time information as prior knowledge in the calibration of radiocarbon dating.

RADIOCARBON CALIBRATION

Analysis is based on the Gigny sequence (Pleistocene, French Jura), which is a highly studied stack of karst deposits (for a synthesis, see Campy et al., 1989). Radiocarbon determinations were carried out on macrofauna for levels IV, VIII, and mixing of macrofauna and microfauna for level V (Evin, 1989; Table 1). The CALPAL2003 curve (Weninger et al., 2002) was used to calibrate these dating results. This spline curve interpolates the extended CALPAL2000 data set and provides standard error for the uncertainties on data and the precision of the spline curve (Weninger et al., 2002). This calibration curve $\mu(\theta)$ relates a ^{14}C measurement to a calibrated age θ according to the probabilistic model (Buck et al., 1996):

$$^{14}C|\theta \sim \mathcal{N}[\mu(\theta), \sigma_\theta^2], \quad (1)$$

where the ^{14}C dating is conditioned (the vertical bar) only on the un-

TABLE 1. RADIOCARBON DETERMINATIONS AND OXYGEN ISOTOPE COMPOSITIONS FROM THE GIGNY CAVE (FRENCH JURA)

Level	^{14}C*	σ	$\delta^{18}O_{(PO_4)}$† (‰)	$\delta^{18}O_{mw}$§ (‰)	σ	Reference in Fig. 1
IV	12,370	460	15.3	−10.0	0.60	A
	13,620	480				A′
V	22,430	500	14.6	−11.2	0.65	B
			15.5	−9.6	0.59	B′
VIII	28,500	1400	15.1	−10.3	0.62	C
	29,500	1400				C′

*Radiocarbon determinations from Evin (1989).
†Oxygen isotope compositions of biogenic apatite from arvicoline teeth.
§Estimations of the oxygen isotope compositions of meteoric waters.

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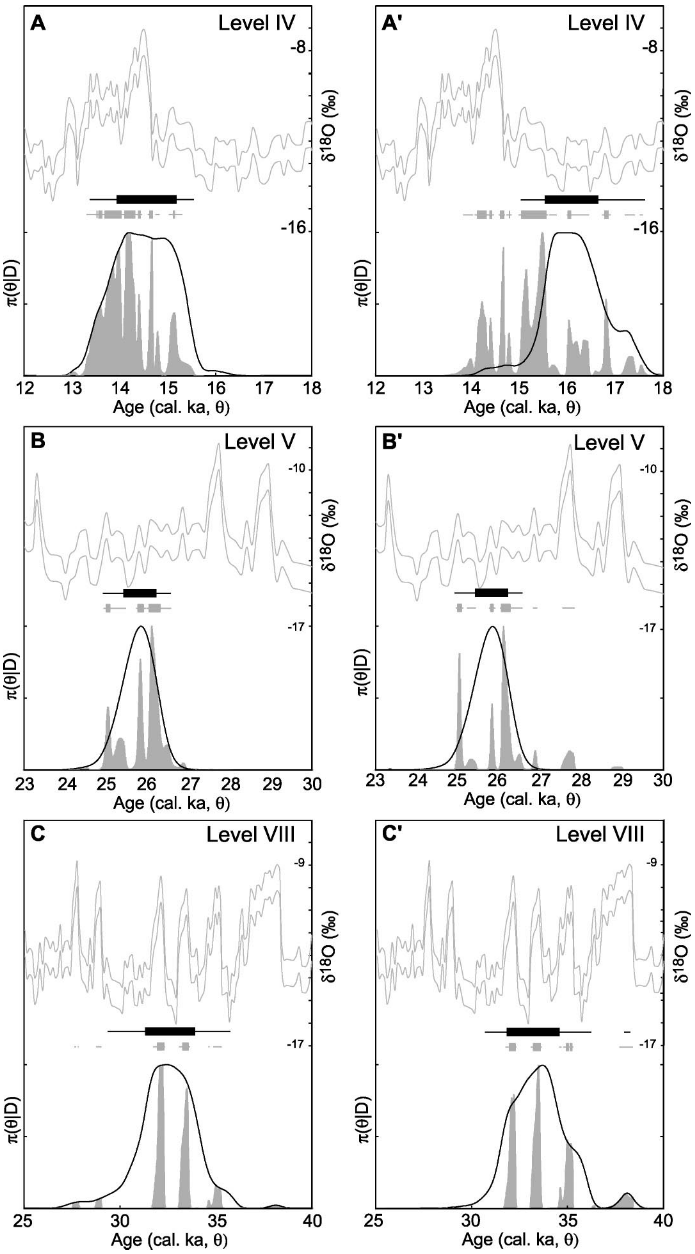

Figure 1. Posterior density of calibrated (cal.) dates of three levels of Gigny cave: $\pi(\theta|D)$, where θ corresponds to calibrated age and D corresponds to either ^{14}C alone, or ^{14}C and $\delta^{18}O$ determinations. Black line corresponds to posterior density given ^{14}C alone. Gray bars correspond to posterior density given both ^{14}C and $\delta^{18}O$ measurements. Black and gray boxes correspond to 1σ (68.2%) and 2σ (95.4%) Highest posterior densities of calibrated age following data set used (either ^{14}C alone or ^{14}C and $\delta^{18}O$). Gray curves correspond to $\pm 1\sigma$ confidence level of $\delta^{18}O$ record (Greenland Ice Sheet Project 2) converted into local values (46.45°N, 5.45°W, and 485 m).

known calibrated age θ, because σ_θ^2 is known and corresponds to the Gaussian propagation of errors of the ^{14}C dating and the precision of the calibration curve:

$$\sigma_\theta^2 = \sigma_{^{14}C}^2 + \sigma_{\mu(\theta)}^2. \tag{2}$$

In the Bayesian calibration, the aim is to obtain, conditional on the observed ^{14}C dates, the distribution of the calibrated age θ (see e.g., Buck et al., 1996; in the context of the radiocarbon calibration or, for more statistical books, Pawitan, 2001; Tanner, 1993). This conditional density function $\pi(\theta|^{14}C)$ of the calibrated age θ given the observed ^{14}C dates is called the posterior density. Posterior density describes updating knowledge about θ given the data and is obtained according to the Bayes theorem (Tanner, 1993):

TABLE 2. HIGHEST POSTERIOR DENSITIES (1σ, 68.2%) OF THE CALIBRATED AGE FROM RADIOCARBON ALONE OR FROM COMBINATION OF RADIOCARBON AND OXYGEN ISOTOPES

Level	Highest posterior densities (HPDs) (yr B.P.)	Method
IV-A	13,940–15,170	^{14}C
	13,560–13,620 13,690–14,020 14,100–14,310 14,390–14,420 14,620–14,680 15,120–15,140	^{14}C, $\delta^{18}O$
IV-A′	15,540–16,640	^{14}C
	14,130–14,310 14,390–14,420 14,610–14,680 15,050–15,560 16,020–16,070 16,790–16,840	^{14}C, $\delta^{18}O$
V-B	25,430–26,210	^{14}C
	25,000–25,090 25,770–25,900 26,040–26,310	^{14}C, $\delta^{18}O$
V-B′	25,430–26,210	^{14}C
	24,990–25,090 25,800–25,860 26,060–26,270	^{14}C, δ^{18}
VIII-C	31,330–33,880	^{14}C
	31,960–32,280 33,280–33,560	^{14}C, δ^{18}
VIII-C′	31,850–34,560	^{14}C
	31,980–32,280 33,320–33,580 34,960–35,050 35,150–35,230	^{14}C, δ^{18}

$$\pi(\theta|^{14}C) = c\, p(^{14}C|\theta)\pi(\theta), \tag{3}$$

where c is the inverse of the marginal density of the data ^{14}C and is a constant of normalization:

$$c^{-1} = m(^{14}C) = \int_\Theta p(^{14}C|\theta)\pi(\theta)\, d\theta. \tag{4}$$

Thus, the posterior density relates especially to $p(^{14}C|\theta)$ and $\pi(\theta)$. The first, $p(^{14}C|\theta)$, is the probability density function of the observed ^{14}C dating conditional on θ, and can be understood as the likelihood $\mathcal{L}(\theta;\, ^{14}C)$ of the calibrated age θ based on the ^{14}C dating because the likelihood is any function proportional to this probability density (Tanner, 1993):

$$\mathcal{L}(\theta;\, ^{14}C) = f(^{14}C)\, p(^{14}C|\theta) \propto p(^{14}C|\theta). \tag{5}$$

The second, $\pi(\theta)$, is the prior distribution of θ and corresponds to the knowledge about θ without knowledge of the ^{14}C dating, for example, the stratigraphic constraints. Thus, the posterior density $\pi(\theta|^{14}C)$ corresponds to the update of the initial knowledge on θ and is proportional to the likelihood times the prior distribution:

$$\pi(\theta|^{14}C) \propto \mathcal{L}(\theta;\, ^{14}C)\pi(\theta). \tag{6}$$

Thus, in the case of uniform prior and without any stratigraphic constraint, the posterior distribution of the calibrated age θ is proportional to the likelihood.

The three dating levels of Gigny show relatively flat posterior densities for their calibrated age based on the radiocarbon measurements (Fig. 1; Table 2). Their highest posterior density (HPD) incorporates several stades and interstades of glacial chronology. For example, the first radiocarbon determination of level IV (Fig. 1A) gives an expected depositional period of the level between the Oldest Dryas and the Allerød interstade. Moreover, the incorporation of stratigraphic information using MCMC did not reduce these uncertainties in this case because large hiatuses are present between successive dating levels (e.g., a hiatus of ~10 k.y. between the deposition of V and IV).

OXYGEN ISOTOPE COMPOSITIONS OF PAST WATERS

On the basis of the present-day equation, the $\delta^{18}O$ values of past meteoric waters can be estimated from the $\delta^{18}O$ values of biogenic apatites (Longinelli, 1984). The teeth of arvicoline rodents, analyzed here, are stratigraphically related to the ^{14}C dating. Basically, some teeth are mixed and analyzed using wet chemistry and mass spectrometry (Navarro et al., 2004). This wet chemistry permits isolation of the PO_4 component of apatite, which is more stable facing diagenetic alteration than the CO_3 component (e.g., Iacumin et al., 1996; see also Zazzo et al., 2004). A fractionation equation can be established between the $\delta^{18}O$ of ingested waters, approximated by the values of meteoric waters from the Global Network for Isotopes in Precipitation (GNIP) database (International Atomic Energy Agency–World Meteorological Organization [IAEA-WMO], 2001), and the $\delta^{18}O$ of phosphate from present-day arvicoline rodents' teeth. Navarro et al. (2004) estimated such an equation and estimated the $\delta^{18}O$ values of past meteoric waters ($\delta^{18}O_{mw}$) for teeth of the Gigny sequence (Table 1). Derivation of the standard errors is based on parametric bootstrap simulation (Efron and Tibshirani, 1993) estimating error on regression parameters based on the analytical error of $\delta^{18}O$ of phosphate and present-day waters.

The GISP2 record (Grootes et al., 1993) can be viewed as a time reference curve of $\delta^{18}O$ of past meteoric waters in the Northern Hemisphere. However, in order to compare these high-latitude values with the estimation in the Gigny area, GISP2 can be translated into local values. The local value (−8.2‰) is obtained from the Oxygen Isotopes in Precipitation Calculator (http://es.ucsc.edu/~gbowen/OIPC_Main.html), which is based on the updated version of the most recently published relationship (Bowen and Wilkinson, 2002; Bowen and Revenaugh, 2003) between oxygen isotope compositions and geographical variables as given in the GNIP database (IAEA-WMO, 2001). The standard error on this local value estimation is 0.38‰ (σ_{MODEL}). This precise estimate of the present day is related to the location of the Gigny cave in western Europe, a particularly well sampled area for $\delta^{18}O$ in precipitation. In poorly sampled areas, standard error can increase to 3.31‰ (Bowen and Revenaugh, 2003).

The GISP2 present-day value was estimated by the average value of the 1978–1987 annual average GISP2 data (Steig et al., 1994). The standard error of this present-day value is 0.55‰ ($\sigma_{GISP2\_PresentDay}$). The GISP2 time scale of Meese et al. (1997) based on the count of annual layers is used. The measurement error of the GISP2 data set is 0.14‰ ($\sigma_{GISP2\_measures}$), considering the standard deviation reported for unique determinations (Greenland Summit Ice Cores CD-ROM, 1997). In order to convert the GISP2 data set into a calibration curve with 1 yr resolution, the $\delta^{18}O$ values are interpolated by using a cubic function.

As for radiocarbon, the $\delta^{18}O_{mw}$ distribution at the calibrated age is presumed to be normal and the standard deviation σ_θ to be equal to the Gaussian propagation of errors of $\delta^{18}O_{mw}$ and the shifted reference record, which incorporates the error on the model, measurement error on GISP2, and error on the estimate of GISP2 present-day values:

$$\sigma_\theta^2 = \sigma_{^{18}O}^2 + \sigma_{MODEL}^2 + \sigma_{GISP2\_measures}^2 + \sigma_{GISP2\_PresentDay}^2. \quad (7)$$

COMBINING RADIOCARBON AND OXYGEN COMPOSITIONS

The posterior density of the calibrated age given the ^{14}C dating and the $\delta^{18}O_{mw}$ values (i.e., the updated knowledge or what is known about θ with knowledge of ^{14}C and $\delta^{18}O$ measurements) is proportional to the product of the likelihood of the calibrated age based on the ^{14}C dating and the $\delta^{18}O_{mw}$ value, times the prior distribution of the calibrated age θ (i.e., what is known about θ without knowledge of ^{14}C and $\delta^{18}O$ measurements):

$$\pi(\theta|^{14}C, \delta^{18}O) \propto \mathcal{L}(\theta; {}^{14}C, \delta^{18}O)\pi(\theta). \quad (8)$$

Because of the independence between $\delta^{18}O_{mw}$ and ^{14}C measurements, the likelihood of calibrated age based on the measurements of $\delta^{18}O_{mw}$ and ^{14}C corresponds to the product of the likelihoods (Buck et al., 1996; Pawitan, 2001):

$$\mathcal{L}(^{14}C, \delta^{18}O|\theta) = \mathcal{L}(\theta; {}^{14}C)\ \mathcal{L}(\theta; \delta^{18}O). \quad (9)$$

Thus, the posterior distribution of the calibrated age is proportional to the product of the two independent likelihoods times the prior distribution of the calibrated age θ:

$$\pi(\theta|^{14}C, \delta^{18}O) \propto \mathcal{L}(\theta; {}^{14}C)\ \mathcal{L}(\theta; \delta^{18}O)\pi(\theta). \quad (10)$$

In the case that the prior distribution of the calibrated age θ is uniform and without any stratigraphic constraint, the posterior density is proportional to the product of the likelihoods. Equation 5 can be also rewritten with the incorporation of posterior density given the $\delta^{18}O_{mw}$ value as the prior distribution of the calibrated age:

$$\pi(\theta|^{14}C, \delta^{18}O) \propto \mathcal{L}(\theta; {}^{14}C)\pi(\theta|\delta^{18}O). \quad (11)$$

Thus, the time information contained in the $\delta^{18}O_{mw}$ data, $\pi(\theta|\delta^{18}O)$, can be viewed as the prior density for the calibration of radiocarbon dating, i.e., the knowledge about θ without the ^{14}C dating.

DUAL CALIBRATION OF GIGNY SAMPLES

Because of the cyclic and flickering nature of the $\delta^{18}O$ record (Dansgaard et al., 1993; Grootes et al., 1993), the likelihood of the calibrated ages based on $\delta^{18}O$ values of meteoric waters shows imprecise regions of confidence. However, a uniform distribution was not obtained. Thus, $\delta^{18}O$ values provide knowledge about the depositional period according to the climate conditions. After incorporating this prior assumption, the posterior densities obtained become radically different from those obtained previously (Fig. 1). The new posterior distributions are more discontinuous and can be extremely multimodal, as a result of the climatic mode occurring during the depositional period. In some cases, posterior distributions given the two isotopic measurements can be shifted into the tail of the posterior distributions determined for ^{14}C alone (e.g., Figs. 1A, 1B), and the 1σ HPDs of the calibrated age considering both isotopic systems correspond to parts of the 2σ HPDs considering the ^{14}C alone.

CONCLUSIONS

The large uncertainties in the radiocarbon calibration can be greatly reduced by taking into account the time information contained in the oxygen isotope compositions of past meteoric waters. Thus, incorporating the $\delta^{18}O_{mw}$ significantly improves the calibration of ^{14}C determinations. However, in the case of large $\delta^{18}O$ uncertainties (e.g., poor sampling area of the GNIP database), the updating of the calibration would be weak because of the weak time information provided by $\pi(\theta|\delta^{18}O)$, which tends to be uniformly distributed. In such a case, the posterior density $\pi(\theta|^{14}C, \delta^{18}O)$ will be proportional to $\pi(\theta|^{14}C)$, and the updating will be null.

A strong initial assumption of the methodology is the use of the present-day relationship between Greenland and the study area. This constancy is relatively unlikely over a long time scale because of probable variations in atmospheric circulations between modern and glacial configurations (North Greenland Ice Core Project [NGRIP] Members, 2004). However, little is know about the variation of geographical gradients: normal (easily introduced in equation 7) or a mixture of switch states. The comparison of deep Greenland ice cores (NGRIP Members, 2004) seems to yield a gradient variation resulting from a mixture of two normal distributions. Such a case, conditional on age, would be incorporated using complex MCMC simulations. Moreover, a general circulation model incorporating isotopic tracers (e.g., Joussaume et al., 1984) would increase this knowledge about the range of variation and the glacial state of the gradient.

Another initial assumption is that the $\delta^{18}O$ from averaged teeth and ^{14}C dating of other material from the same layer represent the same point in time. This assumption could be a problem with high-precision ^{14}C dating. However, the use of techniques such as direct laser fluorination (Lindars et al., 2001), which permits measurements of $\delta^{18}O$ on a small amount of material, associated with larger animals (to avoid the problem of seasonality) could be allowed to make $\delta^{18}O$ and ^{14}C measurement on the same materials, and thus suppress this assumption.

Nevertheless, such a methodology, coupled or not with prior distribution related to stratigraphic constraints by using MCMC simulation, could be used to construct a more precise chronology of local Quaternary sequences with discontinuously laid-down deposits where a depth-based age model is unavailable (e.g., karst deposits) and to permit a better understanding of their depositional dynamics or any other dynamics of derived records (e.g., fossil record). More generally, all isotopes, or any other sources with a sufficiently highly resolved Quaternary record, can be used in the same way to improve the chronological knowledge.

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