BOMB-PULSE DATING OF HUMAN MATERIAL: MODELING THE INFLUENCE OF DIET

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ABSTRACT. The atmospheric testing of nuclear weapons during the 1950s and early 1960s produced large amounts of radiocarbon. This ^{14}C bomb pulse provides useful age information in numerous scientific fields, e.g. in geosciences and environmental sciences. Bomb-pulse dating can also be used to date human material (e.g. in forensics and medical science). Bomb-pulse dating relies on precise measurements of the declining ^{14}C concentration in atmospheric carbon dioxide collected at clean-air sites. However, local variations in the ^{14}C specific activity of air and foodstuffs occur, which are caused by natural processes as well as by various human activities. As ^{14}C enters the human body mainly through the diet, variations of ^{14}C concentration in foodstuffs need to be considered. The marine component of the diet is believed to be of particular importance due to the non-equilibrium in ^{14}C specific activity between the atmosphere and aquatic reservoirs during the bomb pulse. This article reviews the ^{14}C concentration in marine foodstuffs during the bomb-pulse era, and models how the marine component in one's diet can affect the precision of bomb-pulse dating of human material.

INTRODUCTION

Atmospheric testing of nuclear weapons in the late 1950s and early 1960s had a profound impact on the levels of radiocarbon in the atmosphere. The tests approximately doubled the ^{14}C concentration in atmospheric CO_2, and consequently in living matter, by the mid-1960s. In 1963, the majority of nuclear and many non-nuclear states signed the Limited Test Ban Treaty, pledging to refrain from testing nuclear weapons in the atmosphere, underwater, or in outer space. From the peak in 1963, the level of $^{14}CO_2$ has decreased exponentially with a mean life of about 16 yr (Ubelaker and Buchholz 2005), not due to radioactive decay but due to mixing with large marine and terrestrial carbon reservoirs. This "bomb pulse" of excess ^{14}C was recorded in all parts of the living biosphere (Currie 2004). The ^{14}C bomb pulse can be used to obtain age information in geosciences, forensics, and environmental sciences (Harkness and Walton 1969, 1972; Wild et al. 2000; Goodsite et al. 2001). The technique has also been used for retrospective cell dating in humans, in order to provide fundamental insight about the rate of formation of new cells in the human body (Druffel and Mok 1983; Spalding et al. 2005a,b, 2008; Lynnerup et al. 2008).

Bomb-pulse dating of terrestrially living materials relies on precise measurements of the declining ^{14}C concentration in atmospheric carbon dioxide. The ^{14}C activity in atmospheric CO_2 is mainly controlled by 4 major sources:

1. Natural ^{14}C produced by interactions of cosmic rays in the upper atmosphere;
2. Bomb ^{14}C, originally introduced into the stratosphere in the Northern Hemisphere but subsequently stored in vast majority in the oceans and to some extent in the terrestrial biosphere;
3. ^{14}C released by the nuclear industry (such as nuclear power plants, reprocessing facilities for spent nuclear fuel, and ^{14}C-using laboratories);
4. Fossil-fuel CO_2, acting as a ^{14}C-free dilutant—also referred to as the *Suess effect*.

A number of post-bomb atmospheric ^{14}C records are available taking into account that post-bomb atmospheric ^{14}C data varies somewhat regionally (Reimer et al. 2004). These data are collected at clean-air sites, far from the whereabouts of most humans, thus minimizing the effect on the data sets from local releases of ^{14}C from nuclear installations and from dilution of fossil-fuel CO_2. However,

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as pointed out by Reimer et al. (2004), there are also local variations, and hence a local atmospheric ^{14}C data set would be ideal for calibration of a post-bomb ^{14}C measurement of e.g. terrestrial plants. However, when dating human material, other factors also need to be considered.

The transfer of bomb ^{14}C into the human body and the resulting ^{14}C specific activity in human tissues mainly depend on the following 3 factors, according to Nydal et al. (1971):

1. The time between the photosynthesis in vegetal food and its consumption;
2. The diet, particularly its vegetal food content;
3. The residence time of the carbon in different parts of human tissue.

However, most human diets consist not only of terrestrially grown vegetal food and meat products (the latter generally have a longer delay time of ^{14}C from the atmosphere to human consumption than that of vegetal foods, according to Broecker et al. 1959). Fish and seafood are usually also part of the diet, and the importance of this marine source can vary significantly regionally as well as individually.

During the bomb pulse, the ^{14}C specific activity in marine organisms differed significantly from that of terrestrial foods due to the delay time for bomb ^{14}C to enter the oceans and due to the non-equilibrium in ^{14}C specific activity between the atmosphere and aquatic reservoirs. Post-bomb marine data sets exist and have been derived from coral, coralline sponges, fish otoliths, and shell chronologies (Reimer et al. 2004). These data sets show much higher regional variation than the atmospheric ones. This fact, in combination with interindividual amounts of dietary components of marine origin, may lead to variations in ^{14}C specific activity for different individuals. This paper addresses how the marine component of the diet can affect the precision of bomb-pulse dating of human material, like blood and hair samples, by estimating the mean $\Delta^{14}C$ values for local diets at 5 different regions around the globe.

MATERIALS AND METHODS

Post-bomb $\Delta^{14}C$ data for marine organisms ($\Delta^{14}C_{marine}$) were collected from the literature. Five different sites distributed worldwide were used to demonstrate regional variations of ^{14}C in marine organisms: *Barents Sea* (Kalish et al. 2001) (71°N, 40°E, part of the Arctic Ocean, north of Norway); *Georges Bank* (Weidman and Jones 1993) (41°N, east of Massachusetts state, USA); *New Zealand* (Kalish 1993) (39°S, 179°E, Wellington and east coast); *south and southeast Australia* (Kalish 1995, Kalish et al. 1997) (35°S, 139°E); and *United Kingdom* (Cook et al. 1998; Campana et al. 2006) (NE Atlantic, NW England coast). Parts of the UK data are from the vicinity of the nuclear fuel reprocessing plant in Sellafield, to demonstrate the potential influence on the ^{14}C levels in marine diets originating from the nuclear power industry wastes into the sea.

In the case of the Barents Sea data, Kalish et al. (2001) used Arcto-Norwegian cod otoliths to create the post-bomb time series (1950–1990) for the marine environment. In the Georges Bank case, Weidman and Jones (1993) used data derived from growth bands of a 54-yr-old mollusk specimen (Bilvania, *Arctica islandica*) shell, collected live from Georges Bank to produce the local marine $\Delta^{14}C$ data set. Otolith data from squirefish (*Pagrus auratus*) were collected by Kalish (1993) off the east coast of North Island, New Zealand, for the corresponding calculation of the $\Delta^{14}C$ values. For the S and SE Australia study case, fish and otolith ^{14}C data obtained from blue grenadier (*Macruronus novaezelandiae*) as well as from redfish (*Centroberyx affinis*) and used for the local marine $\Delta^{14}C$ data set (Kalish 1995; Kalish et al. 1997). The marine $\Delta^{14}C$ data for the UK west coast originate from dogfish spines (*Squalus acanthias*) sampled in the NE Atlantic (Campana et al. 2006).

In the particular case of Sellafield (UK), ^{14}C activity values of samples from fish and other sea creatures (mussels, flatfish, roundfish, and crustacea), collected by Cook et al. (1998), were used. $\Delta^{14}C$ was calculated following the formula of Stuiver and Polach (1977) and by taking under consideration that A_{SN} is the normalized sample activity and A_{ABS}, which is the absolute international standard activity, is 13.56 dpm/g C (Karlén et al. 1964), equaling 226 Bq/kg C.

$$\Delta^{14}C = \left[\frac{A_{SN}}{A_{ABS}} - 1\right] \cdot 1000\,‰ \quad (1)$$

The atmospheric data $\Delta^{14}C_{atm}$ used for each region were obtained from the Hua and Barbetti (2004) data set. Particularly for the Barents Sea and the UK cases, data for the Northern Hemisphere (NH) zone 1 were used for years before 1970, while for the years after 1970 average values from the locations above 40°N were used. In the Georges Bank case, New York area data were used for years before 1970 while for years after 1970 average values from locations below 40°N were used. For New Zealand, data from Wellington (41°S, 174°E) were used for 1955–59, 1973, 1978–79, 1984, 1987–88 and Southern Hemisphere (SH) averages were used for 1993–97. For S/SE Australia, data from Armidale (30°S 152°E) were used except for period 1978–97 where SH averages were used.

The local human diet's ^{14}C concentration for the different regions was estimated as:

$$\Delta^{14}C_{diet} = X_{\text{marine carbon}} \cdot \Delta^{14}C_{marine} + (1 - X_{\text{marine carbon}}) \cdot \Delta^{14}C_{atm} \quad (2)$$

where $X_{\text{marine carbon}}$ is the fraction of local protein diet derived from seafood, according to the FAO (2008) and $\Delta^{14}C_{marine}$ is the local post-bomb marine data set. The dietary fraction $(1–X_{\text{marine carbon}})$ represents the rest of the protein in the human diet, originating from meat and vegetables, etc. For this fraction, the local atmospheric data set ($\Delta^{14}C_{atm}$) was used, assuming that foodstuffs of terrestrial origin are in equilibrium with the atmosphere of the region. Lag times of ^{14}C entering the terrestrial proteins have not been considered in the calculations.

RESULTS AND DISCUSSION

Table 1 shows the average daily intake of marine foods according to the FAO at the various sites. Figure 1a shows marine post-bomb data ($\Delta^{14}C_{marine}$), atmospheric data ($\Delta^{14}C_{atm}$), and the dietary values ($\Delta^{14}C_{diet}$) for the Barents Sea, calculated using the equations above and the values in Table 1. Since the average marine protein dietary fraction is relatively low (8%), the diet ^{14}C concentrations are rather close to the local atmospheric ^{14}C concentration. Figure 1b shows an estimation of the difference in age between the diet curve and the atmospheric curve, i.e. the approximate error in the age determination that the marine food component can introduce compared to using the atmospheric calibration curve. Some 8% of fish from the Barents Sea in the human diet can alter the date determination in a range from about –2.4 to 1.4 yr maximum compared with the age, coming from the respective atmospheric data. However, an individual consuming twice the average (16% of fish) would give an approximate age alteration range from –2.7 to 2.7 yr.

Table 1 Fish food consumption in the different study cases (FAO).

Location	Fraction of dietary protein fish and seafood consumption, $X_{\text{marine carbon}}$ (FAO 2008)
Barents Sea (Russia, Finland average)	0.08 (time period: 1990–92, 2003–05)
Georges Bank (USA)	0.04 (time period: 1990–97)
New Zealand	0.05 (time period: 1990–92)
S-SE Australia	0.04 (time period: 1990–97)
United Kingdom	0.05 (time period: 1990–2005)

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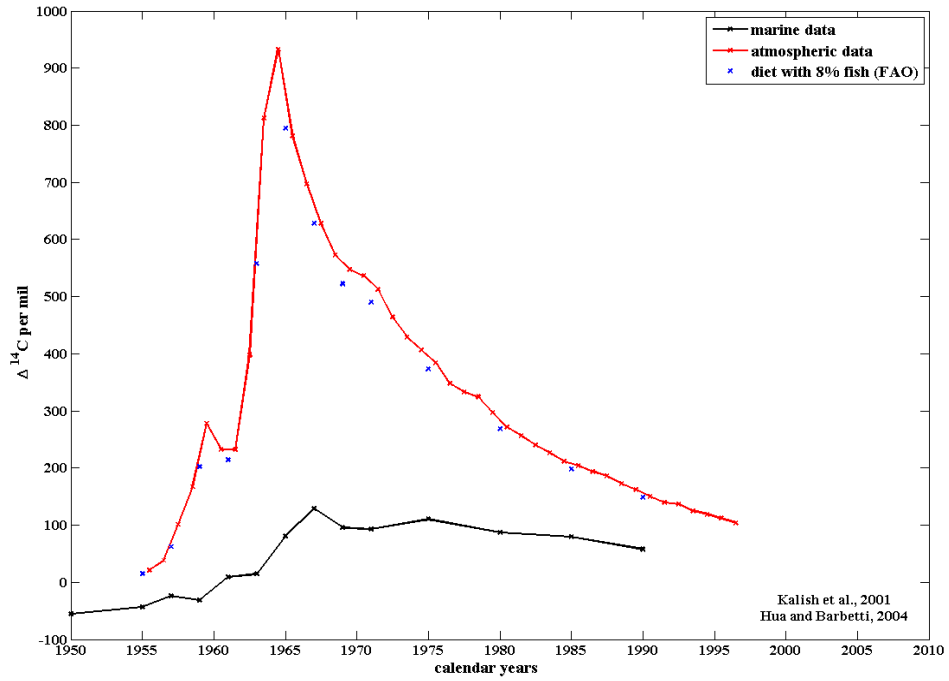

Figure 1a Atmospheric, marine, and dietary curve of ^{14}C data for the Barents Sea

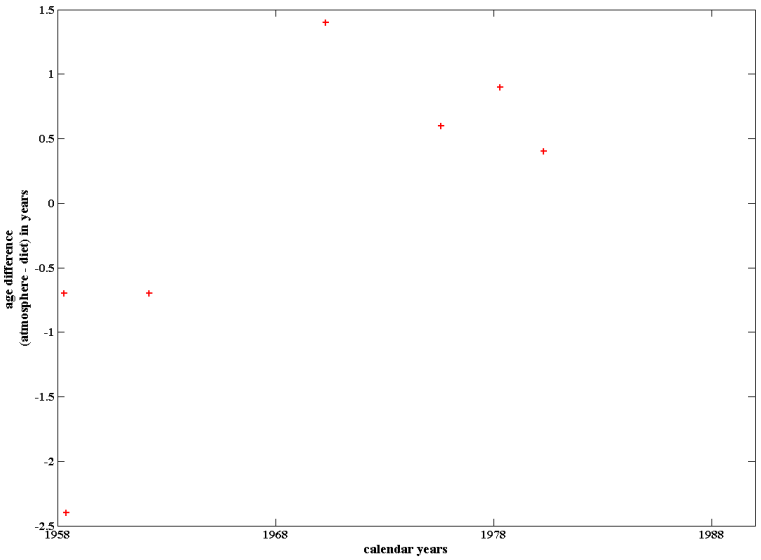

Figure 1b Age alteration between atmosphere and diet in the Barents Sea region, with 8% fish in the diet.

Figures 2–4 show the results of the dietary modeling for Georges Bank, New Zealand, and Australia. These have a lower average consumption of fish than in the Barents Sea, and the age alteration range arising in these cases is given in the Table 2, which also includes the age alteration that results from a diet with fish from Sellafield.

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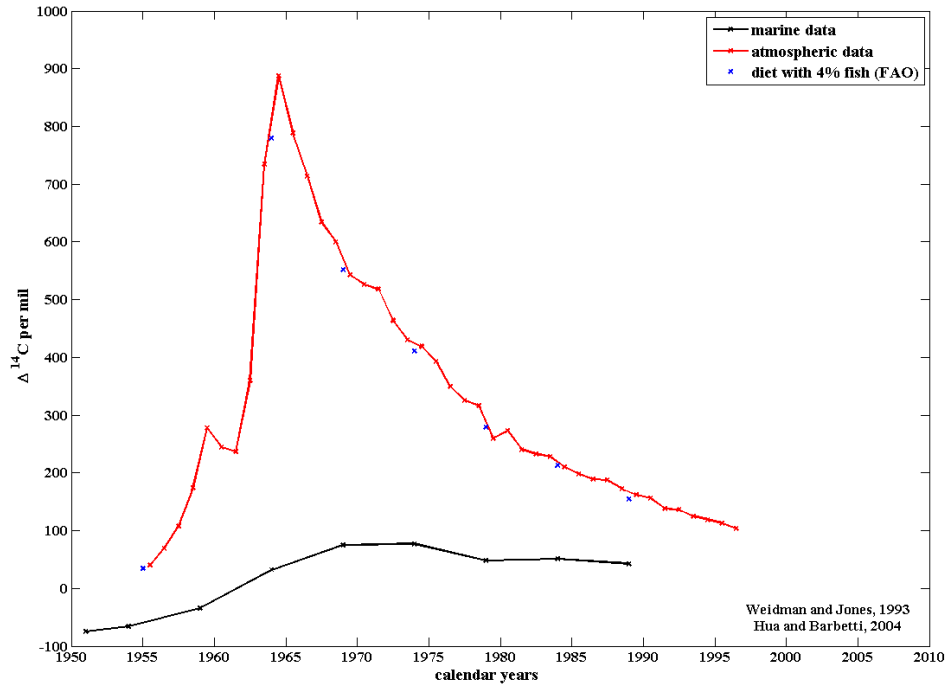

Figure 2 Atmospheric, marine and dietary curve of ^{14}C data for Georges Bank

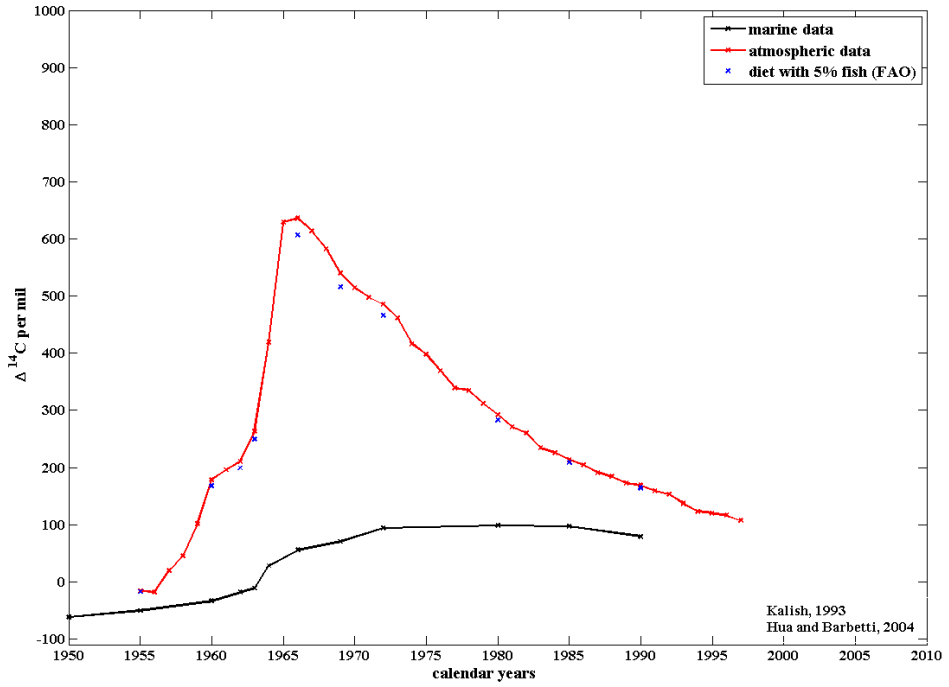

Figure 3 Atmospheric, marine and dietary curve of ^{14}C data for New Zealand

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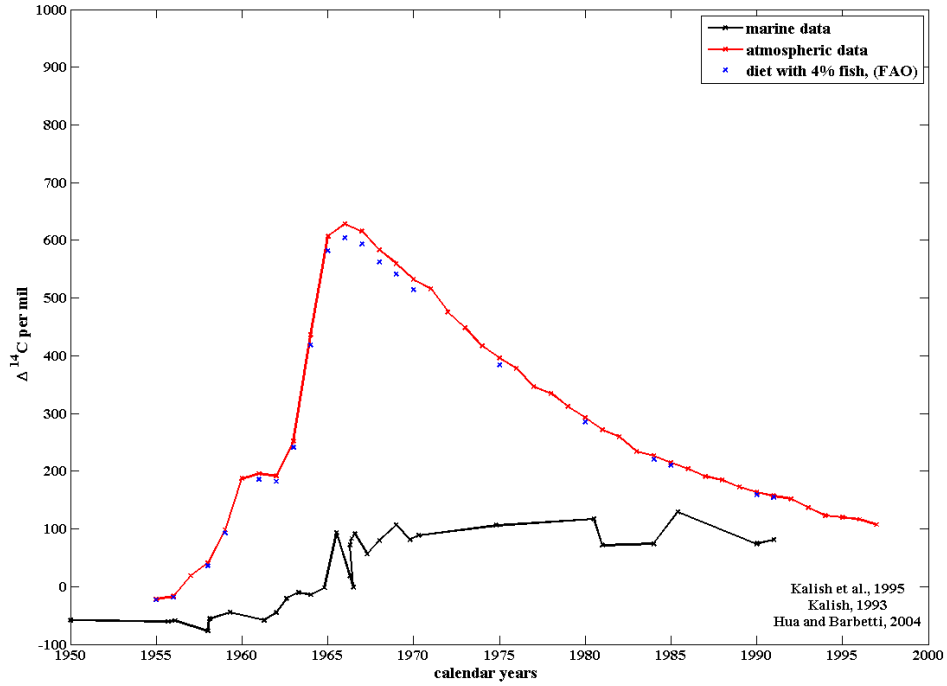

Figure 4 Atmospheric, marine, and dietary curve of ^{14}C data for S and SE Australia

Table 2 Age alteration range (atmospheric age–diet age) in years.

Region	Age alteration range
Barents Sea	–2.4 – 1.4
USA (Georges Bank)	–0.1 – 1
New Zealand	–1.1 – 0.9
S/SE Australia	–2.2 – 1.1
UK (Sellafield), 1995	–26.3

Figure 5 shows the outcome of modeling the data from the UK. $\Delta^{14}C$ values corresponding to a protein diet with 5% fish and seafoods from Sellafield in 1995 ($\Delta^{14}C_{diet}$ = 566‰) were calculated using the mean ^{14}C activity of all marine specimens collected at the Sellafield site (2305 Bq/kg C, corresponding to $\Delta^{14}C_{marine}$ = 9200‰), as reported by Cook et al. (1998) (mussels = 3305 Bq/kg C, flatfish = 2694 Bq/kg C, roundfish = 2543 Bq/kg C, and crustacean = 679 Bq/kg C). The significantly enhanced ^{14}C concentrations in the diet would lead to alteration of the year determination of 26.3 yr compared to the atmospheric data (Table 2).

Based on the modeling results of the diets, it can be concluded that the ^{14}C concentration in one's diet can indeed affect the accuracy of bomb-pulse dating on human samples. In our calculations, we have not taken into account that some of the food might be stored for prolonged times prior to consumption, which can lead to further age prediction alterations compared to the predicted age according to the atmospheric ^{14}C content. To further evaluate the precision of bomb-pulse dating of human material, we plan to retrieve and analyze human blood samples, collected during the bomb pulse and stored at a biobank.

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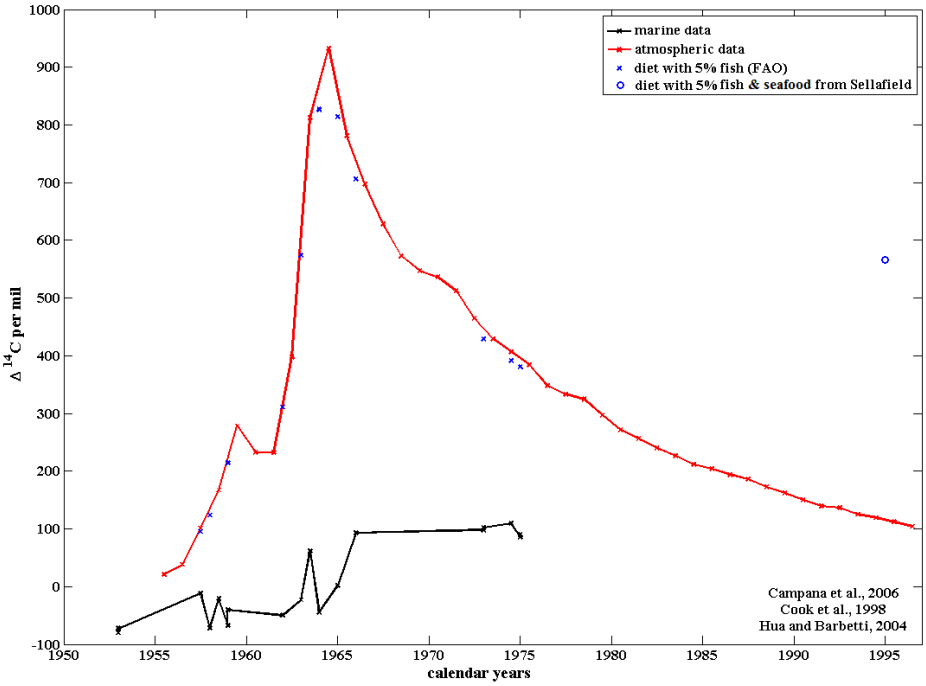

Figure 5 Atmospheric, marine, and dietary curve of ^{14}C data for the UK

CONCLUSIONS

This study shows that a human diet containing a normal fraction of food of marine origin has the potential of influencing bomb-pulse dating by up to a few years, compared to the atmospheric data. For example, 8% of fish from the Barents Sea in the human diet can alter the age determination by around −2.4 yr maximum. For an individual consuming twice the average of fish (16%), the age alteration ranges from about −2.7 to 2.7 yr. The dietary modeling study also shows that the considerably enhanced ^{14}C concentrations, which can be found in fish near nuclear installations (such as the Sellafield nuclear fuel reprocessing plant), can lead to an alteration of age determination of several tens of years compared to the atmospheric data. Thus, for high-precision bomb-pulse dating of human material, it is necessary to consider possible variations of ^{14}C-specific activities in the diet.

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