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# NEW MARINE RESERVOIR CORRECTION VALUES ( $\Delta R$ ) APPLICABLE TO DATES ON NEOLITHIC SHELLS FROM THE SOUTH COAST OF KOREA

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**ABSTRACT.** Shells from Neolithic shell midden sites have been routinely dated in Korea, but they have not been calibrated based on the correction values ( $\Delta R$ ) for the marine reservoir effect (MRE). A lack of proper calibration has left dates on shells incomparable to those on terrestrial samples, and thus unusable in building the chronological sequence of shell middens. Here, we report the two new  $\Delta R$  values of a pre-bomb (pre-1950) blue mussel from the south coast. We applied the two new and the two previously reported  $\Delta R$  values to the three dates on marine shells from the Bibongri shell midden in southeastern Korea. Our  $\Delta R$  adjusted calibration and the comparison to dates on charcoal and bone remains clarify an ambiguity in the stratigraphy and the Early Neolithic chronology at Bibongri. Our contribution is to provide the  $\Delta R$  values that can be further applied to other Neolithic shell middens along the south coast.

**KEYWORDS:** Korea, marine reservoir effect, Neolithic shell midden,  $\Delta R$ .

### INTRODUCTION

Radiocarbon (<sup>14</sup>C) datasets have been used in archaeology not only to build regional chronologies but also to study population dynamics, settlement changes, and the correlations between changes in environments and culture (e.g., Anderson et al. 2011; Crema et al. 2016). A large number of <sup>14</sup>C dates are readily available in site reports, leading to an increase of radiocarbon studies in Korean archaeology since the 2010s. Popular topics include subsistence changes (Bae et al. 2013), temporal variabilities of hunter-gatherer sedentism (Ahn et al. 2015), settlement changes during state formations (Park et al. 2017), and population dynamics (Oh et al. 2017).

In Korea, a considerable number of shell dates is associated with the shell middens dating to the Neolithic period (ca. 8000-3500 cal BP). Over 600 shell middens have been documented in Korea, a large percentage (46%) belonging to the Neolithic period (Ha 2010), and some of well-stratified shell middens have provided key data on establishing the regional chronology. Although marine shells from these Neolithic shell middens are frequently dated materials, counting 120 or more, their <sup>14</sup>C dates are often underutilized or used without proper calibration accounting for the marine reservoir effect (MRE). The MRE contributes to the disparity between the <sup>14</sup>C ages obtained from terrestrial and marine samples. Several factors contribute to this gap, especially upwelling, which occurs when an "older" carbon upwells from the deep ocean and is taken in by marine organisms (Soares and Martins 2010). Recently, Heaton et al. (2020) presented Marine20, an update to the internationally agreed marine radiocarbon age calibration curve. Marine20 models the global influence of the MRE by simulating the ocean/atmosphere/biosphere box-model of the global carbon cycle while also integrating the ice core data on the observed changes in  $CO_2$ . The regional MRE, however, can deviate from the global model due to the variations in the fluvial influx and ocean upwelling (Smittenberg et al. 2004; Cook et al. 2015). By offsetting these



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impacts, the regional correction value ( $\Delta R$ ) can adjust the global pattern of the MRE and thus be used to calibrate the radiocarbon age of marine organisms correctly.

Utilizing marine shell dates in Korea has been difficult in part due to the lack of  $\Delta R$  values. Although several values are available from the surrounding regions (Kuzmin et al. 2001; Southon et al. 2002; Shishikura et al. 2007; Yoneda et al. 2007; Hirabayashi et al. 2017), only two  $\Delta R$  values are available in Korea (Kong and Lee 2005) (Table 1). The provenience of shells used in Kong and Lee's (2005) study, however, is only known as the south coast without further details. As  $\Delta R$  values can be highly variable in coastal regions (Kuzmin et al. 2001; Butler et al. 2009; Thornalley et al. 2011; Napolitano et al. 2019), previously published values from surrounding regions may not be applicable to the Korean Peninsula. Without additional applicable  $\Delta R$  values, marine shell dates have a limited value contributing to the Neolithic chronology. Therefore, the chronology is often built exclusively upon pottery typology and <sup>14</sup>C dates on charcoal. The Bibongri shell midden site (35°24'38"N, 128°38'44"E) is one of such cases in southeastern Korea (Figure 1). As a multi-component site (7800-2800 cal BP), Bibongri provides a wealth of information on subsistence and material culture throughout the Neolithic periods with well-preserved organic remains (Supplementary Material 1) (Gimhae National Museum 2008, 2012). By adding  $\Delta R$  values and <sup>14</sup>C dates, several studies in eastern Eurasia reassessed the regional settlement patterns and the time depth of the sites, including the Neolithic Boisman 2 site in Primorye (Jull et al. 1994; Kuzmin et al. 1994, 2002).

Our study reports two new  $\Delta R$  values. We calculate the weighted mean based on these new  $\Delta R$  values and those published in Kong and Lee (2005) and apply to the calibration of archaeological shells from Bibiongri. Among several cultural layers identified at Bibongri, Shell Layer 1 is the thickest strata, revealing abundant artifacts and organic remains. By comparing the calibrated marine shell dates to those on charcoal and bone fragments from Shell Layer 1, we aim to address the time depth of this cultural layer.

# METHODS

#### Specimen Used

We obtained one pre-bomb blue mussel (*Mytilus edulis*) specimen, live-collected at the Busan Bay of South Gyeongnam Province, South Korea on May 29, 1938 (Table 2; Figure 1). The bivalve specimen was preserved in an 80% aqueous solution of ethanol in an air-tight container at the Fisheries Science Museum located in Busan, South Korea.

Blue mussels inhabit rocky, low, intertidal shores, sheltered harbors, open coasts with hard substrates, or any place with dense masses suitable for their body attachment (Al-Dabbas et al. 1984; Hong 2006; Lee et al. 2006). This marine mollusk is widely used for measuring local pollution levels because of its immobile living habit that results in an exclusive intake of nutrients from the local seawater. Its lifespan varies from three to 24 years, depending on the habitat environment (Seed 1969; Thiesen 1973). Most specimens curated at the Fisheries Science Museum between 1905 to 1943 were collected from the harbor of Busan, including the blue mussel specimen used in this study.

Sample / lab code	R(t) (1σ) cited*	$\Delta R (1\sigma)$ cited**	R(t) (1σ) by Marine20***	$\Delta R$ (1 $\sigma$ ) by Marine20***	Species	Collected vr. (t)	Collected location	Dist. from Bibongri (km)
NZA19427 <sup>a</sup>	127 ± 35	$-160 \pm 35$	130 ± 37	$-296 \pm 35$	Thais clavigera	1942	Southeastern	90
NZA19426 <sup>a</sup>	$170 \pm 45$	$-117 \pm 45$	$173 \pm 46$	$-253 \pm 45$	Venerupis amvgdala	1942	Southwestern coast. Korea	216
CAMS8814 <sup>b</sup>	$228 \pm 60$	$-96 \pm 60$	215 ± 61	$-234 \pm 60$	Gastropoda sp.	1927	Jiaozhou Bay, Oingdao	753
YAUT-021313 <sup>c</sup>	231 ± 27	$-73 \pm 35$	219 ± 28	$-225 \pm 35$	Porites sp.	1929	SE coast of Kikai Island	806
YAUT-021321°	296 ± 25	$-52 \pm 33$	$278 \pm 26$	$-210 \pm 33$	Porites sp.	1911	SE coast of Kikai Island	806
YAUT-021619 <sup>c</sup>	267 ± 34	$-52 \pm 40$	259 ± 34	$-208 \pm 40$	Porites sp.	1920	SE coast of Kikai Island	806
YAUT-018734 <sup>c</sup>	228 ± 38	$-49 \pm 43$	229 ± 39	$-187 \pm 43$	Porites sp.	1945	SE coast of Kikai Island	806
YAUT-021307°	355 ± 25	$-20 \pm 34$	332 ± 26	$-180 \pm 34$	Porites sp.	1902	SE coast of Kikai Island	806
YAUT-021303°	321 ± 58	27 ± 62	320 ± 58	$-117 \pm 62$	Porites sp.	1938	SE coast of Kikai Island	806
TERRA-080905c17 <sup>d</sup>	379 ± 33	$-1 \pm 40$	355 ± 34	$-160 \pm 32$	Haliotis asinina	1901	Amami-Oshima, Kagoshima	823
TERRA-080905c16 <sup>d</sup>	401 ± 31	18 ± 38	376 ± 32	$-141 \pm 30$	Ostreidae sp.	1900	Amami-Oshima, Kagoshima	823
TERRA-080905c15d	474 ± 31	94 ± 38	450 ± 32	$-65 \pm 30$	Malleus malleus	1901	Amami-Oshima, Kagoshima	823

Table 1  $\Delta R$  values within 823 km proximity to the study area, calibrated based on Marine20.

\*Calculated by subtracting <sup>14</sup>C age of marine sample and atmospheric <sup>14</sup>C age using the calibration curve used in the publication.

\*\* $\Delta R$  values cited from original publications, based on Marine98 calibration (Stuiver et al. 1998); Marine04 (Hughen et al. 2004); and Marine13 (Reimer et al. 2013). \*\*\* $\Delta R$  values based on Marine20 (Heaton et al. 2020), cited from the 14CHRONO Marine20 Reservoir Database website (http://calib.org/marine; accessed 2021 Mar 16). Calculated by subtracting <sup>14</sup>C age of marine sample and atmospheric <sup>14</sup>C age in IntCal20 (Reimer et al. 2020).

\*\*\*\*Uncertainty value (1 $\sigma$ ) not presented in the original publication, thus calculated by the authors of this study. References cited: a) Kong and Lee 2005, b) Southon et al. 2002, c) Hirabayashi et al. 2017, d) Yoneda et al. 2007.



Figure 1 The provenience of pre-bomb shell sample and Bibongri archaeological site, and the estimated coastline of Paleo Bibong Bay, which is in a lighter shade than the river channel (modeled after the Gimhae National Museum 2012; 4).

AMS lab code	D-AMS-038966	D-AMS-038967			
<sup>14</sup> C yr BP (conventional age)	510 ± 22	533 ± 24			
Percent modern carbon (pMC, $1\sigma$ )	$93.85 \pm 0.26$	$93.58 \pm 0.28$			
δ <sup>13</sup> C (‰, VPDB)*	-0.24	-0.24			
$R(t) (1\sigma)^{**}$	$347 \pm 25$	$370 \pm 26$			
$\Delta \mathbf{R}$ (1 $\sigma$ )	$-93 \pm 22$	$-70 \pm 24$			
Weighted mean $\Delta R$ (1 $\sigma$ )	$-83 \pm 16$				
Family	Mytil	Mytilidae			
Scientific name	Mytilus edulis (	Mytilus edulis (Linnaeus 1758)			
Common name	Blue mussel; Jinjudamchi (Korean);				
	Murasakiigai (Japanese)				
Specimen registry no.	NFRDI-MS	NFRDI-MS-IS-0000078			
Collection date	May 29	May 29, 1938			
Collection location	Busan Bay, South G	Busan Bay, South Gyeongnam Province			

Table 2 Two <sup>14</sup>C dates from a pre-bomb shell specimen and their  $\Delta R$  values.

\*The  $\delta^{13}$ C values in this table were calculated using isotope ratio mass spectrometry (IRMS) at the Stable Isotope Laboratory, University of Oregon.

\*\*Calculated by subtracting <sup>14</sup>C age of marine sample and atmospheric <sup>14</sup>C age in IntCal20 (Reimer et al. 2020).

#### Shell Preparation

We cleansed the specimen through soaking and gently brushing with deionized water. The exterior organic layers (periostracum) were etched using 10% hydrochloric acid solution and air-dried for 36 hr to remove foreign substances and diagenetically altered carbonate. The shell was microscopically inspected for an intact terminal edge. Axial locations of the terminal growth margin were carefully sampled by hand with 0.05 mm carbide tip and drill bit at the University of Oregon Island and Coastal Archaeology Laboratory (Figure 2).



Figure 2 Pre-bomb shell specimen (*Mytilus edulis*, D-AMS-038966 and -038967) used in this study. Arrows show spots sampled for analysis.

Two sets of particles were sampled from the same specimen (NFRDI-MS-IS-0000078), constituting the two AMS submissions (D-AMS-038966 and D-AMS-038967) (Table 2). The radiocarbon date measurement was processed with the NEC Pelletron 500 kV AMS device at the DirectAMS (D-AMS) facility (see Zoppi 2010).

#### Calculation of $\Delta R$ and the Weighted Mean $\Delta R$

We followed the standard procedure of calculating  $\Delta R$ , used by Kong and Lee (2005) and other studies (Phelan 1999; Yoshida et al. 2010; Nakanishi et al. 2017; Panich et al. 2018). First, we selected a marine shell specimen from the pre-bomb (pre-1950) era to avoid the "bomb effect," which increased the global atmospheric <sup>14</sup>C levels (Nydal 1968; Levin et al. 1985).

$$R_{global}(t) = 14C Age_{ocean} - 14C Age_{atmosphere}$$
(1)

$$R_{marine \ sample}(t) = 14C \ Age_{marine \ sample} - 14C \ Age_{atmosphere}$$
(2)

$$R_{marine \ sample}(t) = R_{global}(t) + \Delta R \tag{3}$$

$$\Delta R = R_{marine \ sample}(t) - R_{global}(t) \tag{4}$$

where

R = Reservoir Age; t = calendar year

Equations (1)–(4) discuss the method of deriving  $\Delta R$  (Jull et al. 2013).  $\Delta R$  is the difference between the reservoir age of marine sample and that of the global ocean age model, the Marine20 calibration curve in this study (Heaton et al. 2020). We used the Calib website (QUB 2021a) for the calculation of  $\Delta R$ . Then we calculated a weighted mean to combine multiple  $\Delta Rs$  into a single value while placing a more weight on the value with a lower error range. The weighted mean is derived by using the equation on the Calib website (see Bevington 1969, QUB 2021b). We apply the weighted mean  $\Delta R$  to the archaeological shell samples from the Bibongri Site for the <sup>14</sup>C age calibration. The calibration was made with OxCal 4.3.2 (Bronk Ramsey 2009, 2017; Bronk Ramsey and Lee 2013), based on IntCal20 atmospheric curve and Marine20 marine curve (Heaton et al. 2020; Reimer et al. 2020).

#### **Bibongri Site**

Bounded by steep hills (400 m asl) to the north and west, Bibongri is located at the bottom of the foothills, overlooking an alluvial plain (3.8 m asl) along the Chengdo-cheon river, a tributary of the Nakdong River (Figure 3). Although Bibongri is located inland today, it sat on a widened bay called Paleo Bibong throughout the Neolithic period and thus Neolithic inhabitants faced an inner bay environment (Gimhae National Museum 2008; Williams et al. 2013). Bibongri's coastal landscape is well documented by several studies. Sea-level and sedimentological studies exhibit that human activities at Neolithic Bibongri were largely affected by seawater dynamics (Hwang 2008, 2012; Hwang et al. 2013). Over 90% of diatoms identified from Shell Layer 1 belong to marine species, including *Coscinodiscus* sp., *Nitzschia cocconieformis*, and *Nitzschia granulata* (Hwang 2008, 2012). Marine shell species, including pacific oyster (*Magallana gigas*) and blood cockle (*Tegillarca granosa*), are also abundant in Shell Layer 1 (Gimhae National Museum 2008; Kaneko 2008). After the site was abandoned, Bibongri transformed into a low-lying



Figure 3 (a) An aerial view of the Bibongri site, showing Shell Layer 1's surface, and (b) a schematic stratigraphic profile (Gimhae National Museum 2008: 14, 193). Stratigraphic layer numbers outside the brackets are copied from the Gimhae National Museum (2008), and numbers inside the brackets are copied from the Gimhae National Museum (2012).

wetland along with sea-level regression and inundation from the Cheongdo-cheon River. As a result, organic materials were well preserved in multiple layers of silty mud, shell, and alluvial sediments.

Bibongri's high pH level of six shell layers preserved faunal remains in excellent condition, including at least 35 vertebrate and molluscan taxa. Two common species of mollusks are blood cockle (*Tegillarca granosa*) and pacific oyster (*Magallana gigas*) (Kaneko 2008; Hwang et al. 2013). Dybowski's sika deer (*Cervus nippon hortulorum*) and Korean wild boar (*Sus scrofa coreanus*) are the most common mammalian taxa, similar to other Neolithic shell midden sites in Korea (Lee 2017). Other mammals, fish, and birds were also identified, including dog (*Canis lupus familiaris*), brown bear (*Ursus arctos*), raccoon dog (*Nyctereutes procyonoides*), Mongolian wolf (*Canis lupus chanco*), Siberian tiger (*Panthera tigris tigris*), wild water buffalo (*Bubalus arnee*), Korean ring-necked pheasant (*Phasianus colchicus karpowi*), duck (*Anatidae* sp.), redlip mullet (*Planiliza haematocheilus*), and spotted sea bass (*Lateolabrax maculatus*) (Kaneko 2008). Artifacts made of organic materials are also preserved intact, including pine dugout canoe and a paddle, basketry, and wooden and bone tools (Gimhae National Museum 2008, 2012).

An earlier study shows domesticated millets became dietary sources by the Middle Neolithic period (ca. 5500–5000 cal BP) (Crawford and Lee 2003). More recent studies based on charred remains and grain impressions on pottery confirm the presence of both foxtail and broomcorn millets in the Early Neolithic context at Tongsamdong (Obata 2013) and Bibongri (Lee et al. 2019; Kwak et al. 2020). Both charred remains and Early Neolithic pottery with millet impressions from Shell Layer 1 at Bibongri push the introduction of millet to the Early Neolithic period (7700–5500 cal BP) (Lee et al. 2019; Kwak et al. 2020). Floated sediments also yielded diverse edible or medicinal plant taxa, including acorn (*Quercus cf. serrata, Q. cf. glauca*), wild walnut (*Juglans sp.*), apricot (*Prunus armeniaca or P. mandshurica*), wild cherry (*P. tomentosa*), wild grape (*Vitis sp.*), spicewood (*Lindera sp.*), hop (*Humulus sp.*), dogwood (*Cornus sp.*), and aster (Asteraceae) (Lee 2008, 2017). Overall, the periods when these shell layers accumulated represent a warmer span with seasonally abundant resources (Gimhae National Museum 2008).

Shell Layer 1 is the most thoroughly analyzed stratum at Bibongri since it is the thickest cultural layer and filled with artifacts and organic remains (Figure 3). Shell Layer 1 yielded ten  ${}^{14}$ C dates on charcoal, bone, and shells published in Gimhae National Museum (2008, 2012), and we added one more  ${}^{14}$ C date on shell (D-AMS-039265) (Table 3).

# **RESULTS AND DISCUSSION**

Our two new  $\Delta R$  values are considerably different from those reported by Kong and Lee (2005). However, they are generally within the ranges reported in the coastal regions of East Asia (Tables 1 and 2). Moderate variations in the  $\Delta R$  values in close proximity are not uncommon (Yoshida et al. 2010; Panich et al. 2018). Several factors may have caused this difference, including handling error during the pre-laboratory and laboratory processes (Kim et al. 2016), an inherent variability in the measurement condition of each sample (Scott et al. 2007), and a possibility of exposure to the freshwater runoff depending on the geomorphology of the sample collection location (Ascough et al. 2005).

 $\Delta R$  values of ours and Kong and Lee's (2005) are applied to calculate the weighted mean  $\Delta R$ . We report two weighted mean  $\Delta R$  values of  $-83 \pm 16$  and  $-134 \pm 100$  (Table 3). The former is

Lab anda	Matarial	Spacios	$\delta^{13}C$	<sup>14</sup> C BP	Weighted	aal DD (1-)****	aal DD (2-)****
	Material	species	(700, VPDB)*	(conventional)		cal <b>DP</b> (10)	cal BP (20)
SNU05-343	Charcoal	—	-27.36*	$5330 \pm 40$	N/A	6191–6168 (11.3%)	6272–6241 (6.4%)
						6152–6111 (19.9%)	6209–5996 (89.0%)
						6082–6007 (37.2%)	
SNU10-1098	Charcoal		-23.39*	$5530 \pm 50$	N/A	6393–6368 (20.2%)	6438–6425 (1.4%)
						6351–6288 (48.1%)	6407-6272 (88.7%)
							6240-6210 (5.4%)
SNU10-1099	Charcoal	—	-18.77*	$5270 \pm 50$	N/A	6176–6146 (13.5%)	6190–5928 (95.4%)
						6116-6044 (30.0%)	
						6020–5989 (13.3%)	
						5969–5942 (11.4%)	
PLD-19846	Bone	Cervus nippon	-23.03*	$4935 \pm 25$	N/A	5702-5698 (2.8%)	5720-5596 (95.4%)
		hortulorum				5660-5599 (65.4%)	
PLD-19844	Bone	Cervus nippon	-22.84*	$4940 \pm 20$	N/A	5705-5695 (6.1%)	5718-5598 (95.4%)
		hortulorum				5660-5601 (62.1%)	
PLD-19845	Bone	Canis lupus familiaris	-13.8*	5640 ± 25	N/A	6480-6475 (2.9%)	6489–6391 (78.1%)
						6450-6395 (62.3%)	6372–6318 (17.3%)
						6363-6358 (3.0%)	
PLD-19843	Bone	Sus scrofa coreanus	-21.52*	5070 ± 25	N/A	5896-5881 (10.3%)	5903-5745 (95.4%)
						5871-5865 (4.2%)	
						5826-5753 (53.7%)	
SNU06-A001	Shell	Tegillarca	-15.21*	$4550 \pm 120$	$-83 \pm 16^{**}$	4831-4499 (68.3%)	5013-4312 (95.4%)
		granosa			$-134 \pm 100^{***}$	4911-4492 (66.9%)	5198-5190 (0.2%)
		-				4480-4465 (1.4%)	5177-4288 (95.1%)
							4283-4277 (0.2%)
SNU10-A013	Shell	Tegillarca	-4.26*	$5100 \pm 50$	$-83 \pm 16^{**}$	5450-5275 (68.2%)	5551-5159 (95.4%)
		granosa			$-134 \pm 100^{***}$	5555-5280 (68.2%)	5670-5065 (95.4%)
Beta-219091	Shell	Tegillarca	-8.3*	$5230 \pm 40$	$-83 \pm 16^{**}$	5579-5415 (68.2%)	5636-5313 (95.4%)
		granosa			$-134 \pm 100^{***}$	5685-5408 (68.2%)	5833–5281 (95.4%)
D-AMS-039265	Shell	Magallana	1.4*	$5727 \pm 28$	$-83 \pm 16^{**}$	6106–5937 (68.2%)	6190–5880 (95.4%)
		gigas			$-134 \pm 100$ ***	6201-5941 (68.2%)	6327-5801 (95.4%)

Table 3 <sup>14</sup>C dates from Shell Layer 1 at the Bibongri site.

\*The  $\delta^{13}$ C values in this table were calculated using accelerator mass spectrometry (AMS), not isotope ratio mass spectrometry (IRMS).

\*\*Based on two  $\Delta R$  values derived from one pre-bomb shell sample in this study.

\*\*\*Based on four  $\Delta R$  values derived from one pre-bomb shell sample in this study and two pre-bomb shell samples from Kong and Lee (2005).

\*\*\*\*This study used OxCal 4.3.2 for <sup>14</sup>C calibration (Bronk Ramsey 2009, 2017; Bronk Ramsey and Lee 2013). Charcoal and animal bone samples were calibrated using the IntCal20 atmospheric curve, and shell samples were calibrated using the Marine20 marine curve (Heaton et al. 2020; Reimer et al. 2020). Lab codes are listed in the at the *Radiocarbon* journal website (Radiocarbon 2021).

based on only the two values from our specimen, while the latter combines ours and Kong and Lee's (2005) (Table 1). A larger variation of  $\Delta R$  between ours and Kong and Lee's (2005) contributes to a greater error range in the weighted mean  $\Delta R$  of the latter.

Although Shell Layer 1 revealed the Yunggimun (appliqué) pottery, a typical style of the Initial Neolithic period (8000–7000 cal BP), a majority of pottery found are typical Early Neolithic wares with impressed, punctated, and incised decorations. Based on these pottery types and <sup>14</sup>C dates on charcoal fragments (Table 3, Figure 4C), Shell Layer 1 was regarded as the Early Neolithic period (7000–5500 cal BP) (Gimhae National Museum 2008, 2012). As Early Neolithic type vessels were in use for over 1500 years, the pottery typology itself cannot answer specific diachronic questions, including how long inhabitants used the area and when it happened within an over 1,500 year span. We can address the occupational span of Shell Layer 1 by comparing  $\Delta R$  adjusted dates on shells (Figure 4A, 4B) with calibrated dates on charcoal and animal bones (Figure 4C).

The stable isotopic analysis on bones from Shell Layer 1, which were also dated, does not raise a concern of sample contamination, collagen degradation, and carbon-exchange issues. The atomic carbon to nitrogen ratios measured from the bone collagens are 3.1 to 3.3 (Kim 2012). As the ratios of modern animals and humans range 2.9 to 3.6, if an archaeological sample falls within this range, that sample is regarded as reliable (DeNiro 1985). Four bones' collagen yields are around 13.2 weight percent collagen and are within the reliable range of 1.0 to 20.0 (van Klinken 1999).

We calibrated dates on archaeological shells with the two weighted mean  $\Delta R$  values, one based on our samples only,  $-83 \pm 16$  (Figure 4A) and the other that combined ours and Kong and Lee's (2005),  $-134 \pm 100$  (Figure 4B). Regardless to which of the two weighted mean  $\Delta R$ values are used, charcoal and animal bone dates are several hundred years older than two shell dates (SNU10-A013, Beta-219091). The other shell date, D-AMS-039265, is roughly contemporaneous to charcoal and animal bone dates (Figure 4, Table 3). The shell date, SNU06-A001, is much younger than all other dates found in the same layer. Also, it has the least precision (i.e., large uncertainty), indicating the possibility that the sample was contaminated during the laboratory procedure or relocated from the layer above. Calibrated date ranges of archaeological shells increase by 100–200 years if we apply the combined weighted mean  $\Delta R$  that includes values published by Kong and Lee (2005) (Figure 4B).

The potential impact of the old-wood effect should be examined (Schiffer 1986; Nolan 2012). Arboreal taxa identified at Bibongri through wood charcoal analysis include sawtooth oak (*Quercus acutissima Carruth.*), Korean red pine (*Pinus densiflora*), mulberry (*Morus* sp.), chestnut (*Castanea* sp.), and maple (*Acer* sp.) (Lee and Oh 2008). Pine and oak can live up to 1000 years under the right conditions. A previous study, however, suggests that the old-wood effect did not produce any consistent difference between the dates on annual seeds and wood charcoal from several Neolithic and Bronze period sites in Korea (ca. 8000–3500 BP) (Hwang et al. 2016). Contrary to the expectation of the old-wood effect, seeds appeared to be about 35 years older than charcoal on average. A contrasting pattern has been observed in historical sites (ca. 1700–1100 cal BP), where the dates on charcoal were older than those on seeds. In historical periods, thicker and older lumbers were likely used to build long-term houses, contributing to the old-wood effect (Hwang et al. 2016).



Figure 4 Comparison of 11 <sup>14</sup>C dates from Shell Layer 1 at the Bibongri site: (a) Dates on shells calibrated by the weighted mean  $\Delta R$  value of  $-83 \pm 16$  derived from the pre-bomb shell in this study (see Table 2). (b) Dates on shells calibrated by the weighted mean  $\Delta R$  value of  $-134 \pm 100$  derived from the pre-bomb shell in this study and Kong and Lee (2005). (c) Calibrated dates on charcoal and terrestrial animal bones. Calibration performed with OxCal 4.3.2 (Bronk Ramsey 2009, 2017; Bronk Ramsey and Lee 2013), based on IntCal20 atmospheric curve and Marine20 marine curve (Heaton et al. 2020; Reimer et al. 2020).

Schiffer (1986) first warned the old-wood effect by examining the dates from the southwest United States. However, Cook and Comstock (2014) identified a much lesser degree of old-wood effects from the Fort Ancient sites (2200–1600 BP) across southwestern Ohio and southeastern Indiana. They found that the decomposition rates of wood are generally very high in the temperate climate there. That is, the availability of old wood is much lower in

the temperate climate than in the arid zone. Southeastern Korea is in a temperate climate zone with distinct seasonal cycles of hot humid summer with monsoon, followed by dry cold winter. This climate condition may reduce the old-wood effect, similar to what Cook and Comstock (2014) observed in the United States.

Diatom analysis also indicates that Bibongri was influenced by saline seawater (Hwang 2008). Salty water is known to promote the decaying of wood (Shupe et al. 2008). Wood probably had a very low chance to survive for a substantive period of time in a seasonally wet condition and a saline water influx at Bibongri. The old-wood effect is less likely to have occurred at Bibongri unless wood materials have a highly durable composition like those in the early historical period in Korea. Accordingly, we can conclude that the age difference between charcoal and shells unlikely resulted from an old-wood effect. The earliest date on charcoal (SNU10-1098) and the latest date on shell (SNU10-A013) are several hundred years apart at the 2 sigma level. The range suggests that Shell Layer 1 was accumulated over an extended period of time between 6400 and 5000 cal BP.

Our data show that the use of marine reservoir correction values ( $\Delta R$ ) on marine shells can improve our understanding of the site chronology. A chronology based heavily on a pottery typology can only suggest that an entire stratum over 1 m thick belongs to a singular "Early Neolithic" period. The stratigraphic sequence and the site's specific duration of use cannot be understood exclusively based on pottery typology or uncalibrated radiocarbon years of marine and aquatic species. With an accurate calibration of marine dates and charcoal, we are able to understand the depth of time span through which multiple generations inhabited the area.

Bibongri is a multi-generational site used by those who shared the Early Neolithic pottery tradition over hundreds of years. Early Neolithic people likely dwelled at or revisited Bibongri to take advantage of its broad-spectrum resources over generations. Archaeobotanical and zooarchaeological studies conducted at the site reflect diverse seasonal marine and terrestrial resources throughout the year (Kaneko 2008; Kwak et al. 2020). Bibongri presented an affluent environment during the period where these shell middens were used. Numerous artifacts that represent diverse activities were recovered from Shell Layer 1, including digging tools, stone knives, axes, adzes, whetstones, grinding stones, spears, fishing hooks, and net sinkers (Gimhae National Museum 2008, 2012). This diverse combination of tools and dwelling structures represent at least multi-seasonal settlements at Bibongri. Diverse seasonal resources, which are reflected in plant and animal remains, were attractive to Bibongri people; they were able to sustain themselves through fishing-shellfishing, hunting, wild plant harvesting, and millet cultivation during the Early Neolithic period (Kwak et al. 2020). A long tradition of such broad-spectrum resource use probably made Bibongri a vital cultural niche over several hundred years, similar to the findings on the east coast of Korea (Lee et al. 2019).

# CONCLUSION AND FUTURE DIRECTIONS

We added two additional  $\Delta R$  values to the Pacific database by dating the pre-bomb (pre-1950) blue mussel shell (*Mytilus edulis*) from the southern coastal region of Korea. As a pioneering case study, we calibrated archaeological shell samples from Shell Layer 1 of the Bibongri Neolithic shell midden, where dates on charcoal are also available. By comparing the dates on charcoal samples with the  $\Delta R$  calibrated shell dates, we demonstrate that Shell Layer 1

represents long-term human activities over several hundred years, and thus improves our understanding of the site chronology. Our Bibongri case study demonstrates that properly calibrated shell dates can play a key role in improving the chronological understanding of Neolithic shell midden in Korea. Moving forward, we will investigate the variation of  $\Delta R$ across the Korean Peninsula and its contributing causes by reporting additional  $\Delta R$  using pre-bomb shell samples and pairing them with the dates on terrestrial samples to build a fine-grained chronology of the Neolithic Period.

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#### SUPPLEMENTARY MATERIAL

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