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# Radiocarbon dating supports bivalve-fish age coupling along a bathymetric gradient in high-resolution paleoenvironmental studies

Supplementary text on methods

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# **DETAILS ON METHODS**

# Location of sampling sites

Table DR1. List of sampling sites with coordinates (WGS84), type of sediment (after Connor et al., 2004) and sedimentation rate.

Station	Latitude	Longitude	Type of sediment	Sedimentation rate (mm/yr)
- 10 m	31.6953° N	34.5588° E	sand	0.4 (Goodman- Tchernov et al., 2009)
- 30 m	31.7100° N	34.5406° E	muddy sand	0.2 (our own unpublished data)
- 40 m	31.7487° N	34.4960° E	mud	2.4 (our own unpublished data)

Grab penetration in the sands at -10 m was likely lower than at the deeper stations. However, this depth is well above the fair-weather wave base and thus a particularly well mixed surface sediment layer that enables fully representative for time-averaging quantification.

# List of dated species

Table DR2. List of samples per depth, including taxa dated and their abundance.

Depth	Mollusk species	Mollusk sample size	Fish species	Fish sample size
- 10 m	Donax semistriatus Poli, 1795	15	<i>Ariosoma balearicum</i> (Delaroche, 1809)	15
- 30 m	<i>Corbula gibba</i> (Olivi, 1792)	15	Gobius auratus Risso, 1810 (1) Gobius paganellus Linnaeus, 1758 (12) Lesueurigobius friesii (Malm, 1874) (1)	14
- 40 m	<i>Corbula gibba</i> (Olivi, 1792)	15	Ariosoma balearicum (Delaroche, 1809) (1) Gobius cobitis Pallas, 1814 (1) Gobius niger Linnaeus, 1758 (1) Lesueurigobius friesii (Malm, 1874) (4) Lesueurigobius suerii (Risso, 1810) (1)	8

All dated species are native to the Mediterranean Sea and recorded alive today.

#### Preparation of samples for radiocarbon dating

To allow for a sufficient mass for radiocarbon analysis, we selected the shells from a 2 mm mesh and otoliths with mass larger than 0.8 mg (because their shape caused many suitable otoliths to pass through the mesh).

Mollusk shells were subsampled by gently breaking and selecting a small fragment. Otoliths were used whole. All samples were cleaned by sonicating and rinsing in deionized distilled water (DDI; 16.3 m Ohm) repeatedly up to three times. Samples were leached with 2 M HCl, with the extent of leaching dependent on sample mass: samples larger than 1 mg were leached to remove about 30% by mass and samples between about 1 and 0.4 mg were leached to remove about 15%. Samples were ultimately rinsed three times with DDI water then dried in a 50 °C oven overnight. They were ground to a fine powder using a small agate mortar and pestle. Between 0.15 and 0.50 mg of the carbonate powder was transferred to serialized (3 hr at 500 °C) borosilicate glass culture tubes (6 mm OD x 50 mm). Samples comprising less than 0.15 mg of recovered powder were not analyzed. The carbonate was combined with 6 to 7 mg of niobium (Nb Puratronic, -325 mesh, 99.99%) powder using a spatula. The tubes were flushed with N<sub>2</sub> gas and capped with Supelco plastic column caps (1/4" OD) to reduce atmospheric exposure until the powder was pressed into targets.

The preparation for standard-precision AMS radiocarbon analysis followed the same cleaning procedure as for mollusks (above). Rather than powdering the carbonate, however, samples were converted to graphite at the Center for Ecosystem Science and Society (ECOSS) laboratory, Northern Arizona University. Following cleaning, between 7 and 8 mg of shell fragments were placed into BD Vacutainer plastic collection tubes (13 x 75 mm) and sealed with red/grey conventional stopper closures. Ambient atmosphere was removed via vacuum and 8 ml of stock phosphoric acid was dispensed into each tube using a small-bore needle. The tubes were placed in a heating block and at 70 °C until no reaction (e.g. bubbling) was visible. The evolved CO<sub>2</sub> was removed via vacuum and cryogenically purified through a chilled ethanol and liquid nitrogen process, and was then converted to graphite by reaction with an Fe catalyst in a hydrogen atmosphere following the methods of Vogel (1992).

The metal plus either carbonate or graphite mixtures were pressed into pre-drilled (0.160" depth) aluminum targets at 400 psi, rotated 90°, and pressed again at 400 psi. The targets were sent to the Keck Carbon Cycle AMS Laboratory at the University of California Irvine for <sup>14</sup>C analysis.

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### Radiocarbon ages calibration

Radiocarbon ages were converted to calendar years using OxCal 4.2 (Ramsey, 2009), Marine13 data (Reimer et al., 2013), and a constant regional marine reservoir correction ( $\Delta$ R) of 3 ± 66 yrs, which is the weighted mean of eight published pre-bomb  $\Delta$ R values from Israel and Lebanon (see Table DR3). For samples younger than 1950 AD, the fraction of modern carbon (F<sup>14</sup>C) was converted to calendar ages using a regional marine calibration curve and the calibration software OxCal v4.2. The post-1950 regional marine curve was constructed using 10 live-collected *Corbula gibba* shells collected along the coast of Israel (see Table DR4). All calibrated ages are reported in calendar years relative to 2016 AD, the year of sample collection (2016 AD = 0 yr).

Table DR3. Regional pre-bomb  $\Delta R$  values for our study sites. These pre-bomb  $\Delta R$  values, listed in the Online Marine Reservoir Correction Database (http://calib.org/marine/), were used for the calculation of a weighted mean  $\Delta R$  value of 3 ± 66 <sup>14</sup>C yr (n=8).

	Location	Latitude	Longitude	Year of	$\Delta R \pm 1\sigma$	References
				collection	( <sup>14</sup> C yr)	
1	Netanya, Israel	34.83	32.17	AD 1937	52 ± 40	Reimer and McCormac, 2002
2	Beirut, Lebanon	35.5	33.87	AD 1929	37 ± 40	Reimer and McCormac, 2002
3	Beirut, Lebanon	35.5	33.87	AD 1929	-52 ± 50	Reimer and McCormac, 2002
4	Israel	34.8482	32.3384	AD 1937	47 ± 40	Boaretto et al., 2010
5	Israel	34.8482	32.3384	AD 1937	-70 ± 50	Boaretto et al., 2010
6	Israel	34.9227	32.6432	AD 1937	-20 ± 50	Boaretto et al., 2010
7	Israel	34.9227	32.6432	AD 1937	75 ± 50	Boaretto et al., 2010
8	Israel	35.0138	32.8431	AD 1937	-115 ± 50	Boaretto et al., 2010

Table DR4. <sup>14</sup>C values of live-collected shells (*Corbula gibba*) measured in this study. All the shell samples were collected at water depth of 50 m or less, and were converted to graphite and measured using the AMS Facility at the University of California Irvine. These measured <sup>14</sup>C values were used to constructed the post-1950 regional marine curve for age calibration.

	Lab ID	Sample ID	Location	Year of collection	F <sup>14</sup> C ± 1σ
1	214201	RC198	Israel, Haifa Bay	AD 1954	0.9468 ± 0.0020
2	214202	RC200	Israel, Ashdod	AD 1960	0.9860 ± 0.0018
3	214203	RC202	Israel, Bat Yam	AD 1962	0.9937 ± 0.0019
4	207180	RC096	Israel, Atlit-Dor	AD 1965	1.0670 ± 0.0020
5	207179	RC098	Israel, Ashdod	AD 1970	1.1241 ± 0.0018
6	207178	RC100	Israel, Palmachim	AD 1977	1.1177 ± 0.0020
7	207177	RC102	Israel, Nizzanim	AD 1988	1.0883 ± 0.0017
8	207176	RC080	Israel, Hadera	AD 2002	1.0499 ± 0.0017

	Lab ID	Sample ID	Location	Year of collection	F <sup>14</sup> C ± 1σ
9	207175	RC092	Israel, Atlit	AD 2016	1.0342 ± 0.0017
10	207174	RC094	Israel, Ashqelon	AD 2017	1.0293 ± 0.0017

#### **Model parameters**

Table DR5. Parameters and their confidence intervals of three models (one-phase exponential, Weibull, and two-phase exponential) fitted to the shell and otolith age-frequency distributions, with AICc values, along the depth gradient off Ashqelon, southern Israel. In **bold** the shell model parameters showing no difference from the otolith ones. The half-life of the one-phase and two-phase exponential models was computed with log(2)/ $\lambda$  and the Weibull hazard (the per-specimen instantaneous probability of loss) with k·r<sup>k</sup>·age<sup>k-1</sup>. The median time to sequestration or burial below the TAZ in the two-phase model is 1/r.

	-10 m	depth	-30 m	depth	-40 m	depth
	Shells	Otoliths	Shells	Otoliths	Shells	Otoliths
One-phase λ	0.0011	0.0031	0.0007	0.0006	0.0053	0.0299
	[0.0006 –	[0.0019 –	[0.0005 –	[0.0004 –	[0.0035 –	[0.0131 –
	0.0028]	0.0063]	0.0011]	0.0010]	0.0099]	0.1013]
Weibull r	0.0958		0.0004	0.0004	0.0089	0.0057
	[0.0008 –	NA	[0.0002 –	[0.0002 –	[0.0020 –	[0.0057 –
	238.9]		0.0035]	0.0666]	1.0106]	0.0417]
Weibull k	0.3188		1.8	2.2	0.7	1.4·10 <sup>7</sup>
	[0.1661 –	NA	[0.5 –	[0.3 –	[0.3 –	[1864.8 –
	2.8·10 <sup>6</sup> ]		2.3·10 <sup>7</sup> ]	2.7·10 <sup>7</sup> ]	3.7·10 <sup>7</sup> ]	17.9·10 <sup>7</sup> ]
Two-phase $\lambda_1$	0.0205	0.0014	0.0010	0.1466	0.0678	0.0696
	[0.0148 –	[0.0011 –	[0.0005 –	[0.0004 –	[0.0395 –	[0.0406 –
	0.4934]	0.0018]	0.0342]	0.1968]	0.1160]	0.1235]
Two-phase λ <sub>2</sub>	0.0006	0.0014	0.0006	0.0006	0.0033	0.0089
	[0.0004 –	[0.0010 –	[0.0004 –	[0.0004 –	[0.0026 –	[0.0065 –
	0.0014]	0.0018]	0.0010]	0.0009]	0.0044]	0.0977]
Two-phase т	0.0008	0.0000	0	0.0033	0.0047	0.0019
	[0.0002 –	[0.0000 –	[0 —	[0 —	[0.0013 –	[0.0001 –
	0.0058]	0.0001]	0.0135]	0.0121]	0.0181]	0.0102]
One-phase half-	274	07.1	130	502	57	10
life (yrs)	274	97.1	430	502	57	10
Two-phase half-						
life of first phase	15	215	301	2	4	4
(yrs)						
Two-phase half-						
life of second	502	215	502	502	91	34
phase (yrs)						
Median time to	1250	NA	NA	303	213	526
sequestration (yrs)	1200			505	215	520

		-10 m depth		-30 m	-30 m depth		-40 m depth	
		Shells	Otoliths	Shells	Otoliths	Shells	Otoliths	
/e bods	one- phase	117.4	101.6	124.6	117.7	93.7	36.1	
gativ elihc	Weibull	113.2	110.9	123.7	116.5	92.6	41.3	
neç loglike	two- phase	110.4	113.7	124.2	116.3	89.5	34	
	one- phase	236.8	211.4	251.1	237.3	189.4	74.2	
AICs	Weibull	231.4	NA	252.4	238.1	190.2	88.9	
<	two- phase	229.1	235.6	256.7	241.0	187.1	80.1	



Figure DR1. The per-specimen probability of loss from the taphonomic active zone (and half-life in the TAZ) and its relationship with postmortem age shows that otoliths and shells at 30 m and at 40 m behave similarly. Otoliths and shells at 10 m are more variable.

#### Accuracy of carbonate-target ages

Four dated shells of *Corbula gibba* (Table DR6) were also dated with the graphitetarget method to assess the accuracy of the carbonate-target ages. The four shells were selected to span the whole age range of the samples. No otolith had enough mass to enable both analyses. Table DR6. Graphite-target and carbonate-target results of *Corbula gibba* samples used to assess the accuracy of the carbonate-target method. Uncalibrated radiocarbon ages are reported in years before present (where 0 BP = AD 1950).

Samplo	Graphite	Graphite	Carbonate	Carbonate
Sample	age	age SD	age	age SD
RC015	435	20	650	60
RC132	4190	25	4140	90
RC168	-632	-15	-540	-46
RC183	2700	25	2730	40

While thousand-year old samples showed almost identical ages, for younger samples (RC015 and RC168; modern to few hundred years of uncalibrated radiocarbon age) the carbonate targets gave older ages than the graphite-target ones.

To estimate the age relationship of the two AMS methods, we used reduced major axis regression supplemented by 10,000-iteration bootstrap as in (Kowalewski et al., 2018) using the script published as appendix 3 of their paper with minor modifications to graphical outputs.

The coefficient of determination is very high ( $r^2 = 0.999$ ), the slope is 0.96 (CI = [0.918, 1.116]) and the intercept is 137.6 years BP (CI = [72.5, 250.7]) (Figure DR2).



Figure DR2. Reduced Major Axis (RMA) regression vs. the hypothesis of perfect agreement between methods (y = x).



Figure DR3. Bootstrap-derived sampling distributions of slope and intercept coefficients for reduced major axis regression of graphite versus carbonate targets (see Figure DR2). A. Slope coefficient. B. Intercept coefficient. Each distribution based on 10,000 replicate bootstrap samples. Symbols: p - two-tailed significance estimates for null hypotheses for slope (b = 1) and intercept (a = 0) coefficients, respectively (based on a percentile approach).

# Correction of shell ages and consequences for time-averaging quantification

The shell radiocarbon ages obtained from carbonate-targets were corrected using the linear model discussed above:

corrected age = carbonate age 
$$\cdot$$
 1.04 - 144

We recomputed the age frequency distribution metrics and model parameters with both uncorrected and corrected carbonate ages (Table DR7 and Table DR8).

Table DR7. Summary statistics and their confidence intervals for the age frequency distributions of mollusk shells using calibrated ages derived from uncorrected and corrected carbonate <sup>14</sup>C analysis. Confidence intervals were computed with a bootstrapping procedure with 10,000 iterations. In green, the corrected values with no difference from the uncorrected ones.

	-10 m		-30 m		-40 m	
	Uncorrected	Corrected	Uncorrected	Corrected	Uncorrected	Corrected
Median age	671	599	1470	1396	207	139
	[54 – 1269]	[5 – 1181]	[735 – 1784]	[194 – 1766]	[19 – 298]	[27 – 191]

	-10 m		-30 m		-40 m	
	Uncorrected	Corrected	Uncorrected	Corrected	Uncorrected	Corrected
Age range	4171	4181	4771	4877	504	393
	[1225 –	[1178 –	[1816 –	[1727 –	[349 – 504]	[203 – 394]
	4186]	4181]	4983]	4997]		
Time-averaging	962	920	1344	1357	292	174 [73 –
(IQR)	[334 – 4118]	[322 – 4158]	[606 – 2521]	[609 – 2589]	[116 – 446]	360]
Skewness	1.646	1.688	0.960	0.986	0.334	0.672
	[0.105 –	[0.135 –	[-0.276 —	[-0.225 –	[-0.447 –	[-0.073 –
	2.784]	2.848]	1.978]	2.046]	1.341]	1.617]

Table DR8. Parameters and their confidence intervals of three models (one-phase exponential, Weibull, and two-phase exponential model) fitted to the shell age-frequency distributions, with AICc values, based on uncorrected and corrected values. In green, the corrected values with no difference from the uncorrected ones. In **bold** the minimum AIC values.

	-10 m	depth	-30 m	depth	-40 m	depth
	Uncorrected	Corrected	Uncorrected	Corrected	Uncorrected	Corrected
One-phase λ	0.0011	0.0011	0.0007	0.0007	0.0052	0.0071
	[0.0006 –	[0.0006 –	[0.0005 –	[0.0005 –	[0.0035 –	[0.0048 –
	0.0027]	0.0033]	0.0011]	0.0012]	0.0098]	0.0126]
Weibull r	0.0928	4622	0.0004	0.0005	0.0083	0.0059
	[0.0008 –	[0 – 1.67·10 <sup>7</sup> ]	[0.0002 –	[0.0003 –	[0.0020 –	[0.0024 –
	205.9436]		0.0033]	0.0061]	1.0228]	0.0349]
Weibull k	0.3194	0.1445	1.8	1.5	0.8	1.2
	[0.1688 –	[0.1060 –	[0.6 –	[0.5 –	[0.3 –	[0.6 –
	5.7·10 <sup>5</sup> ]	0.8678]	2.5·10 <sup>7</sup> ]	2.6·10 <sup>7</sup> ]	3.7·10 <sup>7</sup> ]	3.5·10 <sup>7</sup> ]
Two-phase $\lambda_1$	0.0205	0.1682	0.0009	0.0031	0.0675	0.0284
	[0.0148 –	[0.1187 –	[0.0005 –	[0.0005 –	[0.0401 –	[0.0049 –
	0.4934]	0.2495]	0.0338]	0.0180]	0.1138]	0.0433]
Two-phase $\lambda_2$	0.0006	0.0006	0.0006	0.0006	0.0033	0.0057
	[0.0004 –	[0.0004 -	[0.0004 –	[0.0004 –	[0.0026 –	[0.0039 –
	0.0014]	0.0013]	0.0010]	0.0010]	0.0044]	0.0094]
Two-phase т	0.0009	0.0007	0	0.0010	0.0047	0.0076
	[0.0002 –	[0.0002 –	[0 – 0.0142]	[0 - 0.0065]	[0.0013 –	[0 – 0.0252]
	0.0060]	0.0026]			0.0180]	
One-phase half-	274	274	430	430	58	42
life						
Two-phase half-	15	2	335	97	4	11
life of first phase						
Two-phase half-	502	502	502	502	91	53
life of second						
phase						
Median time to	1111	1429	NA	1000	213	132
sequestration						

	-10 m depth		-30 m depth		-40 m depth	
	Uncorrected	Corrected	Uncorrected	Corrected	Uncorrected	Corrected
One-phase AICc	237.1	235.5	251.1	249.7	189.7	180.5
Weibull AICc	231.5	215.9	252.1	250.7	190.5	182.7
Two-phase AICc	229.3	202.0	256.6	255.0	187.5	185.4

The AFD and model parameters computed with the corrected ages are in most cases not different from those computed with uncorrected ages. Only two differences can be seen at 40 m depth. First, the most supported model is the one-phase exponential instead of the two-phase exponential. The  $\Delta$ AICc between the one-phase and twophase exponential model with uncorrected ages is however very small ( $\Delta$ AICc=2.2). Therefore, the one-phase exponential model was already only marginally less supported than the two-stage one. Second, in the two-stage exponential model,  $\lambda_1$  is lower while  $\lambda_2$  is higher. Still, the half-lives are poorly affected suggesting no major differences in the interpretation of the taphonomic processes behind the observed AFDs.

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