TEMPORAL CHANGES IN RADIOCARBON RESERVOIR AGE IN THE DEAD SEA-LAKE LISAN SYSTEM

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ABSTRACT. The Holocene Dead Sea and the late Pleistocene Lake Lisan were characterized by varying radiocarbon reservoir ages ranging between 6 and 2 ka in the Dead Sea and between 2 ka and zero in Lake Lisan. These changes reflect the hydrological conditions in the drainage system as well as residence time of ¹⁴C in the mixed surface layer of the lake and its lower brine. Long-term isolation of the lower brine led to ¹⁴C decay and an increase in the reservoir age. Yet, enhanced runoff input with atmospheric ¹⁴C brings the reservoir age down. The highest reservoir age of 6 ka was recorded after the sharp fall of the Dead Sea at ~8.1 ka cal BP. The lower reservoir age of zero was recorded between 36 and 32 ka cal BP, when the Lake Lisan mixed layer was frequently replenished by runoff.

INTRODUCTION

Radiocarbon dating of the Dead Sea shallow and deep water masses, as well as its carbonate sediments, depends on the evaluation of the "reservoir age" (RA) of the water, which requires an understanding of the fate of ¹⁴C during the runoff evolution that is the main dissolved inorganic carbon (DIC) input into the lake. Waters flowing over carbonate terrains, like the Judean Mountains west of the Jordan-Arava Valley, or percolating through carbonate aquifers, may contain substantial amounts of "dead" carbon in their DIC. This DIC gets incorporated into the "new" precipitated carbonates, causing an apparent increase in the ¹⁴C ages.

Analyses of fresh and saline waters from the Dead Sea drainage area show that runoff waters are characterized by a high percent of modern carbon values (~100 pMC), while springs that percolate throughout the Cretaceous carbonate aquifers are characterized by significantly lower values (~50 pMC, cf. Talma et al. 1997). Most of the annual bicarbonate supply for aragonite precipitation in the Dead Sea-lacustrine system comes from freshwater, while the Dead Sea brine supplies calcium (Stein et al. 1997; Barkan et al. 2001). Therefore, aragonite layers deposited in the Dead Sea may contain various amounts of ¹⁴C depending on the proportions of water types entering the lake and the nature of their relative mixing with Dead Sea brine (besides possible age effects). The combined effect of these mechanisms determines the reservoir age of the Dead Sea.

The Holocene Dead Sea is a remnant water body after the significantly larger, deeper, and relatively less saline Lake Lisan that existed in the Dead Sea basin during the Last Glacial period (Neev and Emery 1967; Begin et al. 1974; Katz et al. 1977; Stein 2001). The annual nature of sedimentation in the lake (Migowski et al. 2004) and the possibility to obtain both ¹⁴C ages from the aragonite laminae and from associated organic debris opens a way to resolve secular changes in the ¹⁴C RA in the lake. In this paper, we summarize the available ¹⁴C RA data from the late Pleistocene Lake Lisan (for the time interval of 36 to 17 ka BP) and the Holocene Dead Sea (past 10 ka), and discuss their relation to the hydrological-limnological history of the lake system.

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LAKE LISAN AND THE DEAD SEA LIMNOLOGICAL SYSTEMS

Lake Lisan existed between ~70 and 14 ka BP (Kaufman 1971; Haase-Schramm et al. 2004). During the time of highest water level (>170 meters below mean sea level [m bmsl]), the lake extended over about 300 km from the Sea of Galilee in the north to the Hazeva area in the south. The Lisan Formation, which was deposited within the lake and its surrounding fan deltas, consists mainly of chemical precipitates, aragonite and gypsum, and clastic material transported by floods. The aragonite appears in thin (~0.5–1 mm thick) laminae alternating with detrital laminae of similar or larger thickness. The aragonite precipitated most likely from the surface water and is preserved in its primary state due to the dry climate in the Dead Sea basin and the high Mg/Ca ratio of interstitial soluble salts (remnants of the former porewater; Katz and Kolodny 1989; Stein et al. 1997).

The excellent preservation and high U concentrations (~3 ppm) of Lisan aragonite make it useful for ²³⁴U-²³⁰Th dating (Kaufman 1971; Schramm et al. 2000; Haase-Schramm et al. 2004). The U-Th ages established by TIMS on aragonites from the Perazim Valley (PZ1) section lie in the range of ~70 to 14 ka BP (Haase-Schramm et al. 2004).

The average rate of aragonite precipitation during the Lisan period was 1.5 m eq HCO₃ per cm² yr⁻¹, which is 4 to 5 times higher than the rate of the present Dead Sea (Barkan et al. 2001). Such a high precipitation rate required a substantial supply of bicarbonate to the lake, which was furnished by the incoming freshwater that most likely stratified the water column (Stein et al. 1997). The lower density surface layer reached aragonite saturation.

At ~13 ka cal BP, Lake Lisan receded rapidly, arriving to its minimum level of below 500 m bmsl (Stein 2002). The lake (now termed the "Dead Sea") recovered at the beginning of the Holocene, when it stabilized around the elevation of 400 m bmsl with a few fluctuations of up to 30 m (Ken-Tor et al. 2002; Bookman et al. 2004; Migowski et al. 2004). The sedimentary lacustrine section of the Holocene Dead Sea resembles that of Lake Lisan, where sequences of aragonite and silty detritus alternate with thicker sandy and gypsum layers. During the past several hundred years, the Dead Sea has stabilized as a 2-layered meromictic lake, which was disrupted by the 1978 overturn (Stiller and Chung 1984).

¹⁴C RESERVOIR AGES IN LAKE LISAN-DEAD SEA ARAGONITES

The RAs in the late Pleistocene Lake Lisan and the Holocene Dead Sea are evaluated by subtracting the U-Th calendar age of Lisan aragonite or the ¹⁴C age (ka BP) of organic debris (such as wood remains, leaves, twigs) from the measured ¹⁴C age (ka BP) of the aragonite laminae in the same stratigraphic horizon (Table 1). Samples were collected in the Perazim Valley section (PZ1, described by Haase-Schramm et al. 2004), Massada section (beneath the archaeological site, see Bartov et al. 2002), and in the Ze'elim gully (Bookman et al. 2004), and were recovered from a sedimentary core drilled in Ein Gedi spa (Migowski et al. 2004). ¹⁴C data are listed in Table 1 and are illustrated in Figure 1 against the level chronology of the lakes. During the Holocene, the RA displays temporal decrease from an elevated value of 6 ka at ~7.6 ka cal BP (after the Dead Sea sharp fall at 8.1 ka cal BP) to 2.2 ka at present. One pair that was sampled below the high RA peak, at 9.1 ka cal BP, yielded a lower RA of 3.3 ka. It appears that the brine carried an older ¹⁴C memory from the earlier stages of the Holocene and the post-glacial period (when the lake level was at its minimum stand; Stein 2002). When the first aragonite deposited at ~7.6 ka cal BP, the mixed layer accommodated a significant contribution of old carbon from the brine.

Table 1 Lake levels and ¹⁴C ages on aragonite and organic matter and U-Th ages data from Lake Lisan and the Holocene Dead Sea.

Lisan and the			-	Measured			Calibrated	Calendar	
Sample		Lake level	Sample	¹⁴ C age		RA	¹⁴ C age	U-Th age	Data
ID	Location	(m bmsl)a	type	(ka BP)	\pm	(ka)	(cal ka BP)	(ka BP)	source ^b
Lake Lisan									
PZ1-2290	Perazim	280	aragonite	32.8c	1.1	~0a	_	37	1
PZ1 2236-W			wood	35.9	2.7		_	_	
PZ1 2450	Perazim	280	aragonite	30.6	0.6	$\sim 0^{b}$	_	34	1
PZ1 2450-L			leaf	32.6^{d}	1.1			_	
PZ-1 2558	Perazim	280	aragonite	30.5	0.3	0	_	33	2
			wood	30.5	0.3		_	_	
MS-410	Massada	180	aragonite	24.0	0.2	1.5	_	24	3
			wood	22.5	0.2		_	_	
MS-405	Massada	180	aragonite	24.0	0.2	1.7	_	24	3
			wood	22.3	0.2			_	
M-2375	Massada	180	aragonite	22.7	0.2	2.0		24	1
M-2374 W			wood .	20.7	0.2		_	_	_
ID 455	Perazim	200	aragonite	22.0	0.1	1.3	_	23	2
			wood	20.7	0.1		_	_	
MS-179	Massada	200	aragonite	19.5	0.1	1.4	_	21	3
			wood .	18.2	0.1		_	_	
MS-218	Massada	200	aragonite	19.0	0.1	2.0	_	19	3
DEL 0610	. .	• • • •	wood	17.0	0.1		_		
PZ1-3649	Perazim	200	aragonite	14.5	0.1	1.6	_	17	1
			debris	12.9	0.2		_	_	
Dead Sea									
	EGe core	400	aragonite	2.38	0.07	2.2	0.1	_	4
			twigs	0.16	0.03			_	
	EG core	400	aragonite	2.97	0.07	2.2	0.2	_	4
			twigs	0.80	0.03			_	
	EG core	400	aragonite	3.86	0.07	2.3	1.1	_	4
			plant fiber	1.56	0.03			_	
	EG core	370	aragonite	6.09	0.04	2.4	4.1	_	4
			twigs	3.68	0.03			_	
	EG core	400	aragonite	7.92	0.07	2.6	6.2	_	4
			twigs	5.28	0.04			_	
	EG core	400	aragonite	9.85	0.07	3.4	7.4	_	4
			twigs	6.46	0.07			_	
	EG core	<450	aragonite	12.7	1.2	6.0	7.7	_	4
			twigs	6.73	0.04			_	
	EG core	400	aragonite	11.5	1.2	3.3	9.1	_	4
			twigs	8.20	0.04			_	

^am bmsl= meters below mean sea level.

^bData sources: 1=Haase-Schramm et al. 2004; 2=van der Borg et al. 2004; 3=Prasad et al., forthcoming; 4=present study.

^cThe ¹⁴C age was obtained on aragonite from a stratigraphically younger layer; thus, considering short transport time of the wood and the large error on sample PZ1-2236-W, the estimated RA at 2236 is about zero.

^dThe leaf is slightly older than the aragonite; therefore, the reservoir age was estimated to be about zero.

eEG denotes Ein-Gedi.

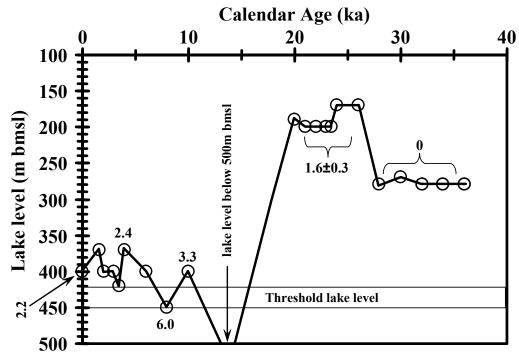


Figure 1 Reservoir ages (RA in ka units) during various water levels of the Last Glacial Lake Lisan and the Holocene Dead Sea. Lake-level data are from Bartov et al. 2002, 2003; Bookman et al. 2004; Migowski et al. 2004.

During the Lisan period (considering the time intervals where aragonite-organic debris pairs are available), the RAs cluster around 2 values: (1) about zero between 36 and 32 ka BP; and (2) 1.6 ± 0.3 ka, between 24 and 20 ka BP. Figure 2 displays the RAs at various lake-level conditions. It appears that the RA is related in a complicated way to the lake level, which is the most prominent hydrological indicator in the lake system. While the early Holocene Dead Sea shows RAs that are significantly higher than Lake Lisan, the mid-late Holocene Dead Sea (past 4000 yr) RAs are not much different from the high-stand Lake Lisan (2.3 ± 0.1 ka compared with 1.6 ± 0.3 ka, respectively). More surprising is the zero RA recorded by Lake Lisan at its "typical" elevation of ~280 m bmsl. These observations indicate that the RA is not a simple function of freshwater bicarbonate supply to the lake, and other factors such as 14 C decay in the water body are involved.

DISCUSSION

Residence Time of ¹⁴C in the Modern Dead Sea

The dynamics of the carbonate system of the Dead Sea were investigated after unusually high rainfall and enhanced runoff in the winter of 1991/92, which came after years of continuous retreat mainly due to anthropogenic interference (Barkan et al. 2001). The intense flooding formed a shallow pycnocline that divided the water column into 2 dissolved inorganic carbon (DIC) reservoirs: the shallow (about 20 m deep) surface mixed layer DIC reservoir opened to the atmosphere, and a large deep water DIC reservoir isolated from the atmosphere. This salinity stratification of the Dead Sea lasted for more than 2 yr. The average DIC value in the Dead Sea is 850 μ mol·kg⁻¹, which is much lower than in the freshwater runoff. The measured $P_{CO_2(liquid)}$ was very high, approximately 2000 μ atm (compared to the atmospheric value of 355 μ atm) at all depths. This high $P_{CO_2(liquid)}$

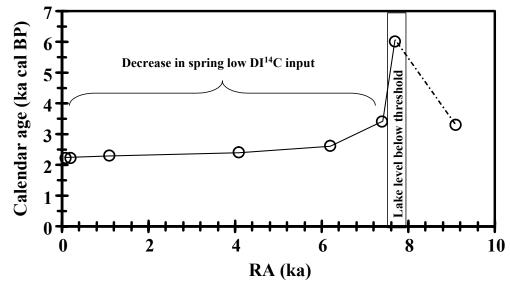


Figure 2 Reservoir age (RA in ka) versus calibrated ¹⁴C age in the Holocene core drilled at Ein Gedi shore. Note the fast decrease from the reservoir age peak at 7.6 ka cal BP, and the following moderate decrease in the reservoir age, reflecting the decrease in DIC supply of springs with low DI¹⁴C (groundwater DIC source with low ¹⁴C content) and the continuous replenishment by runoff water with high DI¹⁴C content (atmospheric ¹⁴C).

represents a DIC excess (above equilibrium with atmospheric CO₂) of about 300 μ mol·kg⁻¹, which is equivalent to 40% of the DIC. The carbon fluxes in the Dead Sea as estimated for the years 1992 to 1994 were (1) HCO₃⁻ influx with floods of 2.1 mol·m⁻²·a⁻¹); (2) CaCO₃ precipitation of 1.4 mol·m⁻²·a⁻¹ that occurred in the mixed layer; and (3) a large CO₂ escape to the atmosphere of 4 mol·m⁻²·a⁻¹ driven by the high $P_{CO_2(liquid)}$.

The overall DIC balance for the mixed layer suggested a net DIC loss of 3.3 mol·m⁻²·a⁻¹, which can potentially deplete the mixed layer DIC within 10 yr and the whole water column within about 100 yr. Earlier DIC data (Neev and Emery 1967; Luz et al. 1997) suggested that these conditions of $P_{CO_2(liquid)} >> P_{CO_2(atm)}$ also prevailed in the deep-water column during the several centuries of continuous meromictic stages which prevailed before the 1978 overturn (Stiller and Chung 1984; Barkan et al. 2001). The existence of such high $P_{CO_2(liquid)}$ over a time period that is longer than the calculated residence time of DIC in the deep-water mass of the Dead Sea suggests that the deep DIC reservoir (the main DIC reservoir of the Dead Sea) remained during the whole meromictic stage. Thus, the ¹⁴C content of the large deep-water reservoir was several hundred years old when ventilated during the 1978 overturn, increasing the RA of the newly formed mixed layer.

When the lake is stratified during meromictic stages, the ¹⁴C content of the surface mixed layer (from which the authigenic carbonate precipitates) may change rapidly. For example, in the modern Dead Sea, the residence time of carbon in the mixed layer is about 8 yr, implying that the RA may approach zero after about 20 yr of continuous stratification. On the other hand, during the holomictic stage (when the lake mixes entirely at least during one period a year), the deep-water column serves as a buffer for abrupt changes in the ¹⁴C content of surface water and, hence, the RA of the lake.

Residence Time of ¹⁴C in the Old Lakes

The high peak of the RA at 7.6 ka cal BP, when the Dead Sea was at one of its lowest stands ~417 m bmsl (Migowski et al. 2004), and the secular decrease in RA that followed this RA peak are key observations for understanding the behavior of ¹⁴C in the old lakes. We suggest that the extremely high RA (6 ka) at 7.6 ka cal BP indicates that i) springs (mainly saline springs having very low ¹⁴C) were the main freshwater input into the Dead Sea during its low-stand, while runoff input (with high ¹⁴C) was virtually shut down; and ii) the Dead Sea remnant brine that filled the deep basin after the strong retreat at 8.1 ka cal BP could have been isolated for several thousand years (Figure 1), causing additional decrease in ¹⁴C content due to decay. When the hydrological system recovered and runoff re-entered the lake, the RA began its continuous decrease towards the present value of 2.2 ka (Figure 2). This decrease is the result of continuous mixing of freshwater with atmospheric ¹⁴C with the Dead Sea brine. At ~3.3 ka cal BP, the Dead Sea level dropped again (Bookman et al. 2004; Migowski et al. 2004). Yet, we see no increase in the RA during the time interval of 4.1 to 1.1 ka cal BP. This may indicate that the threshold lake level is between 420 and 450 m bmsl (Figure 1): when the lake level is above this threshold, runoff is the main DIC supply of the lake; and when the lake level is below this threshold, springs (fresh and saline groundwater sources) are the main DIC source of the lake. Apparently, the level decrease at ~3.3 ka cal BP was not accompanied by long brine isolation that could support an additional increase in RA due to ¹⁴C decay.

During the Lisan time (70 to 14 ka BP), the RA was significantly lower than the Holocene value (Figure 1 and Table 1), suggesting that the main DIC input was runoff and the brine was never isolated for long periods of time from the mixed layer. The lake level during that period was always above the threshold level (Figure 1). The data shows that Lake Lisan had a zero RA value during the time interval that ended just before the lake reached its highest stand (Figure 1). This zero RA value was measured for the time interval that ends at the dating limit of the ¹⁴C method. The lake level at this period was stabilized at 280 m bmsl (Bartov et al. 2002), some 120 m above the present-day level of the Dead Sea. It is possible that the RA of Lake Lisan was zero before its highest stand due to fast replenishment of the surface water with runoff DIC (with high ¹⁴C content). The increase in lake area during the high-stand may have added "new" low 14C DIC sources. It is well documented that during the period of maximum stand (26–24 ka cal BP), Lake Lisan converged with the northern Sea of Galilee (Hazan et al., forthcoming). The high-stand was characterized by a stable layered configuration that persisted for several thousand years during which ¹⁴C content of the lower brine decreased. Thus, some contribution to RA in the mixed layer could come from the brine. In addition, the aguifers formed during the lake's highest stand may have supplied spring waters with low ¹⁴C content, which kept a low RA during the time after the highest stand.

CONCLUSIONS

The Holocene Dead Sea and late Pleistocene Lake Lisan were characterized by varying reservoir ages. In the Holocene Dead Sea, the RA ranges between 2.2 and 6 ka; and in the Lisan, they range from zero to 2.0 ka. These variations reflect the proportions of DIC supplied by freshwater from runoff and springs and periods of brine isolation and mixing with the surface (aragonite precipitating) layer.

The data of the last 36 ka of the lake suggest that the Lisan time was characterized by 2 modes of limnological operation and ¹⁴C content: (1) relatively low lake levels, when fast replenishment of the mixed layer occurred, were characterized by zero RA; (2) long high-stand periods, when the lake

developed a stable 2-layer configuration that allowed ¹⁴C decay in the lower (brine) layer, and particularly, when areas with enhanced spring activity contributed old carbon to the lakes.

The Holocene low-stand showed the highest RA (6 ka) at lake level lower than about 440 m, indicating that saline springs became an important low ¹⁴C DIC source for the lake.

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REFERENCES

- Barkan E, Luz B, Lazar B. 2001. Dynamics of the carbon dioxide system in the Dead Sea. Geochimica et Cosmochimica Acta 65:355–68.
- Bartov Y, Stein M, Enzel Y, Agnon A, Reches Z. 2002. Lake-levels and sequence stratigraphy of Lake Lisan, the Late Pleistocene precursor of the Dead Sea. *Quaternary Research* 57:9–21.
- Bartov Y, Goldstein SL, Stein M, Enzel Y. 2003. Catastrophic arid episodes in the Eastern Mediterranean linked with the North Atlantic Heinrich events. *Geology* 31:439–42.
- Begin ZB, Ehrlich A, Nathan Y. 1974. Lake Lisan, the Pleistocene precursor of the Dead Sea. *Bulletin* 63:1–30
- Bookman R, Enzel Y, Agnon A, Stein M. 2004. Late Holocene lake levels of the Dead Sea. *Geological Society of America Bulletin* 116.
- Haase-Schramm A, Goldstein SL, Stein M. 2004. U-Th dating of Lake Lisan aragonite (late Pleistocene Dead Sea) and implications for glacial East Mediterranean climate change. *Geochimica and Cosmochimica Acta* 68:985–1005.
- Hazan N, Stein M, Agnon A, Marco S, Nadel D, Schwab M, Negendank J, Neev D. Forthcoming. The late Pleistocene-Holocene history of the Sea of Galilee (Lake Kinneret). *Quaternary Research*.
- Luz B, Stiller M, Talma S. 1997. Carbon dynamics in the Dead Sea. In: Niemi TM, Ben-Avraham Z, Gat JR, editors. *The Dead Sea: The Lake and Its Setting*. Oxford Monographs on Geology and Geophysics, Nr 36, Oxford.
- Katz A, Kolodny N. 1989. Hypersaline brine diagenesis and evolution in the Dead Sea-Lake Lisan system (Israel). Geochimica et Cosmochimica Acta 53:59–67.
- Katz A, Kolodny Y, Nissenbaum A. 1977. The geochemical evolution of the Pleistocene Lake Lisan-Dead Sea system. Geochimica et Cosmochimica Acta 41:1609–26.
- Kaufman A. 1971. U-series dating of Dead Sea basin carbonates. Geochimica et Cosmochimica Acta 35:1269– 81
- Ken-Tor R, Stein M, Enzel Y, Agnon A, Marco S, Negendank J. 2002. Precision of calibrated radiocarbon ages

- of earthquakes in the Dead Sea basin. *Radiocarbon* 43(3):1371–82.
- Migowski C, Agnon A, Bookman R, Negendank J, Stein M. 2004. Recurrence pattern of Holocene earthquakes along the Dead Sea transform revealed by varve-counting and radiocarbon dating of lacustrine sediments. *Earth and Planetary Science Letters* 222:301–14.
- Neev D, Emery KO. 1967. The Dead Sea depositional processes and environments of evaporites. *Israel Geological Survey Bulletin* 41. 147 p.
- Prasad S, Vos H, Waldmann N, Goldstein SL, Stein M. Forthcoming. Evidence from Lake Lisan of solar influence on decadal to centennial scale climate variability during MIS 2. Geology.
- Schramm A, Stein M, Goldstein SL. 2000. Calibration of the ¹⁴C timescale to >40 ka by ²³⁴U-²³⁰Th dating of Lake Lisan sediments (Last Glacial Dead Sea). Earth and Planetary Science Letters 175:27–40.
- Stein M. 2001. The sedimentary and geochemical record of Neogene-Quaternary water bodies in the Dead Sea basin—inferences for the regional paleoclimatic history. *Journal of Paleolimnology* 26:271–82.
- Stein M. 2002. The fall and rise of the Dead Sea during the post-glacial and the Younger Dryas event. Geochimica Cosmochimica Acta 12th Annual Goldschmidt Conference. Dayos, A738.
- Stein M, Starinsky A, Goldstein SL, Katz A, Machlus M, Schramm A. 1997. Strontium isotopic, chemical, and sedimentological evidence for the evolution of Lake Lisan and the Dead Sea. Geochimica et Cosmochimica Acta 61:3975–92.
- Stiller M, Chung YC. 1984. Radium in the Dead Sea: a possible tracer for the duration of meromixis. *Limnology and Oceanography* 29:574–86.
- Talma AS, Vogel JC, Stiller M. 1997. The radiocarbon content of the Dead Sea. In: Niemi TL, Ben-Avraham Z, Gat J, editors. The Dead Sea: The Lake and Its Setting. Oxford: Oxford University Press. p 171–83.
- van der Borg K, Stein M, de Jong AFM, Waldmann N, Goldstein SL. 2004. Atmospheric ¹⁴C at 33 ka close to the AD 1950 level as observed in the high-resolution radiocarbon record from U-Th dated sediment of Lake Lisan. *Radiocarbon*, these proceedings.