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THE EFFECT OF ATMOSPHERIC ^{14}C VARIATIONS ON THE ^{14}C LEVELS IN THE JORDAN RIVER SYSTEM

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ABSTRACT. ^{13}C and ^{14}C were measured in the upper Jordan River, in its tributaries which issue from the Mount Hermon aquifer, and in the local atmospheric CO_2 to evaluate the degree to which changes in the atmospheric ^{14}C levels influence the ^{14}C levels in the aquifer and in the Jordan River. The exchange fraction of CO_2 between the river and the atmosphere was calculated for fall 1983 with the two carbon isotopes and it is shown that the value obtained with ^{14}C (0.364) is the more reliable. The ratio of ^{14}C in the Jordan River to that in the atmosphere in 1972 is similar to that in 1983, 0.66 and 0.67, respectively. This implies an approximately constant ratio (0.44) between the ^{14}C level in the Mount Hermon aquifer base-flow and the ^{14}C level in the atmospheric CO_2 . This information can be combined with the known historical fluctuations in the ^{14}C levels of the atmosphere to calculate the ^{14}C fluctuations in the discharge of the Jordan River into Lake Kinneret, backwards in time.

INTRODUCTION

First described by Deevey, Gross, and Kraybill (1954), the hard-water effect in the dating of lake sediments with ^{14}C , is now a well-known phenomenon. The magnitude of this effect remains constant only under stable hydrologic and isotopic conditions. Changes in atmospheric ^{14}C levels as has occurred dramatically since the 1950's because of nuclear testing and in more subdued form throughout the last 8000 years (Clark, 1975; Suess, 1980) can alter the magnitude of the hard-water effect. This alteration, which must be corrected for if valid ^{14}C ages are to be calculated, occurs in several steps. We shall deal with the first two of these: 1) that which occurs in the aquifer prior to the emergence of the bicarbonate in a spring and which is driven by the need for a new balance in ^{14}C levels between the basic aquifer rocks and the changing ^{14}C levels in the dissolved CO_2 ; and 2) that which occurs afterwards in the river that feeds the lake, as the emerged bicarbonate now undergoes exchange with the contemporaneous ^{14}C levels in atmospheric CO_2 . Changes in the magnitude of the hard-water effect caused by processes other than these two will be discussed in a forthcoming article (Kaufman & Stiller, ms in preparation).

In this paper we investigate these first two steps in the Jordan River system north of Lake Kinneret. ^{13}C concentrations and parameters affecting the carbonate equilibrium—pH, alkalinity, and temperature, have also been considered in our investigation.

The Jordan River, one of the principal sources of water for Israel, arises at the confluence of three tributaries fed by the three perennial springs, Snir, Dan, and Hermon (fig 1). The three springs issue from karstic aquifers which are in part calcitic (Dan and Hermon), and in part dolomitic

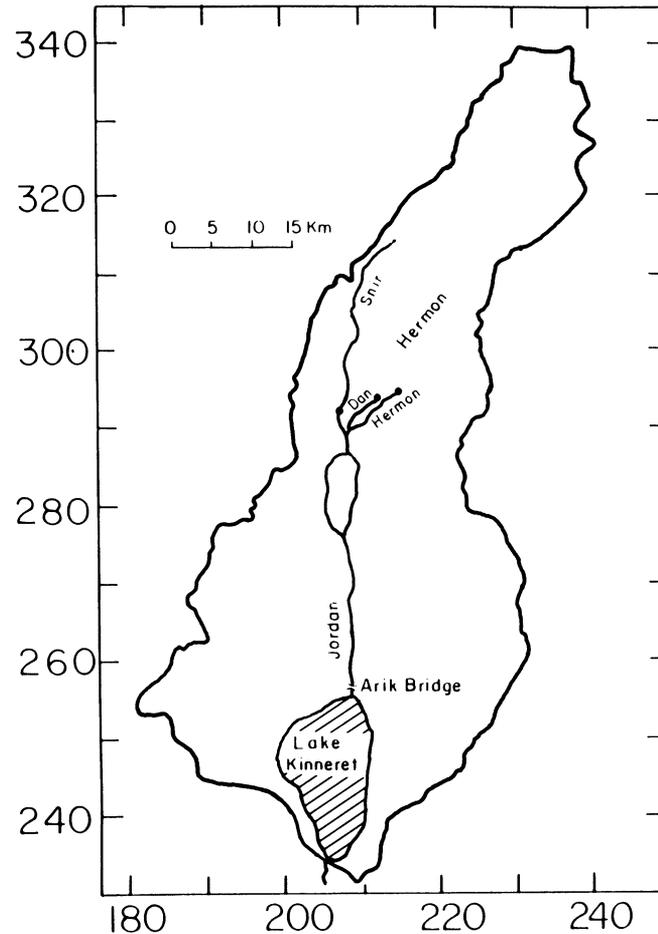


Fig 1. Map of Lake Kinneret watershed with the Jordan River and its tributaries. The Dan and Hermon springs emerge on the southwestern slope of Mt Hermon and the Snir spring emerges on the western slope (Lebanon). The sampling points of the tributaries are shown by (●). Below the confluence of the tributaries, the Jordan River was diverted into two channels for swamp drainage.

(Snir) (Michelson, 1975). Hydrographic and isotopic (tritium and ^{18}O) investigations have shown (Simpson & Carmi, 1983) that the residence time of water in the aquifers that feed these springs is less than 4 yr. The discharge from the Snir and Hermon springs also shows an immediate response to rainfall, but such a response is absent in the Dan spring. The distance between the confluence of the three tributaries and the discharge of the Jordan River into Lake Kinneret is ca 36km.

SAMPLING AND MEASURING TECHNIQUES

Water samples were collected from the three tributaries of the Jordan River and from the river itself (fig 1). The sampling points of the Dan and

the Hermon were very close to their emergence, and the Snir was sampled 34km downstream from its emergence point. The Jordan River was sampled at Arik bridge—near the point of discharge into Lake Kinneret. An atmospheric CO_2 sample was collected at Rehovot, ca 150km south of the upper Jordan River region.

Temperature, pH, and titration alkalinity of the samples were measured on site to a precision of 0.1°C, 0.1 pH units, and 0.1 mmole/L, respectively. Precipitation of BaCO_3 for ^{13}C analysis of the total dissolved carbonate (ΣC) was carried out on site in duplicate, on 1L samples. $\delta^{13}\text{C}$ was measured on a Varian MAT-250 mass spectrometer with an analytical error of $<0.1\text{‰}$; duplicates agreed within the analytical error. For ^{14}C analysis, total dissolved carbonate was precipitated as BaCO_3 from 50L of water within one day after sampling. The ^{14}C activity of the BaCO_3 was measured as described by Carmi, Noter, and Schlesinger (1971). $\Delta^{14}\text{C}_{\Sigma\text{C}}$ was calculated using $\delta^{13}\text{C}$ values measured on the CO_2 prepared for the ^{14}C measurements (Broecker & Olson, 1961).

The values of $\delta^{13}\text{C}_{\text{HCO}_3^-}$ were calculated after accounting for the pH and temperature-dependent distribution of dissolved carbonate among its various species (Riley & Skirrow, 1965) and the isotopic fractionation, ϵ , between them (Mook, 1968; Mook, Bommerson, & Staverman, 1974). The values of $\Delta^{14}\text{C}_{\text{HCO}_3^-}$ were calculated in a similar fashion, considering that the ^{14}C fractionation is twice as large as the ^{13}C fractionation. Despite the recent experimental evidence that these two may differ by a factor of 2.3 (Saliège & Fontes, 1984), the factor of 2 was used for consistency with the conventional calculation of $\Delta^{14}\text{C}$.

RESULTS

Table 1 presents the results of measurements performed on samples collected in 1983 from the Jordan River and its tributaries, and older data from 1971–72 (Stiller, 1974). The ^{14}C level of atmospheric CO_2 measured in 1983 is also given in table 1.

The winter temperatures of the three springs are generally similar at 15°C; but the summer temperature of the Snir tributary at the sampling station is appreciably higher (by 2°C) than that of the other two tributaries because of its exposure to high ambient temperatures during the 34km flow from the emergence point to the sampling station. Similarly, the water of the Jordan River downstream at Arik bridge, after its 36km journey (fig 1) is always warmer than the tributaries.

Each of the three tributaries has its own characteristic pH which does not change appreciably throughout the year. In 1983–84, the pH at Arik bridge was somewhat higher than the pH's of its tributaries. Why the measured pH at Arik bridge in 1971–72 was lower than in 1983–84 is not yet understood.

The relatively high ΣC values in the tributaries (3.3–4.0 mmole/L) reflect the karstic nature of the aquifers that feed them: calcitic for the Dan and Hermon, and dolomitic for the Snir. The weighted average of total dissolved carbon in the three springs is close, within the precision of the measurements, to the value measured at Arik bridge. This suggests that losses

TABLE 1
Isotopic, chemical, and physical data on the Jordan River, its tributaries, and in Rehovot atmosphere

No.	Sample	Date	Discharge $\frac{10^6 m^3}{mon}$	Temp °C	pH	$\frac{\Sigma C}{L}$ <i>mmole</i>	$\delta^{13}C$ ΣC ‰	$\delta^{13}C$ HCO_3^- ‰	$\Delta^{14}C$ ΣC ‰	$\Delta^{14}C$ HCO_3^- ‰
a. Dissolved inorganic carbon										
643B	Snir	2/17/83	23.4	14.8	8.3	4.0	-10.0	-9.9	-201 ± 5	-200
661C		10/19/83	4.0	19.3	8.3	3.5	-9.6	-9.4	-186 ± 17	-186
643A	Dan	2/17/83	8.7	15.8	7.8	3.9	-10.7	-10.3	-434 ± 13	-433
661A		10/19/83	13.3	17.2	7.8	3.3	-10.0	-9.7	-446 ± 12	-445
643C	Hermon	2/17/83	12.8	15.5	7.5	3.5	-11.2	-10.3	-501 ± 5	-499
661B		10/19/83	5.3	16.5	7.3	3.7	-9.6	-8.3	-454 ± 12	-451
371A	Jordan*	12/7/71		17.0	7.7	3.8	-7.1	-6.7	-23 ± 23	-22
		7/22/72		27.0	7.8	3.2	-6.3	-6.0		
		8/23/72		28.0	7.5	3.4	-7.0	-6.4		
661D		10/19/83		22.0	8.5	3.4	-7.4	-7.3	-166 ± 17	-166
b. Atmospheric CO ₂										
651	Rehovot	6/19/83					-10.2	-1.7	+225 ± 25	+242

*Sampled at Arik bridge

or gains of carbon during the flow of the river in 1983 are small. At Arik bridge, no significant variations in ΣC are observed between 1971–72 and 1983, indicating that no greater changes in the carbon balance of the river have taken place in this interval.

The range of $\delta^{13}\text{C}$ values in the three springs is rather small (-11‰ to -9‰). In the Hermon spring, there is an indication that the base flow (represented by October 1983) is more enriched in ^{13}C than is the winter flow (represented by February 1983) which includes the response to rainfall. The bicarbonate at Arik bridge is significantly enriched in ^{13}C compared to the tributaries (by 2‰ to 3‰), and this suggests that exchange with atmospheric CO_2 takes place during the river's flow.

$\Delta^{14}\text{C}$ has distinct values for each of the three tributaries. The discharge of the Dan spring does not respond directly to rainfall and, in fact, no seasonal variation is observed in its ^{14}C levels, which, therefore, represent the baseflow of the aquifer in 1983. In the Hermon spring, the ^{14}C level at the end of the summer is very similar to that of the Dan. This seems reasonable because the baseflows of both springs have the same age distribution. Surprisingly, however, the ^{14}C level in winter 1983 (when the Hermon's monthly discharge has increased up to five-fold by the addition of fresh rain) is distinctly *lower* than that of the summer baseflow with its *older* water. Both of these waters must, therefore, have fallen as precipitation in the unusual period since 1964 when older water had more ^{14}C than did younger water because of the dissipation of the nuclear tests peak (Segl *et al.*, 1983). This constraint on the aquifer residence time (<20 yr) supports the <4 yr residence time discovered independently (Simpson & Carmi, 1983) using tritium. In the Snir, the ^{14}C levels are higher than in the other two springs and no seasonal variation is observed. This indicates that considerable exchange with atmospheric CO_2 occurs between the emergence point of the Snir and our sampling point which is 34km downstream; this exchange apparently masks any minor seasonal variations that may occur in the ^{14}C levels of the spring. The ^{14}C at Arik bridge is enriched compared to the tributaries, which shows that exchange with atmospheric CO_2 significantly affects the levels of ^{14}C in the Jordan River, as observed above for ^{13}C .

DISCUSSION

The carbon isotope data will now be applied to the two modes of change in hard-water effect mentioned in the Introduction: 1) the relationship between changing atmospheric ^{14}C levels and those in the discharge at the spring, which results from processes occurring within the aquifer, and 2) the relationship between changing atmospheric ^{14}C levels and those in the Jordan River which takes place during its flow to Lake Kinneret. The conclusions drawn will be integrated for an estimate of the historic levels of ^{14}C in the Jordan River and its tributaries. We begin by discussing the exchange between the Jordan River and the atmosphere.

The exchange fraction of CO_2 between the atmosphere and the flowing Jordan River, E , can, in principle, be estimated from either ^{13}C or ^{14}C

data as follows:

$$E = \frac{X_{jrd} - X_{trb}}{X_{atm} - X_{trb}} \quad (1)$$

where X is the isotopic composition (either in $\delta^{13}\text{C}$, $\Delta^{14}\text{C}$ units or in concentration units) and the subscripts are: *jrd*–Jordan, *trb*–the combined tributaries, and *atm*–atmosphere.

Of the three terms used to calculate E in the upper Jordan River (Eq 1) two, X_{jrd} and X_{trb} , were obtained by sampling of the Jordan River and its tributaries in October 1983, while the third, X_{atm} , was inferred from an atmospheric CO_2 sample collected 150km away (table 1). The $\Delta^{14}\text{C}$ of this latter sample fits the generally accepted trend of atmospheric ^{14}C levels quite well, but the applicability of its $\delta^{13}\text{C}$ value is far less clear because of the large variability in atmospheric $\delta^{13}\text{C}$ values (Keeling, 1961) which was found to depend on location, plant cover, and exact time of sampling. The effect of this variability on the two carbon isotopes may be demonstrated by considering the extreme case: if X_{atm} is overwhelmed by local plant respiration (with -18‰ fractionation in $\delta^{13}\text{C}$ between the plants and the atmosphere and twice this value for $\Delta^{14}\text{C}$). Here, the effect on the value of $\Delta^{14}\text{C}_{atm}$ would be $<6\%$ of the difference $\Delta^{14}\text{C}_{atm} - \Delta^{14}\text{C}_{trb}$ (Eq 1) measured on 1983 and, therefore, relatively insignificant (the values of X_{trb} are given below). With $\delta^{13}\text{C}$, on the other hand, the corresponding effect would be much larger than the difference $\delta^{13}\text{C}_{atm} - \delta^{13}\text{C}_{trb}$, itself, and therefore very significant. The atmospheric sample was collected far from the Jordan River, during midday and under a completely different plant cover, and it is therefore difficult to extrapolate its $\delta^{13}\text{C}$ value to the atmosphere above the Jordan River. For this reason, although both isotopes qualitatively lead to the same conclusions, we consider the calculated value of ^{14}E to be more reliable than the calculated value of ^{13}E .

In applying Eq 1 to our isotopic measurements, we must remember that dissolved inorganic carbon is distributed among several species, depending on the pH and the temperature of the water, and that each of these species has a distinct isotopic composition determined by the temperature-dependent fractionation between them. The bicarbonate ion is the dominant constituent of the dissolved carbon pool and we used its isotopic composition to follow the exchange of CO_2 between the atmosphere and the river. The isotopic content of the upper Jordan River in fall 1983 at the confluence of the tributaries, X_{trb} , was calculated as a weighted average, the weights being the respective discharges given in table 1. The values obtained for the bicarbonate component are $\delta^{13}\text{C}_{trb} = -9.49\text{‰}$ and $\Delta^{14}\text{C}_{trb} = -400\text{‰}$. From these values, the data for Arik bridge (X_{jrd}) and the atmosphere (X_{atm}) (table 1) we calculate from Eq 1: $^{14}\text{E} = 0.364 \pm 0.023$ and, for the sake of completeness, $^{13}\text{E} = 0.287 \pm 0.019$.

The errors in ^{14}E and ^{13}E , both ca 6.5‰ , were based on analytical errors of 0.1‰ and 10‰ for $\delta^{13}\text{C}$ and $\Delta^{14}\text{C}$, respectively. The former are similar though the latter differ by a factor of 100 because $\Delta^{14}\text{C}_{atm} - \Delta^{14}\text{C}_{trb}$ is ca 80 times greater than $\delta^{13}\text{C}_{atm} - \delta^{13}\text{C}_{trb}$. The discrepancy between ^{14}E and ^{13}E is greater than can be accounted for by the analytical uncertainties and

considering the lower reliability that we ascribe to ^{13}E , we shall restrict all further discussion to ^{14}E .

From our estimate of E, it is possible to calculate the exchange rate along the Jordan River, using the equation of Broecker and Walton (1959), and the stagnant film thickness after Broecker *et al.*, (1980). The relevant parameters and the results are given in table 2. The results are only useful for rough comparison because they are derived values which vary with time and from one part of the river to another. Thus, our 240 ± 30 moles $\text{m}^{-2}\text{yr}^{-1}$ estimate for the CO_2 exchange rate for the Jordan River may be considered to be in good agreement with the 103 ± 50 obtained by Broecker and Walton (1959) for the Truckee River of Nevada (the quoted uncertainties are based on 1σ counting errors). No other estimates of the stagnant film thickness in rivers were found with which to compare our result (table 2). As can be expected for the smaller turbulence, the exchange rates in lakes and oceans are smaller and the stagnant film thickness is much larger (Broecker *et al.*, 1980) than they are in rivers.

The distance between the Snir's emergence point and the confluence point, 34km, is similar to the distance between the confluence point and Arik Bridge, 36km. If similar flow characteristics, *ie*, ratio of average depth to transit time, are assumed for both flow channels, we may use the ^{14}E value obtained for the Jordan River and the October 1983 ^{14}C level of the Snir at the sampling point, to compute $\Delta^{14}\text{C} = -433\text{‰}$ at the emergence point of the Snir. This is a plausible value because of its similarity to the baseflow data at the emergence points of the other two tributary springs in October 1983 (table 1).

We may also combine the 0.364 value of E, computed from the October 1983 data, with the July 1972 $\Delta^{14}\text{C}$ values at Arik bridge (table 1) and the $\Delta^{14}\text{C}_{\text{atm}} = 485\text{‰}$ of the 1972 atmosphere (Segl *et al.*, 1983), to compute the $\Delta^{14}\text{C}$ of the combined tributaries for the baseflow of 1972. The result $\Delta^{14}\text{C}_{\text{trb}} = -315\text{‰}$ is what one would expect because it is higher than the

TABLE 2
Parameters needed to calculate exchange rate of CO_2 and stagnant film thickness in the Jordan River

<i>The CO_2 exchange rate</i>	
Weighted mean ΣCO_2	3.4 mole m^{-3}
Mean depth of river*	0.78 to 0.87m
Transit time	5.3×10^{-3} yr
Exchange rate of CO_2 **	$(226 \text{ to } 253) \pm 30$ moles $\text{m}^{-2}\text{yr}^{-1}$
<i>The stagnant film thickness</i>	
Diffusion coefficient of CO_2 †	$(1.54 \text{ to } 1.74) \times 10^{-5}$ $\text{cm}^2 \text{sec}^{-1}$
Dissolved gaseous CO_2	$(19.3 \text{ to } 2.4) \times 10^{-8}$ moles cm^{-3}
Exchange rate of CO_2 (average)	240 moles $\text{m}^{-2}\text{yr}^{-1}$
Stagnant film thickness	0.4 to 5.2 μ

*Determined for two points along the river's course (Inbar 1977).

**Calculated by Eq 2 of Broecker and Walton (1959)

†In each case, the first value given corresponds to the physico-chemical conditions at the confluence of the tributaries and the second to those at Arik Bridge (36km southwards).

TABLE 3
 ^{14}C in the Jordan River system and its relationship with atmospheric levels
 (m—measured, c—calculated values)

Date	X_{atm}	X_{aquifer}	X_{trb}	X_{jrd}	X_{aquifer}	X_{trb}	X_{jrd}
					X_{atm}	X_{atm}	X_{atm}
10/83	1.242 _m	0.598 _m *	0.598 _m	0.834 _m	0.443	0.482	0.672
7/72	1.487 _m	0.608 _c	0.685 _c	0.978 _m	0.409	0.461	0.657
Pre-bomb	≈1.00	0.43 _c	0.48 _c	0.67 _c			

*Average of the Dan and the Hermon

$\Delta^{14}\text{C}_{\text{trb}}$ of 1983 in about the same proportion that $\Delta^{14}\text{C}_{\text{atm}}$ of 1972 is higher than that of 1983.

Now that the estimate of $E = 0.364$ was found to be consistent with the various aspects of our data, we may proceed to undertake our original task, that of deriving a relationship between X_{jrd} and X_{atm} . By substituting for E in Eq 1 and rearranging:¹

$$\frac{X_{\text{jrd}}}{X_{\text{atm}}} = 0.364 + 0.636 \frac{X_{\text{trb}}}{X_{\text{atm}}} \quad (2)$$

This equation permits us to get X_{jrd} from X_{atm} only if the relationship between the latter and X_{trb} is known. From our measurements, we have two estimates of the ratio $X_{\text{jrd}}/X_{\text{atm}}$, 0.66 and 0.67 in 1972 and 1983, respectively. The fact that these two ratios are so similar though X_{atm} has decreased by 20% in the meantime suggests that the ratio $X_{\text{trb}}/X_{\text{atm}}$ is approximately constant. That this, in fact, is the case is demonstrated by the observation that 1) the measurements of X_{atm} and X_{trb} in 1983 yield $X_{\text{trb}}/X_{\text{atm}} = 0.48$; 2) the measurements of X_{jrd} and X_{atm} in 1972 together with Eq 2 yield $X_{\text{trb}}/X_{\text{atm}} = 0.46$ (table 3). The reason for this near constancy is that the value of this ratio is determined primarily by the extent to which CO_2 bearing ^{14}C -rich recharge waters are chemically neutralized by the basic ^{14}C -free aquifer rock; apparently, this process is reproducible regardless of the atmospheric ^{14}C level.

We now use the ratios found above to calculate the pre-bomb ($\Delta^{14}\text{C}_{\text{atm}} = 0$) ^{14}C levels of the Jordan River at Arik bridge, and of the combined tributaries: $\Delta^{14}\text{C}_{\text{jrd}} = -330\text{‰}$ and $\Delta^{14}\text{C}_{\text{trb}} = -518\text{‰}$. The baseflow of the Mount Hermon aquifer is expected to have a somewhat lower ^{14}C activity than that of the combined tributaries. This is because of exchange with atmospheric $^{14}\text{CO}_2$ during the flow of the Snir from its emergence to confluence with the other two tributaries. We can, however, estimate the pre-bomb ^{14}C level in the baseflows of the aquifers that feed the springs as follows: we express $\Delta^{14}\text{C}_{\text{trb}}$ as an average of the $\Delta^{14}\text{C}$ values of the three springs, weighted by their respective estimated discharge fractions of 0.25 for the Snir, 0.25 for the Hermon, and 0.50 for the Dan. The ^{14}C contributions of the Hermon and the Dan are taken to be $\Delta^{14}\text{C}_{\text{aquifer}}$ but that of the Snir has to

¹Note that for ratios between X's, concentration units must be used:

$$X_i = (1000 + \Delta^{14}\text{C}_i)/1000.$$

be corrected, by use of Eq 1 with $^{14}\text{E} = 0.364$, for exchange with the atmosphere between the emergence of this spring and the confluence. Rearranging the expression for $\Delta^{14}\text{C}_{\text{trb}}$, we get $\Delta^{14}\text{C}_{\text{aquifer}} = \Delta^{14}\text{C}_{\text{trb}} - 0.25^{14}\text{E}\Delta^{14}\text{C}_{\text{atm}}/1 - 0.25^{14}\text{E}$, which gives for $\Delta^{14}\text{C}_{\text{atm}} = 0\text{‰}$, $\Delta^{14}\text{C}_{\text{aquifer}} = -570\text{‰}$ or $X_{\text{aquifer}}/X_{\text{atm}} = 0.43$ (table 3). The ratio $X_{\text{aquifer}}/X_{\text{atm}}$, which is fairly constant (table 3: note that in October 1983 it is based on actual measurements), means that the initial ^{14}C activity of the regional aquifer is $\approx 0.44X_{\text{atm}}$. This is apparently a characteristic feature of the Mount Hermon karstic aquifer, and can be used as initial ^{14}C activity for groundwater dating in the region.

Since $\Delta^{14}\text{C}_{\text{atm}}$ of past millennia is known quite well (Clark, 1975), our final conclusion is that we are now in a position to compute the ^{14}C levels of the Jordan River at its discharge into Lake Kinneret for the last 8000 yr, for a better understanding of the hard-water effect in this lake (Kaufman & Stiller, ms in preparation).

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