GEochronology VIII: Cosmogenic Nuclides

Introduction

As the name implies, cosmogenic nuclides are produced by cosmic rays, specifically by collisions with atoms in the atmosphere (and to a much lesser extent, the surface of the solid Earth). Nuclides so created may be stable or radioactive. Radioactive cosmogenic nuclides, like the U decay series nuclides, have half-lives sufficiently short that they would not exist in the Earth if they were not continually produced. Assuming that the production rate is constant through time, then the abundance of a cosmogenic nuclide in a reservoir isolated from cosmic ray production is simply given by:

\[ N = N_0 e^{-\lambda t} \]

Hence if we know \( N_0 \) and measure \( N \), we can calculate \( t \). Table 12.1 lists the radioactive cosmogenic nuclides of principal interest. As we shall see in the next lecture, cosmic ray interactions can also produce rare stable nuclides, and their abundance can also be used to measure geologic time.

Cosmogenic nuclides are created by a number of nuclear reactions with cosmic rays and by-products of cosmic rays. “Cosmic rays” are high energy (several GeV up to \( 10^{19} \) eV) charged particles, mainly protons, or H nuclei (since, after all, H constitutes most of the matter in the universe), but nuclei of all the elements have been recognized. Only in the last 20 years or so has it been possible to achieve energies as high as several GeV in accelerators: cosmic rays have been a useful source of high energy particles for physicists for decades (the Cornell Electron Storage Ring produces energies up to 15 GeV). A significant fraction originates in the Sun, though these are mainly of relatively low energies. The origin of the remainder is unclear; it is thought that many may originate in supernovae or similar high-energy environments in the cosmos.

The cosmic ray flux decreases exponentially with depth in the atmosphere as these particles interact with matter in the atmosphere. This observation has an interesting history. Shortly after the discovery of radioactivity, investigators noticed the presence of radiation even when no known sources were present. They reasonably surmised that this resulted from radioactivity in the Earth. In 1910, an Austrian physicist named Victor Hess carried his detector (an electroscope consisting of a pair of charged gold leaves; the leaves would be discharged and caused to collapse by the passage of charged particles) aloft in a balloon. To his surprise, the background radiation increased as he went up rather than decreased! It thus became clear that cosmic rays originated from outside, rather than inside, the Earth.

The primary reaction that occurs when cosmic rays encounter the Earth is spallation, in which a nucleus struck by a high energy particle shatters into a number of pieces, including stable and unstable nuclei, as well as protons and neutrons. Unstable particles such as muons, pions, etc. are also created. The interaction of a cosmic ray with a nucleus sets of a chain reaction of sorts as the secondary particles and nuclear fragments, which themselves have very high energies, then strike other nuclei producing additional reactions of lower energy. \(^{14}\text{C}\) is actually produced primarily by reactions with secondary particles, mainly by the \(^{14}\text{N}(n,p)^{14}\text{C}\) reaction involving relatively slow neutrons.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Half-life, years</th>
<th>Decay constant, ( \text{y}^{-1} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>(^{14}\text{C})</td>
<td>5730</td>
<td>(1.209 \times 10^{-3})</td>
</tr>
<tr>
<td>(^{3}\text{H})</td>
<td>12.33</td>
<td>(5.62 \times 10^{-3})</td>
</tr>
<tr>
<td>(^{10}\text{Be})</td>
<td>(1.500 \times 10^{6})</td>
<td>(4.62 \times 10^{-3})</td>
</tr>
<tr>
<td>(^{26}\text{Al})</td>
<td>(7.16 \times 10^{3})</td>
<td>(9.68 \times 10^{-3})</td>
</tr>
<tr>
<td>(^{30}\text{Cl})</td>
<td>(3.08 \times 10^{5})</td>
<td>(2.25 \times 10^{-3})</td>
</tr>
<tr>
<td>(^{28}\text{Si})</td>
<td>276</td>
<td>(2.51 \times 10^{-2})</td>
</tr>
</tbody>
</table>
Carbon-14 is by far the most familiar and useful of the cosmogenic dating schemes. Its usefulness results from its relatively short half-life, a relatively high production rate, and the concentration of carbon in biological material. The short half-life has the advantage of producing accurate dates of young (geologically speaking) materials and events and easy determination by counting the decays. The traditional method of $^{14}$C determination is counting of the $\beta$ rays produced in its decay. $^{14}$C decays without emitting a gamma, which is unfortunate because $\gamma$-rays are more readily detected. Difficulties arise because the rate of decay of a reasonable sample of carbon (a few grams) is low. One of the first problems that had to be solved was eliminating “counts” that arose from cosmic rays rather than the decay of the carbon sample. This is done with a combination of shielding and “coincidence counting”.* Counting techniques make use of either liquid scintillation counters or gas proportional counters. A gas proportional counter is simply a metal tube containing a gas, with a negatively charged wire (the anode) running through it. The emitted beta particle causes ionization of the gas, and the electrons released in this way drift to the wire (the anode), producing a measurable pulse of current. In this technique, carbon in the sample is converted to CO$_2$, and purified (in some cases it is converted to methane) and then admitted into the counting chamber. Several grams of carbon are generally required. In liquid scintillation counting, the carbon is extracted from a sample and converted into CO$_2$, purified, and eventually converted to benzene (C$_6$H$_6$) and mixed with a liquid scintillator (generally an organic liquid). When a carbon atom decays, the beta particle interacts with the scintillator, causing it to give off a photon, which is then detected with a photomultiplier, and the resulting electrical pulse sent to a counter. Liquid scintillation is the new and more sensitive of these two techniques and it has now largely replace gas proportional counting. Liquid scintillation analysis can be done with less than 1 g of carbon.

In the last 15 years or so, a new method, accelerator mass spectrometry (AMS), has been used with considerable success. Greater accuracy is achieved with considerably less sample (analysis requires a few milligrams to 10’s of milligrams). While these instruments are expensive, there are now dozens of

\* Beta detectors are subject to electronic and other noise – in other words pulses that originate from something other than a beta particle. Coincidence counting is a technique that employs two detectors. Only those pulses that are registered on both detectors are counted – other pulses are considered noise.
them in the world. While most radiocarbon dating is still done by beta counting, use of AMS is rapidly increasing.

Because of the way in which it is measured, $^{14}$C is generally reported in units of specific activity, or disintegrations per minute per gram carbon rather than as a ratio to $^{12}$C or $^{13}$C (in SI units, 1 unit of specific activity is equal to 60Beq/g C). Atmospheric carbon, and carbon in equilibrium with the atmosphere (carbon in living plant tissue and the surface of the oceans) has a specific activity of 13.56 dpm/g. This is, in effect, the value of $N_o$ in equation 12.1.

The cosmic ray flux varies latitudinally because of the effect of the Earth’s magnetic field on the paths of charged particles. One would expect, then, the production rate of $^{14}$C to be higher at the poles, and this is indeed the case. However, because the mixing time of the atmosphere is short compared to the residence time of $^{14}$C in the atmosphere, the $^{14}$C concentration in the atmosphere is uniform. By historical convention, radiocarbon ages are reported in years before 1950 (the year the first $^{14}$C age determination; the abbreviation BP thus means not before present, but before 1950) and assuming a half-life of 5568 years (instead of the currently accepted value of 5730 years).

Can we really assume, however, that the atmosphere specific activity today is the same as in the past? From equation 12.1, we can see that knowing the initial specific activity is essential if an age is to be determined. To investigate the variation of the specific activity of $^{14}$C with time in the atmosphere, the specific activity of $^{14}$C in wood of old trees (dendrochronology) has been examined. The absolute age of the wood is determined by counting tree rings. The result of such studies shows that the specific activity has indeed not been constant, but has varied with time (Figure 12.1). There are a number of effects involved. Since 1945 injection of bomb-produced $^{14}$C into the atmosphere has raised the specific activity. Then, over the past hundred years or so, the specific activity has decreased because of addition of “old” (i.e., $^{14}$C-free) carbon in CO$_2$ from fossil fuel burning (the Suess effect). Similar variations occurred further back in time. Indeed, between 33,000 and 45,000 years ago, atmospheric specific activity appears to have been twice what it was in 1950 (Beck et al., 2001). These changes in atmospheric specific activity almost certainly occurred as a consequence of the changing balance between CO$_2$ in the atmosphere and the ocean. Far more CO$_2$ is dissolved in the ocean, particularly in deep water, than in the atmosphere. Shifting CO$_2$ from the atmosphere to the ocean increases the specific activity of $^{14}$C in the atmosphere. These shifts result from variations in (1) the volume of the oceans, (2) changes in ocean temperature (CO$_2$ is more soluble at lower temperature) and (3) changes in marine biological productivity. This last effect is likely the most important. These are termed reservoir effects. There are also several ‘excursions’ over the

![Figure 12.2. Conversion of radiocarbon dates to calendar years. The red curve at left shows the uncertainty histogram for a radiocarbon age of 3000 ± 30 (1σ) years BP. The set of blue curves shows the calibration between radiocarbon age and calendar date. The black histogram at the bottom shows the probably corresponding calendar date for this radiocarbon age.](image)
last 1000 years, known as the deVries events whose origin is unknown, but probably relates to variation in the cosmic ray flux. This results from combination of astronomical factors, including variation in the solar cosmic ray flux, supernovae events, etc., and variations in the geomagnetic field, which can deflect cosmic rays. In any case, a correction must be applied to $^{14}$C dates because of these variations. Calibration of $^{14}$C through dendrochronology has now been done over the last 11,000 years using waterlogged oaks in swamps and bogs of Germany and Ireland and bristlecone pines in the US and is being extended through comparison of $^{14}$C and $^{238}$U/$^{230}$Th ages, for example in corals. Computer programs are available for converting radiocarbon ages to calendar years, for example, the CALIB program available from the University of Washington (http://depts.washington.edu/qil/calib/).

All these factors make the conversion from radiocarbon ages to calendar years somewhat complex. Figure 12.2 illustrates the case of a hypothetical radiocarbon age of 3000 years BP with a 1 uncertainty of 30 years. On the left, is shown the uncertainty histogram in red. The pair of nearly parallel blue curves enclose the uncertainty on the calibration between radiocarbon age and calendar years. The black histogram on the bottom show the probable correspondence of calendar dates to this radiocarbon age. The irregularity of the histogram reflects the irregularity of the calibration curve, which in turn reflects the variation in reservoir effects, cosmic ray flux, etc. mentioned above. In this case, the 2 uncertainty encompasses a range of calendar dates from 1390 BC to 1130 BC.

Most of the applications of $^{14}$C dating are probably in archaeology, but geological applications include volcanology ($^{14}$C dating is an important part of volcanic hazard assessment), Holocene stratigraphy, and oceanography. It is also used to determine the time of prehistoric earthquakes, and thus part of earthquake hazard assessment. In oceanography, the age of bottom water (age meaning the time since the water last equilibrated with the atmosphere) can be determined with $^{14}$C. Typically, this age is of the order of 1000 yrs.

A more recent oceanographic application, which utilizes accelerator mass spectrometry, is the determination of paleo-bottom water ages. The ice ages affected ocean circulation, but the exact nature of the effect is still uncertain. Of particular interest are the changes in the ocean circulation pattern that occurred as the last glaciation ended. Because planktonic foraminifera live in the upper part of the ocean, which is in equilibrium with the atmosphere, one can date a sedimentary stratum by measuring $^{14}$C in planktonic foraminiferal tests. Benthic foraminifera live at the bottom of the ocean, and build their tests from CO$_2$ dissolved in bottom water. Thus a $^{14}$C date of a modern benthic foramin would give the age of the bottom water. By comparing the $^{14}$C ages of planktonic and benthic foraminifera, one may determine the “ages” of bottom waters in the past. This in turn reveals something of paleocirculation patterns.

$^{10}$Be, $^{26}$Al, and $^{36}$Cl

We now consider some of the other nuclides produced by cosmic ray interactions with atmospheric gases. These include $^{10}$Be, $^{26}$Al, and $^{36}$Cl. These nuclides have much longer half-lives than $^{14}$C and thus are be applied to longer time-scale problems, such as Pleistocene chronology and dating of groundwater. Of these, $^{10}$Be has been the most extensively utilized, for several reasons. First, its production rate is higher (10$^2$ – 10$^5$ atoms/cm$^2$/sec versus 10$^{-2}$–10$^6$ for $^{26}$Al and $^{36}$Cl). It also has the advantage over $^{36}$Cl that, once absorbed onto clays, it is relatively immobile, and that while a small amount of $^{36}$Cl will be produced within the earth by neutron capture on $^{35}$Cl (with neutrons arising from fission of U, as well as secondary neutrons produced by $(\alpha,n)$ reactions), there is effectively no internal ‘nucleogenic’ production of $^{10}$Be in the Earth.

$^{10}$Be is created by spallation reactions between cosmic rays and N and O nuclei. Since these are the most abundant nuclei in the atmosphere, the production rate of $^{10}$Be is comparatively high. $^{26}$Al is produced by spallation of $^{40}$Ar, and $^{36}$Cl is produced by $^{40}$Ar$(\alpha,n)$ reactions (probably mainly with secondary protons). Unlike carbon, Be, Cl and Al do not form gases under ambient conditions at the sur-
face of the Earth‡, so residence time of $^{10}$Be, $^{26}$Al, and $^{36}$Cl in atmosphere are quite short. Once they form, they are quickly extracted from the atmosphere by rain. Since we know the cosmic ray flux varies latitudinally, we might expect latitudinal variation in, for example, the $^{10}$Be production rate and flux to the surface of the Earth. Subareal variations $^{10}$Be show the expected latitudinal dependence. However, the distribution in oceans is uniform because $^{10}$Be spends sufficient time in oceans that its distribution is homogenized (the residence time of Be is somewhat uncertain, but seems to be about 4000 years). Be is readily scavenged and absorbed by clay particles, both subareally and in the oceans. The concentration in soils is quite high. In the oceans $^{10}$Be is extracted from seawater by adsorption on suspended clay particles. Because of its low activity, $^{10}$Be is analyzed primarily by accelerator mass spectrometry. The applications include dating marine sediments, paleosols, Mn nodules, etc.

Let’s now consider some examples of how these nuclides can be used for geochronology. We will begin with $^{10}$Be dating of sediments and they go on to show how ages can be improved if we use both $^{10}$Be and $^{26}$Al. We will then consider how $^{36}$Cl and bomb-produced $^3$H are used in hydrology.

Let’s now see how $^{10}$Be can be used for age determination. The relevant equation is

$$^{10}\text{Be} = ^{10}\text{Be}_0 e^{-\lambda t}$$  \hspace{1cm} (12.2)

We assume the rate of deposition of $^{10}$Be in the ocean depends only on the production rate, hence,

$$\left[^{10}\text{Be}\right] \propto \Phi e^{-\lambda t}$$  \hspace{1cm} (12.3)

where $\Phi$ is the production rate. The concentration of $^{10}$Be in a given amount of sediment may nonetheless vary because it can be variably diluted depending on how fast other components in the sediment are deposited. In other words, we must also consider the sedimentation rate. The amount of $^{10}$Be in a given amount of sediment then becomes:

$$\left[^{10}\text{Be}\right] = \frac{\Phi}{a} e^{-\lambda t}$$  \hspace{1cm} (12.4)

where $a$ is the sedimentation rate. If sedimentation rate is constant, then the depth, $d$, of a given horizon in the sediment is given by

$$d = a \times t$$  \hspace{1cm} (12.5)

and

$$t = \frac{d}{a}$$  \hspace{1cm} (12.6)

We can substitute equ. 12.6 into 12.4 to obtain

$$\left[^{10}\text{Be}\right] = \frac{\Phi}{a} e^{-\lambda d/a}$$  \hspace{1cm} (12.7)

Taking the log of both sides, we have:

$$\ln\left[^{10}\text{Be}\right] = \ln\left(\frac{\Phi}{a}\right) - \frac{\lambda}{a} d$$  \hspace{1cm} (12.8)

From this we can see that the slope of a plot of $\ln[^{10}\text{Be}]$ vs. depth will be inversely related to sedimentation rate.

In a more general case, the sedimentation rate will not be constant, but will be a function of time. A general equation will then be

‡ In the laboratory, elemental chlorine is, of course, a gas, but it is so reactive that it quickly reacts with electropositive atoms (e.g., alkalies, metals) in nature to form chlorides.
10Be = \frac{\Phi}{a(t)} e^{-\lambda d/a} \quad 12.9

In a sedimentary sequence such as a piston core, we do not necessarily know \( t \) (indeed, that is what we wish to determine), but we do know the depth in the core, which we know is some function of time. It is convenient then to transform equation 12.8 to a function of depth rather than time:

\[ 10Be(d) = \frac{\Phi}{a(d)} e^{-\lambda d} \quad 12.10 \]

The sedimentation rate \( a \) is simply

\[ a = \frac{d(d)}{dt(d)} \quad 12.11 \]

Substituting for \( a \) and integrating, we obtain:

\[ \int_0^d [10Be](d) \, d(d) = -\frac{\Phi}{\lambda} (e^{-\lambda d} - 1) \quad 12.12 \]

This equation in turn can be solved for \( t(d) \):

\[ t(d) = -\frac{1}{\lambda} \ln \left[ 1 - \frac{\Phi}{\lambda} \int_0^d [10Be](d) \, d(d) \right] \quad 12.13 \]

The integral is simply the sum of the activity of 10Be down the core to any depth \( d \).

This method has been used successful to date sediments. However, since there are a number of ways of determining the age of young marine sediments, a more interesting application is determination of growth rates of manganese nodules, for which age determination is quite difficult.

Somewhat more accurate dates may be achieved when two cosmogenic nuclides are used. For example, 10Be and 26Al. In this case we can write two equations:

\[ ^{10}Be = ^{10}Be_0 e^{-\lambda t} \quad 12.2 \]
\[ ^{26}Al = ^{26}Al_0 e^{-\lambda t} \quad 12.13 \]

Dividing one by the other, we obtain

\[ \frac{^{26}Al}{^{10}Be} = \left( \frac{^{26}Al}{^{10}Be} \right)_0 e^{(\lambda_{26}-\lambda_{10})t} \quad 12.14 \]

The advantage of this approach is that the initial \(^{26}Al/^{10}Be\) ratio should be independent of the cosmic ray flux.

**Cosmogenic and Bomb-Produced Radionuclides in Hydrology**

Determining the age of water in underground aquifers is an important problem because of the increasing demands placed in many parts of the world on limited water resources. A prudent policy for water resource management is to withdraw from a reservoir at a rate no greater than the recharge rate. Determination of recharge rate is thus prerequisite to wise management. Cosmogenic radionuclides are swept out of the atmosphere by rain and into the groundwater system. If we know the concentration of a radionuclide in rainwater, \( N_0 \), and if we can assume that it is not produced within the Earth or lost from solution (this is our closed system requirement), then the ‘age’ of water in an aquifer is determined simply from equation 12.1, where we define ‘age’ as the time since the water left the atmosphere and entered the groundwater system.
In addition to cosmogenic production, $^3$H and other radionuclides were produced in significant quantities during atmospheric thermonuclear tests between 1952 and 1963. The pulse of $^3$H in precipitation during this period is illustrated in Figure 12.3. Tritium is readily detected by beta counting, and therefore bomb-produced tritium is potentially useful for tracing precipitation that fell during the 1950’s and 1960’s through groundwater systems. There are limitations on using it as a geochronological tool because the precipitation history of $^3$H is not well known in most areas. In most cases, tritium has been used simply to distinguish pre-bomb from post-bomb waters. Older ground waters would be essentially free of atmospherically produced $^3$H because of its short half-life (12.3 years), but contain some $^3$H produced in situ by fission-generated neutrons.

$^{14}$C has been used successful for dating groundwater for decades. Since the concentration of $^{14}$C in the atmosphere is uniform, its concentration in precipitation is also uniform. However, there are several problems with $^{14}$C dating. The first is that $^{14}$C is present in water principally as HCO$_3^-$ and CO$_3^{2-}$. Both isotopic exchange reactions with carbonates in soils and the aquifer matrix and precipitation and dissolution of carbonates will alter the concentration of $^{14}$C in groundwater. This, of course, violates the closed-system requirement. A second disadvantage is its relatively short half-life, which restricts the use of $^{14}$C dating to waters less than 25,000 years old. While this is sufficient for shallow, localized groundwater systems, regional systems often contain much older water.

In contrast to $^{14}$C, $^{36}$Cl is essentially conservative in groundwater solutions, and has a half-life suitable for dating a water in regional aquifers as old as 1 Ma. The disadvantage is that it has a much lower production rate in the atmosphere than $^{14}$C, and its analysis requires accelerator mass spectrometry.

In the atmosphere, the primary means of production of $^{36}$Cl are spallation of $^{40}$Ar and neutron capture by $^{36}$Ar. The former process has been estimated to produce about 11 atoms-m$^{-2}$-s$^{-1}$, while the latter produces about half that, for a total production of about 15 atoms-m$^{-2}$-s$^{-1}$. The residence time of $^{36}$Cl in the atmosphere (about 1 week) is not long enough to homogenize its concentration, so the fallout varies with latitude as shown in Figure 12.4.

As usual, dealing with just the number, or concentration, of $^{36}$Cl atoms can have disad-
vantages, and can be misleading. Evaporation, for example, would increase the number of \(^{36}\)Cl atoms. Thus the \(^{36}\)Cl/Cl ratio (Cl has two stable isotopes: \(^{35}\)Cl and \(^{37}\)Cl) is generally used. Stable chlorine can be leached from rocks. This chlorine will be nearly, but not entirely, free of \(^{36}\)Cl. Some \(^{36}\)Cl will be produced naturally by \(^{35}\)Cl capturing neutrons generated by fission and \((\alpha, n)\) reactions on light elements. Further complications arise from the bomb-produced \(^{36}\)Cl. Dissolved chlorine can also capture neutrons. Thus \(^{36}\)Cl will build up in groundwater according to:

\[
N_{36} = \frac{\phi_n f}{\lambda_{36}} (1 - e^{-\lambda_{36} t}) \tag{12.15}
\]

where \(N_{36}\) is the number of \(^{36}\)Cl atoms per mole, \(\phi_n\) is the neutron flux, and \(f\) is the fraction of neutrons captured by \(^{35}\)Cl. The secular equilibrium value, i.e., the concentration at \(t = \infty\) is simply:

\[
N_{36} = \frac{\phi_n f}{\lambda_{36}} \tag{12.16}
\]

This \textit{in situ} production must be taken into account.

Stable Cl derived from sea spray is also present in the atmosphere and in precipitation. Its concentration decreases exponentially from coasts to continental interiors. Thus the initial \(^{36}\)Cl/Cl ratio in precipitation will be variable and must be determined or estimated locally before groundwater ages can be estimated. The age of groundwater may then be determined from:

\[
t = -\frac{1}{\lambda} \ln \left( \frac{C^\text{Cl} \left[ \frac{^{36}\text{Cl}}{\text{Cl}} - \frac{^{36}\text{Cl}}{\text{Cl}}_{\text{se}} \right]}{C_0^\text{Cl} \left[ \frac{^{36}\text{Cl}}{\text{Cl}} - \frac{^{36}\text{Cl}}{\text{Cl}}_{\text{se}} \right]} \right) \tag{12.17}
\]

where \(C^\text{Cl}\) is the chloride concentration and the subscripts “0” and “se” denote initial and secular equilibrium values respectively.

Bentley et al. (1986) used this approach to determine the age of groundwater in the Great Artesian Basin aquifer. The Great Artesian Basin aquifer is one of the largest artesian aquifers in the world and underlies about a fifth of Australia (Figure 12.5). The primary aquifer is the Jurassic Hooray sandstone, which outcrops and is recharged along the eastern edge of the basin. Bentley et al. (1986) sampled 28 wells from the system. They estimated an initial \(^{36}\)Cl/Cl ratio of \(110 \times 10^{-15}\) and a secular equilibrium value of \(9 \times 10^{-15}\) atoms per liter. Some well samples showed evidence of \(^{36}\)Cl addition, probably from upward leakage of water from evaporate-bearing Devonian sediments beneath the aquifer. Other wells, particularly those in the recharge area, showed evidence of evaporation, which increases Cl concentrations before the water penetrated the groundwater system, not surprising in an arid environment. On the whole however, \(^{36}\)Cl ages were comparable to calculated hydrodynamic ages, as illustrated in Figure 12.6.

In a somewhat different application, Paul et al. (1986) have used \(^{36}\)Cl to determine the accumulation time of dissolved salt in the Dead Sea. The Dead Sea is a particularly simply hydrologic system because is has no outlet. In such a simple system, we can describe the variation of the number of \(^{36}\)Cl atoms with time as the rate of input less the rate of decay:

\[
\frac{dN}{dt} = I - \lambda N \tag{12.18}
\]

Figure 12.5. Extent of the Great Artesian Basin aquifer in Australia. From Bentley et al. (1986).
where $I$ is the input rate (precipitation of chloride is assumed negligible). Integration of this equation yields:

$$N = \frac{I}{\lambda} \left(1 - e^{-\lambda t}\right)$$ \hspace{1cm} 12.19

Paul et al. measured $^{36}\text{Cl}/\text{Cl}$ in Mt. Hermon snow, in various rivers in the Dead Sea system, and in saline springs in the Dead Sea basin. These results are summarized in Table 12.2. Using equation 12.19, they estimated an accumulation time of 20,000 years for the salt in the Dead Sea. The Dead Sea basin

<table>
<thead>
<tr>
<th>Site</th>
<th>$^{36}\text{Cl}/\text{Cl}$ $(10^{-15})$</th>
<th>Cl $(\text{mg/l})$</th>
<th>$^{36}\text{Cl}$ $(10^6 \text{ atoms/l})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mt. Hermon Snow</td>
<td>1580±120</td>
<td>1.50</td>
<td>40±5</td>
</tr>
<tr>
<td>Banias River</td>
<td>400±60</td>
<td>11.9</td>
<td>80±15</td>
</tr>
<tr>
<td>Snir River</td>
<td>430±125</td>
<td>11.0</td>
<td>80±20</td>
</tr>
<tr>
<td>Dan River</td>
<td>725±140</td>
<td>10.5</td>
<td>129±25</td>
</tr>
<tr>
<td>Lake Kinneret</td>
<td>49±15</td>
<td>252</td>
<td>210±65</td>
</tr>
<tr>
<td>Jordan River</td>
<td>121±19</td>
<td>646</td>
<td>1,320±210</td>
</tr>
<tr>
<td>Dead Sea</td>
<td>17±2</td>
<td>$2.30 \times 10^5$</td>
<td>$6.6 \times 10^4$</td>
</tr>
<tr>
<td>Ashlag Spring (saline spring)</td>
<td>4±2</td>
<td>$2.6 \times 10^5$</td>
<td></td>
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</tbody>
</table>
The Dead Sea has been estimated to be 15,000 years old based on $^{14}$C. The difference suggests some of the Cl now in the Dead Sea was inherited from its less saline Pleistocene predecessor, Lake Lisan. The data in Table 12.2 also illustrates how a combination of Cl and $^{36}$Cl data can distinguish between addition of Cl from rock leaching and evaporation, both of which are processes that will increase the concentration of Cl. Evaporation should not significantly change the $^{36}$Cl/Cl ratio, while addition of Cl derived from rock leaching should decrease this ratio. There is a general southward (downstream) increase in Cl concentration in the Jordan River–Dead Sea system. It is apparent from the data in Table 12.2 that this is due to both addition of rock-derived Cl and evaporation.

**REFERENCES AND SUGGESTIONS FOR FURTHER READING**


