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**INTRODUCTION**

In addition to the stable isotope  $^9\text{Be}$ , Be is also formed as two cosmogenic isotopes of interest to earth scientists. Cosmogenic Be is formed primarily in the stratosphere from cosmic-ray spallation of oxygen and nitrogen, but some is produced in the troposphere and *in situ* on the surface of the earth. Production of cosmogenic Be was hypothesized by Peters (1955). Soon after, naturally occurring  $^7\text{Be}$  was identified in precipitation by Arnold and Al-Salih (1955) and essentially concurrently and independently by Goel et al. (1956).  $^{10}\text{Be}$  ( $T_{1/2} = 1.5 \times 10^6$  yr) was first detected by Arnold (1956) in marine sediment cores. The  $^{10}\text{Be} / ^7\text{Be}$  production ratio is approximately 0.5 (Lal and Peters 1967).

After its formation in the atmosphere, cosmogenic Be adsorbs electrostatically to aerosols that may be washed out by precipitation and delivered to ecosystems. The amount of cosmogenic Be that reaches the surface of the earth is a function of production rate (cosmic-ray intensity), stratosphere-troposphere mixing, circulation and advection within the troposphere, and efficiency of removal from the troposphere (wet and dry deposition) (Feely et al. 1989). Since the discovery of cosmic-ray produced Be, numerous researchers have gathered data on its (1) production, (2) fluxes to the oceans and terrestrial ecosystems, (3) distribution and inventory in soils, snow, sediments, and vegetation, and (4) geochemical behavior. In 1955, Peters suggested that the relatively long half-life of  $^{10}\text{Be}$  could make it useful for quantifying Tertiary sedimentation rates and other surficial processes. The focus of this chapter is the short-lived radionuclide  $^7\text{Be}$  ( $T_{1/2} = 53.12 \pm 0.07$  days; Jaeger et al. 1996). Due to its short half-life, relative ease of measurement, and well-defined source term,  $^7\text{Be}$  serves as a useful tool for tracing and quantifying environmental processes on the <1 year timescale (Lal et al. 1958; Young and Silker 1980; Krishnaswami et al. 1980; Russell et al. 1981; Turekian et al. 1983; Dutkiewicz and Husain 1985; Olsen et al. 1985; Wallbrink and Murray 1993; Bonniwell et al. 1999), and has applications in meteorology, soil science, sedimentology, geomorphology, hydrology, geochemistry, and nuclear physics.

**ANALYSIS FOR  $^7\text{Be}$** 

$^7\text{Be}$  decays to stable  $^7\text{Li}$  by electron capture ( $\lambda = 4.766 \text{ yr}^{-1}$ ). Eighty-nine and one-half percent of the atoms decay directly to the ground state of  $^7\text{Li}$ , while 10.5% decay first to the excited state of  $^7\text{Li}$ , which decays to ground state  $^7\text{Li}$  via gamma-ray emission at 477.6 keV (Ajzenberg-Selove 1988).  $^7\text{Be}$  activities are normally determined in

environmental samples using gamma spectrometers that detect the 477.6 keV gamma. In earlier investigations,  $^7\text{Be}$  was detected using thallium (Tl) activated sodium-iodide (NaI) detectors (Arnold and Al-Salih 1955; Goel et al. 1956; Walton and Fried 1962; Schumann and Stoepler 1963, and Krishnaswami et al. 1980). However, the relatively low resolution of these instruments requires chemical separation of the Be before the gamma analysis because the NaI(Tl) detector cannot distinguish the  $^7\text{Be}$  photopeak from other radionuclides which decay in the same energy region ( $^{228}\text{Ac}$  at 462 keV;  $^{103}\text{Ru}$  at 497 keV). The techniques used for the purification and concentration of  $^7\text{Be}$  from various samples generally utilize cation exchange columns and/or coprecipitation of Be with Fe hydroxides and other hydroxide phases. Details of these methods are described in the references mentioned above.

More recently, high-purity germanium (Ge) detectors or lithium (Li)-drifted Ge detectors have been employed to determine  $^7\text{Be}$  activity. These detectors generally have high resolution (<2 keV) and high counting efficiencies. In many cases, precipitation, soil, and sediment samples can be analyzed directly for  $^7\text{Be}$ . Larson and Cutshall (1981) describe a method for detecting  $^7\text{Be}$  directly in sediment samples using Marinelli Beakers. They reported a detection limit of approximately  $0.015 \text{ Bq g}^{-1}$  for a 250 g sediment sample counted 200 min (1 Bq = 1 becquerel = 1 disintegration per second, the SI unit for radioactivity. 1 Bq = 27.03 pCi; no concentration equivalent such as  $\text{ng kg}^{-1}$  or  $\text{pg kg}^{-1}$  would be applicable due to the very small number of atoms present in environmental samples). When counting samples high in  $^{232}\text{Th}$ , one should be cautious of the 478.3 keV decay energy of  $^{228}\text{Ac}$ . Although the relative intensity of this photon is only 0.215%, high activities of  $^{228}\text{Ac}$  found in mineral soil and sediment samples can create a disturbance in this region of the spectrum. Surface waters are generally low in  $^7\text{Be}$ , so pre-concentration is necessary for efficient detection. Kostadinov et al. (1988) suggested  $^7\text{Be}$  adsorption on activated charcoal to concentrate  $^7\text{Be}$  from surface waters.

For over five decades,  $^7\text{Be}$  has received attention from scientists investigating the possible variability in a radioactive nuclide's decay constant. Emilio Segrè (1947) first suggested that the decay constant of a nuclide that decays by electron capture should be influenced by the density of its electrons. He suggested that, for a light element such as  $^7\text{Be}$ , this change could be "appreciable", and by putting  $^7\text{Be}$  in various chemical compounds, a slight change in half-life could potentially be observed. A few years later, Leininger et al. (1949; 1951) reported that  $[\Delta\lambda/\lambda] \times 10^{-3}$  for  $\lambda(^7\text{Be}) - \lambda(^7\text{BeF}_2)_{\text{hex}} = 0.84 \pm 0.10$ . Between 1950 and 1970, several researchers reported a deviation of  $\lambda$  for  $^7\text{Be}$  in different Be compounds (Table 1). These reports received little attention from the scientific community because the differences in  $\lambda$  were detected using NaI detectors or ionization chambers. The analytical error of these instruments approached the magnitude of changes in  $\lambda$  they were trying to resolve. In 1999, using a high purity intrinsic Ge detector, Huh (1999) reported that the half-lives of  $^7\text{Be}$  in  $\text{Be}(\text{OH}_2)^{+2}$ ,  $\text{Be}(\text{OH})_2$ , and  $\text{BeO}$  are 53.69, 53.42, and 54.23 d ( $\pm 0.1\%$ ), respectively. Changes in the chemical form of  $^7\text{Be}$  may not be the only variable affecting its decay constant. Hensley et al. (1973) found that the decay constant of  $^7\text{Be}$  in  $\text{BeO}$  increased slightly under high pressure in a diamond-anvil press. Liu and Huh (2000) found similar results as Hensley et al. (1973), but they noted that the rate of increase of  $\lambda$  with pressure in a  $\text{Be}(\text{OH})_2$  gel decreased with increasing pressure.

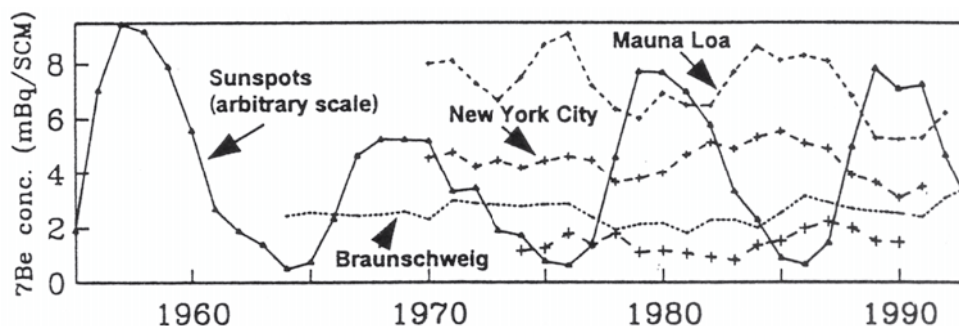
### **PRODUCTION AND DELIVERY OF $^7\text{Be}$ TO THE EARTH'S SURFACE**

Production of  $^7\text{Be}$  depends on the cosmic-ray flux, which varies with latitude, altitude, and solar activity. Lal et al. (1958) calculated production rates for cosmogenic Be. Production in the troposphere increases by a factor of approximately 1 to 3 from the equator to the poles. In the stratosphere, production can increase by a factor of 4 to 5

from the equator to the poles. Production is greatest between 12 and 20 km altitude (depending on latitude), decreasing nearly exponentially by about 3 orders of magnitude to the earth's surface (Lal et al. 1958; Bhandari et al. 1970). Cosmogenic Be production varies with the 11-year solar cycle. Solar activity maximums result in increased deflection of cosmic rays from the solar system (Lal and Peters, 1967) that decreases the cosmic-ray flux to the earth, and thus decreases  $^7\text{Be}$  production. Several authors have demonstrated an inverse relationship between cosmogenic Be concentrations in air and on the surface of the earth with solar activity (Fig. 1) (Beer et al. 1990; Hötzl et al. 1991; Durana et al. 1996). Concentration fluctuations in surface air resulting from the 11-year solar cycles are generally on the order of 15 to 25% (Koch and Mann 1996) (Fig. 1).

**Table 1.** Variation in the decay constant of  $^7\text{Be}$  in various chemical compounds. All values are  $\Delta\lambda/\lambda \times 10^{-3}$ . Modified from Jöhlige et al. (1970).

Source	Leininger et al. (1949); Seg & Wögel (1951)	Krka et al. (1953)	Bahez et al. (1956)	Jöhlige et al. (1970)	Hu (1999)
$\lambda(\text{BeO}) - \lambda(\text{BeF}_2)_{\text{hex}}$	0.69±0.03	0.609±0.055			
$\lambda(\text{BeO}) - \lambda(\text{BeF}_2)_{\text{am}}$				1.130±0.058	
$\lambda(\text{Be}) - \lambda(\text{BeF}_2)_{\text{hex}}$	0.84±0.10	0.741±0.047			
$\lambda(\text{Be}) - \lambda(\text{BeF}_2)_{\text{am}}$			1.2±0.1		
$\lambda(\text{BeO}) - \lambda(\text{Be})$	-0.15±0.09	-0.131±0.051			
$\lambda(\text{BeO}) - \lambda(\text{Be}(\text{OH})_2)$					-15.177±0.1

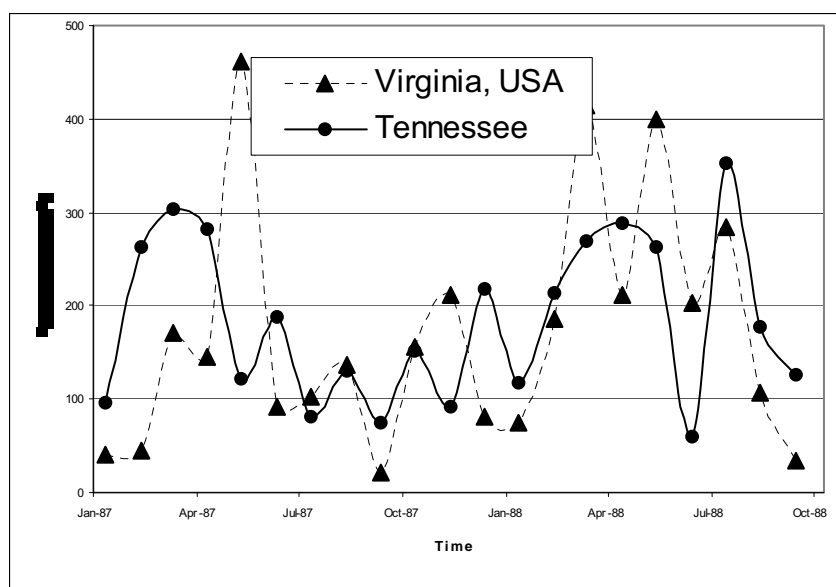


**Figure 1.** Annual variation of  $^7\text{Be}$  in surface air plotted with solar activity, 1955-1993.  $^7\text{Be}$  units are millibecquerels  $\text{m}^{-3}$  at standard temperature and pressure. Figure used by permission of the editor of *Journal of Geophysical Research*, from Koch et al. (1996), Fig. 2, p. 18,653.

Because the stratospheric residence time of aerosols (approximately 14 months [Reiter, 1975]) exceeds the half-life of  $^7\text{Be}$  by more than six-fold, equilibrium activity is assumed to occur. In the troposphere, however, production of  $^7\text{Be}$  is significantly lower, and the residence time of  $^7\text{Be}$  is much shorter (22 to 48 days [Bleichrodt 1978; Durana et al. 1996]) due to rapid wash-out. This results in a fairly high concentration gradient between the stratosphere and the troposphere, with tropospheric air generally containing one to two orders of magnitude less  $^7\text{Be}$  ( $\text{Bq m}^{-3}$ ) than the stratosphere. Rama and Honda (1961) and Bhandari and Rama (1963) reported  $^7\text{Be}$  activity for air sampled at various points in the atmosphere. Stratosphere air sampled at 18-20 km ranged from approximately 0.16 to 0.58  $\text{Bq } ^7\text{Be m}^{-3}$ . Troposphere air ranged from approximately 0.005 to 0.02  $\text{Bq m}^{-3}$ . Dutkiewicz and Husain (1985) found that  $^7\text{Be}$  activity in lower stratospheric air averaged approximately 0.17  $\text{Bq m}^{-3}$ , while the upper troposphere averaged 0.02  $\text{Bq m}^{-3}$ .

While the activity of  $^7\text{Be}$  in the stratosphere remains fairly constant, concentrations in the troposphere and near-surface air, and thus the amount of  $^7\text{Be}$  available to ecosystems exhibit seasonal fluctuations (Feely et al. 1989).  $^7\text{Be}$  concentrations in near-surface air generally range from 0.001 to 0.007 Bq m<sup>-3</sup> (Feely et al. 1989; Dueñas et al. 1999). Stratosphere-troposphere exchange can increase  $^7\text{Be}$  concentrations in the troposphere and near-surface air. Husain et al. (1977) correlated high concentrations of  $^7\text{Be}$  at Whiteface Mountain, New York, USA with stratospheric air masses as indicated by potential vorticities. Vizee and Singh (1980) correlated high concentrations of  $^7\text{Be}$  in surface air with low-pressure troughs. Maximum mixing between the stratosphere and the troposphere generally occurs in the spring of each year at mid-latitudes, and near-surface air at the middle latitudes generally has higher concentrations of  $^7\text{Be}$  at this time (Feely et al. 1989). Intense thunderstorms may also mix stratospheric air downward, thus increasing the amount of  $^7\text{Be}$  available for scavenging by precipitation (Dingle 1965; Noyce et al. 1971). Several authors have suggested that vertical mixing within the troposphere also governs the concentration of  $^7\text{Be}$  in rain and near-earth surface air (Feely et al. 1989; Baskaran 1995; Durana et al. 1996; Dueñas et al. 1999). Warming of the earth's surface during the spring and summer will increase convection, which would transport  $^7\text{Be}$  from the upper troposphere to near-surface air. At latitudes  $>60^\circ$ , seasonal variations in surface air result from the transport poleward of mid-latitude air. Arctic sites commonly have peak  $^7\text{Be}$  air concentrations in the late winter or early spring, probably a result of the arrival of mid-latitude tropospheric air high in  $^7\text{Be}$  (Feely et al. 1989). Sites with a pronounced seasonal variation in rainfall amount show an inverse relationship between rainfall rate and  $^7\text{Be}$  concentration in air, which demonstrates that washout of  $^7\text{Be}$  can have a significant impact on surface air concentration (Feely et al. 1989). Koch and Mann (1996) used a singular value decomposition (SCD) technique to analyze the spatial and temporal variability of near-surface air concentrations of  $^7\text{Be}$ . They found that up to 47% of the monthly air concentration variability could be explained by 4 "modes" which correspond to the 11-year solar cycles, El Niño Southern Oscillation cycles (2 to 7 years), annual, and semi-annual cycles. Zanis et al. (1999) measured  $^7\text{Be}$  at an alpine research station in Switzerland from 4 April 1996 to 1 January 1997, and compared this with meteorological parameters and composite 500 hPa geopotential height maps. They concluded that downward transport of air from the upper troposphere and wet scavenging of aerosols were main processes controlling the  $^7\text{Be}$  concentrations measured at the site. Papastefanou (1991) found that a supernova explosion in 1987 increased the  $^7\text{Be}$  activity in air by up to four times the normal activity found in temperate air masses. Nuclear detonations do not contribute to  $^7\text{Be}$  production.

Arnold and Al-Salih (1955) were the first to report cosmic-ray-produced  $^7\text{Be}$  in wet precipitation. They collected large volumes of rain and snow in Indiana and Illinois from October 1953 until April 1954.  $^7\text{Be}$  was concentrated from the samples by adding a stable Be carrier and co-precipitating  $\text{Be}(\text{OH})_2$  along with  $\text{Fe}(\text{OH})_3$  and other hydroxides at a pH of 9. The precipitate was then purified to contain only BeO and counted using a 2.54 cm NaI (TI) crystal scintillation spectrometer with an overall counting efficiency of 0.99%.  $^7\text{Be}$  activities ranged from  $0.16 \pm 17\%$  to  $2.40 \pm 8\%$  Bq L<sup>-1</sup>, with a volume-weighted mean of approximately 0.95 Bq L<sup>-1</sup>. While the Arnold and Al-Salih (1955) paper was in press, Goel et al. (1956) independently demonstrated the presence of  $^7\text{Be}$  in Bombay, India. During the months of July, August, and September of 1955, they documented  $^7\text{Be}$  in monsoon rain with a volume-weighted mean activity of 0.36 Bq L<sup>-1</sup>. Since then, numerous researchers have reported the activity of  $^7\text{Be}$  in wet-precipitation, with annual volume-weighted means generally ranging from 0.75 to 2.75 Bq L<sup>-1</sup>. Baskaran et al. (1993) tabulated the volume-weighted  $^7\text{Be}$  activities in precipitation reported between 1983 and 1993. Some studies prior to 1980 did not acidify their precipitation samples



**Figure 2.** Seasonal variation of <sup>7</sup>Be flux at two sites. Data plotted from Olsen et al. (1985).

during collection, which may have resulted in poor recovery of the <sup>7</sup>Be from the precipitation collectors due to adsorption.

As with tropospheric air, the concentration and flux of <sup>7</sup>Be in rainfall varies seasonally as well (Fig. 2). Maximum fluxes and ecosystem inventories are often recorded in the spring to early summer (Olsen et al. 1985; Brown et al. 1989; Dibb 1989; Bachuber and Bunzl 1992; Baskaran 1995). Brown et al. (1989) and Harvey and Matthews (1989) determined that <sup>7</sup>Be activities in rainfall can vary by a factor of greater than 20 between individual rainstorms. This suggests that individual storms can potentially dominate <sup>7</sup>Be deposition. Therefore, to obtain an accurate flux of <sup>7</sup>Be to the earth's surface, samples must be collected from every storm for at least 1 full year. For investigators who have sampled at least 12 months and acidified their samples in the collectors, annual atmospheric flux of <sup>7</sup>Be observed from bulk atmospheric deposition collectors has ranged from approximately 1000 to 6500 Bq m<sup>-2</sup> (Turekian et al. 1983; Olsen et al. 1985; Dominik et al. 1987; Schuler et al. 1991; Wallbrink and Murray 1994). Vogler et al. (1996) reported an annual flux at Lake Constance, Germany of 2250 Bq m<sup>-2</sup>. Deposition was highest during the summer and related strongly to rainfall (mm d<sup>-1</sup>). Harvey and Matthews (1989) measured <sup>7</sup>Be for a year on the Hokitika, South Island, New Zealand. The annual precipitation was about 2.8 m. Wet deposition accounted for about 97% of the total deposition that was 6350 Bq m<sup>-2</sup>, and the <sup>7</sup>Be flux was proportional to the precipitation volume. Dry deposition apparently contributes <10% of <sup>7</sup>Be inventories (Brown et al. 1989; Harvey and Matthews (1989); Todd and Wong (1989); Walbrink and Murray 1994; Benitez-Nelson and Buesseler 1999; Kaste 1999, Kaste et al. 1999).

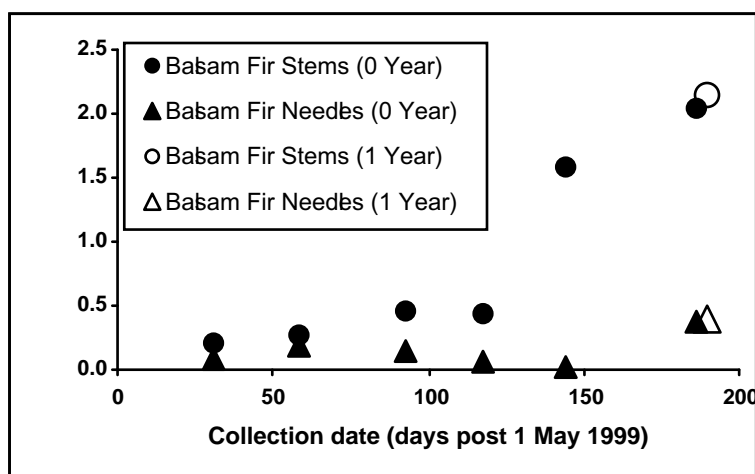
<sup>7</sup>Be is delivered to ecosystems primarily as Be<sup>+2</sup> in slightly acidic (pH<6) rainfall. The Be<sup>+2</sup> ion is extremely competitive for cation exchange sites because of its high charge density. As <sup>7</sup>Be<sup>+2</sup> comes in contact with soils and vegetation, it is rapidly sequestered by exchange surfaces. Forest canopies may decrease the amount of <sup>7</sup>Be that reaches soils and streams, in contrast to the increases observed for most other analytes (Kaste et al. 1999). After deposition, most <sup>7</sup>Be decays in the watershed, but some may be exported in particulate or dissolved form.

**Table 2.** Terrestrial inventories of  $^7\text{Be}$ .

<i>Cbch du</i>	<i>Loñ</i>	<i>Apw ñe</i>	<i>Shñññd (n )</i>	<i>Tñ ñventyBq m<sup>2</sup></i>	<i>Sñce</i>
7/82	Delaware, USA	39°N	marsh + overlying grass	207±13%	Olsen et al. (1985)
7/84	Oak Ridge, TN, USA	36°N	soil + overlying grass	673±3%	Olsen et al. (1985)
1/85	Wallops Island, VA, USA	38°N	marsh + overlying grass	673±7%	Olsen et al. (1985)
1/85	Wallops Island, VA, USA	38°N	unvegetated marsh	107±17%	Olsen et al. (1985)
9/88	Black Mt, Australia	35°S	soil + overlying grass (3)	200±10%	Wallbrink & Murray (1996)
9/88	Black Mt, Australia	35°S	alluvial bare soil (3)	130±20%	Wallbrink & Murray (1996)
5/89	Black Mt, Australia	35°S	soil + overlying grass (3)	400±10%	Wallbrink & Murray (1996)
5/89	Black Mt, Australia	35°S	alluvial bare soil (3)	155±15%	Wallbrink & Murray (1996)
5/96	Idaho, USA	44°N	soil and overlying vegetation (6)	139±16%	Bonniwell et al. (1999)
6/-8/98	Maine, USA	45°N	forested soil (8)	165±40%	Kaste (1999)
12/98	Maine, USA	45°N	bog core + overlying vegetation	554±26%	Kaste (1999)

### $^7\text{BE}$ DISTRIBUTION IN VEGETATION AND SOILS

$^7\text{Be}$  surface inventories (activity of  $^7\text{Be}$  per unit area regardless of its vertical distribution) generally range from 100 to 700 Bq m<sup>-2</sup>. Table 2 summarizes some terrestrial inventories of  $^7\text{Be}$  for soil and wetland profiles. Vegetation may be a very important sink for  $^7\text{Be}$ , and a significant portion of the  $^7\text{Be}$  surface inventory may reside in grasses and forest canopies. Olsen et al. (1985) collected 2 marsh cores in early January 1985 just one meter apart. The vegetated core site had approximately six times the inventory of  $^7\text{Be}$  found in the unvegetated core site, with approximately 40% of the  $^7\text{Be}$  activity in the grass. Wallbrink and Murray (1996) determined the inventory of  $^7\text{Be}$  from soil+grass cores from Black Mountain, Australia from 1988 to 1989. They found  $^7\text{Be}$  inventories to double in the same plot in less than one year, and attribute the increase to grass growth (50 to 90 cm increase) which would increase the efficiency of precipitation scavenging. Russell et al. (1981) determined that White Pine (*Pinus strobus* L.) needles from MA, USA contained approximately 1.9 Bq  $^7\text{Be}$  g<sup>-1</sup> ash, and found approximately 6 to 9 Bq  $^7\text{Be}$  g<sup>-1</sup> ash in twigs from the same tree. They concluded that in a climate with 1.2 meters of rain/year, the forest canopy typically retains 5 to 20% of the open field flux of atmospherically deposited radionuclides. Wallbrink and Murray (1996) calculated that a Eucalypt (*Eucalyptus*) forest canopy retained 4 to 27% of the total  $^7\text{Be}$  inventory calculated from fallout measurements. Kaste et al. (1999) reported dried red spruce (*Picea rubens* Sarg.) needles containing approximately 0.15 Bq  $^7\text{Be}$  g<sup>-1</sup>, and estimated that over 150 Bq  $^7\text{Be}$  m<sup>-2</sup> could reside in the forest canopy (ca. 50% of the total surface inventory).  $^7\text{Be}$  accumulates over time on vegetation, and is balanced by wash off (precipitation) and decay. Leaves apparently reach steady-state (adsorption = decay) with respect to atmospherically delivered  $^7\text{Be}$  before twigs and bark (Fig. 3) (Russell et al. 1981; Norton and Perry, unpublished data 1999).



**Figure 3.**  $^7\text{Be}$  activity on 0 year balsam fir (*Abies balsamea* L. Mill) needles and stems compared with activities on 1 year old balsam fir needles and stems. (Norton and Perry, unpublished data).

$^7\text{Be}$  may be a useful tracer for studying erosional processes (Murray et al. 1992). However, such investigations require a detailed understanding of the depth distribution of  $^7\text{Be}$ . Wallbrink and Murray (1996) found  $^7\text{Be}$  no deeper than 2 cm in bare soil, grassland soil, and eucalyptus forest soil in Australia. Olsen et al. (1985) reported  $^7\text{Be}$  was retained in the upper 2 cm of soil in Tennessee, USA and a marsh core collected from Wallops Island, Virginia, USA. However, a second marsh core collected in the summer from Wallops Island had  $^7\text{Be}$  as deep as 10 cm. They suggested that the depth of the groundwater table could influence the depth distribution of  $^7\text{Be}$ . In central Idaho, Bonniwell et al. (1999) reported that the entire  $^7\text{Be}$  inventory was retained in the top 1.2 cm of a soil profile. Kaste (1999) found that the  $^7\text{Be}$  residing in the upper 8 cm of the organic horizon at the Bear Brook Watershed in Maine, USA (64 to 300 Bq m<sup>-2</sup>) plus that predicted in the canopy matched the surface inventories predicted by input measurements. Apparently the drainage structure, moisture status of the soil, and overlying vegetation govern the inventory and depth distribution of  $^7\text{Be}$ .

### $^7\text{Be}$ IN FRESHWATERS

You et al. (1989) determined partitioning coefficients ( $K_d = [\text{Bq kg}^{-1}]/[\text{Bq L}^{-1}]$ ) for  $^7\text{Be}$  between river water and various substrates in the laboratory, and found that most mud, silt, and clay minerals have a  $K_d$  near 10<sup>5</sup> L kg<sup>-1</sup> under neutral to alkaline conditions (pH > 6; see Fig. 9 in Vesely et al., this volume). They reported that the  $K_d$  decreases by four orders of magnitude as pH decreases from 6 to 2, and found no significant effect of DOC on the adsorption of  $^7\text{Be}$ . Hawley et al. (1986) and You et al. (1989) both found that  $K_d$  varies inversely with the amount of suspended particles in freshwater. Li et al. (1984) suggested that higher concentrations of suspended particles increase particle aggregation, thereby decreasing the effective exchange surface area g<sup>-1</sup> of suspended matter. Olsen et al. (1986) and Bonniwell et al. (1999) determined field  $K_d$  values for  $^7\text{Be}$  in freshwaters to be >10<sup>4</sup>. Vogler et al. (1996) reported that  $K_d$  in the water column of Lake Constance, Germany ranged from 10<sup>5.3</sup> to 10<sup>6</sup>.

$^7\text{Be}$  mobility may be enhanced in natural waters by the formation of soluble fluoride (F) and organic acid complexes (Vesely et al. 1989; Vesely et al. this volume). If free F anions are available in the pH range 4.5 to 7,  $\text{BeF}^+$  may be a significant fraction of the total Be species in solution. This would decrease the charge density of the total  $^7\text{Be}$  in solution, and reduce its adsorption to the solid phase. Below pH 4, aluminum (Al) competition for F decreases Be-F complexes (Vesely et al. 1989). Above pH 7, Be

mobility decreases as hydroxide complexes of Be become significant and Be is adsorbed by sediment and precipitating iron (Fe) and Al hydroxides. Vesely et al. (1989) suggested that Be complexes with fulvic acids may increase Be mobility in surface waters by reducing the fraction of the  $\text{Be}^{+2}$  in solution

Despite numerous publications documenting the production of  $^7\text{Be}$  and the flux to the surface of the earth, there are relatively few studies documenting its mobility and export from watersheds. Dominik et al. (1987) found  $^7\text{Be}$  activity in filtered streamwater from Switzerland to be  $<1.1 \times 10^{-3} \text{ Bq L}^{-1}$ , but found fluvial suspended matter to have an activity of up to  $0.49 \text{ Bq g}^{-1}$ , averaging  $0.132 \text{ Bq g}^{-1}$ . Bonniwell et al. (1999) reported that dissolved Be in a mountain stream in central Idaho was below detection limit ( $<3 \times 10^{-4} \text{ Bq L}^{-1}$ ), but suspended sediment generally had  $0.05$  to  $1 \text{ Bq g}^{-1}$ . Kaste (1999) found that unfiltered streamwater draining an artificially acidified catchment (pH 4.7) in central Maine had up to  $1 \text{ Bq L}^{-1}$  during a large snowmelt/rain event.  $^7\text{Be}$  was up to  $0.75 \text{ Bq L}^{-1}$  in unfiltered streamwater from a nearby stream with  $20 \text{ mg DOC L}^{-1}$  during high discharge. Olsen et al. (1986) reported activity of dissolved  $^7\text{Be}$  in the lower Susquehanna and Raritan Rivers eastern USA ranging from approximately  $0.002$  to  $0.003$  and  $0.002$  to  $0.014 \text{ Bq L}^{-1}$ , respectively.  $^7\text{Be}$  activities on suspended particles ranged from  $\sim 0.26$  to  $0.44 \text{ Bq g}^{-1}$ . In both the Raritan and the Susquehanna, dissolved  $^7\text{Be}$  activities ( $\text{Bq L}^{-1}$ ) were higher when the total particle concentration ( $\text{mg L}^{-1}$ ) was higher. In a  $^7\text{Be}$  budget for the James River, Virginia, USA, Olsen et al. (1986) determined that less than 5% of the  $^7\text{Be}$  in the estuary was from the rivers, and suggested that soils and vegetation in watersheds sequester most of the  $^7\text{Be}$  delivered by rainfall. Cooper et al. (1991) studied  $^7\text{Be}$  export during a snowmelt event in an arctic watershed. Even though  $^{18}\text{O}/^{16}\text{O}$  ratios indicated that  $>85\%$  of the discharge during the melt was from the snowpack, over 90% of the  $^7\text{Be}$  inventory in the snowpack remained in the watershed after the event.

The efficiency of removal of  $^7\text{Be}$  from the water column by particulates may vary among lakes. Krishnaswami et al. (1980) found that the  $^7\text{Be}$  inventory in a sediment core from Lake Whitney closely matched the atmospheric flux of  $^7\text{Be}$ . They suggested that if sediment focusing effects could be ruled out, the residence time of  $^7\text{Be}$  in the lake was very short because of its very high reactivity with particles in the water column. However, in Lake Zurich, Switzerland, Schuler et al. (1991) found that  $^7\text{Be}$  fluxes into sediment, measured by sediment traps, accounted for only 30% of the measured atmospheric flux of  $^7\text{Be}$ . They proposed that the residence time of  $^7\text{Be}$  in the lake was greater than its mean life ( $1/\lambda = 76.6$  days). Wan et al. (1987) determined the residence time of  $^7\text{Be}$  in Greifensee, Switzerland to be approximately 75 days using calculated atmospheric fluxes and measured  $^7\text{Be}$  inventories in sediment. Vogler et al. (1996) observed that the concentration of total  $^7\text{Be}$  in Lake Constance, Germany varied with atmospheric input and particle flux. The proportion of  $^7\text{Be}$  associated with particles varied with biological productivity, and the residence time calculated with a particle-scavenging model ranged from 40 to 340 days. Steinmann et al. (1999) reported a similar residence time of 50 to 230 days for  $^7\text{Be}$  in the water column at Lake Lugano in Switzerland. They found that  $\log K_d$  for  $^7\text{Be}$  on colloids was approximately 6, while  $\log K_d$  on particulates ( $>1 \mu\text{m}$ ) was between 4 and 5. Dominik et al. (1989) calculated the residence time of  $^7\text{Be}$  in Lake Geneva to be 60 to 1100 days, using four different methods. They suggested that the most accurate method to determine residence time of  $^7\text{Be}$  is to compare its atmospheric flux with  $^7\text{Be}$  fluxes in sediment traps. Residence times calculated from  $^7\text{Be}$  inventories in bottom sediments were generally within the range of those calculated by sediment traps (400 to 930 days), but the authors warned that small scale heterogeneity could make sediment inventory calculations unreliable.

$^7\text{Be}$  inventories from lake sediment cores range from 35 to  $875 \text{ Bq m}^{-2}$ , with  $^7\text{Be}$



activities in the upper few centimeters of the cores generally ranging from 0.02 to 0.55 Bq g<sup>-1</sup> (Krishnaswami et al. 1980; Wan et al. 1987; Dominik et al. 1989). Krishnaswami et al. (1980) found <sup>7</sup>Be as deep as 3 cm in Lake Whitney, Connecticut, USA, while Wan et al. (1987) reported <sup>7</sup>Be no deeper than 1 cm in a lake core from Greifensee, Switzerland. Wan et al. (1987) found that <sup>7</sup>Be inventories in cores ranged by a factor of 4, and were higher where sedimentation rates were higher. Schuler et al. (1991) found a strong seasonality for <sup>7</sup>Be fluxes in sediment traps in Lake Zurich, with highest fluxes in July and August over three consecutive years. Maximum <sup>7</sup>Be atmospheric flux and maximum particle flux also occurred during this time period.

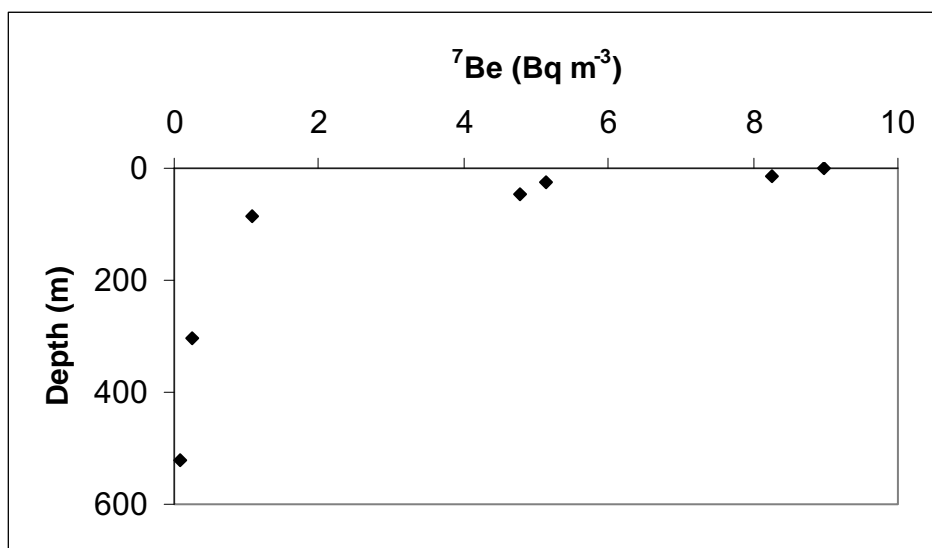


Figure 4. Depth-distribution of <sup>7</sup>Be in the Pacific Ocean. Data Plotted from Silker (1972).

### <sup>7</sup>BE IN THE MARINE ENVIRONMENT

Young and Silker (1980) compiled a significant amount of data on dissolved <sup>7</sup>Be in the Atlantic and Pacific Oceans. They reported that <sup>7</sup>Be activities in seawater generally ranged from 0.0017 to 0.0117 Bq L<sup>-1</sup>. Total inventories ranged from approximately 80 to 600 Bq m<sup>-2</sup>. They found that concentrations and inventories were highest in areas associated with high rainfall, and increased with latitude. Silker (1972) reported on the depth distribution of <sup>7</sup>Be in the North Pacific Ocean (Fig. 4), and determined that <sup>7</sup>Be distribution generally reflected the temperature profile. In cases where there was a strong thermocline, <sup>7</sup>Be activity was relatively uniform in the mixed (upper) layer, and concentrations decreased rapidly below the thermocline.

Since Young and Silker (1980), several authors have reported <sup>7</sup>Be activities in seawater (Table 3). Comparing the activities of dissolved <sup>7</sup>Be and K<sub>d</sub> values reported for marine waters with the few data available for freshwater, it appears that <sup>7</sup>Be can be partitioned favorably into the aqueous phase in the marine environment. Considering the strong pH dependence of Be partition coefficients (You et al. 1989; Vesely et al. 1989; Vesely et al., this volume) the high pH of seawaters relative to many freshwaters should decrease <sup>7</sup>Be in the aqueous phase. Dibb and Rice (1989a) found that sorption of <sup>9</sup>Be in laboratory experiments increased with increasing salinity. The wide range of dissolved <sup>7</sup>Be and K<sub>d</sub> values indicates that several variables are likely governing the solubility and partitioning of Be in seawater including pH, salinity, concentrations of suspended matter concentrations (especially Fe) and DOC, competing cations, and the residence time of the suspended load.

**Table 3.** Dissolved and particulate  $^7\text{Be}$  activities in seawater.

Loth [USA uss ndvud] (me cbctd)	Ddd $^7\text{Be}(\text{Bq L}^{-1})$	$^7\text{Be}$ n pnded mtr [(Bq % fu $^7\text{Be}$ )]	$K_d$	Snded u ( $\text{mg L}^{-1}$ )	Srce
Sequim Bay, WA	0.0009±50%	0.26±24%, 50	5.4	3.55	Bloom & Crecelius (1983)
Sequim Bay, WA	0.0009±38%	0.28±78%, 24	6.0	1.07	Bloom & Crecelius (1983)
Gironde Estuary, France	0.0004 to .002	0.007-0.41	4.5- 5	n.a.	Martin et al. (1986)
Chesapeake Bay, MD (Spring)	0.013±3% to 0.020±2%	0.41-0.47±2%, 36	4.4- 4.5	11 to 24	Olsen et al. (1986)
Chesapeake Bay, MD (Fall)	0.001±33% to 0.003±14%	0.17±11% to 0.43±4%, 20 to 54	4.8- 5.3	4 to 6	Olsen et al. (1986)
Chesapeake Bay, MD (Year)	<0.0008 to 0.048±2%	<0.007-2.5, 1 to 86	2.8- 6	1 to 121	Dibb & Rice (1989b)
Sabine-Neches Estuary, TX	0.003±15% to 0.035±4%	0.08-0.55±9%, 7 to 57	3.2- 4.9	8 to 92	Baskaran et al. (1997)

Bloom and Crecelius (1983) determined that  $^7\text{Be}$  in seawater had a “limited affinity” for suspended matter, and the percent  $^7\text{Be}$  adsorbed to particulates is directly proportional to suspended load. Using laboratory experiments with  $^7\text{Be}$  and natural “detritus”, they demonstrated that high-suspended loads ( $20 \text{ mg L}^{-1}$ ) had approximately 50% of the total  $^7\text{Be}$  adsorbed to suspended matter. At approximately  $1 \text{ mg L}^{-1}$  suspended matter, only 20% of the total  $^7\text{Be}$  in the water column was adsorbed. They also showed that  $^7\text{Be}$  adsorbed similarly to detritus and amorphous  $\text{Fe}(\text{OH})_3$ , while adsorption to the algal culture *Pavlova lutheri* (monochrysis) was significantly lower. Although the percent  $^7\text{Be}$  adsorbed to suspended matter was directly proportional to suspended load concentrations, the  $K_d$  was inversely proportional to suspended load concentrations for their field data (Table 3). Martin et al. (1986) also noted that the  $K_d$  was lowest in the area with the maximum suspended load. Li et al. (1984) found that  $K_d$  for  $^7\text{Be}$  and several other particle-reactive nuclides in seawater was inversely proportional to the suspended load concentrations, in agreement with Hawley et al. (1986) and You et al. (1989) for freshwater systems.

Olsen et al. (1986) found  $^7\text{Be}$  activities in filtered river-estuarine and coastal waters along the northeastern USA seaboard ranging from  $1.1 \times 10^{-3}$  to  $2 \times 10^{-2} \text{ Bq L}^{-1}$ , with total (unfiltered)  $^7\text{Be}$  reaching  $3 \times 10^{-2} \text{ Bq L}^{-1}$ . Most of the  $^7\text{Be}$  was dissolved, typically 60 to 90%. A major factor controlling the concentration of  $^7\text{Be}$  on suspended matter was the amount of time the particles remained in the water column.  $^7\text{Be}$  activities on suspended particles were highest in high-energy areas, and lowest in low energy areas with high sedimentation rates. Dissolved  $^7\text{Be}$  averaged 60% of total  $^7\text{Be}$  in Chesapeake Bay (Dibb and Rice 1989a). However, activities of  $^7\text{Be}$  on suspended matter reached  $2.5 \text{ Bq g}^{-1}$ , and partition coefficients ranged by over 3 orders of magnitude up to  $\log K_d = 6$ . They concluded, as did Olsen et al. (1986), that rapid variations in the supply of particulates in estuaries and bays may not permit equilibrium to be reached between dissolved  $^7\text{Be}$  and the solid phase.

Olsen et al. (1986) determined  $\log K_d$  for river-estuarine and coastal waters from the northeastern USA coast to range from 3.85 to 5.3, with a median of 4.6. They found a positive correlation between  $K_d$  and the Fe concentration of the suspended matter, and suggested that the precipitation of Fe may be an important factor governing the

scavenging of  $^7\text{Be}$ . Baskaran et al. (1997) reported relatively low  $K_d$  values for  $^7\text{Be}$  in the Sabine-Neches estuary, Texas, USA. They found  $\log K_d$  to range from 3.18 to 4.94. There was no significant correlation between suspended load and  $K_d$ . Baskaran et al. (1997) attributed their low  $K_d$  values to the high concentrations of DOC (5 to 20  $\text{mg L}^{-1}$ ) and suggested that DOC could play a significant role in the fate and the residence times of particle-reactive nuclides such as  $^7\text{Be}$  in the water column.

Inventories of  $^7\text{Be}$  in near-shore marine sediments range from  $<30$  to  $6000 \text{ Bq m}^{-2}$ , with  $^7\text{Be}$  activities ranging up to  $\sim 0.65 \text{ Bq g}^{-1}$  (Krishnaswami et al. 1980; Olsen et al. 1986; Dibb and Rice 1989b; Canuel et al. 1990, and Summerfield et al. 1999). Maximum inventories of  $^7\text{Be}$  in marine sediment commonly occur in the spring, and in areas where fine particles are accumulating rapidly. Canuel et al. (1990) examined the spatial and vertical distribution of  $^7\text{Be}$  in sediments of Cape Lookout Bight, NC, USA. They sampled 10 cores in September, 1987, and found  $^7\text{Be}$  inventories ranging from approximately 650 to  $1375 \text{ Bq m}^{-2}$  in a  $<30 \text{ m}^2$  area.  $^7\text{Be}$  penetration reached a maximum depth in the sediment profile during the summer months.  $^7\text{Be}$  activity reached  $0.033 \text{ Bq g}^{-1}$  13 cm deep in the core. The authors attributed this seasonal pattern in  $^7\text{Be}$  depth distribution to the formation of methane bubble tubes during the summertime. Olsen et al. (1986) found that  $^7\text{Be}$  reached a depth of 8 cm in coastal sediments of the eastern USA, but generally was retained in the upper 4 cm. Sneed (1986) determined  $^7\text{Be}$  and  $^{210}\text{Pb}$  in cores from a dredging site with a high sedimentation rate in New York City harbor. She found  $^7\text{Be}$  to a depth of 4 cm in an area where the sedimentation rate was approximately  $7 \text{ cm yr}^{-1}$ .

### APPLICATIONS OF $^7\text{Be}$

A detailed understanding of the transport and residence time of aerosols in the atmosphere is desired because particles in the micron and sub-micron range probably play a significant role in climate change and trace metal cycling.  $^7\text{Be}$  is a very useful tracer in atmospheric transport studies because it (1) has a short half-life, (2) is produced at a relatively constant rate, (3) rapidly attaches to aerosols after formation, and (4) has a large concentration gradient between the stratosphere and troposphere (Bhandari and Rama 1963; Junge 1963; Dutkiewicz and Husain 1979, 1985; Viezee and Singh 1980). In the past decade in particular,  $^7\text{Be}$  measurements in the atmosphere have been used to test and validate chemical transport models in the atmosphere (Brost et al. 1991; Rehfeld and Heimann 1995; Koch et al. 1996). Dibb and Jaffrezo (1993) used  $^7\text{Be}$  and  $^{210}\text{Pb}_{\text{ex}}$  (decay product of  $^{222}\text{Rn}$ ,  $T_{1/2} = 22.3 \text{ yr}$ ) to investigate short term "communication" between the atmosphere and the Greenland Ice Sheet. They found that concentrations of  $^7\text{Be}$  and  $^{210}\text{Pb}$  in fresh snow did not have the spring and fall peaks that ground level aerosol concentrations displayed. On the basis of these data, Dibb and Jaffrezo (1993) suggested that caution must be used when inferring atmospheric chemistry from fresh snow.

The potential for using  $^7\text{Be}$  in atmospheric transport studies is particularly great when it is coupled with a nuclide with a contrasting source function and/or a different half-life. The initial production ratio of  $^{10}\text{Be}/^7\text{Be}$  is approximately 0.5 (Lal and Peters 1967). As air moves away from the region of maximum production, the  $^{10}\text{Be}/^7\text{Be}$  ratio on aerosols in the air mass will increase, because  $^7\text{Be}$  has a much shorter radioactive half-life ( $T_{1/2} = 53 \text{ days}$ ) than  $^{10}\text{Be}$  ( $T_{1/2} = 1.5 \text{ m.y.}$ ). Thus the  $^{10}\text{Be}/^7\text{Be}$  ratio in the atmosphere can be used as a "clock" to determine air mass age, and has been used to indicate the intrusion of stratospheric air into the troposphere (Raisback et al. 1981; Dibb et al. 1994; Rehfeld and Heimann 1995; Koch and Rind 1998). Because the residence time of aerosols in the troposphere is fairly short (ca. 1 month [Durana et al. 1996; Bleichrodt 1978]) compared to those in the stratosphere (about 1 year), stratospheric air masses characteristically have a relatively high  $^{10}\text{Be}/^7\text{Be}$  ratio. Dibb et al. (1994) found that the

$^{10}\text{Be}/^7\text{Be}$  ratio was nearly constant at 2.2 throughout the year in northwestern Canada. They concluded that a significant portion of the aerosols in the arctic troposphere must be derived from the stratosphere, likely a result of vertical mixing between the two air masses rather than stratospheric injections. Koch and Rind (1998) used a General Circulation Model (GISS GCM) to suggest that the spring maximum in the  $^{10}\text{Be}/^7\text{Be}$  ratio observed by Dibb et al. (1994) in the lower stratosphere may have resulted from the transport of equatorial air to higher latitude during the previous winter.

The contrasting source terms of  $^7\text{Be}$  and  $^{210}\text{Pb}$  make the  $^7\text{Be}/^{210}\text{Pb}$  ratio a useful tool for investigating air mass sources and vertical transport within the troposphere (Dibb et al. 1992; Baskaran et al. 1993; Rehfeld and Heimann 1995; Koch et al. 1996; Graustein and Turekian 1996, and Benitez-Nelson and Buesseler 1999). While  $^7\text{Be}$  is produced primarily in the upper atmosphere,  $^{210}\text{Pb}$  is derived from the decay of  $^{222}\text{Rn}$  gas ( $T_{1/2}=3.8$  days) that is emitted primarily from the surface of the continents. Therefore,  $^{210}\text{Pb}$  concentrations in air should decrease with altitude and distance from land. However,  $^7\text{Be}$  concentrations in air will be uniform over land and sea, always increasing with altitude. High  $^7\text{Be}/^{210}\text{Pb}$  ratios may be an indication of stratosphere-troposphere exchange, or an air mass with components originating from the open ocean. Benitez-Nelson and Buesseler (1999) found that the average  $^7\text{Be}/^{210}\text{Pb}$  ratio in rainfall from Woods Hole, MA and Portsmouth, NH (USA) was 12.6 and 20.1, respectively. These data suggest that Woods Hole received air from a more continental source than Portsmouth during the sampling period. The efficiency and rate of vertical transport (convection) within the troposphere will also affect the  $^7\text{Be}/^{210}\text{Pb}$  depositional ratio. Increased vertical transport should increase the amount of  $^7\text{Be}$  delivered to the ground, and increase the rate at which  $^{222}\text{Rn}$  moves upwards. That is,  $^7\text{Be}$  is enriched in the lower troposphere while  $^{210}\text{Pb}$  is diluted. Baskaran (1995) found that the  $^7\text{Be}/^{210}\text{Pb}$  ratio (measured in depositional fluxes) was highest in the summertime (14.4 to 23.9) and lowest in the winter (12.8 to 13.7). He attributed the increased  $^7\text{Be}/^{210}\text{Pb}$  ratios during the warmer months to decreased stability of the troposphere (increased convection).

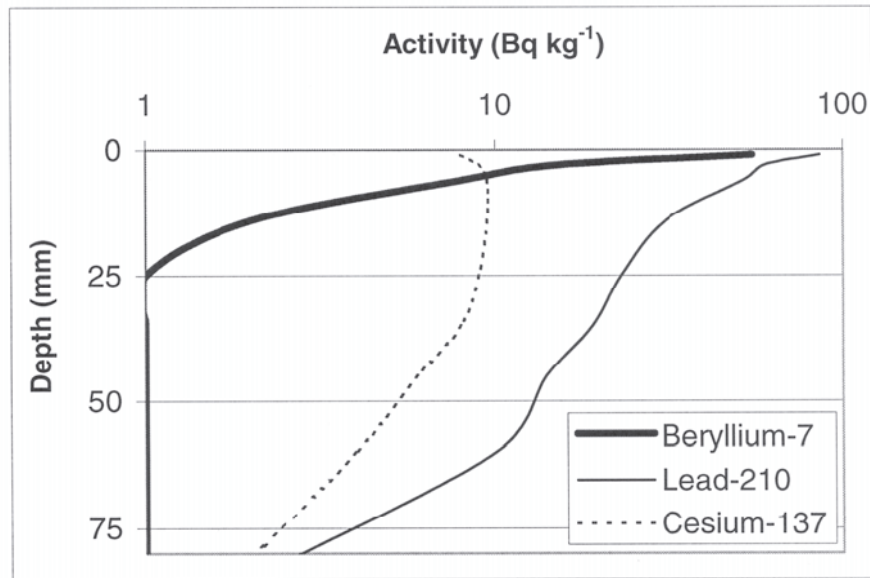
$^7\text{Be}$  has useful applications in sedimentological studies. Olsen et al. (1989) used the ratio of short-lived  $^7\text{Be}$  to longer-lived excess  $^{210}\text{Pb}$  ( $^{210}\text{Pb}_{\text{ex}}$ ) to quantify the amount of re-suspended sedimentary material in coastal waters.  $^7\text{Be}$  is very low on bottom sediments relative to its activity on suspended particulates, while  $^{210}\text{Pb}_{\text{ex}}$  is nearly the same on bottom sediments and suspended particulates. Using  $^7\text{Be}/^{210}\text{Pb}_{\text{ex}}$ , they determined that re-suspended bottom sediments may account for more than 80% of the suspended matter in the Savannah Estuary. Canuel et al. (1990) concluded that in coastal sediments unaffected by bioturbation,  $^7\text{Be}$  is a useful tracer of sediment accumulation. They found that sediment cores taken before and after storm events can be used to quantify the impacts of individual storms by identifying “new” pulses of sediment. Fitzgerald et al. (2001) used this technique to determine monthly sediment deposition and re-suspension rates in PCB-rich sediments in the Fox River system in Wisconsin, USA. They found that short-term accumulation rates determined by  $^7\text{Be}$  were up to 130 times higher than those determined with  $^{137}\text{Cs}$ , indicating the dynamic nature of sediment transport in this impounded system. Sommerfield et al. (1999) used  $^7\text{Be}$  as a tracer of fine-grained river sediment on the northern California, USA continental margin. They found that  $^7\text{Be}$  was present in shelf and slope deposits only after periods of extremely high river discharge, and determined that storms and floods play a significant role in sedimentary processes in modern environments in their study area.  $^7\text{Be}$  has been used to calculate sediment accumulation rates in shallow marine environments (Sneed 1986). Canuel et al. (1990) and Dibb and Rice (1989b) found that short-term sediment accumulation rates determined with  $^7\text{Be}$  were comparable to longer-term sediment accumulation rates determined from  $^{210}\text{Pb}$ . Dibb and Rice (1989b) suggested that the general agreement

between the  $^7\text{Be}$  and  $^{210}\text{Pb}$  derived sedimentation rates could indicate that the processes operating on the Chesapeake Bay sediments have remained relatively constant for the past 100 years. Steinmann et al. (1999) used  $^7\text{Be}$  to calculate the coagulation rate of colloids in the epilimnion of Lake Lugano. They measured the sedimentary flux of  $^7\text{Be}$ , as well as concentrations of dissolved, colloidal, and particulate  $^7\text{Be}$  in the epilimnion. Based on the 2.2 to 16.3 day residence time of  $^7\text{Be}$  in the colloidal fraction, they reported coagulation rates of 0.06 to 0.46  $\text{d}^{-1}$ .

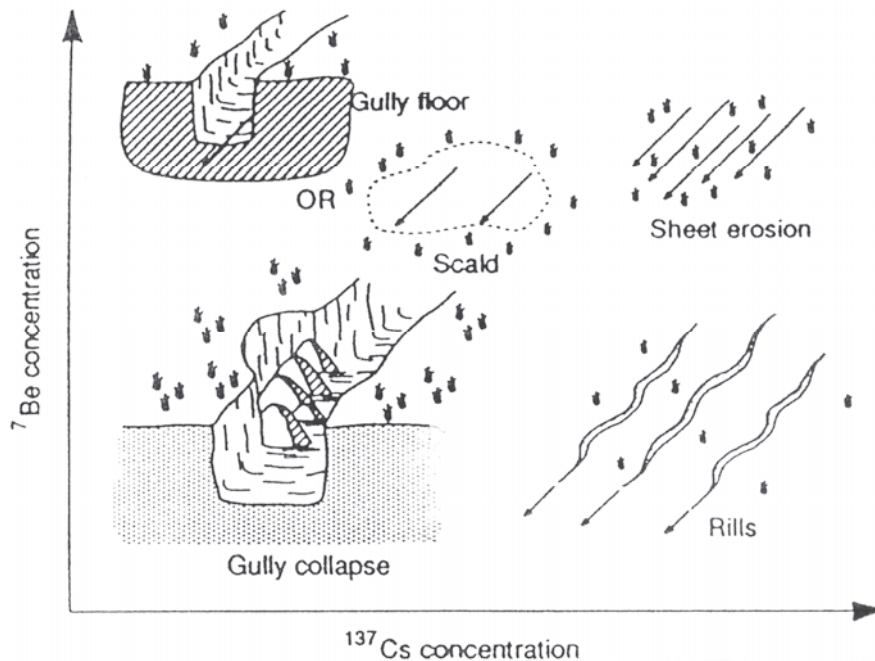
$^{234}\text{Th}$  ( $T_{1/2} = 24$  days; derived from decay of  $^{238}\text{U}$ ) can be used in conjunction with  $^7\text{Be}$  to investigate sedimentation processes in areas with a salinity gradient. Both nuclides are particle-reactive, but the source of  $^{234}\text{Th}$  is  $^{238}\text{U}$ , which is positively correlated with salinity. Feng et al. (1999a, 1999b) utilized the  $^{234}\text{Th}_{\text{ex}}/^7\text{Be}$  activity ratios on suspended and bottom sediments as a tracer of particle sources and transport in the Hudson River estuary, where  $\text{Th}_{\text{ex}}$  is the total Th activity of the sediment minus that supported by  $^{238}\text{U}$ . By using the activity ratio of these two nuclides, the apparent effects of grain size, composition, and concentration on adsorption are minimized. They analyzed sediment during different flow regimes and tidal cycles to determine the relative importance of local sediment re-suspension and advection of sediments through the estuary. They found that both radionuclides were removed from the water column on time scales from <1 to 13 days. They also noted that the  $^{234}\text{Th}_{\text{ex}}/^7\text{Be}$  tended to be equal or greater than that predicted by local equilibrium (*in situ* production of  $^{234}\text{Th}_{\text{ex}}$ , atmospheric inputs of  $^7\text{Be}$ , scavenging, particles settling and re-suspension) during low flow, which can be explained by transport of particles up-estuary by estuarine circulation. During high flows they observed lower activity ratios, indicating the importance of re-suspension. Feng et al. (1999b) reported that during the course of tidal cycles, the  $^{234}\text{Th}_{\text{ex}}/^7\text{Be}$  values were comparable in suspended particles from surface and bottom water with different salinities, which indicated that particles are mixed fairly well in the water column.

The strong affinity of  $^7\text{Be}$  for small particles and its steady production make  $^7\text{Be}$  a unique tool for hydrologists and geomorphologists. Walling et al. (1999) used  $^7\text{Be}$  measurements to document event-based soil redistribution. They found that the spatial patterns of  $^7\text{Be}$  and  $^{137}\text{Cs}$  (fission product,  $T_{1/2} = 30.2$  yr) total inventories were similar. By comparing the inventories of these nuclides to a reference inventory (taken in a stable area where neither erosion or aggradation is occurring) before and after a storm, they could identify areas of soil loss and accumulation on short (a few days) and medium (a few decades) time scales. Bonniwell et al. (1999) used  $^7\text{Be}$  and other fallout nuclides to trace sediment and determine erosion rates in a small watershed in Idaho. They used  $^7\text{Be}$  as an indicator of “new” sediment during snowmelts, by assuming that “old” sediment that gets re-suspended in the water column would have a negligible concentration of  $^7\text{Be}$ . Bonniwell et al. (1999) calculated erosion rates in the watershed by comparing the total export of radionuclides with the measured inventories in the soils.

The different depth distributions of  $^7\text{Be}$ ,  $^{210}\text{Pb}_{\text{ex}}$  and  $^{137}\text{Cs}$  (fission product,  $T_{1/2} = 30.2$  yr) can be used to fingerprint the soil depth from which suspended sediment was originally removed (Burch et al. 1988; Walling and Woodward 1992; Wallbrink et al. 1999) (Fig. 5). Because  $^7\text{Be}$  is often concentrated in the uppermost soil horizons, Murray et al. 1992 suggested using  $^7\text{Be}$  as a tracer of topsoil movement. For example, suspended sediment with a high activity of  $^7\text{Be}$  (and/or  $^{210}\text{Pb}_{\text{ex}}$ ) could indicate an initial source area at the top of the soil profile, while sediment with a lower activity of  $^7\text{Be}$  (and/or  $^{210}\text{Pb}_{\text{ex}}$ ) and high activity of  $^{137}\text{Cs}$  could have been derived from deeper in the profile. Wallbrink and Murray (1993) extended the use of these fallout nuclides to identify the type erosion process (sheet flow, rill or gully erosion) removing soil from the landscape (Fig. 6).



**Figure 5.** Depth-distribution of fallout radionuclides applicable for erosion studies. Generalized from Wallbrink et al. (1999).



**Figure 6.** Generalized  $^7\text{Be}$  and  $^{137}\text{Cs}$  labeling of sediment (after Burch et al. 1988). This concept is based on the theory that sediment retains the radionuclide signature originally obtained when it was soil. Used by permission of the editor of *Hydrological Processes*, from Wallbrink and Murray (1993), Fig. 1c, p. 298.

### SUMMARY AND CONCLUDING REMARKS

$^7\text{Be}$  is a short-lived naturally occurring radionuclide with a unique source term and a relatively constant production rate. After  $^7\text{Be}$  is formed in the atmosphere, it rapidly adsorbs to aerosols. Because the residence time of aerosols in the stratosphere is greater than the half-life  $^7\text{Be}$  by more than a factor of six, steady-state (production = decay)

equilibrium maintains nearly constant concentrations above the tropopause. Tropospheric concentrations of  $^7\text{Be}$  are more dynamic and are generally 1 to 2 orders of magnitude lower than in the stratosphere due to wash out and decay. Near surface air exhibits fluctuations on decadal, semi decadal (4 to 7 years), annual, and semi-annual timescales. The short-term variations of  $^7\text{Be}$  concentration in near surface air are a result of variation in production, wash out by precipitation, vertical mixing between and within the stratosphere and troposphere, and horizontal air mass movement/mixing within the troposphere.  $^7\text{Be}$  terrestrial inventories and ecosystem fluxes (measured by bulk atmospheric deposition collectors) vary seasonally due to variation in tropospheric  $^7\text{Be}$  and precipitation amount.  $^7\text{Be}$  is deposited on ecosystems primarily by wet deposition; dry deposition is apparently <10% of the total flux.

$^7\text{Be}$  deposited from rainfall rapidly adsorbs to vegetation, soils, and sediment, and in many cases will remain adsorbed to the solid phase until decay. In the soil profile,  $^7\text{Be}$  is generally retained in the upper layers. Most of the movement of  $^7\text{Be}$  within and from watersheds appears to be in particulate form. The residence time of  $^7\text{Be}$  in the water column of lakes is uncertain, reports range from a few weeks to a few months. The partitioning of  $^7\text{Be}$  in the marine environment is also poorly understood, with reported  $K_d$  values ranging over three orders of magnitude. In estuaries, a significant portion of the  $^7\text{Be}$  may be in dissolved form, especially in the presence of DOC. Sediment inventories of  $^7\text{Be}$  in both lakes and shallow ocean water exhibit seasonal variations.

$^7\text{Be}$  studies have provided scientists with much insight into atmospheric, geochemical, erosional, sedimentological, and nuclear processes. Its decay energy is fairly easy and inexpensive to measure. Therefore some of the more recently developed applications of  $^7\text{Be}$ , particularly in atmospheric, erosion, and sedimentation studies, have significant potential for evaluation of environmental problems. However, most of the applications of  $^7\text{Be}$  require a very detailed understanding of the distribution and geochemistry of  $^7\text{Be}$ , which is still somewhat incomplete in terrestrial ecosystems. In particular, more information is needed on the depth distribution of  $^7\text{Be}$  in soils with respect to different drainage conditions, bulk density, and amount of macropore flow channels. Also, the partitioning of  $^7\text{Be}$  and its residence time in lakes and the ocean is poorly understood. The effect of competing ligands and DOC on the adsorption behavior needs further documentation, as is evident in the wide range of  $K_d$ s reported (three orders of magnitude) in estuaries and tidal bays. With further studies on the geochemical behavior (see Vesely et al., this volume) and distribution of  $^7\text{Be}$  under various conditions, it will be possible to take full advantage of this nuclide in earth science investigations.

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**James M. Kaste, Stephen A. Norton, Charles T. Hess**

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