

# Cosmogenic $^{10}\text{Be}$ : A critical view on its widespread dominion in geosciences

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The radionuclide  $^{10}\text{Be}$  (half-life = 1.5 my), produced naturally in the Earth's atmosphere by nuclear interactions of cosmic rays, was sought in ocean sediments in the late fifties, considering its potential usefulness as a radiotracer for dating sediments.  $^{10}\text{Be}$  was discovered independently by two groups, one in India and the other in the USA, and used only for dating marine sediments and manganese nodules until the seventies. Subsequently, as a result of a technical advance resulting in the improvement in the sensitivity of measurement of  $^{10}\text{Be}$  by about a factor of  $10^6$ , there was a global rush to measure this nuclide in most materials participating in the physical, chemical and biological processes in the dynamic geosphere. This paper outlines the reasons for this "isotope rush", and the lessons learned from these studies. I also present my personal views of the special attractive features of this nuclide on the one hand, and on the other, the pitfalls or the wrong message this nuclide could convey!

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## 1. Introduction

The search for the radionuclide  $^{10}\text{Be}$  (half-life = 1.5 my), expected to be produced by nuclear interactions of cosmic rays in the atmosphere, began in India at the Tata Institute of Fundamental Research (TIFR), Bombay, following a theoretical paper by Peters (1955), in which he stated that conditions for observing the resulting activity of this nuclide in various regions of the Earth are favorable, and that it might be useful for measuring sedimentation rates and other geophysical surface changes during the tertiary. The potential of  $^{10}\text{Be}$  as a dating tool also led J. R. Arnold, who, with W.F. Libby, had successfully developed the cosmogenic radiocarbon method for applications in archaeology (Anderson *et al* 1947; Libby *et al* 1949), to look for it in his laboratory at the University of Chicago. The two groups, at the TIFR and the Univ. of Chicago, unaware of each other's researches on  $^{10}\text{Be}$ , looked for the radionuclide in marine sediments from the eastern Pacific Ocean and found a measurable, unambiguous signature of its activity, also at about the same time (Arnold 1956; Goel *et al* 1957). Interestingly, both groups also looked for another

cosmic ray produced isotope,  $^7\text{Be}$  (half-life = 53 days), this time Arnold's work preceding that of the TIFR group by about a year (Arnold and Al-Salih 1955). The remarkable stories of the independent searches for the cosmogenic  $^{10}\text{Be}$  by the two groups in distant continents have been written by Arnold (1991) and Lal (1991a).

The driving force for the two groups was quite correctly the urge to find a tool for dating continental and marine sediments in the time window extending to the past 5–10 my. Naturally synthesized radionuclides of half-lives  $< 10^8$  y are extinct today. No other natural radionuclides of half-life in the million-year range, except for those continually produced in nuclear interactions of cosmic rays, are available for this purpose. The longest-lived daughter nuclide in the U and Th series is  $^{230}\text{Th}$ , which has a half-life of 75,200 y. The cosmogenic  $^{10}\text{Be}$  did serve as a dating tool in the million-year range, but its measurements were difficult, requiring samples containing  $> 2 \times 10^{12}$  atoms, to result in activity of  $> 1$  disintegration per minute ( $^{10}\text{Be}$  is a beta emitter, with  $E_{\text{max}} = 0.555$  MeV). Extraction of  $^{10}\text{Be}$  from large amounts of sediments ( $\sim 100$  g) involved highly selective and

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time-consuming radiochemical methods. Its studies were therefore limited to a few tough experimenters such as Somayajulu (1977), who studied it in marine sediments with a view to determine sediment accumulation rates. The work of Somayajulu also led to a very important realization, namely that  $^{10}\text{Be}$  could be used to determine the growth rates of marine manganese nodules, for which he determined growth rates of the order of mm/My, a thousand times slower than the slowest accumulating red clay sediments in the Pacific Ocean (Somayajulu 1967). Only in these marine accretions and in other authigenic phases, were the  $^{10}\text{Be}$  activity levels high enough to permit its measurement by beta-counting, as was done until the early eighties (Amin *et al* 1966, 1975; Somayajulu 1977; Sharma and Somayajulu 1982; Somayajulu *et al* 1984a).

## 2. The great isotope rush of the '80s

In the late seventies, two groups, one in Canada (Nelson *et al* 1977) and one in the USA (Bennett *et al* 1977), independently showed that  $^{14}\text{C}$  could be detected at natural levels by using electrostatic tandem accelerators. In this technique, sample carbon atoms are sputtered using caesium ions and accelerated in the tandem as negative ions to MeV energies, and the  $^{14}\text{C}$  atoms are directly identified in the mass spectrometer. Thus it becomes possible to identify  $^{14}\text{C}$  ions in the beam, and measure their abundance relative to stable carbon isotopes, without having to wait for the  $^{14}\text{C}$  atoms to decay. The AMS technique works efficiently for several nuclides ( $^{14}\text{C}$ ,  $^{41}\text{Ca}$ ,  $^{36}\text{Cl}$ ,  $^{26}\text{Al}$ ,  $^{10}\text{Be}$  and  $^{129}\text{I}$ ), permitting detection of  $\sim 10^6$  atoms in each case (cf. Elmore and Phillips 1987), provided the isotope/element ratio is  $< 10^{-14}$ , yielding a gain factor over isotope detection by the decay-counting method, of  $1/10^6\lambda$ ; the disintegration constant,  $\lambda$ , is expressed in ( $\text{min}^{-1}$ ) to correspond to the detection limit of 1 dpm by the counting method. The 4–6 orders of magnitude higher isotope measurement sensitivity meant a large reduction in sample sizes, from 10–100 g to (1–10) mg, and in turn reopening a host of investigations carried out earlier using the decay-counting method, for a deeper introspection. The decrease in sample size meant:

- economy on precious samples, e.g. the extraterrestrial and air-filter samples;
- making measurements not possible earlier with the sample sizes available, e.g. nuclide time series in ice cores;
- making adequate number of measurements to characterise geochemical entities, e.g. marine sediments and weathered soil profiles;
- measuring concentrations of cosmogenic nuclides produced *in situ* in terrestrial solids.

The principal considerations in choosing a radio-tracer are:

1. its half-life,
2. its chemical behavior,
3. knowledge of its source function(s), and
4. ease of its detection.

The interpretation of the tracer data would have to explicitly take into account the first three parameters, of course, and importantly whether supplementary data on other elements/isotopes could be brought to bear in constructing robust geophysical/geochemical models of transport/mixing. These are important but generally complex issues needing careful attention. Historically, geochemists have always rushed in at the availability of techniques to follow a tracer through different reservoirs, and only later on, they have examined the critical issues 1 thru 3 in the light of the experimental data. This strategy rests on two premises: (i) opportunities must be seized and (ii) all tracer data are useful. In studies of global scale phenomena it is generally difficult to forecast the tracer behavior, and it may seem prudent to carry out an experiment first and to think later, with the data in hand. This approach gets justified often, with occasional big surprises.

With this background it is easy to see why there was a great isotope rush in the eighties, when the AMS technique became available in the USA, Canada and Europe to the scientific community. The first wave of AMS studies dealt with measurements of  $^{10}\text{Be}$  in extraterrestrial samples, in marine sediments and in manganese nodules. There are far too many published papers for one to cite here; only a few references are given here as examples. These studies confirmed earlier data acquired the old fashioned way, but generated a new extensive data base. The next wave concerned acquisition of new data on  $^{10}\text{Be}$  not obtainable using the decay counting method:

1. In wet precipitations (Somayajulu *et al* 1984b; Monaghan *et al* 1985/86),
2. in polar ice cores (Raisbeck *et al* 1981; Beer *et al* 1984),
3. in oceanic water columns (Kusakabe *et al* 1982; Measures and Edmond 1982),
4. in marine sediments and manganese nodules (Sharma and Somayajulu, 1982; Ku *et al* 1982; Eisenhauer *et al* 1994; Sharma *et al* 1987; von Blanckenburg *et al* 1996),
5. in weathered soil profiles (Pavich *et al* 1986; Barg *et al* 1997),
6. in aeolian dust deposits (Heller *et al* 1993; Gu *et al* 1996, 1997),
7. in terrestrial rocks (Yiou *et al* 1984; Nishiizumi *et al* 1993), and
8. in diverse terrestrial samples where  $^{10}\text{Be}$  could be measured (Raisbeck and Yiou 1984; Bourles *et al* 1984; Valette-Silver *et al* 1987).

Several of these studies also combined measurements of other cosmogenic radionuclides,  $^{36}\text{Cl}$  and  $^{26}\text{Al}$ , but  $^{10}\text{Be}$  was the principal nuclide studied. Note here the important achievement of AMS in obtaining time series of  $^{10}\text{Be}$  in polar ice cores. In 1955, the author processed about 200,000 gallons of ice in Kashmir to measure its concentration. This measurement was imprecise because of a weak  $^{10}\text{Be}$  signal (Lal 1991a).

Armed with the new data, we can now look at our gains in geosciences, focussing our attention on  $^{10}\text{Be}$ , the most widely studied cosmogenic tracer today, next to cosmogenic  $^{14}\text{C}$ . Natural radiocarbon is present in all living organic matter at a ratio  $^{14}\text{C}/^{12}\text{C} = 1.17 \times 10^{-12}$ , with the result that 50 mg of living carbon has  $2.5 \times 10^6$  atoms, easily detectable with the AMS. The dynamic carbon in the carbon cycle has specific activities within about 20% of this value, and is easily measurable in samples of  $< 1$  mg C. Samples of “age” up to  $\sim 30$  ky, “age” defined as the time of isolation from the carbon cycle, can be studied with the AMS.

In the case of  $^{10}\text{Be}$ , the  $^{10}\text{Be}/^9\text{Be}$  ratios in the principal dynamic reservoirs in which it is diluted with its stable isotope  $^9\text{Be}$ , namely soils, groundwaters, oceans, biogenic and authigenic matter in the hydrosphere, are several orders of magnitude higher than  $10^{-14}$ , the lower range for  $^{10}\text{Be}$  measurement using the AMS. Furthermore, since beryllium is particle-active (You *et al* 1989; Anderson *et al* 1990), i.e. it is associated with particles in aqueous systems by virtue of its preferential attachment to inorganic and organic surfaces, it attaches first to surficial solids, and is subsequently widely distributed among the dynamic constituents of the geosphere. (For a review of the mixing histories of  $^{10}\text{Be}$ , reference is made to McHargue and Damon 1991; and Morris 1991). The resulting concentrations of  $^{10}\text{Be}$  in surficial soils and in marine sediments (CaCO<sub>3</sub>-free basis) are about  $10^7$ – $10^9$   $^{10}\text{Be}$  atoms/g with  $^{10}\text{Be}/^9\text{Be}$  ratios in the range of  $10^{-8}$ – $10^{-7}$ . Tens of mg, or a gram at most, suffice to measure  $^{10}\text{Be}$  in all recent authigenic materials, a great convenience in its study. Note that although its production rate is about two orders of magnitude lower than that of  $^{14}\text{C}$ , the comparable ease of measurement in the case of  $^{10}\text{Be}$  arises because (i) the dilution factor (with its stable isotope) is much smaller for  $^{10}\text{Be}$ , and (ii) its longer half-life results in its accumulation in the geosphere over longer periods of time.

Thus, it is seen that  $^{10}\text{Be}$  can be measured in tens of mg samples of continental materials from the dynamic exchange reservoirs, and in authigenic materials on the ocean floor. In the case of  $^{14}\text{C}$  it can be measured in milligram size carbon samples from the carbon cycle reservoirs. In the earlier days of applications of radiotracers, major attention was directed toward development of techniques for their sensitive measurements. Today, the primary focus is on identification of principal tracer pathways, on their measurement

in different reservoirs of importance, and on building robust models. From studies of tracers, we hope to learn of their pathways through the geophysical and geochemical discontinuities. They teach us about the behaviors of natural systems. With this realization, geoscientists rushed to measure their distribution in space and time. Therein lies the justification for the great isotope rush of the eighties, which continues at a fairly high level of activity in the nineties, for both  $^{14}\text{C}$  and  $^{10}\text{Be}$ .

For some of us,  $^{10}\text{Be}$  has become our *raison d'être*, inasmuch as some of us develop their researches around one or more stable isotopes, e.g.  $^{18}\text{O}$ ,  $^{13}\text{C}$ , or radionuclides, e.g. the radioactive decay schemes, e.g.  $^{147}\text{Sm}$ – $^{144}\text{Nd}$ ,  $^{40}\text{K}$ – $^{40}\text{Ar}$ , or the daughter nuclides in the U,Th series. *Studies of  $^{10}\text{Be}$  have assumed the stature of an industry operated by geoscientists, for characterizing physical, chemical and biological systems.*

### 3. $^{10}\text{Be}$ : Its comparison with $^{14}\text{C}$ as a radiotracer

*Isotopes, like everything else, are not created equal.* The cosmogenic  $^{14}\text{C}$  finds very diverse applications, because it is mixed within the dynamic carbon cycles, and serves as a tracer for (i) mixing time scales of carbon within the carbon cycle reservoirs, and (ii) for dating the time since a sample containing carbon was separated from any of the carbon cycle reservoirs, with the  $^{14}\text{C}/^{12}\text{C}$  ratios providing robust constraints on mass transfer/mixing processes. This is so because the exchange of  $^{14}\text{C}$  between, and the flux of  $^{14}\text{C}$  out of, carbon cycle reservoirs occurs along with the stable carbon isotopes  $^{12}\text{C}$  and  $^{13}\text{C}$ , with which it is well mixed in organic/inorganic compounds in the carbon cycle reservoirs. It is therefore expected that the pathways of stable C and  $^{14}\text{C}$  are the same in all carbon cycle reservoirs except for any chemical isotopic fractionation, which can be properly taken into account.

The greatest handicap of  $^{10}\text{Be}$  as a tracer is that its pathway through the atmosphere, hydrosphere and lithosphere neither “relate” to, nor are “parallel” to, those of beryllium’s stable isotope,  $^9\text{Be}$ . In the atmosphere and in wet precipitations,  $^{10}\text{Be}$  and  $^9\text{Be}$  pathways are mostly not “related”. In the oceans,  $^{10}\text{Be}$  is primarily introduced via wet precipitations, whereas  $^9\text{Be}$  sources are “run-off” and “aeolian” transport of dust/soils. On the other hand, the single-most unique attribute of  $^{10}\text{Be}$  over  $^{14}\text{C}$  is its half-life.  $^{14}\text{C}$  covers only the recent period in the Quaternary, while the half-life of  $^{10}\text{Be}$  equals the duration of the Quaternary and should permit addressing questions within the recent Tertiary, back to about 7–8 my, taking five tracer half-lives as the range of its applicability.

By virtue of its longer half-life,  $^{10}\text{Be}$  considerably extends the range of time in “dating” continental and marine sediments, and in the study of geomorphic

processes. These should in fact be considered principal applications of the cosmogenic radiotracer  $^{10}\text{Be}$ . Note here, however, that since the  $^{10}\text{Be}$  dates must be based on some initial concentrations which cannot be estimated accurately (compare the case of  $^{14}\text{C}$ , where dates are based on the known initial  $^{14}\text{C}/^{12}\text{C}$  ratios), the uncertainties in  $^{10}\text{Be}$  age estimates are large,  $> (10-20)\%$ .

Limitations in the use of cosmogenic  $^{10}\text{Be}$  produced in the Earth's atmosphere as a tracer often arise due to our inability to model a geophysical/geochemical reservoir. In the simplest case, robust tracer models can be constructed if the radiotracer is "conservative", meaning that it strictly follows the pathways of the reservoir matrix. Such situations are rare indeed, for example, the rather unique case of  $^3\text{H}$ , which would be expected to faithfully track water molecules within and between watery reservoirs; the handicap of  $^3\text{H}$  is its short half-life, 12.3 y. The non-availability of conservative radiotracers is not an obstacle, however, because often it becomes possible to use non-conservative tracers very effectively. The cosmogenic  $^{14}\text{C}$  and  $^{32}\text{Si}$  are two examples of non-conservative tracers, which are very useful for studying large-scale vertical mixing and biological processes in the oceans.

In view of its chemical behavior, the principal handicap in the case of  $^{10}\text{Be}$  is often the lack of knowledge of its source functions—because it is fractionated from the atmosphere soon after its formation in the atmosphere, due to attachment on aerosols. Thereafter it is eventually removed from the lower atmosphere principally by wet precipitations. However, based on our knowledge of the large-scale circulation of air, and aerosol scavenging processes, we have a fair idea of the geographic and seasonal pattern of its fallout (Lal and Peters 1967). These considerations help determine the oceanic injection pattern of  $^{10}\text{Be}$ .

Interestingly,  $^{10}\text{Be}$  studies provide information even when timescales involved in its transport/mixing are very short, say, less than a few thousand years. In these applications,  $^{10}\text{Be}$  is being treated as a "stable" tracer; i.e., its concentrations in a reservoir are related to one or more processes of interest which control its movement to, within and out of the reservoir(s). The application of  $^{10}\text{Be}$  in studies of atmospheric circulation, aerosol scavenging by wet precipitations, climatic variations in precipitation, changes in its atmospheric production rates (due to temporal changes in cosmic ray intensity) and biological/gravitational processes occurring in the oceans, are all examples where the half-life of  $^{10}\text{Be}$  does not enter in the models. Thus,  $^{10}\text{Be}$  serves as a "label" for air, aerosols, water and soils. In studies of soils, marine sediments and manganese nodules, its half-life is of principal consequence in models which, in their simplest form, assume that  $^{10}\text{Be}$  is quickly removed to the sediment/nodule in short time periods ( $< 1000$  y) after production in the atmosphere. Some interesting applications

of  $^{10}\text{Be}$  have been discussed by Brown (1987). Special mention is made here to studies of soil erosion and island-arc volcanism (see also Morris, 1991).

The applications of cosmogenic *in situ* produced  $^{10}\text{Be}$  in surficial rocks are relatively straightforward. The cosmic ray intensity at sea level and at mountain altitudes are well known, and intensity changes associated with changes in solar activity and geomagnetic field intensity can be modeled fairly well (Lal 1988). Its production rate in quartz, which is widespread in terrestrial rocks, is also well known (Lal 1991b). In rocks exposed at sea level and at high latitudes, and undergoing steady-state erosion at the rate of 1 cm/1000 y, one expects a concentration of  $\sim 3 \times 10^5$   $^{10}\text{Be}/\text{g}$  quartz. Ten grams quartz are needed to get a good  $^{10}\text{Be}$  signal with the AMS. Again, it must be noted here that in short term geophysical processes,  $< 10^4$  y,  $^{10}\text{Be}$  serves as a stable cosmogenic tracer, and complements other radiotracers of shorter half-lives (Lal 1991b). For a sample of geomorphic problems which can be addressed using cosmogenic *in situ*  $^{10}\text{Be}$  and other nuclides, reference is made to Lal (1991b) and Nishiizumi *et al* (1993).

We discussed earlier that although the source function of atmospheric  $^{10}\text{Be}$  is known, it does not track air masses faithfully because of its attachment onto aerosols. This should not be considered as a serious problem invalidating its use as a tracer. In fact, an important facet of tracers is their chemical behavior, which only enhances their value provided meaningful models can be constructed. With the acquisition of a large data base of concentrations of  $^{10}\text{Be}$  in polar ice cores, it becomes clear that the crude scavenging models which do not explicitly consider air circulation patterns, vapor fluxes and tracer chemical behaviors, are not useful. Since several cosmogenic nuclides are available, well constrained models can be constructed. Similarly, one realizes that oversimplified models of incorporation of atmospheric  $^{10}\text{Be}$  in soils are not useful. One has to explicitly consider  $^{10}\text{Be}$  losses due to surficial run-off and erosion. Furthermore, it is necessary to include other tracers to constrain the models (cf. Barg *et al* 1997).

Before concluding our discussion on  $^{10}\text{Be}$  applications, we would like to point out that although the pathways of  $^{10}\text{Be}$  and  $^9\text{Be}$  are generally quite different, the  $^{10}\text{Be}/^9\text{Be}$  ratios in terrestrial reservoirs (excluding the atmosphere and polar ice) lie within a narrow range (Barg 1992; Barg *et al* 1997). The extensive  $^{10}\text{Be}$  data in ocean waters, in continental and marine sediments, and authigenic minerals including the manganese nodules, shows that whereas their  $^{10}\text{Be}$  concentrations are spread over several orders of magnitude, the ratios remain within an order of magnitude (Barg 1992; von Blanckenburg *et al* 1996; Barg *et al* 1997). As should become evident, this makes it possible to consider viable models for mass fluxes and chemical/biological processes in which beryllium isotopes participate.

#### 4. Epilogue

Cosmogenic <sup>10</sup>Be is continually produced on the Earth: in the atmosphere and in surficial matter, principally in nuclear interactions of cosmic rays with oxygen, with a production rate that makes it easy to measure its concentration in diverse materials participating in biogeochemical cycles. Its half-life and its chemical behavior make it an ideal tracer to study large-scale circulation of aerosols in the atmosphere and a wide range of particles in the oceans (aeolian and biogenic). The chronology of accumulation of matter in the hydrosphere can be determined from its measured concentrations in these deposits, back to 7–8 my. The rate of <sup>10</sup>Be production in the atmosphere and in surficial materials can be determined for the cosmic ray fluxes as measured during the past five decades. The radiotracer <sup>10</sup>Be is thus unique in several respects, but it should be realized that interpretation of data on its concentrations in the geosphere requires suitable models to appropriately consider the various processes which control its transport through the Earth's dynamic reservoirs. Studies of <sup>10</sup>Be have often made us realize that we know so little about how the atmosphere, oceans and the lithosphere evolve through an ever-changing Earth. Occasionally, <sup>10</sup>Be time-series data have revealed large temporal oscillations, which must be interpreted as either due to changes in its atmospheric production rate (due to modulation of cosmic ray flux by Sun, changes in the geomagnetic dipole moment, or short term increases due to a nearby supernova, etc.), or due to climatic fluctuations leading to an appreciable change in its concentrations from the “normal”. These data have been interpreted differently by researchers (cf. Raisbeck *et al* 1987). From the foregoing, it should be obvious that it is necessary to first understand the transport pathways of <sup>10</sup>Be, and its sensitivity to climatic forcing, before drawing any firm conclusions about past changes in cosmic ray intensity.

Studies of cosmogenic nuclides began in India at TIFR, Bombay, with its first success of discovering <sup>10</sup>Be in 1956, which was followed by the discovery of nine more cosmogenic nuclides within a few years (Lal and Peters 1967). Those were exciting days of new searches and rewards, for members of a group which was christened the “geocosmophysics” group. It stood out as a dynamic entity by demonstrating its capacity to discover new facts about nature. The group derived its energy from synergistic processes within the group, composed of scientists working in diverse fields, freelance. It was composed of personnel who had made their mark in studies of cosmic rays and elementary particle physics. Prof. K. Gopalan joined the group in 1970, and infused everyone with new possibilities which could be addressed using the mass spectrometer. Our resources were limited, although TIFR scientists were better endowed than other groups in India. For

this group there were no limits—not even the availability of an instrument, because its scientists had the confidence to make them with their own hands, literally, which K. Gopalan did: he made a mass spectrometer tube by carving out a mould in a graphite block, freehand. This mass spectrometer is still functional, at the Physical Research Laboratory (PRL), Ahmedabad. As mentioned earlier, members of this group used to bounce ideas off each other; Dr. Gopalan played his role actively, both as a catcher and a bouncer. To this day we are occasionally showered with his new ideas, many of which we have not yet been able to put to practice to date. To this modest idea-man, a creative and hard working colleague, I dedicate this article. Prof. Gopalan helped build state-of-the-art programs in climatology and geochronology at the Physical Research Laboratory (PRL) during his stay there from 1973–1984. Even after having moved to the National Geophysical Research Institute (NGRI), he continues to help PRL and NGRI develop good programs and instruments at the cutting edge of earth science research.

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