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Development of cosmogenic nuclear methods for the study of soil erosion and formation rates

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The importance of studying soil formation and erosion rates in geomorphology, biological production and carbon sink source functions cannot be over-emphasized. In this paper we discuss two quantitative nuclear methods for the study of soil formation rates using the cosmogenic radionuclide ^{10}Be and for determination of soil erosion rates using the cosmogenic radionuclide ^{10}Be . The radionuclide ^{10}Be (half-life = 1.5 m.y.) produced by cosmic rays in the earth's atmosphere, and *in situ* cosmogenic ^{14}C (half-life = 5730 yr) have both been detected in soils/rocks using accelerator mass spectrometry¹⁻⁵. ^{10}Be concentrations have been studied in several soil samples; they have yielded useful but qualitative ideas about soil dynamics. For determining *soil formation rates*, we propose studying the specific activities of ^{10}Be in soil minerals with respect to its stable element ^9Be . In our model, backed by some laboratory tests and analyses of samples analysed earlier for ^{10}Be , we expect that the $^{10}\text{Be}/^9\text{Be}$ ratios in soil minerals formed at different depths would constitute a fairly quantitative measure of the soil formation rate. For measuring *soil erosion rates*, we propose to study the *in situ* produced ^{14}C activity in quartz in surface soil samples. The *in situ* radiocarbon method is ideal⁶ for study of erosion rates of 10^{-3} to $5 \times 10^{-2} \text{ cm yr}^{-1}$. Soil renewal rates and 'low' soil erosion rates fall in the bracket 5×10^{-3} to $2.5 \times 10^{-2} \text{ cm yr}^{-1}$ as pointed out by Revelle⁷. Appreciable aeolian contributions of quartz may occur in certain soils, but this does not constitute any problem since the aeolian quartz can be separated from the *in situ* generated quartz by grain-size separation.

It is generally recognized that a significant step forward would be to quantify soil erosion and formation rates (cf. Birkeland⁸). Soil formation by weathering of rocks and minerals is one of the most important processes

operating on the surface of the earth⁹. Soil studies are important in geomorphology, in helping deciphering climatic/vegetation changes during the Quaternary and older periods. Further, the carrying capacity of the earth for human beings must depend on its potential for sustainable biological production. Revelle⁷ has emphasized that, to meet future human food and fiber demands and to predict global climatic changes due to increasing carbon dioxide, it is now necessary to have a much better understanding of the processes of soil formation. This knowledge is essential for predicting future changes in soil characteristics as well as to chart out a proper strategy for soil management. Studies of soil dynamics are also very relevant to the source and sink functions of carbon.

The characteristic processes involved in surface weathering of rocks and soil evolution have been elucidated by a number of recent studies¹⁰⁻¹², but generally a quantification of 'time', and therefore rate, is missing in these investigations. Several attempts have, however, been made to introduce time-scales in soil processes using ^{14}C produced in the atmosphere by cosmic ray neutrons, U-Th series nuclides and the TL method. These methods do provide some information about soil processes and time-scales, but do not yield the required information.

To date, the studies of cosmogenic ^{10}Be in soils have been most successful (cf. Brown¹). The half-life of ^{10}Be is 1.5 m.y.; this nuclide is produced in the atmosphere¹³ by cosmic ray interactions with N, O nuclei, and introduced into the soil with rain-water which scavenges atmospheric ^{10}Be . Brown and Pavich, and their colleagues^{1-2,14} were able to document the retention of ^{10}Be by soils and removal of soil ^{10}Be by

erosion. Rates of saprolite formation have been obtained^{8,12} from material balance studies; this information compares reasonably well with deductions based on ¹⁰Be studies¹⁵. A serious problem with ¹⁰Be is that it is found to be mobile within highly acidic soils (pH ≤ 5), making it difficult to quantitatively model erosion/formation rates in some soils, such as highly acidic oxisols.

Pavich *et al.* presented a simple equation that related ¹⁰Be soil inventories to time-averaged deposition flux, radioactive decay and soil surface erosion. They argued that loss in solution has little effect on observed inventories.

In this paper, we discuss the feasibility of using the two cosmogenic radionuclides ¹⁴C and ¹⁰Be in soils to study soil erosion/formation rates. In the case of ¹⁴C, we are interested in studying the ¹⁴C produced *in situ* in quartz in soils (and not the atmospheric ¹⁴C fixed in the soil). In the case of ¹⁰Be we are interested in the atmospheric ¹⁰Be which was studied by Brown and Pavich (*op. cit.*), but with a difference; we would study ¹⁰Be/⁹Be ratios in authigenic silicates and oxides. The rationale for the two studies is explained below in some detail.

Soil erosion rates

We propose to study ¹⁴C produced *in situ* in quartz in soil by nuclear interactions of cosmic rays. Quartz has a tight crystal structure, and one can unambiguously measure *in situ* ¹⁴C produced in it just as *in situ* ¹⁴Be was studied in quartz by Nishiizumi *et al.*¹⁶ *In situ* ¹⁴C has been found in rocks by Jull *et al.*⁴, in ice¹⁷ and in quartz extracted from rocks by Lal *et al.*⁵. For a review of *in situ* cosmogenic nuclides, see Lal¹⁸. The *in situ* concentration of ¹⁴C is a measure of the rate of erosion since erosion continuously changes the rate of production at any given point. From surface, the production rate decreases by a factor of *e* at a depth of ~ 60 cm. For a constant steady-state erosion, the isotope concentration at the surface of a soil chronosequence reaches a stationary value (cf. Lal and Arnold¹⁹), of *N* (atoms g⁻¹):

$$N = \frac{P}{\lambda + \mu e}, \quad (1)$$

where *P* is the nuclide production rate (atoms/g sec) at surface, λ the nuclide disintegration constant (sec⁻¹), μ the absorption coefficient for cosmic rays in the rock (~ 1/60 cm⁻¹), and *e* the erosion rate (cm sec⁻¹). The value of 60 cm for 1/ μ is based on mean absorption distance of 150 g cm⁻² for cosmic rays¹³ and an *ad hoc* value of 2.5 g cm⁻³ for the density of soil.

In situ ¹⁴C is ideal for estimating erosion rates in the vicinity of $e \sim \lambda/\mu$ (see eq. 1), say for values of

$10^{-3} < e < 3 \times 10^{-2}$ cm yr⁻¹. Soil renewal rates and 'low' soil erosion rates fall in the bracket 1 to 5 tons ha⁻¹ yr⁻¹, as pointed out by Revelle⁷. This corresponds to erosion rates of 5×10^{-3} to 2.5×10^{-2} cm yr⁻¹, well in the optimum range for ¹⁴C. The production rate of ¹⁴C at sea-level and latitudes ≥ 50° is estimated to be ~ 20 atoms ¹⁴C/g quartz yr; the corresponding value at 2000 m altitude is ~ 10² atoms ¹⁴C/g quartz yr. For details reference is made to Jull *et al.*⁴ and Lal⁶. Quartz samples of the order of 50 g would have to be studied to measure the *in situ* ¹⁴C in soils exposed near sea-level; the corresponding value would be an order of magnitude smaller for soils at 2000 m altitude.

Implicit in eq. (1) is the assumption that the soil studied formed *in situ* by weathering of rock, and that no addition of soil/quartz occurred during its evolutionary history. Deposition of fluvially transported soil and aeolian quartz are examples of addition. The fluvially transported material will carry a ¹⁴C 'memory' of its past history, but one can check for the magnitude of this transport by examining the depth variation in the concentration of ¹⁴C. The *in situ* produced ¹⁴C is expected to show an exponential depth variation with a mean distance of 60 cm. Similarly, it is possible to eliminate any problems caused by admixtures of aeolian quartz in the soil profile. The aeolian quartz component can be largely discriminated against by selecting quartz of > (10–20) μm. In fact it would be useful to study the ¹⁴C concentrations of fine- and coarse-grained quartz to determine the aeolian component in a soil sample.

Rates of formation of soils/soil minerals

For determining the *rates of formation of soils*, we will use the cosmogenic radionuclide ¹⁰Be (half-life 1.5 m.y.) produced in the atmosphere by interactions of cosmic rays with the atmospheric nuclei nitrogen and oxygen. After production in the atmosphere, ¹⁰Be soon attaches to aerosols. It is then removed from the atmosphere by wet precipitations. Based on experiments with rain-water, it is known that cosmogenic ⁷Be is in solution¹³. The same behaviour would be expected of ¹⁰Be. We postulate that most of the ⁹Be is released in solution from feldspar and other beryllium-bearing minerals in the upper acidic soil layers. After entering a soil-solution, ⁹Be and ¹⁰Be would soon preferentially be scavenged by an *in situ*-formed aluminium silicate or some hydroxide, notably Al(OH)₃ and/or Fe(OH)₃. A direct parallel to this is given by our recent experiments²⁰ based on passing sea-water through an interwoven mesh of fine iron and aluminium metal fibers. This matrix is found to be very efficient for picking up a large number of cations and anions. We

have checked on the trapping efficiency of this system for a number of species, including PO_4 and bacterial particles; the result is close to one hundred per cent trapping even at space velocities of ~ 1000 (space velocity is defined as the number of column volumes passed through the column, per hour). Our experiments revealed that the reason for this is that the redox reactions in the column produce appreciable amounts of colloidal $\text{Al}(\text{OH})_3$ which is (i) a very efficient scavenger, and (ii) very refractory. As long as the colloidal hydroxides (or silicates) are not mineralized in a soil layer, there occurs an efficient downward flux of ^{10}Be and ^9Be through this layer, but as the mineralization progresses, the downward flux decreases. Any beryllium that is scavenged will remain tightly adsorbed and that not scavenged by oxyhydroxides will be mobile. We would expect that $\text{Fe}(\text{OH})_3$ and $\text{Al}(\text{OH})_3$ would both serve as efficient scavengers, but there may be basic differences in their scavenging rates. One would expect that the $^{10}\text{Be}/^9\text{Be}$ ratio would be the same in different authigenic minerals in which beryllium is adsorbed efficiently. The same would be expected for *in situ* aluminium silicates, e.g. kaolinite formed in the soil horizon.

In its essentials, our ^{10}Be soil model is based on a homogenization of the rain-water ^{10}Be in the A (and E?) horizons of the soil profile with the dissolved ^9Be . The low pH in these horizons ensures a good supply of leached ^9Be in soil solution and a minimal removal of ^{10}Be and ^9Be to hydroxides. The lower soil horizons then receive the dissolved radiotracer ^{10}Be and ^9Be in a certain ratio. For soil waters in the deeper sections where pH exceeds 6, no significant addition of ^9Be would be expected to the soil water. However, both ^{10}Be and ^9Be would be removed from the soil solution and incorporated into authigenic silicates and hydroxides/oxides. Some of the minerals formed may redissolve due to leaching action of the soil water but the residual authigenic minerals will retain the $^{10}\text{Be}/^9\text{Be}$ ratios fixed in the A-horizon.

We have carried out a number of tests to check on the validity of the above model, e.g. depth variations in the ^9Be concentrations of soil water and of the total soil sample. We determined ^9Be concentrations in a number of samples where ^{10}Be concentrations have been measured earlier^{2,14,21,22}. We have studied the total ^9Be concentrations in a total of ~ 50 soil samples from (i) three cores M4, M6 and M8 from Yugoslavia (Pavich, unpublished), (ii) one profile¹⁴ from China Hat formation in Merced Terrace, N. Calif., (iii) soil samples (40–150 cm depth) from the Atlantic coast, N. America^{2,14}, five soil samples from the N. American river basins²³, and samples (20–80 cm depth) from the Tatun bauxite profile from Taiwan²². As stated earlier, the ^{10}Be concentrations have been reported for these samples by the respective authors (references cited). The results are plotted in

Figures 1–3 for all the samples. The following observations can be made from the limited data so far:

- (i) The ^{10}Be – ^9Be concentration plots (Figures 1–3) show a positive correlation over a wide range of ^{10}Be concentrations $(60\text{--}2500) \times 10^6 \text{ atoms g}^{-1}$. The slope, γ , in the relation

$$C(^{10}\text{Be}) = \text{const.} \times [C(^9\text{Be})]^\gamma \quad (1)$$

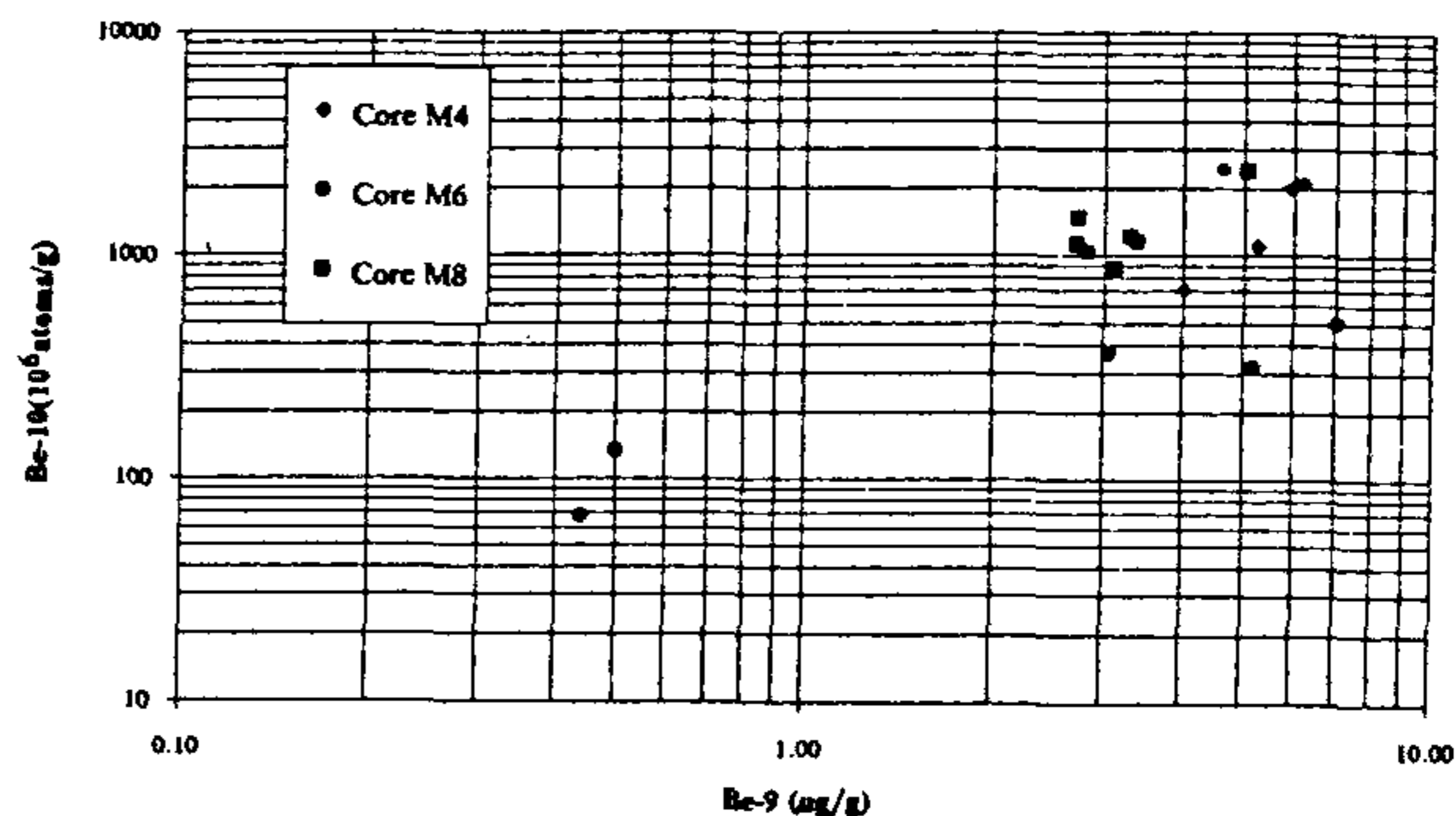


Figure 1. ^{10}Be and ^9Be concentrations in Yugoslavia soils; core M4, core M6, and core M8.

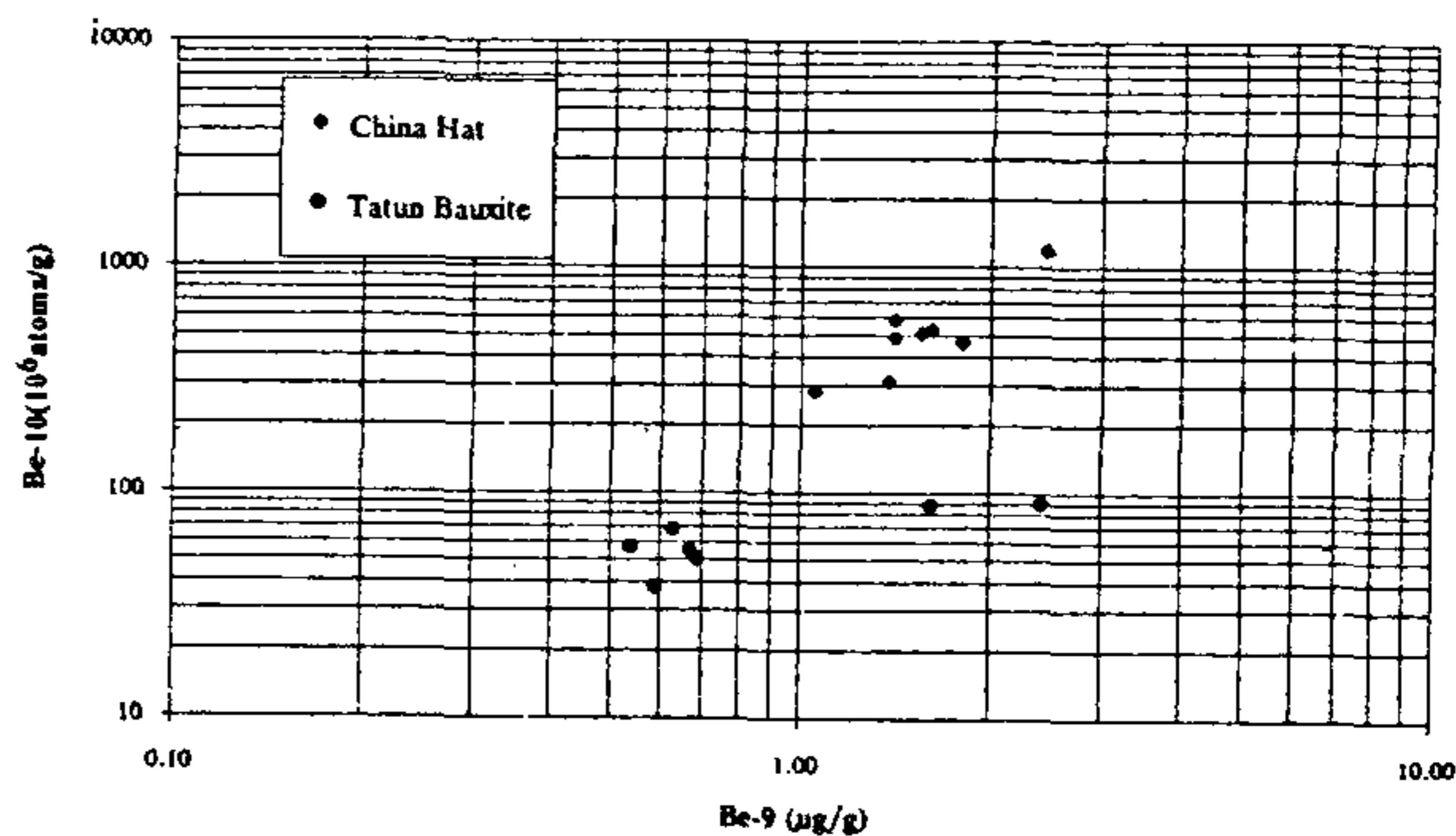


Figure 2. ^{10}Be and ^9Be concentrations in China Hat soils and Tatun bauxite.

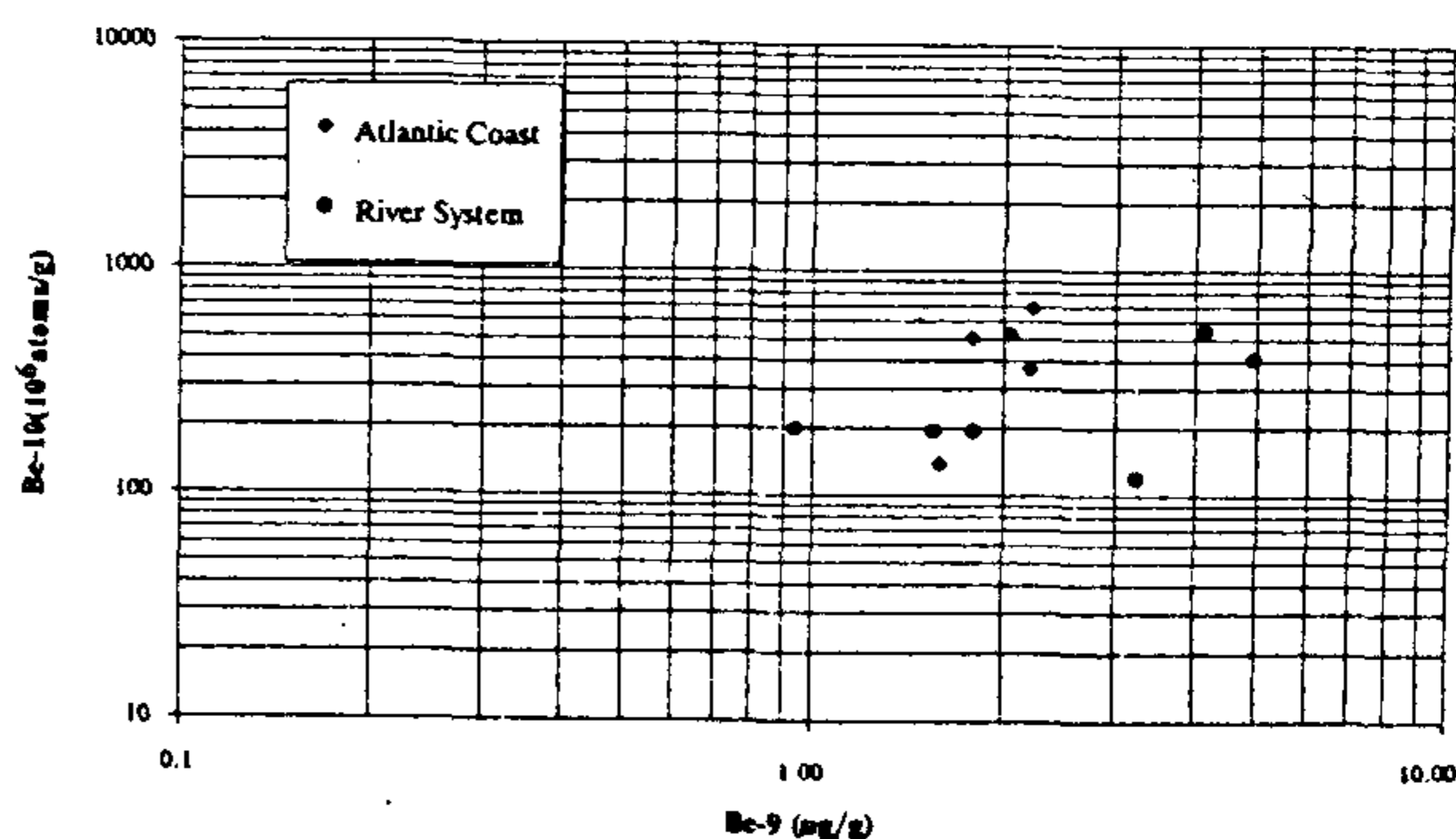


Figure 3. ^{10}Be and ^9Be concentrations in soils from Atlantic coast and river system.

is of the order of ~ 2 for the Yugoslavia soil samples M4 and M8 and ~ 1 for M6. The value of γ is ~ 1.5 for the China Hat samples. (Note that the $^{10}\text{Be}/^9\text{Be}$ ratios in soils are about two orders of magnitude lower than those in the sea water and in marine manganese nodules²⁴.

(ii) The absolute ^{10}Be concentrations differ by $>40\times$ in the samples studied, but within a core, the variations are small, usually within a factor of 2–3. The standard deviation on the mean varies from 25–60%.

(iii) The corresponding range in the standard deviation on the mean ^9Be concentration is 15–45%. Absolute ^9Be concentrations range between 0.6 and 6 ppm.

Within a core the $^{10}\text{Be}/^9\text{Be}$ ratios, analogous to ^9Be concentrations, show smaller variations. The results for two cores where several samples were analysed are given in Table 1.

The observed smaller scatter in $^{10}\text{Be}/^9\text{Be}$ ratios (Table 1) makes a good case for the model discussed above for the incorporation of ^{10}Be and ^9Be in approximately the same ratio in the soil minerals at the time of their formation. The observed slopes, γ in eq. (1) also support the model. Rewriting eq. (1) in terms of the ratio, R [$^{10}\text{Be}/^9\text{Be}$]:

$$R [^{10}\text{Be}/^9\text{Be}] = \text{constant} \times [C(^{10}\text{Be})]^\alpha, \quad (2)$$

$$\text{where } \alpha = (\gamma - 1)/\gamma. \quad (3)$$

The slope α in eq. (3) lies between 0.3 and 0.5 corresponding to $\gamma = 1.5$ –2. This is also expected because of the progressively larger ^{10}Be decay in the older samples.

Thus, we see that the results obtained so far do support the model of a nearly uniform $^{10}\text{Be}/^9\text{Be}$ ratio at formation of the soil. It should be noted here that in our present studies, we have analysed ^9Be in the whole soil sample, and not in the soil minerals. We would expect a much smaller scatter in the $^{10}\text{Be}/^9\text{Be}$ ratios in the soil minerals.

Our model explains well the large variability observed in the depth profiles of ^{10}Be concentrations in total soil samples. The concentration minimum usually observed in the upper 50 cm is apparently due to the high mobility of ^{10}Be in low pH soil solutions. The intermediate broad maxima at depths of ~ 1 –1.5 m signify incorporation of ^{10}Be and ^9Be in authigenic minerals. The low ratios at depths, close to the bedrock are indicative of young soil layers with insufficient incorporation of ^{10}Be and ^9Be . Thus, the results above seem to validate the potential usefulness of $^{10}\text{Be}/^9\text{Be}$ ratio for determining ages of soil minerals. The success of the proposed method of dating soils would depend totally on how well ^{10}Be is retained in the mineral

Table 1. Mean concentrations of ^{10}Be and ^9Be in two cores

Sample code (Number of samples)	Mean concentrations*		Mean ratio, $^{10}\text{Be}/^9\text{Be}$ (10^{-9} atom/atom)
	^{10}Be (10^6 atoms g^{-1})	^9Be (10^{-6} g g^{-1})	
Yugoslavia ¹ Core, M8 (7)	1400 \pm 510	3.3 \pm 0.81	6.0 \pm 1.3
China Hat ² (8)	540 \pm 280	1.58 \pm 0.41	4.9 \pm 1.2

*The errors quoted are 1 σ values.

1 ^{10}Be data are from Pavich (private communication)

2 ^{10}Be data are from Pavich *et al.*¹⁴.

phases of interest. The principal question is whether ^{10}Be in clays, gibbsite, etc. could be considered to be a closed system. If so the $^{10}\text{Be}/^9\text{Be}$ ratios (and absolute ^{10}Be concentrations) would provide a dating method analogous to the radiocarbon dating:

$$T = T_m \ln(R),$$

where T is the age of the mineral, T_m is the mean life of ^{10}Be (2.16×10^6 yr), and R is the ratio of $^{10}\text{Be}/^9\text{Be}$ at formation to that at the time of measurement. In a sample of 10 m.y. age, for instance, even a 50% error in the initial ratio would result only in an error of ca. 0.8 m.y. in the age estimate.

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We have great pleasure in dedicating this paper to the memory of late Prof. Birbal Sahni, one of the founders of modern palaeobotany, in the year of his birth centenary.

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