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A comparison on the accumulation characteristics of ⁷Be and ¹³⁷Cs in lake sediments and surface soils in western Yunnan and central Guizhou, China

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Abstract

The uplifting of the Himalayas has resulted in the development of a three-tiered landform in Southern China, the Yunnan-Guizhou Plateau is the middle tier of this landform on the eastern slope of the Himalayas. Lake Hongfeng and Lake Baihua in central Guizhou are about 1000 km away from Lake Erhai in western Yunnan. Sediment cores were collected from Lakes Erhai and Lugu in west Yunnan and Lakes Hongfeng and Baihua in central Guizhou, along with 28 surface soil cores from the watersheds of these lakes. The accumulation characteristics of ⁷Be and ¹³⁷Cs show that: (1) ¹³⁷Cs activities in the soil cores of central Guizhou were higher than that in the western Yunnan. The activities and maximum penetrative depth of ⁷Be were similar for the two regions. The activity ratio of ⁷Be/¹³⁷Cs in surface soil reached 100–1000 in western Yunnan, but only 10–100 for central Guizhou region. (2) ⁷Be inventories in soil cores at most sampling sites of central Guizhou were low, reflecting severe soil erosion. ⁷Be inventories in the soil cores of Lake Lugu watershed in summer-autumn were higher than that in Lake Hongfeng/Baihua watershed and also higher than that in Lake Erhai watershed. This could be related to high ⁷Be precipitation in summer and altitude. (3) ⁷Be inventories in sediment cores of Lake Erhai, Lake Lugu and Lake Baihua were 237 ± 73 , 322 ± 19 and 783 ± 44 Bq m⁻², respectively. Based on activity values that were corrected to deposition, prior to 1986, ¹³⁷Cs inventories in these lakes were 519 ± 26 , 937 ± 13 and 3704 ± 56 Bq m⁻², respectively. (4) Modeling indicated that ⁷Be and ¹³⁷Cs accumulations in Lake Hongfeng and Lake Baihua were dominated by watershed erosion, and controlled by the retention factor in Lake Erhai and Lake Lugu, but the ¹³⁷Cs

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inventory ratios obtained in the sediment core and from direct atmospheric fallout were similar, and the ratios were smaller for ⁷Be. (5) ⁷Be inventories from direct atmospheric fallout in Lake Erhai, Lake Lugu and Lake Baihua were 0.07 ± 0.02 , 0.29 ± 0.02 and 0.08 ± 0.01 Bq m⁻², respectively. Prior to 1986, ¹³⁷Cs inventories were 0.11 ± 0.01 , 0.22 ± 0.01 and 0.37 ± 0.01 Bq m⁻², respectively, showing that ¹³⁷Cs deposition has a regional difference. This phenomenon may reflect that uplift of the Himalayas has a screening effect on the fallout of globally distributed atmospheric pollutants in western Yunnan, China. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: ¹³⁷Cs and ⁷Be; Precipitation and accumulation; Lake sediments; Surface soils; Yunnan-Guizhou Plateau of China

1. Introduction

The uplifting of the Himalayas has resulted in the development of a three-tiered landform in Southern China, and produced variable effects on global and regional climate change and air pollutant diffusion. The Yunnan–Guizhou Plateau on the eastern slope of the Himalayas is located in the geologic transfer zone between the Tibetan–Yunnan fold belt and the Yangtze craton. The Yunnan–Guizhou Plateau has large altitude gradients and complex landforms with a subtropical climate influenced by both the southeastern (Pacific Ocean) and southwestern (Indian Ocean) monsoons. To understand the effects of the Himalayan uplift on recent environmental change in the Yunnan–Guizhou Plateau, it is important to comparatively study the distribution and accumulation of fallout radio-nuclides (⁷Be and ¹³⁷Cs) in lake sediments and watershed surface soils throughout the Plateau.

Fallout radionuclides have a given input function and decay and have tracing value for geochemical processes in the environment on variable time scales (Wan, 1988). ⁷Be, half-life of 53.3 days, is produced in the atmosphere as a spallation product of O and N nuclei by bombardment of cosmic rays. ⁷Be can be rapidly delivered to sediment after being transported into lake waters (Krishnaswami and Lal, 1978), and has a mean residence time of 77 days (Robbins and Eadie, 1982; Wan et al., 1986, 1987). ⁷Be can be used as a tracer for seasonal particle mixing and transport. ⁷Be atmospheric deposition is related to precipitation (Schuler et al., 1991; Baskaran et al., 1993), therefore, ⁷Be can trace aquatic particle movement (Santschi, 1986; Wan, 1988; Baskaran et al., 1997; Feng et al., 1999a,b). It is a good tracer for seasonal particle mixing and transport (Bai et al., 1996; Blake et al., 1999; Walling et al., 1999).

¹³⁷Cs has been produced by nuclear testing, and has a half-life of 30.2 years. Its flux to the earth's surface shows a dependence on nuclear testing density, region and latitude (Wan, 1988). Discrepancy between ¹³⁷Cs vertical distribution characteristics and depositional fluxes is probably caused by ¹³⁷Cs diffusion in early-diagenesis processes, but this does not affect the locations of the peaks (Wan, 1988). Sediment accumulation rates calculated by the 1963, 1975 and 1986 time markers, respectively, show good consistency. Using fallout ¹³⁷Cs as a geochemical tracer of soil erosion has yielded good results in the tracing rates of soil erosion (Brown et al. 1981; Ritchie and McHenry 1990; Walling and Quine, 1991; Walling and He, 1999; Ritchie and Rasmussen, 2000).

We explore the regional differences in the atmospheric pollutants in the eastern slope of the Himalayas here, based on the comparison of the accumulation of ⁷Be and ¹³⁷Cs in sediments and soils.

2. Materials and methods

2.1. Study areas

Based on the authors' previous work, Lake Erhai and Lake Lugu in western Yunnan, and Lake Hongfeng and Lake Baihua in central Guizhou were selected as study areas in this paper (Bai and Wan, 1998, 1999; Wan, 1999; Xu et al., 1999).

Lake Erhai, the largest fault lake in the western Yunnan Plateau, is located in the north of Dali $(100^{\circ}5' - 17' \text{ E}, 25^{\circ}35' - 58' \text{ N})$. The surface of the lake is 1974 m above mean sea level, an area of 249.8 km² and a 2656 km² watershed. It has an average depth of 10.5 m, a maximum depth of 20.9 m, a volume of 2.88×10^9 m³, a water residence time of 2.75 years and an annual water discharge of 8.13×10^8 m³ (Wan et al., 1988). The Lake Erhai area has an average annual precipitation of 1060 mm, temperature of 15 °C, and potential evapotranspiration of 1970 mm. Lake Erhai lies in the eastern foot of the strongly uplifting Mt. Diancangshan. Its basin has an eroded geomorphic landscape with an asymmetric valley sharply incised by streams. The watershed of Lake Erhai is mainly underlain by sedimentary and metamorphic rocks, specifically carbonate and siliciclastic rocks and gneisses. Soil types are mainly Ultisols, Aquults and Fluvents. Forest coverage is 7%. Because of the influence of the carbonate bed rock and the subtropical monsoon, the lake water has the characteristics of (i) weak alkalinity (pH=8.0-8.5); (ii) low salinity; (iii) medium hardness (~8) as solutes in lake water are dominated by Ca^{2+} , Mg^{2+} and HCO_3^{-} , which represent more than 60% of the total equivalent anions and cations; (iv) ion concentrations $Ca^{2+} > Mg^{2+} > Na^+ > K^+$, and $HCO_3^- \gg SO_4^{-2} > Cl^-$ (Wan et al., 1988).

Lake Lugu is located in Ninglang county in the western Yunnan Plateau $(100^{\circ}47' \text{ E}, 27^{\circ}42' \text{ N})$ with a mean lake water height of 2685 m above sea level. It has an average depth of 40.4 m, a maximum depth of 73.2 m, water area of 50.5 km², a watershed area of 171.4 km², a volume of $20.2 \times 10^8 \text{ m}^3$, a water residence time of 18.5 years and an annual water discharge of $1.1 \times 10^8 \text{ m}^3$ (Wan et al., 1988). The Lake Lugu area has an average annual precipitation of 926 mm, average annual temperature of 8 °C, and potential evapotranspiration of 1270 mm. It is a semi-closed deep fault lake in the Jinshajiang River drainage. Lake Lugu watershed is mainly made up of limestone, mud stone and sandstone. Soils mainly include Alfisols, Oxisols, Entisols and Mollisols. Forest coverage is 47.6%.

Lake Hongfeng, the largest artificial lake in central Guizhou, was built in 1960 near Guiyang, capital of Guizhou province $(106^{\circ}26' \text{ E}, 26^{\circ}31' \text{ N})$. It has a surface area of 32 km², a volume of $2.97 \times 10^8 \text{ m}^3$, a drainage basin of 1551 km², a mean depth of 9.3 m, a maximum depth of about 50 m, water flux of $11.26 \times 10^8 \text{ m}^3 \text{ a}^{-1}$, a residence time of the water of 0.26 a, lake water height of 1233 m above sea level. It belongs to the drainage system of the Wujiang River, a tributary of the Yangtze River. Lake Hongfeng-Baihua watershed is mainly underlain by the Trias Limestone, with a well-developed karst landscape. The land is covered by Ultisols and Oxisols. Due to significant surface soil

erosion, a "rock desertification" landscape is in evidence. Hilltops are bare, thin soils remain in concave ditches and cracks and mountain slopes are covered with sparse grass and shrubs. Lake Baihua is another artificial lake 6 km downstream of Lake Hongfeng (Fig. 1).

Both western Yunnan and central Guizhou have a subtropical climate that is affected by the Southeast Monsoon (the Pacific Monsoon) and Southwest Monsoon (the Indian Monsoon). The climate is moderate with seasonal change. The annual average precipitations in western Yunnan and central Guizhou are 1000 and 1200 mm, respectively.

2.2. Sampling and analytical methods

A sediment core from Lake Hongfeng (HF880818-1, overlying water depth of 38 m) was collected in August 1988. Sediment cores of Lake Baihua (BH941003-2, overlying water depth of 20 m) and Lake Erhai (EH940713-3-2, overlying water depth of 16 m) and Lake Lugu (LG940706-2, overlying water depth of 60 m) were collected in October, 13 July and 6 July 1994, respectively, using the sediment collector described in Yuan et al. (1993). All the sediment cores were sampled from the deepest parts of the lakes. Cores were 26-90 cm long, surfaces of the cores were not disturbed and the interface water was clear. The cores were immediately sectioned at 0.5-1.0-cm intervals. All wet samples were weighed and freeze-dried. After weighing the dry samples were ground to pass a 0.15-mm sieve for analyses. The porosity and mass depth of each layer were calculated.



Fig. 1. Location of the studied areas.

Surface soil samples in Lake Hongfeng/Baihua watershed were collected before and after spring rains and in autumn of 1994 and spring of 1995. Soil cores in Lake Hongfeng watershed were collected at the Karst Experimental Station near the lake. The landform at the station is comprised of six terraces from up to down slope which were man-made in the early 1960s. HF-2 to HF-7 and site HF-1 were collected on the top of the mountain, with forest coverage. HF/pz was sampled on a bare red earth hill at the village Puzi. All terraces were depositional sites for detritus eroded mountain slopes. Samples LS-1, LS-2 and LS-3 were collected moving from the top to the foot of Mt. Liangsuotun.

Surface soil samples from the watersheds of Lakes Erhai, and Lugu were collected in the summer of 1994. Soil cores in the Lake Erhai watershed were collected on a mountain slope below 2100 m. The samples were EH-1, EH-2 and EH-3 from up-slope, middle and mountain foot, respectively. Soil samples in Lake Lugu watershed were collected from near the mountain slope (LG) and Zhudi (ZD), i.e., samples LG-1 to LG-4 are sited from the top to the foot of the slope which is situated on the SW bank of the Lake, near Luoshuicun. Specifically, LG-1 was located on a shallow plateau of the mountain (elevation 2800 m); LG-2 and LG-3 were sited on the middle and down slopes, respectively. LG-4 was on the bank of the lake; and LG/zs was collected from a mountain slope near Zuosuo situated on the eastern bank of Lake Lugu. ZD-1 and ZD-2 were sampled on a mountain slope and a depression at the mountain foot, respectively.

All sampling sites were uncultivated. Each soil core, with an area of 0.18 m^2 , was sectioned into four to five slices and the sample weights were recorded. Samples were reweighed after freeze-drying for calculation of the mass depth of each layer. The mass depth of the cores is about $0.3-0.5 \text{ g cm}^{-2}$, and amounts to 3-5 mm in situ. The samples were ground for analysis. Whole samples and standards were analyzed using an H-P Ge detector coupled to a multi-channel analyzer (Canberra, S-100) under the same geometric condition. ⁷Be peak energy is 477 keV, and ¹³⁷Cs is at 661.6 keV. The counting efficiency was calibrated with standards provided by the Institute of Atomic Energy, Chinese Academy of Sciences, and these analytical data were compared to the results analyzed in the Laboratory of Oceanographic and Environmental Research (LOER) of Texas A&M University at Galveston, USA.

3. Results and discussion

3.1. Distribution characteristics of ⁷Be and ¹³⁷Cs in surface soils

From Fig. 2-1, all 28 soil profiles show ⁷Be activity as a logarithmic decrease with depth. This vertical profile distribution is directly related to ⁷Be source, penetration and decay. From the boundary soil downward, ⁷Be activities decrease gradually because of soil particle mixing and decay. At a mass depth of $0.3-0.5 \text{ g cm}^{-2}$, ⁷Be activities are $\leq 10 \text{ Bq} \text{ kg}^{-1}$. Fig. 2-1 shows the vertical profiles of ⁷Be for three sampling events (before and after rain in spring, summer–autumn), respectively. They show that ⁷Be has a similar vertical distribution, but the curves are much different in slopes and intercept.

The vertical distribution of ¹³⁷Cs activity in soils differs from that of ⁷Be. ¹³⁷Cs, a fallout radionuclide, is input to soils similar to ⁷Be. However, because ¹³⁷Cs has a longer





Fig. 3. ¹³⁷Cs vertical profiles in the soil cores at sites HF-1 and HF-2.

half-life (30.2 years) and it has time for penetration and mixing in soils. Fig. 3 shows the 137 Cs vertical profiles in cores at HF-1 and HF-2 to 40 cm. HF-1 is located in forests with undisturbed soils. 137 Cs activity at the topsoil is 26–31 Bq kg⁻¹, decreasing gradually with depth and reaches background level below 15 cm (mass depth 16 g cm⁻²). 137 Cs activity in the topsoil was only 4–7 Bq kg⁻¹, increasing to 11 Bq kg⁻¹ with soil depth and then decreases rapidly, at soil depth below 27 cm.

Since ⁷Be only exists within a soil mass depth of $0.3-0.5 \text{ g cm}^{-2}$, a comparative discussion of ¹³⁷Cs and ⁷Be could only be focused on surface soils. Fig. 2-2 shows that ¹³⁷Cs activities in soils of central Guizhou are higher than that in the western Yunnan. ¹³⁷Cs profiles of topsoil cores show that most curves trend near parallel, but some curves such as LS-1, LG-1, LG-3, LG/zs, ZD-1 and ZD-2 obviously decrease with soil depth (Fig. 2-2); whereas EH-3 appears to increase with soil depth (Fig. 2-2). These increases or decreases in ¹³⁷Cs activity result from the mixing and diffusion in soils, and erosion of the

Fig. 2. Vertical profiles of the activities of ⁷Be and ¹³⁷Cs and their ratios in the surface soils of the Yunnan– Guizhou Plateau. First line: ⁷Be profiles; second line: ¹³⁷Cs profiles; third line: the ratio of ⁷Be/¹³⁷Cs, with: (a) sampling in Lake Hongfeng watershed in Spring (before rain) 1994; (b) sampling in Lake Hongfeng watershed in Autumn 1994; (c) sampling in Lake Hongfeng watershed in Spring (after rain) 1995; (d) sampling at Liangsuotun hill in Spring 1994; (e) sampling in Lake Erhai watershed in Summer 1995; (f) sampling in Lake Lugu watershed and its upstream Zhudi in Summer 1994.

topsoil or accumulation of low ¹³⁷Cs concentration soils. Fig. 2-3 shows that most vertical profiles of the activity ratio between ⁷Be and ¹³⁷Cs (⁷Be/¹³⁷Cs) tended to decrease in the soil cores. The ⁷Be/¹³⁷Cs ratio in the topsoil reached 100–1000 in western Yunnan, but only 10–100 for the central Guizhou region. This suggests that ⁷Be and ¹³⁷Cs have a regional difference in deposition and/or accumulation.

3.2. ⁷Be and ¹³⁷Cs accumulation in soils

3.2.1. Vertical distribution model

The net input of fallout nuclides (F_{in}) in soils is controlled by atmospheric deposition $(F_{in(d)})$, and the input from eroded soil particles $(F_{in(a)})$, and the output with soil particle transport (or water flow) $(F_{out(e)})$. So, the net input of the fallout radionuclides in soils can be expressed as:

$$F_{\rm in} = F_{\rm in(d)} + F_{\rm in(a)} - F_{\rm out(e)} \tag{1}$$

The vertical distribution of radionuclides in soils is controlled by net input and penetrative mixing and radioactive decay. If the penetrative mixing is regarded as a diffusive process, the changes in radionuclides activity (c) with time (t) in a soil profile (z) have been described as (Robbins and Edgington, 1975):

$$\frac{\partial}{\partial Z} \left(D \frac{\partial}{\partial Z} \rho C \right) - P \left(\frac{\partial}{\partial Z} \rho C \right) - \lambda \rho C = \frac{\partial}{\partial t} \rho C \tag{2}$$

where D is the mixing coefficient (cm⁻² a⁻¹), t is the mixing time (year), ρ is the soil density in situ (g cm⁻²), P is the accumulation rate (+) or erosion rate (-) of soil particles (g cm⁻² a⁻¹), and λ is the radioactive decay constant of ⁷Be (4.74 a⁻¹) and ¹³⁷Cs(=0.023 a⁻¹).

Assuming that ⁷Be activities are constant at a given site in specific season, i.e., (C/t)=0; D, P and ρ remain constant:

$$Z = 0, \ C_{(z)} = C_0^* \tag{3}$$

$$Z = \infty, \ C_{(z)} = 0. \tag{4}$$

solving Eq. (2) and obtaining,

$$C_{(z)} = C_0^* \mathrm{e}^{\alpha z} \tag{5}$$

where

$$\alpha = \frac{P - (P^2 + 4D\lambda)^{\frac{1}{2}}}{2D} \tag{6}$$

On a sampling site, assuming there is no erosion or accumulation of soil particles, and mixing penetration is dominant, i.e., P=0, we can obtain,

$$D = \frac{\lambda}{\alpha^2} \tag{7}$$

$$\alpha = -\left(\frac{\lambda}{D}\right)^{\frac{1}{2}} \tag{8}$$

$$C_{(z)} = C_0^* e^{-Z(\frac{z}{D})^{\frac{1}{2}}}$$
(9)

In model (9), if Z is 0, the activity is defined as the apparent activity (C_0^*) . When radionuclide activity tends to zero, the soil depth is regarded as the maximum penetrative depth (Z).

3.2.2. ⁷Be apparent activity, maximum penetrative depth and inventory in soil cores

On the basis of the above model, ⁷Be apparent activity and maximum penetrative depth in the soils were calculated using the equation to imitate the curves from Fig. 2-1a-f. Results from the model are given in Table 1.

⁷Be apparent activities in the 28 soil profiles varied greatly, ranging from 119 to 1584 Bq kg⁻¹. This variation was affected by surface soil erosion or accumulation and by penetration and mixing in the soils. Affected by seasonal change and precipitation, ⁷Be apparent activities in the soil cores of the Hongfeng Karst Station were higher in the spring than in autumn, and larger in pre-rain than in the post-rain period of spring (Table 2). ⁷Be maximum penetration depths in the undisturbed soils were 0.3–0.5 g cm⁻² (analytical datum line \geq 10 Bq kg⁻¹), corresponding to the ⁷Be activity results of the fourth soil layers. ⁷Be penetrative depths at the same sites of the Hongfeng Karst Station (HF-1, HF-2, HF-4 and HF-6) in the autumn were larger than in the spring (Fig. 2-1a). Furthermore, during the same spring, ⁷Be maximum penetration depths in soils were obviously greater in the postrain than in the pre-rain. ⁷Be apparent activity and maximum penetrative depths in the soil of the soil soils were obviously greater in the soil of the western Yunnan were similar to the central Guizhou (Table 1).

3.2.3. ⁷Be inventory in the soils of western Yunnan and central Guizhou

Data on ⁷Be inventory in the soils of western Yunnan and central Guizhou are listed in Table 1.

Table 1 shows that the average ⁷Be inventory in all the 28 soil profiles was 335 Bq m⁻², ranging from 154 to 1065 Bq m⁻². The mean ⁷Be inventory at the 10 sampling sites in Lake Erhai and Lake Lugu watersheds in western Yunnan was 445 Bq m⁻², and 274 Bq m⁻² at the 18 sites in Lake Hongfeng/Baihua watershed. ⁷Be inventory in western Yunnan was higher than that in the central Guizhou, and a distinct difference appeared in the Lake Lugu watershed.

Precipitation in Lake Hongfeng watershed occurs from June to November, comprising 60-70% of the total annual precipitation. Therefore, ⁷Be atmospheric flux and its inventory of soils in summer and autumn should be higher than in spring and winter. The seven sampling sites in the Lake Hongfeng watershed are within 1 km² area, ⁷Be flux

| Area | Sampling no. | Apparent activity ($\times 10^3$ Bq kg ⁻¹) | | | Max. penetrative depth (cm) | | | Inventory (Bq m ⁻²) | | |
|-----------------|--------------|---|-----------------------|-------------------|-----------------------------|-----------------------|-------------------|---------------------------------|-----------------------|-------------------|
| | | Spring (pre-rain) | Spring (post-rain) | Summer- autumn | Spring (pre-rain) | Spring (post-rain) | Summer– autumn | Spring (pre-rain) | Spring (post-rain) | Summer- autumn |
| Central Guizhou | HF-1 | 906 | 370 | 321 | 0.16 | 0.28 | 0.27 | 263 ± 7 | 217 ± 4 | 208 ± 5 |
| | HF-2 | 726 | 251 | 248 | 0.22 | 0.26 | 0.35 | 321 ± 8 | 178 ± 4 | 224 ± 4 |
| | HF-3 | 494 | | | 0.20 | | | 226 ± 8 | | |
| | HF-4 | 386 | | 363 | 0.21 | | 0.25 | 194 ± 5 | | 229 ± 4 |
| | HF-5 | 645 | | | 0.25 | | | 337 ± 8 | | |
| | HF-6 | 688 | 440 | 571 | 0.34 | 0.42 | 0.41 | 501 ± 13 | 419 ± 5 | 539 ± 7 |
| | HF-7 | 249 | | | 0.24 | | | 159 ± 8 | | |
| | HF/pz | | 227 | | | 0.42 | | | 262 ± 5 | |
| | LS-1 | 119 | | | 0.45 | | | 154 ± 11 | | |
| | LS-2 | 199 | | | 0.34 | | | 198 ± 11 | | |
| | LS-3 | 450 | | | 0.28 | | | 300 ± 17 | | |
| Western Yunnan | EH-1 | | | 788 | | | 0.33 | | | 541 ± 15 |
| | EH-2 | | | 514 | | | 0.27 | | | 380 ± 15 |
| | EH-3 | | | 466 | | | 0.20 | | | 184 ± 11 |
| | LG-1 | | | 1584 | | | 0.36 | | | 1065 ± 2 |
| | LG-2 | | | 560 | | | 0.24 | | | 341 ± 1 |
| | LG-3 | | | 422 | | | 0.27 | | | 308 ± 1 |
| | LG-4 | | | 947 | | | 0.26 | | | 480 ± 1 |
| | LG/zs | | | 486 | | | 0.30 | | | 361 ± 1 |
| | ZD-1 | | | 539 | | | 0.30 | | | 359 ± 11 |
| | ZD-2 | | | 685 | | | 0.30 | | | 430 ± 21 |

Table 1 ⁷Be apparent activity and maximum penetrative depth and inventory in the soils

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 Table 2

 Data on ⁷Be and ¹³⁷Cs accumulation in Lake Erhai, Lake Lugu, Lake Hongfeng and Baihua sediment cores

 Lake name
 Core no.
 ⁷Be
 ¹³⁷Cs

 Repetertive
 Inventory
 Pack mass
 Accumulative rate
 Pack activity
 Tatal

| Lake name | Core no. | Be | | Lo Cs | | | | | | |
|---------------|--------------|---------------------------|------------------------------------|--|--|---|--|---|--|--|
| | | Penetrative depth (cm) | Inventory (Bq m ⁻²) | Peak mass depth (g cm ⁻²) | Accumulative rate $(g \text{ cm}^{-2} a^{-1})$ | Peak activity (Bq kg ⁻¹) | Total inventory (Bq m ⁻²) | Pre-1986 inventory (Bq m ⁻²) | | |
| | | | | | | | | | | |
| Lake Erhai | EH940713-3-2 | 2 | 237 ± 73 | 1.455 | 0.047 ± 0.002 | 48.3 ± 2.7 | 590 ± 27 | 519 ± 26 | | |
| Lake Lugu | LG940706-2 | 2 | 322 ± 19 | 1.354 | 0.045 ± 0.004 | 104.4 ± 4.5 | 1111 ± 13 | 937 ± 13 | | |
| Lake Baihua | BH941003-2 | 2 | 783 ± 44 | | | | | | | |
| Lake Hongfeng | HF880818-1 | | | 4.112 | 0.17 ± 0.01 | 252.6 ± 15.1 | 3713 ± 56 | 3704 ± 56 | | |

should be identical. Variation of ⁷Be inventory at the different sites reflects changes in soil erosion or accumulation. ⁷Be inventory was lower in the rainy season than in the dry season, which means that increasing precipitation intensity could raise the erosive rate. However, at the other sampling sites, the values are higher in autumn than in spring. This indicates that in the study of eroded or accumulative soil particles traced by beryllium-7, more attention should be paid to the comparison of ⁷Be inventories at different sites in the same season than that in different seasons. These data show that ⁷Be inventories in the soil cores at most sampling sites in central Guizhou were relatively low, which reflects serious soil erosion in the region. EH-1 is situated on a plain at the top of Mt. Diancangshan with dense forest and has little erosion. EH-2 and EH-3 were on the middle slope and foot, and slope with little vegetation, the soil erosion was serious, ⁷Be inventories at the two sites were lower than EH-1.

If radionuclide fluxes are identical, radionuclide inventory in soils depends on its activity and penetrative depth. Fig. 4 shows that ⁷Be inventories are closely related to their apparent activity and maximum penetrative depth, i.e.,

When $C_0^* = (1141 \pm 300) e^{-4.24Z}$, ⁷Be inventory in the soils is low, <230 Bq m⁻²; When $C_0^* = (2139 \pm 450) e^{-5.24Z}$, ⁷Be inventory in the soils is moderate, 230–400 Bq m⁻²; When $C_0^* > (2139 \pm 450) e^{-5.24Z}$, ⁷Be inventory in the soils is high, >400 Bq m⁻².

Based on the above levels of ⁷Be inventories of the 18 soil cores in the central Guizhou, 11 belong to "low", 4 to "moderate" and 3 to "high"; but for 10 soil cores in the western Yunnan, 4 belongs to "high", 5 to "moderate" and 1 to "low". These also indicate that



Fig. 4. Scattering distribution of the apparent activity and maximum penetrative depth of ⁷Be in the soil cores of the Yunnan–Guizhou Plateau.

⁷Be deposition and accumulation was different between the two regions. Seasonal differences in ⁷Be deposition and accumulation show that ⁷Be inventory in the soils of Lake Lugu watershed during the summer–autumn were higher than that in the Lake Hongfeng/Baihua basin. It also was higher than that in the Lake Erhai watershed.

3.3. ⁷Be and ¹³⁷Cs accumulative characteristics in the sediment cores

3.3.1. ⁷Be accumulation in sediment cores

⁷Be was mainly distributed within the top 2 cm of sediments in these lakes, identical to other lakes and bayou (Wan et al., 1986, 1987). However, due to the difference in input source and action, ⁷Be penetrative depth in soils differs from lake sediments (Bai and Wan, 1999). ⁷Be inventories in the sediments of Lake Erhai, Lake Lugu and Lake Baihua were 237 ± 73 , 322 ± 19 and 783 ± 44 Bq m⁻², respectively (Table 2).

Study on ⁷Be geochemical speciation has shown that more than 79% of ⁷Be was bound to the organic and the Fe–Mn fraction (Bai et al., 1997). The ⁷Be average inventory of 10 soil cores in the watersheds of Lake Erhai and Lake Lugu is 445 Bq m⁻² (184–1065 Bq m⁻²), but only 274 Bq m⁻² (154–539 Bq m⁻²) for the 18 soil cores in Lake Hongfeng/ Baihua watershed (Bai and Wan, 1999). ⁷Be inventory was much higher in the lake sediment core of Baihua than in the soil cores of the catchment, indicating that ⁷Be in soils could be transported into the lake sediment. However, ⁷Be inventory in the sediment cores of Erhai and Lugu were less than that in the soils of their watersheds, suggesting little soil erosion in the basins of Lake Erhai and Lake Lugu.



Fig. 5. Vertical distribution of ¹³⁷Cs in the sediments of Lake Erhai, Lake Lugu and Lake Hongfeng.

3.3.2. ¹³⁷Cs accumulative characteristics in sediment cores

There is a similar vertical distribution of ¹³⁷Cs in the sediment cores from Lake Erhai and Lake Hongfeng (Fig. 5). In the Lake Erhai sediment core, the peak of ¹³⁷Cs maximum accumulation was at a mass depth of 1.455 g cm⁻² and the sub-peaks of accumulation were at mass depths of 0.370 and 0.888 g cm⁻². In the Lake Hongfeng sediment core, the peak of ¹³⁷Cs maximum accumulation was at a mass depth of 2.286 g cm⁻². The ¹³⁷Cs peak of maximum accumulation occurred in 1964; the sub-peaks were related to the global fallout deposition in 1975 and the Chernobyl accident (1986).

The sediment dating using ¹³⁷Cs is identical to the result from ²¹⁰Pb, showing that there was a constant sedimentation in the past three decades. Although the ¹³⁷Cs vertical profile in Lake Lugu sediment core indicates that the top sediment has been mixed, its dating results are in agreement with ²¹⁰Pb (Wan, 1999; Xu et al., 1999). Although ¹³⁷Cs dating in the sediment cores of the three lakes in the two regions gives reliable sedimentation chrono-sequences, its activities in the corresponding layers of the three lake sediment cores show distinct differences. Corrected to deposition time, ¹³⁷Cs activity at the peak of the maximum accumulation in the sediment core of Lake Erhai is 48.3 ± 2.7 Bq kg⁻¹ and 252.6 ± 15.1 Bq kg⁻¹ in the sediment core of Lake Hongfeng. The peak of ¹³⁷Cs activity in Lake Lugu is 104.4 ± 4.5 Bq kg⁻¹, only 41% of that in Lake Hongfeng (Table 2).

Usually, if a radionuclide has stable deposition or erosion in a basin, the higher the sedimentation rate is, the lower the activity is in the corresponding layers. On the basis of the ¹³⁷Cs dating, the sedimentation rate of Lake Erhai (0.047 g cm⁻² a⁻¹) remains stable in the past three decades. The 0.17 ± 0.01 g cm⁻² a⁻¹ rate in Lake Hongfeng is 3.6 times that in Lake Erhai. The fact that the sedimentation rate differs from the ¹³⁷Cs activity in the sediment reflects the variations of ¹³⁷Cs inventories among the lakes.

There is an apparent difference in the ¹³⁷Cs inventories in the sediment cores among Lake Erhai, Lake Lugu and Lake Hongfeng. Expressed by the data corrected to deposition time, ¹³⁷Cs inventories in the sediment cores from prior to 1986 among Lake Erhai, Lake Lugu and Lake Hongfeng are 519 ± 26 , 937 ± 13 and 3704 ± 56 Bq m⁻², respectively. ¹³⁷Cs inventory in Lake Hongfeng is 7.1 times of that in Lake Erhai, and 4 times of that in Lake Lugu (Table 2). This suggests that ¹³⁷Cs accumulation in the lake sediment of western Yunnan is obviously lower than that in the central Guizhou area. Moreover, within the soil cores of HF-1 and HF-2 (Fig. 3), ¹³⁷Cs inventories were 1036 and 1843 Bq m⁻², respectively. However, ¹³⁷Cs inventories in the soils of Lake Erhai watershed were less than 1000 Bq m⁻².

3.3.3. Model analysis of ⁷Be and ¹³⁷Cs in sedimentation

Accumulation of fallout radionuclides in lake sediments is controlled by direct precipitation, hydrogeologic conditions, and surface soil erosion. To understand the precipitation–accumulation characteristics of ¹³⁷Cs and ⁷Be in western Yunnan and central Guizhou regions, a model analysis of sedimentation was made (Table 3). A model was constructed to distinguish radionuclide distribution relationship between direct atmospheric precipitation, basin erosion, and sedimentation (Wan and Santschi, 1987; Wan et al., 1987). The model is used in this study to calculate the ratio of accumulation

| Nuclide | Lake name | λ_{s} (a^{-1}) | $\tau_{s} \text{ (day)}$ | fr | Fe | fr/fe | $F = f_r + f_e$ $= I_c / I_a$ | $I_{\rm c} ({\rm Bq} \ {\rm cm}^{-2})$ | $I_{\rm a} \\ ({\rm Bq} \ {\rm cm}^{-2})$ |
|-------------------|---------------|--------------------------|--------------------------|------|------|-------|-------------------------------|--|---|
| ¹³⁷ Cs | Lake Erhai | 0.18 | 2.05×10^{3} | 0.32 | 0.17 | 1.9 | 0.49 | 0.052 ± 0.003 | 0.11 ± 0.01 |
| | Lake Lugu | 0.04 | 8.21×10^{3} | 0.37 | 0.06 | 6.6 | 0.42 | 0.094 ± 0.001 | 0.22 ± 0.01 |
| | Lake Hongfeng | 0.73 | 5.01×10^{2} | 0.19 | 0.81 | 0.2 | 1.00 | 0.370 ± 0.006 | 0.37 ± 0.01 |
| ⁷ Be | Lake Erhai | 2.13 | 1.71×10^{2} | 0.28 | 0.05 | 5.2 | 0.34 | 0.024 ± 0.007 | 0.07 ± 0.02 |
| | Lake Lugu | 0.53 | 6.86×10^{3} | 0.09 | 0.02 | 5.3 | 0.11 | 0.032 ± 0.002 | 0.29 ± 0.02 |
| | Lake Baihua | 10.01 | 3.65×10^{1} | 0.40 | 0.62 | 0.6 | 1.02 | 0.078 ± 0.004 | 0.08 ± 0.01 |
| | | | | | | | | | |

Table 3 Model simulation of accumulation–precipitation of ¹³⁷Cs and ⁷Be in the lake sediments in western Yunnan and central Guizhou^a

^a In the model calculation, solid–liquid distribution coefficient K: ¹³⁷Cs: 4×10^3 cm³ g⁻¹, cited from Robbins et al. (1977); ⁷Be: 5×10^4 cm³ g⁻¹, cited from Wan et al. (1987). λ_s is the first-order removal rate constant with respect to sedimentation (a⁻¹); τ_s is the residence time of the radionuclide in the lake with respect to sedimentation (= $1/\lambda_s$, day); f_r and f_e are the retention factor and erosion factor, respectively; I_a is the radionuclide inventory from direct atmospheric fallout (Bq cm⁻²); I_c is the radionuclide inventory in sediment core (Bq cm⁻²); $f = I_c/I_a$.

and precipitation of radionuclide in the lake sediments, then based on the measured inventories to determine the regional difference in direct atmospheric fallout.

The model analysis has shown that ⁷Be accumulation in the sediments of Lake Erhai and Lake Lugu was affected by atmospheric precipitation. However, in Lake Baihua, the erosion factors' influence on ⁷Be accumulation is larger than that from precipitation, the influence factor ratio of the precipitation/erosion is only 0.6. The inventory ratios of that obtained in sediment core to that from direct atmospheric fallout in Lake Erhai and Lugu were only 0.34 and 0.11, respectively; but it was 1.02 in Lake Hongfeng. ⁷Be inventories from direct atmospheric fallout in Lakes Erhai, Lugu and Baihua were 0.07 ± 0.02 , 0.29 ± 0.02 and 0.08 ± 0.01 Bq cm⁻², respectively. This means that ⁷Be atmospheric fluxes in the watersheds of Lake Erhai and Lake Hongfeng/Baihua were similar, but Lake Lugu basin was higher.

¹³⁷Cs accumulation in Lake Hongfeng was dominated by erosion, being 4.3 times the retention. ¹³⁷Cs accumulations in Lakes Erhai and Lugu were controlled by direct atmospheric fallout. The retention factors were 1.9 and 6.6 times of the erosion factors in Lake Erhai and Lake Hongfeng, respectively. However, the inventory ratios of that obtained in the sediment cores to that from direct atmospheric fallout in Lake Erhai and Lake Erhai and Lake Lugu by model calculation were similar, they were 0.49 and 0.42, respectively. This suggests that the difference in the measured accumulation of ¹³⁷Cs in the lakes mainly resulted from the regional difference in atmospheric precipitation. The inventory ratios (I_c/I_a) in Lake Hongfeng were two times of that in Lake Erhai and Lake Lugu, reflecting the important impact from erosion. ¹³⁷Cs inventories of direct atmospheric fallout in the Lake Erhai, Lake Lugu and Lake Hongfeng regions before 1986 were 0.11 ± 0.01, 0.22 ± 0.01 and 0.37 ± 0.01 Bq cm⁻², respectively. They show that ¹³⁷Cs inventories from direct atmospheric fallout in the western Yunnan are much lower than that in the central Guizhou region. ¹³⁷Cs from direct atmospheric fallout in the Lugu watershed.

3.4. Regional differences in the ⁷Be and ¹³⁷Cs atmospheric precipitation

⁷Be fallout distribution and accumulation characteristics could be affected by latitude and altitude. In a given region, ⁷Be from atmospheric flux is mainly affected by precipitation (Baskaran et al., 1993), its maximum value appears in summer, and could be several times that of winter and spring (Schuler et al., 1991). All sediment cores in this study were collected in summer and autumn. So, the higher inventory of ⁷Be could have originated from direct atmospheric fallout. In the Lake Lugu region, this would mainly be the result of increased precipitation during sampling.

¹³⁷Cs flux to the earth's surface depends on the intensity of nuclear testing, region and latitude. ¹³⁷Cs vertical profiles in the sediment cores of these lakes correspond to the global fallout chrono-sequences. Therefore, the difference in ¹³⁷Cs from direct atmospheric fallout between west Yunnan and central Guizhou could not be explained by the nuclear test intensity. Since the variation in rain is just 20% between the two regions, ¹³⁷Cs atmospheric fallout should be close. Consequently, the reason for less ¹³⁷Cs fallout in west Yunnan than that in central Guizhou could only be explained by regional fallout differences. This difference is identical to the global fallout of the ¹³⁷Cs inventories reported by UNSCEAR (United Nations, 1969) and Agudo (1998).

4. Summary and conclusion

From the accumulation and distribution characteristics of ⁷Be and ¹³⁷Cs in the sediments and soils in western Yunnan and central Guizhou, the following were found.

(1) The activity ratios of ${}^{7}\text{Be}/{}^{137}\text{Cs}$ in the topsoil reached 100–1000 in western Yunnan, but only 10–100 for the central Guizhou region, suggesting that the two radionuclides have a regional difference in deposition.

(2) The apparent activities and maximum penetrative depth of ${}^{7}\text{Be}$ in the soils of the two regions were similar, indicating that the two regions have similar erosion and ${}^{7}\text{Be}$ transportation with surface soil movement.

(3) There was a difference in the ⁷Be fallout and accumulation between the two regions. ⁷Be inventories in the soil cores in the Lake Lugu watershed in the summer and autumn were higher than that in Lake Hongfeng/Baihua watershed and also higher than that in Lake Erhai watershed. This was probably related to high precipitation in summer.

(4) ⁷Be inventories in the sediment cores of Lake Erhai and Lake Lugu were 237 ± 73 and 322 ± 19 Bq m⁻², respectively, corresponding with the ⁷Be inventories in the surface soils of their basins. ⁷Be inventory in the sediment of Lake Baihua was 783 ± 44 Bq m⁻², which was higher than the average value in the soil cores, showing that watershed erosion had an influence on the accumulation of ⁷Be in the sediments.

(5) On the basis of the activity values that were corrected to deposition, prior to 1986, 137 Cs inventories in Lakes Erhai, Lugu and Hongfeng were 519 ± 26 , 937 ± 13 and 3704 ± 56 Bq m⁻², respectively. These show that the accumulated amount of 137 Cs in the lake sediments in western Yunnan was distinctly lower than that in central Guizhou.

(6) The model simulation indicated that ⁷Be and ¹³⁷Cs accumulations in Lakes Hongfeng and Baihua were dominated by watershed erosion. But in Lakes Erhai and

Lugu, it was mainly controlled by atmospheric precipitation. The proportion of the influence factors from retention and erosion was similar, with a similar ratio for ¹³⁷Cs and a lower ratio for ⁷Be. ⁷Be inventories from direct atmospheric fallout in lakes Erhai, Lugu and Hongfeng regions were 0.07 ± 0.02 , 0.29 ± 0.02 and 0.08 ± 0.01 Bq m⁻², respectively. But for ¹³⁷Cs prior to 1986, these were 0.11 ± 0.01 , 0.22 ± 0.01 and 0.37 ± 0.01 Bq m⁻², respectively. ¹³⁷Cs is regarded as a global diffusion pollutant, the regional variations in the ¹³⁷Cs inventories from direct atmospheric fallout in the western Yunnan and central Guizhou regions reflect a screening effect of west wind airflow during transport. It is inferred that the uplifting Himalayas serve as a barrier to the fallout of global atmospheric pollutants in the western Yunnan–Guizhou plateau.

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