Modes of Occurrence and Geological Origin of Beryllium in Coals from the Pu'an Coalfield, Guizhou, Southwest China

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Abstract: The concentration, modes of occurrence and geological origin of beryllium in five workable coal beds from the Pu'an Coalfield of Guizhou were studied using the inductively coupled-plasma mass spectrometry (ICP-MS), floating and sinking experiments (FSE) and sequential chemical extraction procedures (SCEP). The results show that the average concentration of beryllium in coals from the Pu'an Coalfield is 1.54 μ g/g, much lower than that in most Chinese and worldwide coals. Beryllium in the Pu'an coals was not significantly enriched. However, it should be noted that the No. 8 coal bed from the study area has a high concentration of beryllium, 6.89 μ g/g, three times higher than the background value of beryllium in coal. Beryllium in coal mainly occurs as organic association and has predominantly originated from coal-forming plants when its concentration is relatively low. The concentration of beryllium concentration of coal is similar to its background value, and in addition to coal-forming plants, beryllium is mainly derived from detrital materials of terrigenous origin. When beryllium is anomalously enriched in coal, it mainly occurs as organic association and is derived from volcanic tonsteins leached for a long geological time and then adsorbed by organic matter in peat mire.

Key words: coal, beryllium, mode of occurrence, geological origin, Guizhou

1 Introduction

It is both theoretically and practically significant to study hazardous trace elements in coal in fully and probably utilizing coal resources and environmental protection (Sun et al., 2002; Sun, 2003; Luo et al., 2004; Wang et al., 2004; Dai and Ren, 2006b). Beryllium is one of the toxic elements in coal, because its halides and oxides have strong toxicity. Beryllium can lead to pathologic variations of the lung when it is adsorbed by human bodies. Beryllium can be easily volatilized and it is the important source of Be in the atmosphere. Owing to its environmental sensitivity, beryllium was included in the 189 hazardous atmosphere pollutants in the "Supplementary Laws of Cleaning Air of USA" in 1990. Some researchers have studied the concentration and modes of occurrence of beryllium in coal (Kolker and Finkelman, 1998; Huang et al., 1999; Ren et al., 1999, 2004; Zeng et al., 2000; Shao et al., 2003; Dai et al., 2004a, 2004b, 2005a, 2005b, 2006a).

The study results show that the concentration of beryllium in coals from the world generally varies from 0.1 μ g/g to 15 μ g/g, with an average of 1.5–2 μ g/g. The

crustal abundance of beryllium is 2.8 µg/g (Taylor, 1964), thus the enrichment factor of beryllium in most coals is generally less than 1. There are no direct methods to determine the mode of occurrence of beryllium in coal, due to its low content in coal and its low atomic number. The modes of occurrence of beryllium in coal were deduced from the relationship between beryllium concentrations and ash yields. Bai et al. (2004) revealed that beryllium mainly occurs in inertinite and illite. The most possible occurrence of beryllium in coal is organic association, and beryllium occurring in clay minerals was not excluded; however, such confidence level is as low as 4 (Finkelman, 1995). There is rare literature related to the geological origin of beryllium enrichment of coal. In this paper, the modes of occurrence and geological origin of beryllium in five workable coal beds from the Pu'an Coalfield were studied using floating-sinking experiments and sequential chemical extraction procedures.

2 Geological Setting

The Pu'an Coalfield is located in western Guizhou (Fig. 1) and consists of the main Upper Permian coal-bearing strata including the Longtan Formation (P_2l) , and the

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Fig. 1. Location and Late Permian paleogeography of the Pu'an Coalfield (modified from Shao et al. (2003), Dai et al. (2004a) and Hao (2000)).

Changxing Formation (P_2ch). The Longtan Formation is the main coal-bearing formation in the study area and consists of sandstone, siltstone, mudstone, limestone and coal, with a total thickness of about 320 m. Its sedimentary environment varies from lagoon and tidal flat through lower delta plain to tidal flat and carbonate subtidal flat. The thickness of the Changxing Formation ranges 84–154 m, and its sedimentary environment is predominantly of the shallow-sea and littoral delta facies.

3 Methods of Study

There are five main workable anthracite coal beds $(R_{o, ran} = 2.59\% - 2.64\%)$, including the Nos. 1, 2, 8, 11 and

Table 1 Occurrence of beryllium in coal and its SCEP methods

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17 beds in the Longtan Formation of the Pu'an Coalfield, with the thickness being 1.68 m, 2.41 m, 0.76 m, 1.95 m and 1.29 m, respectively.

The five representative coal-seam channel samples were taken from mined areas, excluding dirt partings thicker than 3 cm. The collection of samples was in accordance with the Chinese Standard for Collecting Channel Samples GB482-1985.

The samples were crushed and ground to 200 meshes and ultimate analyses were made on the basis of the Chinese Standards GB/T 212-2001, GB/T-214-1996 and GB/T-215-2003. Vitrinite reflectance and maceral compositions were analyzed using a Leitz MPV-III photometer system.

The samples were dried at 105° C in an oven for 8 h, and ignited at 750° C for 5 h in a muffle furnace, and then the ashing recovery was calculated. Residual ash (200 mg) from the ignition was first wetted in concentrated superpure HNO₃, followed by the addition of HF (2 mL) and HCl (6 mL), allowing the samples to digest and evaporate, and the residue was finally dissolved in 1 mL of 5 M HNO₃. The mixture was left still until the samples were fully digested and evaporated to dryness. The samples were then diluted to 100 mL in distilled and de-ionized H₂O, and then analyzed with the inductively-coupled

plasma mass spectroscopy (ICP-MS) as described by Dai et al. (2003c).

On the basis of coal density, the samples were divided into ten grades: <1.4 g/cm³, 1.4–1.5 g/cm³, 1.5–1.6 g/cm³, 1.6–1.8 g/cm³, 1.8–2.0 g/cm³, 2.0–2.2 g/cm³, 2.2–2.4 g/cm³, 2.4–2.6 g/cm³, 2.6–2.8 g/cm³ and >2.8 g/cm³ (Fig. 2). The concentration of beryllium in the above floatingsinking products was determined by using the ICP-MS. The modes of occurrence of beryllium were studied with the sequential chemical extraction procedures (SCEP) that were designed by Dai et al. (2004a). We slightly modified the SCEP methods and their procedures were given in Table 1.

 Number	Modes of occurrence	Extraction method
 Ι	Water soluble state	10 g coal sample + 60 mL water, 25°C, 48 h
II	Ion exchangeable state	Residue + 60 mL NH_4AC , 25°C, 48h
III	Organic-bonded state	Residue of II + 1.47 g/cm ³ CHCl ₃ , floating dried at 40°C, 1:1 HNO ₃ and HClO ₄ added, 10 h
IV	Carbonate mineral state	Residue of II + 1.47 g/cm ³ CHCl ₃ , sinking + 20 mL 0.5% HCl, 1 h
V	Silicate minerals state	Residue of III + 2.89 g/cm ³ CHBr ₃ , floating + 1:1 HNO ₃ and HF, 10 h
VI	Sulfide mineral state	Residue of III + 2.89 g/cm ³ CHBr ₃ , sinking + 1:1 HNO ₃ , 10 h



4 Results and Discussion

4.1 Coal chemistry and coal petrology

The ultimate analysis, vitrinite reflectance and maceral compositions of the five workable coal beds (Nos. 1, 2, 8, 11 and 17) from the Pu'an Coalfield of Guizhou were given in Table 2.

With an exception of the No. 8 coal bed, other four workable coal beds are considered high-sulfur coals and the sulfur is mainly pyritic. The SEM-EDX results show that the organic sulfur content of vitrinite in the No. 2 coal bed varies from 0.41% to 0.96%, with an average of 0.71% (based on 33 test spots), in accordance with the ultimate analysis.

The total sulfur content of the No. 8 coal bed is 0.78%. No pyrite particles were observed either under the optical microscope or under the SEM-EDX, indicating that the sulfur is mainly organic.

The results of the XRD and optical microscope show that minerals in the Nos. 1, 11 and 17 coal beds are mainly composed of pyrite, quartz and a trace amount of kaolinite. The minerals in the No. 2 coal bed are mainly made up of pyrite and kaolinite, while those in the No. 8 coal bed are dominated by kaolinite. It should be noted that pyrite in the Nos. 1, 11 and 17 coal beds occurs in framboidal, massive, cell-fillings, isolated euhedral and nodular forms, indicating that pyrites in the coals are being of syngenetic or early diagenetic origin (Chou 1997; Dai et al. 2000, 2002b, 2003a, 2006e).

Pyrite in the No. 2 coal bed occurs as fracture-fillings, indicating an epigenetic origin (Zhang, 1999; Dai et al.,



Fig. 2. Distribution of beryllium in sinkingfloating products of the Pu'an coals. A: <1.4 g/cm³; B: 1.4–1.5 g/cm³; C: 1.5–1.6 g/cm³; D: 1.6-1.8 g/cm³; E: 1.8-2.0 g/cm³; F: 2.0-2.2

2003a). The δ^{34} S value of pyrite in the No. 2 coal bed is -4.8‰, in accordance with the δ^{34} S range of pyrites of lowtemperature hydrothermal origin reported by Zhang (1999), but in sharp contrast to that of low-organism

(bacteria and algae) and syngenetic sedimentary origins as reported by Dai et al. (2002b).

 g/cm^{3} ; J: >2.8 g/cm³

Kaolinite in the five workable coal beds occurs as lens and dispersive fine particles, and quartz is distributed in collodetrinite. No cell-filling kaolinite and quartz were observed in these coals, indicating that kaolinite and quartz are mainly from detrital materials of terrigenous origin.

The Pu'an coals have a high content of inertinite, and vitrinite varies from 51.2% to 55.8%. No liptinite macerals were observed in the Pu'an coals.

In should be noted that there is a stable gray to grayblack tonstein layer with a thickness of 5 cm in the No. 8 coal bed, which is 20 cm far from the roof of the latter. The XRD results show that kaolinite is the dominant mineral of the tonstein. Some accessory minerals of volcanic origin occurred in the tonstein layer, for examples, quartz with sharp edges and β -quartz paramorhp, followed by sanidine with sharp edges and a trace amount of zircon and apatite crystals. The mineral associations in the tonstein layer in the No.8 Coal are very similar to those in tonsteins of the Permo-Carboniferous strata from northern China and the late Permian strata from southwestern China (Zhou et al., 2000), indicating their original materials are acid volcanic ashes.

4.2 Beryllium concentration of coals from the Pu'an Coalfield

Swaine (1990) estimated the concentration of world coals, varying from 0.1 µg/g to 15 µg/g. A study by Finkelman (1993) showed that the average beryllium

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Coal bed	Thickness (m)			Proximat	e analysis					Mace	eral compo	osition			R _{o, ran}
		A_d	V_{daf}	S _{t, d}	$S_{p,d}$	S _{s, d}	S _{o, d}	V	Ι	Ру	Q	СМ	Cal	Т-М	
No. 1	1.68	18.4	9.45	2.14	1.38	0.06	0.70	53.9	32.4	2.3	8.6	2.2	0.6	13.7	2.62
No. 2	2.41	21.6	9.34	3.21	2.52	0.05	0.64	51.2	33.7	3.4	2.3	9.4	bdl	15.1	2.64
No. 8	0.76	20.7	9.68	0.78	0.11	0.05	0.62	51.7	33.0	0.6	2.6	11.3	0.8	15.3	2.59
No. 11	1.95	22.4	9.70	3.56	2.81	0.12	0.63	54.3	29.8	3.2	9.8	2.5	0.4	15.9	2.61
No. 17	1.29	26.8	10.41	2.87	2.17	0.08	0.62	55.8	27.0	2.6	12.9	1.7	bdl	17.2	2.64

Note: A – ash; V – volatile matter; S_t – total sulfur; S_p – sulfide sulfur; S_s – sulfate sulfur; S_o – organic sulfur; V – vitrinite; I – inertinite; Py – pyrite; Q – quartz; CM – clay minerals; Cal – calcite; T–M, total minerals; $R_{o,ran}$ – mean random reflectance of vitrinite; d – on a dry base; daf – on a dry and ash-free base; bdl – below the detection limit.

concentration of US coals is 2.2 μ g/g, with the maximum value of 330 μ g/g. The average concentrations of beryllium in coals from North China and Guizhou Province are 2.05 μ g/g and 2.2 μ g/g, respectively (Dai et al., 2004a, 2005b). On the basis of 1195 samples, Tang and Huang (2004) suggested that the Be concentration of Chinese coals varies from 0.5 μ g/g to 8 μ g/g, with an average of 1.9 μ g/g.

On the basis of the thickness proportions (20.8%, 29.8%, 9.4%, 24.1%, and 15.9%, respectively) and Be concentrations of the five coal beds from the Pu'an Coalfield, the weighted average concentration of Be in Pu'an coals is calculated to be 1.54 μ g/g (Table 3). Beryllium in the Pu'an coals is not significantly enriched as compared with that in coals from Guizhou, North China, China, and the world. However, the No. 8 coal bed has a high concentration of Be (6.89 μ g/g), three times higher than that in common coals. Thus, the modes of occurrence and geological origin of Be in the No. 8 bed should be paid great attention.

Seven benches were collected from mining faces of the No. 8 coal bed of the Pu'an Coalfield. The benches from top to bottom of the No. 8 bed are Bench-1 to Bench-7. The Be concentrations and ultimate analyses of the benches are presented in Table 3. Bench-3 underlying the tonstein layer has the highest Be concentration, 12.66 μ g/g. Moreover, the concentrations of Be are decreasing from Bench-3 to Bench-7. Bench-1 and Bench-2 overlying the tonstein layer have low concentrations of Be, and the beryllium concentration is higher in Bench-2 than in Bench-1 (Table 4).

4.3 Floating–sinking experiments of beryllium in the **Pu'an** coals

On the basis of published data concerning beryllium in coal, the modes of occurrence of beryllium were studied by indirect methods. Beryllium in coal may occur either in organic matter or in minerals (Dai et al., 2005b).

Beryllium generally occurs as organic association when it significantly enriched in coal and is mainly distributed in minerals when its concentration is close to the background value of coal (Yudovich et al., 1985). A study by Yudovich et al. (1985) shows that the amount of beryllium related to organic matter is close to that distributed in minerals in most coals. Beryllium in the Late Permian coals from western Guizhou is positively correlated to ash yields, indicating an inorganic affinity (Dai et al., 2005b).

Table 5 and Fig. 2 give the concentrations of beryllium in the products of floating-sinking experiments of five workable coal beds from the Pu'an Coalfield. The following characteristics of beryllium behaviors during floating-sinking experiments can be drawn as follows:

(1) Beryllium in the No. 2 and No. 11 coal beds that have a low beryllium concentration ($<1 \ \mu g/g$) is predominantly distributed in the density range of $<1.8 \ g/cm^3$ and has the highest value in the range of $1.5-1.6 \ g/cm^3$ and the lowest value ($<0.55 \ \mu g/g$) in the range of $>1.8 \ g/cm^3$.

(2) Beryllium in the No. 1 and No. 17 coal beds that have a medium beryllium concentration (1.25 μ g/g and 1.21 μ g/g, respectively) is mainly distributed in the density range of <1.86 g/cm³ and 2.4–2.6 g/cm³.

(3) Beryllium in the No. 8 coal bed which has a high beryllium concentration is mainly distributed in the density range of less than 1.6 g/cm^3 and is relatively low in the density range of less than 1.6 g/cm^3 .

Because the density of organic matter, clay minerals and sulfide minerals are generally distributed in the range of <1.8 g/cm³, 2.4–2.6 g/cm³ and about 5.0 g/cm³, respectively, it can be concluded that 1) beryllium is mainly related to organic matter when its concentration is low; 2) beryllium is related both to organic matter and minerals when its concentration is medium; 3) beryllium is predominantly related to organic matter when its concentration is anomalously enriched.

Table 3 Beryllium concentrations in five workable coal beds from the Pu'an Coalfield, as compared with coals from Guizhou, North China, China, USA, and the world $(\mu g/g)$

Coal bed	No. 1	No. 2	No. 8	No. 11	No. 17	Guizhou ^{a)}	North China ^{b)}	China ^{c)}	USA ^{d)}	World ^{e)}
Concentration	1.25	0.89	6.89	0.74	1.21	2.2	2.05	1.9	2.2	1.5-2

Note: a) - from Dai et al. (2005b); b) - from Dai et al. (2004a); c) - from Tang and Huang (2004); d) - from Finkelman (1993); e) - from Swaine (1990).

Bench No.	Thickness (cm)	Concentration (µg/g)	A _d (%)	$V_{daf}(\%)$	$S_{t,d}(\%)$
Bench-1	10	1.66	23.5	9.75	0.74
Bench-2	10	2.21	22.6	9.63	0.78
Tonstein layer	5	10.24	Nd	Nd	Nd
Bench-3	10	12.66	20.6	10.51	0.79
Bench-4	10	9.53	21.8	9.85	0.82
Bench-5	10	10.62	19.7	10.22	0.75
Bench-6	10	5.67	22.5	9.75	0.80
Bench-7	11	2.23	20.8	9.96	0.79

Table 4 Beryllium concentrations and ultimate analysis of benches and tonstein of the No. 8 coal bed from the Pu'an Coalfield

Note: Nd - not detected.

4.4 Sequential chemical extraction procedures of beryllium in the Pu'an workable coals

Table 6 shows the results of sequential chemical extraction procedures. The following can be deduced from the SCEP results:

(1) The concentrations of Be in water-soluble, ionexchangeable and carbonate associations are very low or below the ICP-MS detection limit.

(2) The No. 1 and No. 17 coal beds have medium concentrations of beryllium that mainly occurs in organic and silicate associations.

(3) The No. 2 and No. 11 coal beds have a low concentration of beryllium that is mainly distributed in organic association, with concentrations of 1.33 μ g/g and 1.11 μ g/g, respectively.

(4) The No. 8 coal bed has a very high concentration of beryllium that is mainly related to organic association, with a concentration of 9.85 μ g/g, which is in accordance with the floating-sinking results.

The results of sinking-floating experiments and sequential chemical extraction procedures show that 1) beryllium mainly occurs as organic association when its concentration is low in the Pu'an coals; 2) the amount of beryllium related to organic association is close to that in inorganic matter when beryllium concentration is close to its background value of coal, and inorganic beryllium is mainly distributed in clay minerals; 3) beryllium predominantly occurs in organic matter when it is anomalously enriched in coal.

4.5 Origins of beryllium in coal

On the basis of data of coal chemistry, coal petrology and coal geochemistry, it can be deduced that there are three sources of beryllium in the Pu'an coals.

(1) Coal-forming plants. Beryllium can be detected in most plants and land plants (ash base) contain an average beryllium content of 4.1 μ g/g (Dai et al., 2002a; Tang and Huang, 2004; Ren et al., 2006). Beryllium is mainly derived from the coal-forming plants when the coal contains a low concentration of beryllium. In addition, the vitrinite group can

adsorb more beryllium than the inertinite and liptinite groups. The No. 2 and No.11 coal beds contain low concentrations of beryllium that is mainly associated with organic matter based on the data of floating–sinking experiments and sequential chemical extraction procedures, and thus it can be concluded that beryllium in the two coals is predominantly derived from coal–forming plants.

(2) Volcanic ashes. Sometimes beryllium in coal is related to volcanic ashes (Dai et al., 2003b). Crowley et al. (1989) found that tonsteins can lead to enrichment of beryllium in coal.

Beryllium is generally enriched in thin coal beds (< 1 m), as beryllium prefers to combine organic matter in thin coal beds (Dai et al., 2002a, 2006c, 2006d). The tonstein layer and its underlying coal benches contain higher Be than its overlying ones, and beryllium concentration decreases from top to bottom benches underlying the tonstein (Table 4). Thus, it can be concluded that the high content of beryllium in coal mainly results from tonstein that is derived from volcanic ashes. The enrichment of beryllium in coal is closely related to the leaching of volcanic ashes, and peat is considered to be the geochemical barrier for beryllium enrichment. After beryllium is leached from the tonstein by groundwater, it will be adsorbed by organic matter in peat mire.

(3) Supplies of detrital materials. In addition to the coalforming plants, detrital materials of terrigenous origin are the important sources for beryllium in most coals. A study

Table 5 Beryllium contents in the floating-sinking products of the Pu'an coals (µg/g)

Coal bed	No. 1		No. 2		No. 8		No. 11		No. 17	
Density (g/cm ³)	Recovery (%)	Content								
<1.4	11.8	1.55	10.5	1.32	8.7	8.24	11.4	0.85	8.8	1.67
1.4-1.5	14.6	1.48	12.6	1.24	14.6	9.57	13.6	0.98	13.5	1.54
1.5-1.6	23.5	1.82	22.4	1.36	20.6	10.55	20.5	1.05	22.4	1.62
1.6-1.8	15.6	1.46	18.6	0.89	22.9	2.65	19.4	0.76	16.9	0.95
1.8-2.0	1.6	0.55	5.8	0.35	6.2	0.76	6.2	0.55	1.8	0.54
2.0-2.2	2.8	0.64	1.8	0.42	2.5	0.45	2	0.48	3.2	0.43
2.2-2.4	8.1	0.58	10.5	0.41	8.4	0.98	9.6	0.35	9.6	0.64
2.4-2.6	12.6	1.85	10.4	0.38	9.7	2.41	8.6	0.45	14.1	1.72
2.6-2.8	5.8	0.71	4.5	0.39	5.6	0.85	5.6	0.22	5.6	0.62
>2.8	3.6	0.26	2.9	0.30	0.8	0.39	3.1	0.31	4.1	0.38

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Table 6 Results of sec	mential chemical extr	action procedures o	f the Pu'an	coals (ug/g)
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Coal bed	No. 1		No. 2		No. 8		No. 11		No. 17	
Occurrence	Recovery (%)	Content								
Water-soluble	0.5	bdl	0.2	bdl	0.6	0.21	0.6	bdl	0.4	bdl
Ion-exchangeable	0.5	bdl	0.6	bdl	0.4	bdl	bdl	bdl	1.1	0.14
Carbonate	4.6	0.22	0.8	bdl	1.7	bdl	1.1	bdl	bdl	bdl
Organic	75.9	1.75	77.1	1.33	73.8	9.85	72.8	1.11	68.1	1.71
Silicate	14.6	1.54	15.5	0.41	21.6	1.58	18.7	0.35	24.6	1.59
Sulfide	3.9	0.28	5.8	0.31	0.9	0.45	6.8	bdl	5.8	0.24

Note: bdl – below detection limit.

by Dai et al. (2005b) shows that the positive relationship between beryllium concentration and ash yields indicates the inorganic affinity of beryllium in Guizhou coals. Additionally, beryllium is also positively correlated to lithophile elements (such as As, Si, Ga, Hf, Zr and Th), indicating that beryllium is mainly from detrital materials of terrigenous origin. Minerals in the No. 1 and No. 17 coal beds are mainly composed of quartz and kaolinite of detrital materials of terrigenous origin, and beryllium in the two coal beds predominantly occurs in silicate associations. The concentrations of beryllium in sulfide associations of the No. 1 and No. 17 coal beds are very low (Table 6), although the two beds contain syngenetic pyrite. Moreover, minerals of hydrothermal origin and tonstein layers were not observed in the two beds, indicating that beryllium mainly comes from detrital materials of terrigenous origin.

In addition, pyrite veins originating from low– temperature hydrothermal fluids are well developed but not enriched in beryllium in the No. 2 coal bed from the Pu'an Coalfield. The results of sequential chemical extraction procedure show that beryllium occurring in sulfide associations is only 0.31 μ g/g in content. Studies by Dai et al. (2002a, 2005a) also show that the veined quartz and veined ankerite originating from siliceous and iron-rich calcic low-temperature hydrothermal fluids did not lead to enrichment of beryllium in the Late Permian coals from the Dafang Coalfield, western Guizhou Province.

5 Conclusions

(1) The weighted average concentration of Be in the Pu'an coals is 1.54 μ g/g, lower than that in most Chinese and worldwide coals. Beryllium is on the whole not enriched in the Pu'an coals. However, the beryllium content in the No. 8 coal bed of the Pu'an Coalfield is as high as 6.89 μ g/g, three times higher than its background value of coal.

(2) Beryllium mainly occurs in organic matter and is derived from coal-forming plants when it is low in coal.

(3) The amount of beryllium related to organic association is close to that in inorganic matter when beryllium concentration is close to its background value of coal. In addition to coal-forming plants, detrital materials

of terrigenous origin are the dominant source of beryllium in coal.

(4) Beryllium is mainly distributed in organic matter when it is anomalously enriched in coal. In such a case, beryllium is predominantly from tonsteins that are derived from volcanic ashes. Beryllium will be adsorbed by organic matter in peat mire after it is leached out from tonsteins by groundwater.

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