Contamination of the environmental ecosystems by trace elements from mining activities of Badao bone coal mine in China

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Abstract Bone coal, as a main mining object, can be used by local inhabitants as daily fuel and by local industrial enterprises as industrial fuel in Pinglin County, Shaanxi Province, China. This study reports how the environmental ecosystems have been polluted around the Badao bone coal mine. Geochemical samples (e.g. rock, water, soil, edible plant and animal) were collected. Bone coal from the Badao mine contains Se up to 75 μg/g Se and 28 μg/g Se in ashes after its combustion, with higher contents of other trace elements. Bone coal and its ash seem to be the main geochemical source of trace elements in soils and plants, which may cause contamination of the local environmental ecosystems. Three ways by which soils have been contaminated by these trace elements derived from bone coal are proposed in this paper. Radishes and beans have the ability to accumulate Mo and Se from soils. There is no obvious difference in concentrations of Cu, Cr and F in each plant from the two areas.

Keywords Baodao coal mining · Trace elements · Contamination ways · Ecosystem · Shaanxi Province · China

Received: 19 February 2002 / Accepted: 8 January 2003

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Introduction

Pollution of environmental ecosystems by trace elements from black shales and activities related to coal mining and combustion is a world-wide problem (Zheng and others 1992; Benvenuti and others 1997; Foos 1997; Sharmasarkar and others 1998; Loukola-Ruskeeniemi and others 1998; Wright and others 1998; Qi and others 1999; Xiao and others 1999; Zhu and Zheng 1999; Yan and others 1999; Liu 2000; Zheng and Wang 2000; Liorens and others 2001). Pollution of the environmental ecosystem by different trace elements is associated with different types of ore deposits or rocks with abundant trace elements (Lee and others 1998; Wen and Qiu 1999; David 2002; Heikkinen and others 2002; Kim and others 2002; Milu and others 2002; Goodarzi and others 2003; Wang and others 2003). For example, two reservoirs in western Oregon contain mercury-contaminated sediments and fish as a result of historical mercury mining in the Cottage Grove Lake watershed and mercury amalgamation used in gold mining in the Dorena Lake watershed (Ambers and Hygelund 2001), whereas *Polygonum microcephalum* and *Rumex hastatus* as representatives of typical high-elevation copper flowers grow extensively in copper mining areas of Yunnan Province, southwest China, and contain high concentrations of copper (Tang and Fang 2001). Waters discharged from coal mines in the Upper Silesia of Poland have caused contamination of rivers and their sediments because saline waters discharged from the coal mines in the southern parts of Poland contain increasingly high barium and radium (Pluta 2001). Furthermore, barite nanocrystals are common and abundant in the troposphere over the Upper Silesian Industrial Region, Poland. Jablonska and others (2001) documented that the presence of barite nanocrystals (very fine-grained) over this area is the result of combustion of Ba-enriched coals (up to 4,260 ppm Ba). However, more than 20 elements are concentrated in bone coals in southern China (Zhang and others 1987). Do bone coal mining activities cause trace elements to be released from bone coal, thus leading to contamination of the local environmental ecosystems? The aim of this study is to determine how the environmental ecosystems are polluted by trace elements released from bone coals near the Badao bone coal mine at Pinglin County, Shaanxi Province, China. At the same time, this study will characterize the mechanism of contamination of the environmental

DOI 10.1007/s00254-003-0768-3

ecosystems by trace elements and make comparisons with naturally lithogenic contamination by black shales and combined artificial and lithogenic contamination pertaining to bone coal mining activities.

Landscape

The studied area is in southern Shaanxi Province in the central China, where there is the prevailing subtropical mountain climate of Middle Asia. In general, the area is more than 2,000 m above sea level with an undulation ranging from 2,000–2,700 m. There are only two main seasons in a year: wet and dry. The wet season lasts from March to November, with heavier rainfall from July to October. The annual precipitation generally varies from 600 to 1,000 mm with a maximum value of 1,323 mm and a minimum value of 550 mm. The annual average temperature is about 14 °C and the monthly average temperature is above 4 °C. There are about 240 frost-free days a year. Vegetation type is attributed to the mixing of evergreen-broadleaf pines and oak forests. Mulberry, tea, wheat, rice, corn and vegetables are grown on terraces on the gentle slopes of mountains and river sides. Soils are dominated by brown forest soil and yellow brown mountain soil. Underground water comes from fracture-interlayer water discharged from bedrocks. The concentrations of underground water are middle grades. Hydraulic discharges of fountain outflows are less than 1.5–2.5 tons/h with the mineralization degree being less than 1.0 g/l in the water from this area.

As shown in Fig. 1, the Badao bone coal mine is one of the most important coal mines in Pingli County, Shaanxi Province. Bone coals are mined via mining adits where there are also ventilation and draining feeders. Cultivated land is generally distributed on the northern, shaded slopes of mountains. Agricultural crops comprise corn, potatoes and beans. Vegetables mainly include cabbage, radish, and chilli. Livestock comprises pigs and chickens. As shown in Fig. 1, the Early Paleozoic black shale, which is of wide occurrence in the areas of South China, is one of the most important rock types in the studied area. More than 20 elements are concentrated in the black shales and bone coals in southern China (Zhang and others 1987; Fang and others 1995). Generally, bone coal is measured at 8–15 cm in thickness, so thin as not to be found in some places. However, there are a series of bone coal layers of more than 1 m in thickness in localized places. According to Zhang and others (1987), bone coals in the Early Paleozoic black shales are a kind of black caustobiolith with a calorific capacity amounting to more than 3,347 J/g. Poor bone coals have a calorific capacity ranging from 3,347–5,020 J/g, good-bone coals have a calorific capacity ranging from 5,020–2,552 J/g, and the best-bone coals have a calorific capacity of more than 12,552 J/g. Bone coals from the Qinling bone coal zone (zone I in Fig. 1) belong to the best bone coal with a calorific capacity of 20,920–29,288 J/g. These bone coals, as a main mining object, can be used by both local inhabitants as daily fuels and local

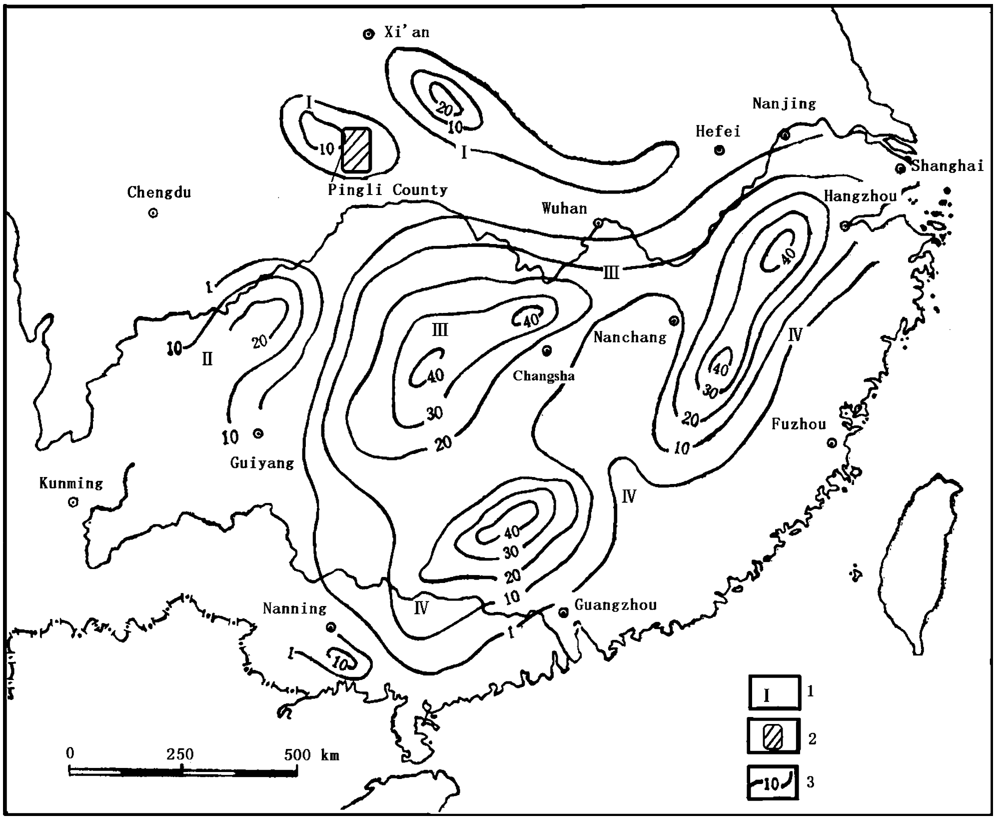

Fig. 1
Location of the studied area and the Lower Cambrian bone coals in China (modified after Zhang and others 1987). *1* Bone coal zones and their numbers; *I* Qinling zone; *II* Yangtze zone; *III* Southern China zone; *2* location of the area studied; *3* thickness of bone coal measures (in m)

industrial enterprises as industrial fuels, indicating that there is a potential contamination problem threatening the environmental ecosystems that should be evaluated.

Materials and methods

Geochemical samples (rock, water, soil, edible plants and animals) were collected from two areas, the Badao bone coal mine and the Baxian black shales. Edible plant samples included corn, potato, *Amorphophallus konjac* K.·Kock, radish, radish leaf, and bean from the studied area. Livestock samples included chickens and pigs.

Rock samples were crushed to 0.25 mm or less in size by means of a jaw crusher, then quartered, and pulverized to 0.095 mm or less in size in a high-Al-ceramic crusher. Surface soil samples were collected at depths of 10–30 cm. Soil samples were placed in cotton bags to air-dry, then crushed in the cotton bags with a stick of 0.275 mm or less in size, quartered, and pulverized to 0.095 mm or less in size in a high-Al-ceramic crusher. Edible plant samples were placed in cotton bags for partial air-drying prior to the removal of all moisture in an oven at a temperature of less than 40 °C in case some more easily transient constituents were released from edible plants at >40 °C. The dried plant samples were crushed to 0.20 mm or less in size by means of a GWF-1 multiple crusher and then processed by water enzyme decomposition (Fang and others 2002).

Each of the samples was analyzed by two-banded atomic fluorescence spectroscopy with a detection limit of 0.01 ppm for Se. Edible plant samples were analyzed by oscillography-polarography (JP-II), with a detection limit of 0.05 ppm for Mo and 0.10 for V, and by atomic absorption spectroscopy. Data quality was controlled by inserting reagent blanks, duplicate samples, and Chinese National Standard Samples into each batch. All the samples were analyzed at the Analysis Center of Trace Elements, The Geological Exploration Center of Non-ferrous Metals (Chinese National Class-A Analytic Center).

Results and discussion

Concentrations of trace elements in bone coals, ashes and black shales

Wang and others (1999) reviewed advances in the study of Se distribution, occurrence and source in coals and their ashes, reporting a mean value of 3.64 μg/g Se in Chinese coals, which is higher than that of world coals with a mean value of 3 μg/g Se (Minkin and others 1984) and American coals with a mean value of 1.8 μg/g Se (Finkelman 1993). As shown in Fig. 2, bone coals from the Badao bone coal mine contain Se ranging from 75–41 μg/g with a mean value of 57.2 μg/g, which is much higher than that of the world coals, Chinese coals, and American coals. The Se contents are more than 27 times higher than those of feed coals with 1.5 mg/kg Se for a 1,050 MW power plant in NE Spain (Liorens and others 2001). Therefore, these bone coals used by local inhabitants as daily fuels and local industrial power plants as industrial fuels could cause Se contamination in the local environmental ecosystems.

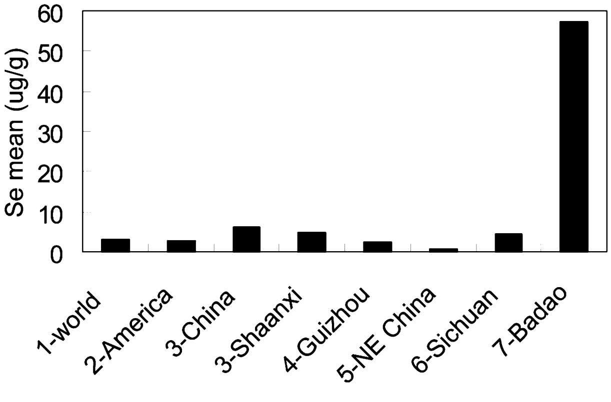

Fig. 2
Selenium mean values of coals from different areas throughout the world. References: *1* Minkin and others (1984); *2* Finnkelman (1993); *3* Ren and others (unpublished); *4*, *5*, *6* Wang and others (1999); *7* this study

The mean concentrations of Se, Mo, Cu, Zn, Cr, V, and Ni in bone coals are significantly higher than those in black shales from the study area (Table 1). However, the mean concentrations of F with a maximum content of 1,610 μg/g and Mn in black shales are higher than those in bone coals. The concentrations of these trace elements are the highest in bone coals, which may be the main geochemical source of trace elements, such as the sources of V, Mo and Se (Fig. 3), in soils and plants in the study area.

The combustion of bone coals leaves a large amount of ash. The coal ashes contain 28 μg/g Se, indicating that more than 50% of Se from bone coals after their burning is released into the atmosphere over the studied area. The selenium content of the ashes is more than 17 times that of fly ashes with 1.6 mg/kg Se from a 1,050 MW power plant in NE Spain (Liorens and others 2001), and is much higher than those of the following four ashes derived from coal combustion in the USA, i.e., 3.3 mg/kg in bed ashes, 16 mg/kg in fly ashes, 11 mg/kg in scrubber sludges, and 2.3 mg/kg in by-products (Wright and others 1998). Therefore, the ashes in the studied area can also cause Se contamination in the local environmental ecosystems. However, the contents of Mo, Cu, Zn, Cr, Ni, and V in the ashes are higher than those in raw bone coal. It is suggested that these elements are concentrated largely in the ashes. The case in this study is similar to that pertaining to those trace elements from feed coal and fly ashes in a 1,050 MW power plant in NE Spain, for the fly ashes contain more elevated contents of the trace elements than the feed coal (Liorens and others 2001). On the one hand, daily combustion can provide a certain amount of Se in the atmosphere. On the other hand, the ashes are another main geochemical source of trace elements in the area because bone coal is one of the most important fuels for local inhabitants. Agricultural utilization of the bone coal ashes may, at a high environmental risk, pollute soils and

Table 1
Contents of trace elements (μg/g) in bone coals and black shales from the Badao and Baxian areas

Area	Rock type	Se	Mo	Cu	Zn	Cr	V	Ni	Mn	F
Badao bone coal mine	Bone coal	61	506	335	1,350	248	2,840	600	100	420
	Bone coal	60	288	220	1,100	230	2,370	305	100	305
	Bone coal	49	472	330	1,350	320	3,000	550	100	720
	Bone coal	41	472	330	1,100	155	2,200	450	150	320
	Bone coal dust	75	420	750	2,270	342	3,520	600	150	780
	Mean	57.2	432	393	1,434	259	2,786	501	120	509
	Ash of bone coal	28	460	450	1,920	387	3,300	700	200	550
	Soil	6.5	102	108	493	100	775	155	960	–
	Soil	7.6	98	167	516	158	1,380	114	590	–
	Mean	7.1	100	138	505	129	1,078	135	775	–
	Water from the mine (μg/l)	1.1	–	0.01	5	0.5	–	–	0.1	–
	Water from a stream (μg/l)	0.3	–	0.01	0.01	3	0.1	–	–	–
Baxian black shale area	Carboniferous shale	36	250	210	850	180	2000	–	100	1,230
	Black chertic shale	0.67	2.5	5	25	15	40	–	350	580
	Black chertic shale	0.12	1.7	15	50	65	61	–	450	460
	Black chertic shale	0.05	1.2	40	85	100	110	–	300	900
	Black chertic shale	0.05	0.75	10	65	70	61	–	300	950
	Black chertic shale	0.05	1.1	10	75	75	70	–	100	1,610
	Mean	6.2	43	48	192	84	390	–	267	955
	Soil	0.26	1.7	25	80	85	108	–	750	610
	Soil	0.17	1.5	40	80	80	97	–	850	300
	Mean	0.22	1.6	33	80	83	103	–	800	455

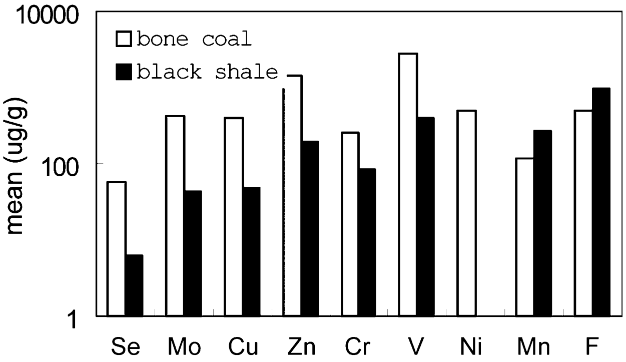

Fig. 3
Mean concentrations of trace elements in Badao bone coals and Baxian black shales

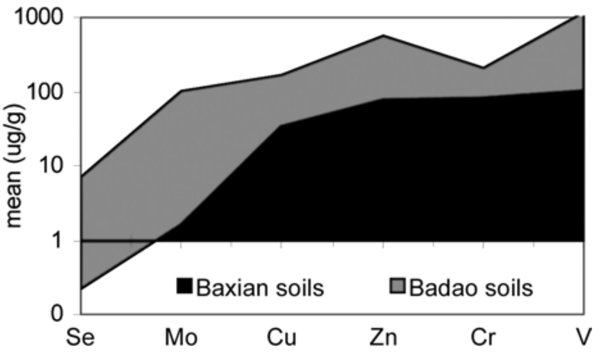

Fig. 4
Mean concentrations of trace elements from soils in the Badao and Baxian areas

crop plants because the levels of Se, Mo, Cu, Zn, Cr, Ni and V are still high.

Concentrations of trace elements in soils and waters

As can be seen in Table 1, percolating water from the Badao bone coal mine contains 1.1 μg Se/l and has a pH of 7, while water from a stream running through the mine contains as much as 0.3 μg Se/l with a pH of 6–7, indicating that the Se content of stream water is depleted because of dilution, although water discharged from the mine contains relatively high Se. Cu, Zn, Cr and V contents are very low in waters from the area even though the elements are concentrated mainly in bone coals. Thus, Se is much more readily dissolved and transported in waters discharged from the bone coal mines than Cu, Zn, Cr and V. Therefore, water-soluble Se can provide soils and plants with sufficient Se via leaching by underground or surface waters in the coal mining areas.

Similar to trace elements in bone coals, Se, Mo, Cu, Zn, Cr, V and Ni are enriched in soils of the Badao bone coal mining area, but much less than those in bone coals (Table 1). The mean concentrations of Se, Mo, Cu, Zn, Cr and V in soils of the Badao bone coal mining area were significantly higher than those in soils of the Baxian black shale area (Table 1, Fig. 4), and the former is more seriously contaminated than the latter.

There are three ways in which soils are contaminated by these trace elements originating from bone coals. First, soils were derived from their parental bedrocks, including carbonaceous limestones, black shales, and bone coal-bearing rocks, in which these trace elements are enriched. As a result, these soils inherited trace elements from their parental bedrocks. Second, dust and ashes of bone coals are used as fertilizers by the local inhabitants, thus polluting the soils with trace elements. Third, percolating water from the Badao bone coal mine may provide some of the trace elements, especially Se.

Concentrations of trace elements in plants and animals

Selenium concentrations in edible plants from the study area vary over a wide range for each species (see Table 2). Selenium contents are high, for example, 2.58 μg/g in bean, 2.29 μg/g in corn, and 2.75 μg/g in radish. The highest Se concentrations are produced in radish leaves, which contain 10 μg/g. Se concentrations in *Amorphophallus konjac* K.·Kock, egg, pork, and potato are at normal levels. Like Se, Mo concentrations in edible plants from the study area also vary over a wide range in each species. Mo contents are elevated in potato, *Amorphophallus konjac* K.·Kock, and radish. Absorption of Mo was found in bean with 43.5 μg/g and in radish leaf with 62.5 μg/g, as both are Mo-tolerable species. Radish leaf and bean may have some potential for phytostabilization of Se- and Mo-polluted soils because they both have the ability to accumulate Mo and Se and survive well in Se- and Mo-contaminated soils. There was no obvious difference in the concentrations of Cu, Cr or F in each plant from the two areas.

Selenium concentrations of 2.29 μg/g in corn from the Badao bone coal mining area are ten times those (0.23 μg/g) from the Baxian black shale area. Se and Mo contents in *Amorphophallus konjac* K.·Kock from the Badao bone coal mining area are higher than those from the Baxian black shale area. It is indicated that bone coals have greater potential to pollute edible plants than black shales.

Conclusions

Bone coals from the Badao bone coal mine have high concentrations of trace elements, with mean values of 57.2 μg/g Se and 432 μg/g Mo, whereas ashes from the combustion of bone coals contain 28 μg/g Se. The contents of Mo, Cu, Zn, Cr, Ni, and V in the ashes are higher than those in raw bone coals. Therefore, these bone coals used by local inhabitants as daily fuels and local industrial enterprises as industrial fuels can cause contamination by trace elements in the local environmental ecosystems. Bone coals and their ashes may be the main geochemical source of trace elements, and may act as the source of V, Mo and Se in soils and plants in the study area.

As viewed from the above discussion, it seems that the pollution may be caused by artificial or lithogenic factors. High concentrations of Se, Mo, Cu, Zn, Cr, Ni, and V were found in soils near the Badao bone coal mining area, which were higher than those in soils of the Baxian black shale area. Three ways in which soils may be contaminated by trace elements derived from bone coals can be established. First, soils were derived from their parental bedrocks, including carbonaceous limestones, black shales, and bone coals, in which these trace elements are enriched. As a result, these soils inherited trace elements from their parental bedrocks. Since bone coals have a greater potential to discharge trace elements into soils near the mining area than black shales due to their lithogenic contamination under supergene geological conditions, their lithogenic contamination would be intensified by bone coal mining activities. Second, local inhabitants use the bone coal dusts and ashes after combustion to fertilize soils and plants, which pollute the soils with trace elements. Agricultural utilization of the ashes, as a result of artificial contamination caused by disposing solid dumps discharged from mining activities of bone coal, may cause an increased risk of soils and crop plants because Se, Mo, Cu, Zn, Cr, Ni and V still remain at higher levels. Third, the presence of high levels of Se in bone coals suggests that selenium in the water is related to the chemistry of the rocks. Percolating water from the Badao bone coal mine may provide some of the trace elements, especially Se, as a consequence of lithogenic contamination by bone coals under supergene geological conditions.

Owing to the pollution of soils where edible plants grow, some edible plants have also been contaminated by trace elements available in the polluted soils. Higher concentrations of Se and Mo are detected in bean and radish leaf.

Table 2
Contents of trace elements (μg/g) in edible plants from Badao and Baxian areas

Area	Plant species	Se	Mo	Cu	Zn	Cr	V	F
Badao bone coal mine	Potato	0.15	4.03	5.5	12	0.12	0.45	9.4
	Potato	0.05	1.25	5.3	13	0.19	0.62	10
	Amorphophallus konjac K. Kock	0.79	4.02	5.8	27	0.22	0.98	11
	Amorphophallus konjac K. Kock	0.57	3.15	6.2	58	0.7	1.31	12
	Corn	2.29	1.25	2	19	0.63	0.98	81
	Egg	0.7	0.11	0.83	14	0.5	0.003	9.4
	Pork	0.64	0.36	1.3	84	0.15	0.3	16
	Bean	2.58	43.5	9.5	32	0.76	0.52	16
	Radish	1.81	6.25	3	32	0.28	0.52	14
	Radish	2.75	13.8	3	40	0.32	0.71	11
	Radish leaf	10	62.5	7.3	61	3	2.6	22
Baxian black shale	*Amorphophallus konjac* K. Kock	0.13	0.82	3.8	43	1	0.38	10
	Corn	0.23	–	–	170	–	–	–

Varying trends of Se and Mo in corn and *Amorphophallus konjac* K.·Kock from the Badao bone coal mining area and the Baxian black shale area indicated that bone coals have greater potential to pollute edible plants than black shales.

Acknowledgements The authors wish to thank the local government of Pingli County, Shaanxi Province, China, for its assistance in fieldwork. The State Key Project on Fundamental Research Planning of China (grant no. 2001CB409805), the China National Non-ferrous Metal Industry Corporation (grant nos. CNNC95-D-25 and 93E01), and Visiting Scholar Fund of LODG of the Institute of Geochemistry, Chinese Academy of Sciences, provided joint financial support for this project. Special thanks are due to CNNC for giving the authors the opportunity to complete this research.

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