Mobility of some potentially toxic trace elements in the coal of Guizhou, China

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Abstract The mobility of 10 potentially toxic trace elements (PTTE), As, Cd, Cr, Cu, Hg, Ni, Pb, Se, Tl, and Zn from 32 coals of the Longtan Group formed in Permian Period in Guizhou Province, China was investigated using sequential extraction procedures. The results demonstrate that PTTEs such as Hg, As, Se, Cd, Cu, and Pb have the highest mobility at surface conditions, and the average extractable fractions of them are 86%, 95%, 79%, 76%, 69%, and 69% of the total amount in coal, respectively. The elements in coal with the lowest leachability include Tl, Cr, and Ni, and the average extractable fractions of them are 30%, 20%, and 29% of the total amount in coal respectively. Zinc has an intermediate behavior, and the average leachable fraction of it accounts for 46% of the total amount in coal. The results demonstrate that mobility of PTTE in coal depends on the speciation of these elements. The elements associated with sulfates, carbonates, sulfides and some organic matter in coal show the highest extraction rates during the weathering process, while elements with silicate affinities are inert at surface conditions.

Key words Coal Potentially toxic trace elements · Speciation · Mobility

Introduction

Due to the special geochemical environment produced during peatification and coalification processes, many trace elements, especially potentially toxic trace elements (PTTE), can be enriched in coal. Organic matter and diagenetic minerals can act as enrichment traps for these trace elements (Swaine 1990). After coal was extracted and hauled to the ground, changing its physical and chemical condition, sulfides in coal such as pyrite formed at reducing condition begin to decompose when they meet groundwater or are exposed to air, and produce a great amount of acid mine drainage (AMD). This kind of AMD has a strong leaching potential, and can bring a large amount of trace elements, especially PTTE in coal into the environment, which cause serious environmental pollution. This kind of pollution can not only occur at coal outcrops, but at coal heaps, coal mine wastes and coal cleaning wastes as well. Therefore, in coal producing areas, this kind of pollution is very common. Guizhou Province is one of the largest coal production areas in China. Due to the high pyritic sulphur content of the coals and acidification of the precipitation in this area, AMD occurs in most coal mining areas in Guizhou and consequently there is potential leaching of PTTE.
Sequential extraction tests are used to elucidate the mobility and speciation of trace elements in soil, industrial wastes and other solid materials (Tessier and others 1979, 1982; Wadge and Hutton 1987; Feng and others 1997). In the present study, the chemical mobility of PTTE such as As, Cd, Cr, Cu, Hg, Pb, Ni, Se, Tl, and Zn in coals of the Longtan Group formed in the Permian Period in Guizhou Province was investigated by means of sequential extraction tests.

Experimental

Sampling

The main coal-bearing stratum in Guizhou Province is the Longtan Group from the Permian Period. Thirty-two coals from the main mining seams of 19 coal mines which belong to four coal basins, namely Shuicheng, Liuzhi, Panjiang and Guiyang coal basin, were sampled. All samples were gathered by means of underground channel sampling. All samples were bituminous coals except one, (LDN-38) gathered from Guiyang coal basin is an anthracite coal.

Analytical method

The sequential extraction procedures used in this study were adopted from the procedures used by Querol and

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Table 1
Scheme of sequential extraction of PTTEs in coal

Step	Leaching method	Modes
1	A 4-g coal sample was weighed into a 50-ml polyethylene centrifugal tube, and 20 ml of deionized and distilled water was added in. Vibrate the tube for 15 h at room temperature, and then centrifuge it. The solution was filtered and 2.5 ml of HNO_3 was added in. The volume was made up to 50 ml by adding in deionized and distilled water. The residue was dried at 40 °C.	Water-extractable
2	20 ml of 1 mol/l HN_4Ac (pH = 7)[a] was added into the residue above. The following procedure were the same as that of step 1.	Exchangeable
3	20 ml of 1 mol/l HN_4Ac (pH = 5)[a] was added into the residue above. The following procedure were the same as that of step 1.	Carbonate-bound
4	The residue above was oxidized with 2.5 ml HNO_3 in a Teflon-lined stainless-steel pressure bomb for 15 h at 95 °C. After centrifugation, the solution was filtered. The volume was made up to 50 ml by adding in deionized and distilled water.	Organically and sulfide-bound

[a] The pH values of NH_4Ac solution were adjusted to 7 or 5 with 25% of HAc and 25% of NH_4OH

Table 2
The instrument conditions for analyzing As, Se, Pb, Cd and Tl using transverse heated graphite atomizer coupled with AAS

Elements	Diluent	Wavelength (nm)	Slit (nm)	Background correction	Carrier gas	Drying		Ashing		Atomization		Cleaning	
						Temp. °C	Time (s)	Temp. °C	Time (s)	Temp. °C	Time (s)	Temp. °C	Time (s)
As	3% H_2SO_4	193.7	0.7	Zeeman	Ar	110/130	30/20	1100	10	2000	4	2400	2
Se	0.2% HNO_3	196.0	2.0	Zeeman	Ar	110/130	30/20	1100	10	1900	4	2400	2
Pb	0.2% HNO_3	283.8	0.7	Zeeman	Ar	110/130	30/20	500	10	1600	4	2400	2
Cd	0.01 MHCL	228.8	0.7	Zeeman	Ar	110/130	30/20	300	10	1400	4	2400	2
Tl	0.5% HNO_3	276.8	0.7	Zeeman	Ar	110/130	30/20	400	10	2000	4	2400	2

others (1996) for sequential extraction of coal, and which in turn were based on the leaching sequence used by Fernandez-Turiel and others (1994) for that of fly ash. The leaching sequence is summarized in Table 1.

The analyses of As, Se, Pb, Cd, Tl in the leachates were performed by graphite furnace atomic emission spectrometry with the analytical conditions listed in Table 2; those of Cr, Cu, Zn, Ni in the leachates were carried out by flame atomic emission spectrometry with the analytical conditions shown in Table 3; and Hg was analyzed using double stage amalgamation coupled with cold vapor absorption spectrometry (Feng and others 1998). The matrix influences were tested by producing and analyzing leachate blanks and those with addition of certain amount of the elements interested. It was found out that there were matrix influences when As and Se were analyzed. After using of matrix modifier [5 μg Pd + 3 μg $Mg(NO_3)_2$] during analysis of As and Se, no interference effects were observed. The AAS spectrometer used in the studies is a PE5100PC (Perkin Elmer, USA).

The determination procedures of bulk content of each element in coal are as follow: 0.1 g of the coal sample was digested with 3-ml concentrated HNO_3 and 0.5 ml 40% HF in a Teflon-lined stainless steel pressure bomb at 140° C for 24 h, the resulting faintly yellow solution was diluted into 25 ml by adding deionized and distilled water, and the analyses of the elements in the solution were the same as above.

Table 3
The instrument conditions for analyzing Cr, Cu, Zn and Ni using flame atomizer coupled with AAS. A-AC Air-Acetylene flame

Elements	Wavelength (nm)	Slit (nm)	Diluent	Flame type
Cr	537.9	0.7	H_2O	A-AC rich
Cu	324.8	0.7	1% HNO_3	A-AC lean
Zn	213.9	0.7	1% HCl	A-AC lean
Ni	232.0	0.2	1% HNO_3	A-AC lean

Table 1 lists the speciation of the elements leached during each leaching step. Since the components that are decomposed with nitric acid extraction are weathered easily under natural conditions over long periods, and elements retained in the residue from the last extraction step are unavailable under natural leaching conditions (Querol and others 1996), the sum of element concentrations in the sequential extraction procedure can be assumed to be the mobile fraction under natural leaching conditions over long periods, and the ratio of the sum with the total

content of each element represents the magnitude of mobility of the element under a surface environment.
The major mineral transformations during the leaching experiments were investigated by X-ray diffractometry (XRD) of the residues obtained from each sample in each leaching step. From the XRD patterns for each residue, the net intensities of selected reflections for each mineral phase (proportional to its concentration in the residue) were calculated. The ratio of mineral phases with respect to quartz, which is not chemically affected by the leaching sequence, enabled the XRD intensity data to be referred to a constant internal standard. This method enabled the detection of changes in mineral phases during a leaching sequence. The XRD background intensity was used to define the evolution of the content of amorphous organic matter in coal (Background intensity at $2\phi=25$, Querol and others 1996).

Conclusion and discussion

The major mineral transformations during the sequential extraction procedure

Six coal samples were chosen (the samples were LD-04, LM-06, SL-12, SD-14, PH-34 and LDN-37, respectively). Mineralogical analyses of the original samples and the residues obtained from each leaching step were performed by X-ray diffractometry (Table 4). Studies showed that even though there was little difference in mineral composition among the samples, the major mineral compositions of the samples are silicate, such as quartz, kaolinite, illite, and plagioclase etc., carbonate such as calcite and dolomite, sulfide such as pyrite and marcasite, sulfate such as gypsum. From Table 4, it can be seen that the major mineral transformations observed through the

Table 4
The mineral distribution in original coal samples and residues of the sequential extraction of the six coal samples (the unit is counts s^{-1}). O original sample; W water-leached residue; Ac7, $NH_4Ac(pH=7)$ leached residue; Ac5, $NH_4Ac(pH=5)$ leached residue; NA nitric acid leached residue; C calcite; D dolomite; F feldspars; G gypsum; I illite; K kaolinite; M marcasite; P pyrite; Q quartz; GB represents the XRD background intensity at 25° of 2θ; < dl less than XRD detection limit

Sample		Q	P	M	C	K	F	I	BG	P/Q	M/Q	C/Q	BG/Q	
	O	2236	231	246	1833	< dl	340	283	603	0.1	0.1	0.8	0.3	
	W	2341	164	251	1454	< dl	226	< dl	611	0.1	0.1	0.6	0.3	
LD-04	Ac-7	2087	159	223	1102	< dl	234	< dl	583	0.1	0.1	0.5	0.3	
	Ac-5	3014	193	382	< dl	< dl	300	195	886	0.1	0.1	0	0.3	
	NA	2730	< dl	< dl	< dl	261	300	280	733	0	0	0	0.3	
		Q	P	M	C	F	K	BG		P/Q	M/Q	C/Q	BG/Q	
	O	967	318	459	352	286	605	494		0.3	0.5	0.4	0.5	
	W	1156	312	368	238	318	673	540		0.3	0.3	0.2	0.5	
LM-06	Ac-7	990	370	403	< dl	287	556	540		0.4	0.4	0	0.5	
	Ac-5	876	482	720	< dl	370	696	347		0.6	0.8	0	0.4	
	NA	1284	< dl	< dl	< dl	310	727	550		0	0	0	0.4	
		Q	P	C	D	K	BG		P/Q	C/Q	D/Q	BG/Q		
	O	2456	220	1364	326	1219	556		0.1	0.6	0.1	0.2		
	W	2103	224	1111	258	1124	431		0.1	0.5	0.1	0.2		
SL-12	Ac-7	2900	178	998	355	1128	567		0.1	0.3	0.1	0.2		
	Ac-5	3425	237	< dl	< dl	1071	705		0.1	0	0	0.2		
	NA	3333	< dl	< dl	< dl	890	727		0	0	0	0.2		
		Q	P	M	C	K	G	BG		P/Q	M/Q	C/Q	G/Q	BG/Q
	O	1199	189	261	699	576	462	374		0.2	0.2	0.6	0.4	0.3
	W	1663	145	399	846	577	< dl	466		0.1	0.2	0.5	0	0.3
SD-14	Ac-7	1054	157	246	235	494	< dl	400		0.2	0.2	0.2	0	0.4
	Ac-5	987	258	500	< dl	609	< dl	369		0.3	0.5	0	0	0.4
	NA	1601	< dl	< dl	< dl	512	< dl	677		0	0	0	0	0.4
		Q	P	C	K	G	BG		G/Q	P/Q	C/Q	BG/Q		
	O	2971	185	608	1430	204	433		0.1	0.1	0.2	0.15		
	W	3086	237	825	1601	< dl	500		0	0.1	0.3	0.16		
PH-34	Ac-7	3111	218	840	1531	< dl	450		0	0.1	0.3	0.14		
	Ac-5	4126	261	< dl	1576	< dl	583		0	0.1	0	0.14		
	NA	4329	< dl	< dl	1324	< dl	510		0	0	0	0.12		
		Q	P	M	C	K	F	G	BG	G/Q	P/Q	M/Q	C/Q	BG/Q
	O	1847	401	667	443	375	326	166	352	0.1	0.2	0.4	0.2	0.2
	W	1933	392	481	538	415	361	< dl	432	0	0.2	0.3	0.3	0.2
LDA-37	Ac-7	2248	454	613	343	446	380	< dl	458	0	0.2	0.3	0.2	0.2
	Ac-5	1902	655	704	< dl	334	403	< dl	307	0	0.3	0.4	0	0.2
	NA	2891	< dl	< dl	< dl	287	300	< dl	383	0	0	0	0	0.1

Table 5

The results of analyses of Hg, As, Se, Cd, Tl, Cr, Ni, Pb, Cu, and Zn in each extraction step of 32 coal samples. W water extractable; Ac7 exchangeable; Ac5 carbonate bound; NA organic matter and sulfide bound; SUM sum of four leacheable modes; BULK Total concentration in coal; Leachable(%): ratio of total leachable fractions with respect to bulk; LL-01, LS-02, LD-03, LD-04, LD-05, LM-06 sampled from Luizhi coal basin; SW-07, SW-08, SX-09, SX-10, SL-11, SL-12, SD- 13, SD-14, SH-15, SH-16, SW-17, SDL-18, SM-19, SM-20 sampled from Shuicheng coal basin; PS-21, PS-22, PY-23, PY-24, PL-25, PT-27, PT-28, PH-31, PH-34 sampled from Panjiang coal basin; LDA-37 and LDN-38 sampled from Guiyang coal basin

Elements		LL-01	LS-02	LD-03	LD-04	LD-05	LM-06	SW-07	SW-08	SX-09	SX-10	SL-11	SL-12	SD-13	SD-14	SH-15	SH-16
	W	0.004	0.0007	0.0008	0.017	0.002	0.0004	0.0001	0.0002	0	0.007	0.002	0.0003	0.001	0.0005	0.0003	0.0002
	Ac7	0.002	0.001	0.001	0.004	0.0004	0.002	0.001	0.0008	0.001	0.001	0	0.0009	0.001	0.002	0.0004	0
	Ac5	0.0019	0.0005	0.0004	0.0003	0.0004	0.0003	0	0.0005	0.0002	0	0	0.0003	0.0002	0	0	0
Hg(ug/g)	NA	2.42	0.445	0.295	0.148	0.301	0.379	2.169	0.676	0.329	0.271	0.155	1.251	0.544	0.891	0.264	0.895
	SUM	2.4279	0.4472	0.2972	0.1693	0.3038	0.3817	2.1701	0.6775	0.3302	0.279	0.157	1.252	0.5462	0.8935	0.2647	0.8952
	Bulk	2.67	0.657	0.368	0.138	0.276	0.504	2.11	0.958	0.357	0.318	0.169	1.207	0.612	0.911	0.394	0.992
	Leachable%	90.9	68.1	80.8	122.7	110.1	75.7	102.8	70.7	92.5	87.7	92.9	103.8	89.2	98.1	67.2	90.2
	W	0.046	0	0	0	0.15	0	0	0	0	0	0	0	0	0	0	0
	Ac7	0	0.066	0	0	0.034	0	0	0	0	0	0	0	0	0	0.007	0
	Ac5	0.024	0.017	0	0	0.048	0	0.029	0	0		0	0	0	0	0	0
As(ug/g)	NA	11.38	7.44	5.14	8.86	13.67	9.42	9.45	9.47	7.36	4.19	2.7	5.92	21.21	10.63	8.46	5.48
	SUM	11.45	7.523	5.14	8.86	13.902	9.42	9.479	9.47	7.36	4.19	2.7	5.92	21.21	10.63	8.467	5.48
	Bulk	10.43	10.05	5.92	10.18	16.45	12.03	11.98	11.68	7.65	7.22	3.33	10.43	21.65	13.03	6.7	5.78
	Leachable%	109.8	74.9	86.8	87.0	84.5	78.3	79.1	81.1	96.2	58.00	81.1	56.8	97.9	81.6	126.4	94.8
	W	0.028	0.044	0.014	0.08	0.029	0.003	0.008	0.017	0.029	0.081	0.229	0.226	0.678	0.016	0.001	0.014
	Ac7	0.064	0.015	0.015	0.032	0.029	0.017	0.038	0.05	0.028	0.034	0.043	0.054	0.073	0.022	0.017	0.028
	Ac5	0.014	0.016	0	0.024	0.058	0.019	0	0.026	0.025	0.052	0.055	0.087	0.096	0.032	0.091	0.059
Se(ug/g)	NA	1.41	0.16	0.085	1.05	3.937	1.351	1.312	1.947	1.717	2.237	2.145	2.007	2.517	3.059	2.607	1.387
	SUM	1.516	0.235	0.114	1.186	4.053	1.39	1.358	2.04	1.799	2.404	2.472	2.374	3.364	3.129	2.716	1.488
	Bulk	1.75	0.48	0.35	1.2	4.05	1.5	1.53	2.05	2.18	3.33	2.98	3.88	4.03	3.25	3.3	2.1
	Leachable%	86.6	48.9	32.6	98.8	100.1	92.7	88.8	99.5	82.5	72.2	82.9	61.2	83.5	96.3	82.3	70.9
	W	0.0027	0.0003	0.0011	0.0025	0.003	0.0007	0.0065	0	0.0017	0.0035	0.001	0.001	0.0027	0	0.00075	0.001
	Ac7	0.019	0.007	0.005	0.008	0.205	0.023	0.02	0.011	0.003	0	0.11	0.01	0.005	0.027	0.005	0.009
	Ac5	0.179	0.006	0.002	0.1	0.029	0.123	0.05	0.056	0.058	0.031	0.07	0.021	0	0.219	0.006	0.003
Cd(ug/g)	NA	0.052	0.023	0.12	0.01	0.147	0.149	0.035	0.087	0.019	0.061	0.058	0.03	0.045	0.059	0.03	0.019
	SUM	0.2527	0.0363	0.1281	0.1205	0.384	0.2957	0.1115	0.154	0.0817	0.0955	0.239	0.062	0.0527	0.305	0.04175	0.032
	Bulk	0.23	0.225	0.14	0.138	0.52	0.23	0.105	0.145	0.128	0.091	0.42	0.085	0.065	0.483	0.045	0.035
	Leachable%	109.9	16.1	91.5	87.3	73.8	128.6	106.2	106.2	63.8	104.9	56.9	72.9	81.1	63.1	92.8	91.4
	W	0	0	0	0.011	0.035	0.005	0	0	0	0	0.005	0.01	0.025	0	0.001	0
	Ac7	0.0154	0.021	0	0.219	0.013	0.019	0.061	0	0	0.013	0.03	0	0.062	0.005	0.103	0
	Ac5	0.07	0	0	0.082	0	0	0	0.008	0	0	0.008	0.056	0	0	0.15	0.061
Tl(ug/g)	NA	0.019	0.05	0	0.11	0.11	0	0	0	0.16	0	0	0.086	0	0	0	0
	SUM	0.1044	0.071	0	0.422	0.158	0.024	0.061	0.008	0.16	0.013	0.043	0.152	0.087	0.005	0.254	0.061
	Bulk	0.3	0.4	0.8	0.73	0.4	0.3	0.4	0.5	0.15	0.25	0.18	0.58	0.33	0.05	0.25	0.2
	Leachable%	34.8	17.75	0	57.8	39.5	8	15.25	1.6	106.7	5.2	23.9	26.2	26.3	10	101.6	30.5
	W	0.23	0	0	0	0.055	0	0	0	0	0	0	0	0	0	0	0
	Ac7	1.03	0.21	0.13	0.82	0.12	0.1	0.09	0.07	0.07	0.36	0.13	0.13	0.02	0.02	0	0.05
	Ac5	0	0	0	0.04	0	0	0	0	0	0	0	0.4	0	0	0.29	0
Cr(ug/g)	NA	12.67	9.63	6.4	7.5	9.47	12.25	9.16	15.17	25.16	18.19	2.66	17.1	0	1.25	2.24	0.25
	SUM	13.93	9.84	6.53	8.36	9.645	12.35	9.25	15.24	25.23	18.55	2.79	17.63	0.02	1.27	2.53	0.3
	Bulk	68	52.5	20.49	33.76	55.45	41.25	183.25	80.75	80.75	78.25	12.98	36.75	5.99	13.6	23.49	13
	Leachable%	20.5	18.7	31.9	24.8	17.4	29.9	5.0	18.9	31.2	23.7	21.5	47.9	0.3	9.3	10.8	2.3
	W	0.148	0.046	0.062	0.156	0.138	0.4	0.166	0.029	0.024	0.019	0.032	0.059	0.027	0.047	0.128	0.021
	Ac7	1.59	0.64	0	0	1.56	1.75	0.01	0	0	0	0	0.31	0.07	0.08	0.41	0.29
	Ac5	2.17	0.39	0.33	1.51	0.77	4.37	0.71	2.28	0.29	0.19	0.35	1.83	0.73	0.83	2.31	1.34
Ni(ug/g)	NA	3.55	0	0	0	0	7.25	0	14.78	16.09	22.28	6.32	10.4	10.75	5	2.03	0
	SUM	7.458	1.076	0.392	1.666	2.468	13.77	0.886	17.089	16.404	22.489	6.702	12.599	11.577	5.957	4.878	1.651
	Bulk	90	75	1.06	9.59	8.44	32.5	2.5	42.5	45	87.5	16.58	25.5	25.48	12.36	22.26	10
	Leachable%	8.3	1.4	36.9	17.4	29.2	42.4	35.4	40.2	36.4	25.7	40.4	49.4	45.4	48.2	21.9	16.5
	W	0.39	0.136	0.077	0.059	0.151	0.044	0.23	0.081	0.085	0.073	0.081	0.096	0.219	0.046	0.064	0.067
	Ac7	0.006	0	0	0	0	0	0	0	0.004	0.016	0.048	0	0.04	0	0	0
	Ac5	0.77	0.51	0.48	1.49	1.15	0.72	1.32	2.57	1.59	2.44	2.35	4.17	4.47	1.31	1.7	2.5
Pb(ug/g)	NA	14.91	16.25	11.7	5.28	8.96	3.02	7.9	11.68	11.21	11.06	15.74	8.07	13.68	3.75	4.33	6.25
	SUM	17.2	23.98	12.63	7.8	11.83	8.78	9.68	15.8	19.85	14.33	41.34	13.83	37.48	13.6	6.48	7.75
	Bulk	16.076	16.896	12.257	6.829	10.261	3.784	9.45	14.331	12.889	13.589	18.219	12.336	18.409	5.106	6.094	8.817
	Leachable%	93.5	70.5	97.0	87.6	86.7	43.1	97.6	90.7	64.9	94.8	44.1	89.2	49.1	37.5	94.0	113.8
	W	0.635	0	0	0.055	9.96	0.046	0.08	0.016	0	0	0	0	0	0	0	0
	Ac7	0.68	0	0	0.17	0	0.03	5.21	3.75	0.1	0	0	4.02	0.19	0	0	0.37
	Ac5	3.68	0.94	1.2	1.44	1.22	5.7	11.9	19.02	7.48	3.08	0.92	10.8	3.87	0.07	1.07	3.56
Cu(ug/g)	NA	38.52	16.25	15.7	13.35	17.98	62.5	34.65	46.02	56.05	30.71	18.16	76.05	38.56	18.75	23.81	13.75
	SUM	43.515	17.19	16.9	15.015	29.16	68.276	51.84	68.806	63.63	33.79	19.08	90.87	42.62	18.82	24.88	17.68
	Bulk	45	55	21.9	18.1	30.96	85	77.5	85	77.5	50	31.2	107.5	42.5	27.56	31.78	27.5
	Leachable%	96.7	31.2	77.2	82.9	94.2	80.3	66.9	80.9	82.1	67.58	61.2	84.5	100.3	68.3	78.3	64.3
	W	0.7	0.325	0.275	0.213	8.713	0.225	0.138	0.138	0.55	0.163	0.1	0.125	0.25	0.225	0.138	0.175
	Ac7	0.75	0.79	0	3.16	0.64	0	5.36	0.18	0	0	0.95	1.82	1.82	2.2	0.15	0.83
	Ac5	5.2	0.31	1.12	0.68	0.57	4.69	11.53	6.15	1.48	1.88	3.69	4.9	2.96	7.64	3.61	1.43
Zn(ug/g)	NA	45.68	8.03	19.44	6.24	7.22	24.65	10.18	22.48	13.44	32.87	5.71	21.13	11.04	15.4	13.9	3.53
	SUM	52.33	9.455	20.835	10.293	17.143	29.565	27.208	28.948	15.47	34.913	10.45	27.975	16.07	25.465	17.798	5.965
	Bulk	62.5	22.25	30.25	17.5	25	69.75	37.75	33	37.5	47.5	37.5	67.5	50	40	45	35
	Leachable%	83.7	42.5	68.9	58.8	68.6	42.4	72.1	87.7	41.3	73.5	27.9	41.4	32.1	63.7	39.5	17.0

Table 5
Continued

Elements		SW-17	SDL-18	SM-19	SM-20	PS-21	PS-22	PY-23	PY-24	PL-25	PT-26	PT-27	PH-28	PH-31	PH-34	LDA-37	LDN-38
	W	0.0002	0.0003	0.0003	0	0.0004	0.0001	0.0002	0.0003	0.001	0.0002	0	0	0	0.0002	0	0.0002
	Ac7	0	0.0004	0.0004	0.0003	0.0006	0	0.0004	0.0003	0.0008	0.0008	0.0004	0.0026	0.0003	0.0003	0.0004	0.0015
	Ac5	0.0002	0	0.0002	0	0.0006	0.0002	0	0.0003	0.0003	0	0	0	0	0.0003	0.0096	0
Hg(ug/g)	NA	0.958	0.259	0.264	0.148	0.321	0.247	0.239	0.454	0.121	0.117	0.107	0.16	0.328	0.224	0.279	0.18
	SUM	0.9584	0.2597	0.2649	0.1483	0.3226	0.2473	0.2396	0.4549	0.1231	0.118	0.1074	0.1626	0.3283	0.2248	0.289	0.1817
	Bulk	0.864	0.374	0.277	0.143	0.418	0.286	0.325	0.651	0.169	0.155	0.096	0.134	0.504	0.235	0.322	0.188
	Leachable%	110.9	69.4	95.6	103.7	77.2	86.5	73.7	69.9	72.8	76.1	111.9	121.3	65.1	95.7	89.8	96.6
	W	0	0	0	0.105	0	0	0.039	0	0.009	0	0	0	0	0	0	0
	Ac7	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0
	Ac5	0	0	0	0	0	0	0	0	0.016	0	0	0.043	0	0	0	0
As(ug/g)	NA	7.51	2.1	5.53	4.48	2.65	3.83	4.31	26.19	0.21	2.91	1.23	3.17	3.37	11.42	12.11	3.97
	SUM	7.51	2.1	5.53	4.585	2.65	3.83	4.349	26.19	0.235	2.91	1.23	3.213	3.37	11.42	12.1	3.97
	Bulk	6.88	2.33	5.43	4.38	1.9	3	4.13	19.08	0.23	3.53	1.3	4.3	4	12.38	12.68	2.93
	Leachable%	109.2	90.1	101.8	104.7	139.5	127.7	105.3	137.2	102.2	82.4	94.6	74.7	84.3	92.2	95.6	135.5
	W	0.023	0.061	0.057	0.053	0.026	0.112	0.136	0.046	0.031	0.158	0.046	0.035	0.203	0.073	0.037	0
	Ac7	0.02	0.045	0.024	0.069	0.032	0.05	0.031	0.024	0.02	0.027	0.005	0.005	0.039	0.043	0.011	0.031
	Ac5	0.045	0.073	0.036	0.117	0.02	0.086	0.054	0.03	0.057	0.08	0.075	0.056	0.071	0.105	0.06	0.009
Se(ug/g)	NA	1.717	2.137	1.065	3.707	0.637	1.437	1.247	1.507	0.917	1.847	1.717	1.607	1.598	1.978	2.687	1.547
	SUM	1.805	2.316	1.182	3.946	0.715	1.685	1.468	1.607	1.025	2.112	1.843	1.703	1.911	2.199	2.795	1.587
	Bulk	1.93	3.03	1.35	4.38	1.03	2.08	1.75	2.3	1.5	2.95	2.58	2.9	2.13	2.05	3.95	2.78
	Leachable%	93.5	76.4	87.6	90.1	69.4	81.0	83.9	69.9	68.3	71.6	71.4	58.7	89.7	107.2	70.8	57.1
	W	0	0	0.0014	0.00075	0	0	0.0011	0	0.0005	0.0045	0.002	0.0014	0.00025	0	0.0013	0
	Ac7	0.005	0.002	0.008	0.016	0	0.003	0.007	0.002	0.045	0.005	0.016	0.003	0.008	0.005	0.011	0.014
	Ac5	0.001	0.002	0.007	0.02	0	0.005	0.05	0.007	0.017	0.001	0.05	0.009	0.02	0.03	0.1	0.049
Cd(ug/g)	NA	0.034	0.057	0.025	0.1	0.001	0.044	0.043	0.034	0	0.036	0.038	0.025	0.032	0.035	0.136	0.195
	SUM	0.04	0.061	0.0414	0.13675	0.001	0.052	0.1011	0.043	0.0625	0.0465	0.106	0.0384	0.06025	0.07	0.2483	0.258
	Bulk	0.048	0.139	0.075	0.21	0.001	0.068	0.12	0.055	0.07	0.464	0.106	0.131	0.094	0.056	0.504	0.709
	Leachable%	83.3	43.9	55.2	65.1	100	76.7	84.25	78.2	89.3	10.0	100	29.3	64.1	125	49.3	36.4
	W	0	0	0	0.01	0	0	0	0	0	0	0	0	0	0	0	0
	Ac7	0	0.042	0.087	0.056	0.047	0.074	0	0.173	0.016	0.004	0	0	0.052	0	0	0.053
	Ac5	0.099	0.071	0	0	0.005	0	0	0	0	0.029	0	0.005	0.038	0	0.068	0
Tl(ug/g)	NA	0	0	0	0	0	0	0.0001	0	0	0	0	0	0	0	0	0.01
	SUM	0.099	0.113	0.087	0.066	0.052	0.074	0.0001	0.173	0.016	0.033	0	0.005	0.09	0	0.068	0.063
	Bulk	0.18	0.1	0.4	0.38	0.18	0.45	0.0001	0.65	0.55	0.2	0.38	0.25	0.23	0.33	0.35	0.73
	Leachable%	55	113	21.75	17.4	28.9	16.4	100	26.6	2.9	16.5	0	2	39.1	0	19.4	8.6
	W	0	0	0	0.068	0	0	0	0	0.041	0.861	0.001	0	0	0	0	0
	Ac7	0.02	0.02	0.04	0.11	0.08	0.04	0.04	0.04	0.01	0	0	0	0	0	0	0.02
	Ac5	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0.24	0
Cr(ug/g)	NA	0.53	3.81	6.59	7.23	3.69	9.8	21.05	15.74	0.69	8.24	40.43	14.17	3.59	6.67	4.34	0
	SUM	0.55	3.83	6.63	7.408	3.77	9.84	21.09	15.78	0.741	9.101	40.431	14.17	3.59	6.67	4.58	0.02
	Bulk	42.5	23.65	33.6	45.67	53.3	37.5	64.13	73.92	2.01	72.93	92.21	82.59	31.3	12.25	61.25	5.17
	Leachable%	1.3	16.2	19.7	16.2	7.1	26.24	32.9	21.3	36.9	12.5	43.8	17.2	11.5	54.4	7.5	0.4
	W	0.012	0	0.199	0.006	0.006	0.006	0	0	0.032	0.103	0.083	0.008	1.405	0.027	0.828	0.272
	Ac7	0.3	0.1	0.26	0.52	0.51	0.1	0.48	1.36	0.13	0.08	0.01	0	0.16	0.19	1.82	0.47
	Ac5	0.62	0.45	0.69	1.26	0.36	0.5	1.34	2.69	0.18	0.51	0.56	0.38	0.73	0.85	2.69	0.54
Ni(ug/g)	NA	6.11	5.71	8.55	0	0	6	7	12.75	0	8.66	15.94	4.89	5.83	5.82	21.38	3
	SUM	7.042	6.26	9.699	1.786	0.876	6.606	8.82	16.8	0.342	9.353	16.593	5.278	8.125	6.887	26.718	4.282
	Bulk	42.5	15.9	20.769	9.22	8.26	27.5	34.96	48.25	1.31	28.18	50.786	23.71	37.5	30	122.5	13.42
	Leachable%	16.6	39.4	46.7	19.4	10.6	24.0	25.2	34.8	26.1	33.2	32.7	22.3	21.7	22.9	21.8	31.9
	W	0.057	0.072	0.334	0.089	0.106	0.266	0.388	0.084	0.184	0.141	0.376	0.385	0.177	0.075	0.051	0.066
	Ac7	0	0	0	0.002	0.065	0	0	0	0.291	0	0.162	0	0.35	0.339	0	0
	Ac5	1.2	2.52	2.63	4.62	0.53	2.26	1.7	1.1	1.27	2.15	3.15	1.89	9.83	8.76	1.18	1.68
Pb(ug/g)	NA	4.7	7.39	18.66	20.54	3.99	10	8.98	6.8	0.89	7.94	5.92	10.97	21.97	17.92	5.8	8.95
	SUM	5.957	9.982	21.624	25.251	4.691	12.526	11.068	7.984	2.635	10.231	9.608	13.245	32.327	27.094	7.031	10.696
	Bulk	6.13	49.78	108.2	147.3	8.25	12.23	11.58	11.33	7.08	15.58	12.13	14.28	374.2	132.1	8.9	14.1
	Leachable%	97.2	20.1	19.9	17.1	56.9	102.4	95.6	70.5	37.2	65.7	79.2	92.7	8.6	20.5	79	75.8
	W	0	0	0.143	0	0.027	0.099	0.243	0	0	0.036	0.162	0.134	0	0	0.081	0
	Ac7	1.3	0.46	0	3.35	0	1.86	0.73	0.77	0	0.87	0.1	0	0	0	0	0
	Ac5	4.52	3.16	0.51	4.46	0.27	5.73	3.15	5.19	0.15	3.67	2.12	2.52	1.32	0.8	0.82	0.14
Cu(ug/g)	NA	13.37	17.24	9.95	44.37	4.99	48	26.93	51.49	3.98	41.71	20.71	17.96	10.96	6.97	33.6	7.95
	SUM	19.19	20.86	10.603	52.18	5.287	55.689	31.053	57.45	4.13	46.286	23.092	20.614	12.28	7.77	34.501	8.09
	Bulk	30	36.5	19.75	51	32.5	90	38	77	6.87	74.9	33.52	27.09	20	17.5	42.5	40
	Leachable%	63.97	57.2	53.7	102.3	16.3	61.9	81.7	74.6	60.1	61.8	68.9	76.1	61.4	44.4	81.2	20.2
	W	0.25	0.2	0.188	0.175	0.225	1.388	0.575	0.2	0.138	0.15	0.225	0.138	0.163	0.038	0.188	0.163
	Ac7	7.21	1	0.16	0.89	1.17	0.83	0.46	6.41	5.22	0.97	3.13	0.51	0.41	0.34	0.26	0.45
	Ac5	10.67	2.02	0.45	3.57	8.88	2.31	2.46	3.07	2.54	3.8	9.09	2.73	0.91	1.45	1.65	2.61
Zn(ug/g)	NA	10.87	7	3.7	48.56	0	9.7	12.93	25.73	0	9.03	7.88	18.42	4.44	2.54	31.19	25.48
	SUM	29	10.22	4.498	53.195	10.275	14.228	16.425	35.41	7.898	13.95	20.325	21.798	5.923	4.368	33.288	28.703
	Bulk	55	47.5	35	77.5	45	40	55	47.5	35	37.5	42.5	52.5	37.5	32.5	67.5	50
	Leachable%	52.7	21.5	12.8	68.6	22.8	35.6	29.9	74.5	22.6	37.2	47.8	41.5	15.8	13.4	49.3	57.4

leaching sequences are as follows: (1) dissolution of gypsum during the deionized and distilled water extraction; (2) dissolution of calcite and dolomite during NH_4Ac (pH = 7,and pH = 5) extraction; (3) dissolution of pyrite and marcasite during nitric acid extraction, and a little amount of organic matrix was dissolved in some sample at the same time.

The mobility of potentially toxic trace elements in coal

Table 5 shows the results of the analysis of the leachates obtained from the sequential extraction. From Table 5 it can be seen that the elements such as Hg, As, Se, Cd, Cu, and Pb have the highest mobility at surface conditions; and the average extractable fractions of Hg, As, Se, Cd, Cu, and Pb are 86%, 95%, 79%, 76%, 69%, and 69% of the total amount in coal, respectively. The elements in coal with the lowest leachability include Tl, Cr, and Ni; the average extractable fractions of them are 30%, 20%, and 29% of the total amount in coal, respectively. Zinc has an intermediate behavior, and the average leachable fraction of it accounts for 46% of the total amount in coal.

From Table 5, it can be seen that the amount of Hg, As, Se, Cu, and Pb in leachates from nitric acid accounts for most part of the bulk contents of these elements in coal (average 85.4% of total Hg, 94.8% of total As, 71.8% of total Se, 59.6% of total Cu, and 54.7% of total Pb in coal are found in the nitric extraction). As stated above, all of the pyrite and marcasite in coal are dissolved in all samples, and only a little amount of organic matrix is dissolved in part of the samples during nitric acid extraction. From the properties of elemental geochemistry of these elements, it can be seen that they all have a sulfide affinity, therefore, Hg, As, Se, Cu, Pb, and Zn mainly exist in sulfide minerals in coal. Of course, the possibility that there is a little amount of these elements existing in organic matter cannot be excluded. The leachable fraction in the nitric acid extraction of Cd accounts for 41.1% of the total amount of Cd in coal, and that 1 mol/l of NH_4Ac(pH = 5) extraction makes up to 23.5% of total amount. In addition, there is about 36.7% of the total amount of Cd in coal existing in the residue of the nitric acid extraction. This shows that Cd may exist in silicates, carbonates and sulfides in coal. On an average, 54% of the total amount of Zn in coal is in the residues of the nitric extraction, while another 32.1% is found in nitric acid leachates. Therefore, Zn in coal mainly exists in sulfide minerals and silicates. On the average, 70% of the total amount of Tl, 80% of total amount of Cr, and 71% of total amount of Ni in coal exist in silicates. Since Cr is a lithophile element, it could exist in both primary and secondary silica minerals in coal. For Ni and Tl, they are sulphophile elements, and would be supposed to enter sulfide phases during peatification and coalification processes if they existed in primary minerals, therefore, in these coal samples most Ni and Tl could exist in secondary silica minerals which were brought in coal from surrounding rocks.

From the discussion above, the conclusion can be made that the mobility of PTTEs in coal depends on the speciation of these elements. The elements associated with sulfates, carbonates, sulfides and some organic matter in coal show the highest extraction rates during the weathering process, while elements with silicate affinities are inert at surface conditions. Since the speciation of PTTEs in coal varies among coals in different areas, and among the coal seams even in the same area, the mobility of these elements in one coal differs from that in another coal greatly.

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