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Distribution and Accumulation of Metals in Soils and Plant from a Lead–Zinc Mineland in Guangxi, South China

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Abstract Six kinds of metals were measured in soils and plant from the Siding Pb–Zn mineland, South China, to investigate the pollution and restoration technique. The mean Pb, Zn, Fe, Mn, Cu and Cr contents in soils were 5,215 ± 642, 13,352 ± 1,242, 24,755 ± 2,475, 438 ± 52, 67 ± 38 and 68 ± 31 mg kg^{-1}, respectively. The results revealed the mean contents of Pb and Zn exceeded the third level of China standard of soil (GB15618-1995) for 10.4 and 26.7 times, respectively. The soil of Siding Pb–Zn mineland has heavily been subjected to Pb and Zn pollution. 22 plant species from 13 families were found colonizing. There were great variations of metal contents in plant species with Pb 1.58–1496 mg kg^{-1}, Zn 7.56–204,256 mg kg^{-1}, Cu not detected (ND)–286 mg kg^{-1}, Fe 83–25,972 mg kg^{-1}, Mn 1.02–160 mg kg^{-1} and Cr ND–152 mg kg^{-1}. Of the six metals, Fe content was the highest, followed by Pb and Zn, which were similar to the situation of the soil. The local pioneer species, *Pteris vittata,* was observed a higher accumulation and translocation capability for Pb, which could be chosen as pioneer of phytoremediations to restore Pb/Zn mineland.

Keywords Metal pollution · Phytoaccumulation · Agricultural restoration

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The metal pollution are of ubiquity which has become one of the most seriously problems in mineland (Navas-Acien et al. 2007). Bad soil structure, lack of organic matter and nutrient element, and higher metal content altogether lead to environmental restoration to be hard. While, some special metallophytes can grow well in soil polluted by metals for the reason of natural selection, which therefore take the crucial function on eco-restoration of the polluted land, although many studies on eco-restoration of metal-mined wastelands were conducted (Chehregani et al. 2009; Chen et al. 2008), reports on eco-restoration of Pb/Zn minelands are scanty. Guangxi province, one of the most well developed karst areas, ranks the fourth of China in terms of Pb/Zn mining. There surface soil is in serious shortage, and further deteriorated by expanding soil erosion and rocky desertification (Bhagure and Mirgane 2010). Agricultural reclamation of mineland has long been a tradition in China, but its risk has been overlooked. The trend is more popular in Guangxi because of poverty and severe lack of cultivated land. Restoration techniques for metal-mined wasteland and assessment of the agricultural restoration pattern are equally important for Guangxi area.

Siding Pb–Zn Mine is located at longitude 109°31′17″–109°31′37″E, latitude 25°02′31″–25°02′54″N, 124 km north-east from Guilin city, South China. This mine had been extracted (close-cast mining) since 1960 and basically closed in 2004. Reclamation of destroyed land was conducted in the early 1990s, mainly by planting crops and vegetable for income generation. Over 10 years of rehabilitation by natural or artificial processes has allowed a good vegetation cover (>70%) over the most of the mineland. The landform is hilly with laterite soil, the regional vegetation is subtropical evergreen broadleaf forest.

Materials and Methods

Field survey and sampling were conducted in October 2008. 22 plant species from 13 families species encountered were recorded and their relative abundances were estimated visually and described as 1–5 (from very rare to very abundant). The mineland was divided into seven subareas in terms of the dominant plants colonized and mineland purpose (Fig. 1): (1) A natural restoration area (soil sampling site: S1), the soil surface has slumped in many plots due to the impact of groundwater while mining. Many natural *Rhizoma imperatae* and *Arthraxon bispidus* were collected; (2) The area around smelt factory was set as S2. The wastewater from smelt factory was drained into a small river which is about 10 m apart from the factory. The *Phragmites communis* Trin and *Acorus calamus* L. were collected on the shoreside; (3) The area polluted by wastewater from a drainage tube located on the hill adjacent to La Qiong village (S3), the wastewater flowed along the hill and led to form a zone about 300 m^2 with grey and black sludge. Only *Pteris vittata* and *Rosa laevigata* were collected near the zone; (4) The flat ground lies at the foot of the hill above (S4). A dense shrub belt is canopy closed, where *Equisetum ramosissimum* and *P. vittata* were collected. (5) Tailing dump (S5), there four types of plant, *Taraxcum mingolicum*, *Ficus tikoua*, *Miscanthus floridulus* (Labill) Warb and *Ageratum conyzoides* were gathered. (6) Hole tailing dump (S6), ounce tailings still remained. The *Flos buddlejae* and *Pteris multifida* were collected. (7) Artificial agricultural restoration field (S7). The *Brassica chinensis*, *Zea mays* L. and *Arachis hypogaea* L. were collected. For each sampling site, surface soils (0–20 cm) were sampled separately. The 3–5 subsamples were merged into one single sample and three parallel samples were collected. Samples of dominant plants for each area, including some agronomic species, were collected. All soil and plant samples were sealed with polythene bags and transported into laboratory.

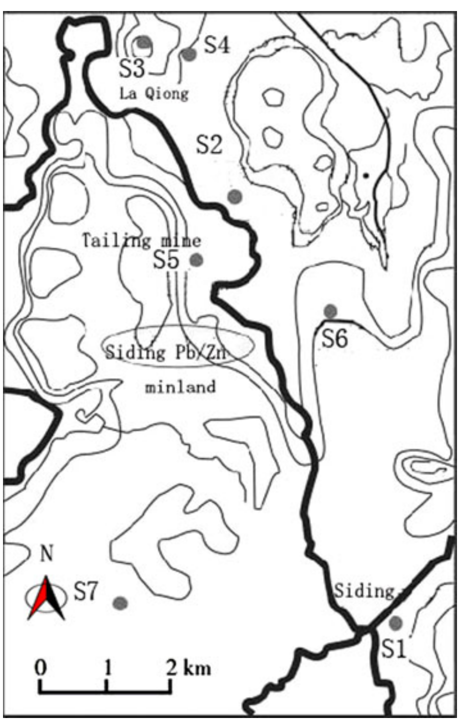

Fig. 1 The schematic drawing of sampling subareas and positions. *S1* a natural restoration area; *S2* the area around smelt factory; *S3* wastewater drainage in La Qiong country; *S4* the foot of the hill near the wastewater drainage; *S5* tailing dump; *S6* hole tailing dump; *S7* artificial agricultural restoration field

Soil samples were air-dried, ground and passed through 100 mesh plastic sieve. Plant samples were gently rinsed with deionized water. Dried plant tissues were ground into fine powder. Soil samples were digested with concentrated HCl + concentrated HNO_3 + HF + $HClO_4$ (10:5:5:3, v/v) and plant tissues digested with concentrated HNO_3 + $HClO_4$ (10:3–5, v/v). The total metal concentrations (Pb, Zn, Fe, Mn, Cu, and Cr) in the digestates were then determined by PE-AA700 Atomic Absorption Spectrophotometer. Analytical limits of detection were 0.03 mg L^{-1} for Zn, 0.06 mg L^{-1} for Cu and Mn, 0.1 mg L^{-1} for Fe and 0.2 mg L^{-1} for Pb and Cr. The analytical recovery range of six kinds of metal was 98.7%–106.4%, which was measured up to the analytical demand (QA/QC). Data with replicates were presented as mean ± standard deviation (SD).

Results and Discussion

Descriptive statistics of metal contents in soils of Siding mineland and comparison with those in background and China standard of soil (GB15618-1995) were shown in Tables 1 and 2. The mean contents descended in the order: Fe > Zn > Pb > Mn > Cr > Cu. In S2, the contents of Cu and Cr (146 and 110 mg kg^{-1}) were higher than those in other subarea, Mn content in S4 was the highest (846 mg kg^{-1}), so Cu, Cr and Mn contens did not exceeded the second level of China standard. Pb and Zn values in S2 were obviously higher than other subareas with 36 and 65 times of the third level of China standard respectively, in other subarea, Pb and Zn also exceeded the third level of standard for 3.6–13 and 6.5–53 times, respectively. Cu and Cr contents were lower than the second level of the standard. The mean contents of metals in soils all exceeded local background values except Fe. Pb and Zn contents were critically high, representing 267 and 2,577 times of local background values respectively. On the whole, the soils of Siding mineland were not polluted by Fe and Mn, but Pb and Zn values exceeded the third level of China

Table 1 Descriptive statistics of metal contents in soils of Siding mineland (mg kg^{-1})

	S1	S2	S3	S4	S5	S6	S7	Range	Mean ± SD
Pb	1,800	18,212	5,138	2,500	2,500	6,500	1,700	1,700–18,212	5,215 ± 642
Zn	3,236	32,375	14,041	4,500	9,375	26,500	3,440	3,236–32,375	13,352 ± 1,242
Fe	9,153	50,654	36,427	16,700	14,425	37,852	9,168	9,153–50,654	24,755 ± 2,475
Mn	214	310	596	846	630	246	212	212–846	438 ± 52
Cu	36	146	109	48	42	56	33	33–146	67 ± 38
Cr	50	110	67	59	100	46	45	45–110	68 ± 31

Table 2 Comparison of metal contents with those in background and China standard (GB15618-1995) (mg kg^{-1})

Metal	Pb	Zn	Fe	Mn	Cu	Cr
This study (mean ± SD)	5,215 ± 642	13,352 ± 1,242	24,755 ± 2,475	438 ± 52	67 ± 38	68 ± 31
Guangxi soil background	19.5	5.18	24,500–50,300	176	23.1	65.3
National soil background	23.6	67.7	NA	103-342	20	53.9
GB15618-1995 (2nd level)	250	200	NA	NA	150	150
GB15618-1995 (3rd level)	500	500	NA	NA	400	NA

NA not ruled, *n*: sample number ($n = 21$), *SD* standard deviation

standard. The soil of Siding Pb–Zn mineland has heavily been subjected to Pb and Zn pollution, which were much more serious than those previously reported (Rizo et al. 2011; Zhou et al. 2007; Lu and Bai 2010). The whole pollution trends were: the smelt factory > hole tailing dump > the area of drainage tube > tailing dump field > agricultural restoration field > natural restoration area. Both the mineral residue and tail mine heap were the reason of the most serious pollution occurred around the area of smelt factory. The plant grew well in tail dam field and the soil was contaminated slightly, showing there the soil has been improved greatly due to the natural phytoaccumulation over several years (Nouri et al. 2009; Lone et al.2008).

The metal contents from 16 plant samples are presented in Table 3. There were great variations in metal contents with Pb 1.58–1,496 mg kg^{-1}, Zn 7.56–204,256 mg kg^{-1}, Cu not detected (ND)–286 mg kg^{-1}, Fe 83–25,972 mg kg^{-1}, Mn 1.02–160 mg kg^{-1} and Cr ND–152 mg kg^{-1}. Of the six metals, Fe content was the highest, followed by Pb and Zn, which were similar to the situation in the soil. Compared with the normal content for metals in plants (Chen et al. 2008), only Mn content in all plant studied measured up. Pb and Zn values in all plants were overrun with a peak value for Pb in the root of *R. laevigata* and a peak value for Zn in the root of *P. vittata*, but Pb and Zn contents in caudexes and leaves of the two plants was below the mark as hyperaccumulators (Backer and Brooks 1989).

Bioaccumulation coefficient (BAC) is defined as a ratio of metal content in plants compared to that in soil, which reflects the bioaccumulation capability of the plant to metals in soil (Mani et al. 2007). Biological transfer coefficient (BTC) is a ratio of metal content in upper ground to that in the root of a plant. It shows the capacity of transferring metal from the root to the caudexes and leaves. Table 4 presented the BAC and BTC of the 14 dominant plants. The BAC for 6 metals was all less than 1 except *P. vittata*, which for Cr was 2.27. Generally, metal accumulated far more in plant roots than caudexes or leaves. Meanwhile, the metal contents in leaves were often higher than those in caudexes. Probably it is an avoidance strategy to the high level toxic metals by defoliation. Pb values in *P. vittata*, *T. mingolicum* induced a stronger transferring capability on multiplicate metals (Zn, Mn, Cu and Cr), suggesting in case both the BAC and BTC were larger than 1, the plant had phytoaccumulation potential to contain metal elements (Salt et al. 1995). Such status was accorded by *P. vittata* to Cr in present study, in addition to considering the low Cr content in soil, hence it might illuminate a bioaccumulation plant on Cr, *P. vittata*.

In planning the restoration of metal-mined wasteland, agricultural utilization is usually the last option in the developed countries. Probably owing to the poverty and the extreme shortage of cultivable lands in Guangxi, growing agronomic plants on the reclaimed mineland is more common than other parts of China. The major edible plants grown in this artificial agricultural restoration field include peanut, corn, orange tree and other vegetables. Worries arose because there were usually no protective treatments in place before planting and no monitoring of toxic metals was conducted before they enter the food chain. Pb and Zn contents in the planted cabbage detected were 76 and 5

Table 3 Metal contents of 16 dominant plant species in Siding mineland (mg kg^{-1})

Species	Localization	Tissue	Pb	Zn	Fe	Mn	Cu	Cr
Rhizoma imperatae	S1	Root	759	3,989	6,433	50	44	32
		Shoot	190	1,516	1,544	33	12.4	24
Phragmites communis Trin	S2	Root	1,480	1,311	1,374	8.2	23	8.5
		Shoot	93	1,463	745.5	7.1	17.1	9.1
		Leaf	578	500	2,528	10.0	17.6	14.3
Pteris vittata	S3	Root	468	20,425	24,950	160	286	142
		Shoot	730	2,538	7,110	56	47	152
Rosa laevigata	S3	Root	1,285	13,400	939	24	26	4.2
		Shoot	256	587	775	18.1	16.5	14.8
		Leaf	126	550	936	41	13.4	26
Alchornea trewioides	S3	Root	378	638	1,298	30	15.6	8.5
		Shoot	340	562	720	25	18.7	6.3
		Leaf	164	662	649	31	12.3	20
Equisetum ramosissimum	S4	Root	152	1,250	9,497	53	18.5	33
		Shoot	103	575	8,263	28	8.3	9.3
Cydonia oblonga Mill	S4	Root	959	2,325	4,590	59	12.9	140
		Shoot	111	300	525	17.2	7.6	7.2
Taraxcum mingolicum	S5	Root	23	125	179	4.7	9.0	4.4
		Shoot	36	264	88	10.6	21.8	37
		Leaf	86	875	158	15.8	13.2	15.7
Ficus tikoua	S5	Root	515	1,700	1,344	45.5	13.6	33
		Shoot	92	338	99	11.0	3.9	4.6
		Leaf	126	525	73.3	47	3.7	5.0
Miscanthus loridulus	S5	Root	217	1,231	2,371	34	16.2	31
		Shoot	324	1,163	1,711	34	7.1	51
Ageratum conyzoides	S5	Root	1,496	4,079	546	51	24	33
		Shoot	703	1,550	519	40	11.5	22
Flos buddlejae	S6	Root	793	2,500	1,393	30	17.5	9.9
		Shoot	495	1,438	1,023	20	14.1	4.4
		Leaf	305	1,300	1,768	30	15.8	23
Pteris multifida	S6	Root	278	16,175	25,972	104	45	78
		Shoot and leaf	537	2,650	2,931	43	13.8	8.1
Brassica chinensis	S7	Root	54	344	69	24	5.6	3.8
		Shoot and leaf	15.2	110	60	5.2	1.6	1.0
Zea mays L.	S7	Root	10.9	123	189	5.6	1.7	ND
		Seed	1.2	23.0	70	1.1	ND	ND
Arachis hypogaea L.	S7	Seed	4.4	46.0	83	2.4	1.2	ND
		Leaf	1.6	7.6	10.5	1.0	ND	ND
Normal content			0.1–41.7	1–160	350	1–700	0.4–46	0–8.4

Data were mean value of the two parallel measurements

ND not detected

times higher than the prescribed limits by China food safety standard (0.2 mg kg^{-1} for Pb, 20 mg kg^{-1} for Zn) respectively. Moreover, corn and peanut also have been analyzed, Pb values in edible parts were 1.2 mg kg^{-1}, 4.4 mg kg^{-1}, Zn values were 23.0 mg kg^{-1}, 46.0 mg kg^{-1}, respectively, which all went beyond the prescribed limits by China Food Safety Standard. Combined with some reports from other restoration minelands in Guanngxi, it was considered unfit for planting edible vegetables and crops currently in the mineland. In the primary process of mineland restoration, planting some economic tree species and ornamentals could be appropriate.

Table 4 Bio-accumulating coefficient (BAC) and biological transfer coefficient (BTC) of the dominant species in Siding mineland

Species	Pb		Zn		Fe		Mn		Cu		Cr	
	BAC	BTC	BAC	BTC	BAC	BTC	BAC	BTC	BAC	BTC	BAC	BTC
Rhizoma imperatae	0.13	0.25	0.47	0.38	0.19	0.24	0.16	0.67	0.35	0.28	0.49	0.75
Phragmites communis Trin	0.03	3.89	0.02	0.38	0.05	1.84	0.03	1.22	0.12	0.76	0.13	1.68
Pteris vittata	0.14	1.56	0.18	0.12	0.19	0.29	0.09	0.35	0.432	0.17	2.27	1.08
Rosa laevigata	0.02	0.10	0.04	0.04	0.03	1.00	0.07	1.74	0.12	0.51	0.4	6.35
Alchornea trewioides	0.03	0.43	0.05	1.04	0.02	0.50	0.05	1.03	0.11	0.79	0.30	2.38
Equisetum ramosissimum.	0.06	0.68	0.13	0.46	0.5	0.87	0.03	0.52	0.17	0.45	0.16	0.28
Cydonia oblonga Mill	0.06	0.12	0.07	0.13	0.03	0.11	0.02	0.29	0.16	0.59	0.12	0.05
Taraxcum mingolicum	0.05	3.70	0.10	7.00	0.01	0.88	0.02	3.39	0.35	1.47	0.16	3.56
Ficus tikoua	0.07	0.25	0.06	0.31	0.01	0.05	0.07	1.03	0.10	0.27	0.04	0.15
Miscanthus loridulus	0.17	1.49	0.12	0.94	0.12	0.72	0.05	0.99	0.19	0.44	0.46	1.67
Ageratum conyzoides	0.37	0.47	0.17	0.38	0.04	0.95	0.06	0.78	0.30	0.48	0.20	0.67
Flos Buddlejae	0.20	0.39	0.05	0.52	0.05	1.27	0.12	1.01	0.28	0.90	0.50	2.32
Pteris multifida	0.08	1.93	0.10	0.16	0.08	0.11	0.17	0.41	0.25	0.31	0.18	0.10
Brassica chinensis	0.01	0.28	0.03	0.32	0.01	0.90	0.02	0.22	0.05	0.28	0.02	0.27

Zea mays L. (no shoot and leaf data) and *Arachis hypogaea* L. (no root data) were omitted in the table because their BAC and BTC values were not available

Phytoremediation mainly includes phytoextraction (removal of metals from soil through hyperaccumulators) and phytostabilization (immobilize and/or detoxify metals in soil). So far most Pb hyperaccumulators were found in mining wastelands or metal-contaminated sites. In addition to the metal concentrations suggested by Baker and Brooks (1989) to qualify a hyperaccumulator, once BAC and BTC are larger than 1, potential metal hyperaccumulators can be defined (Mani et al. 2007). Accumulators tend to accumulate more metals in soils with higher metal concentrations before they reach the maximum accumulation potential. For an effective restoration of mine wasteland, identification and characterization of native plant species on mineland is essential to develop phytostabilization technologies. A desired species for revegetating mine spoils should best grow well on poor and harsh soils and develop the vegetation cover in a relatively short time, accumulating biomass rapidly. In addition, shortage in roots is most beneficial for phytostabilization of the metal contaminants which are available. In south China, pioneer herbal species *C. dactylon, D. sanguina, L. cyclindrica* var. major, *Sesbania rostrata, Paspalum notatum, P. distichum, Vetiveria zizanioides* (Zhou et al. 2007) have been proven suitable for reclamation of Pb/Zn mine wasteland. Woody species including *Castanea mollissima, Pinus massoniana, P. elliottii, Casuarina equisetifolia, Eucalyprus robusta* (Chehregani et al. 2009) and legumes *Leucaena leucocephala* and *Acacia auriculiformis* have demonstrated good results in the restoration of mine soils as well as tailings dams after some amelioration (Sun et al. 2010).

In this study, although *P. vittata* was not qualified for a hyperaccumulator, it was still demonstrated a high accumulation and transfer ability for Pb. The *P. vittata* contained a high Pb content in the caudexes and leaves with 729.9 mg kg^{-1} and the BTC value was 1.56. On the other hand, The *P. vittata* extracted most of the six kinds of metal in every dominant plant species sampled in this study, whch implied *P. vittata* has a strong resistibility to the multiplicate metal pollution. It was documented the ciliate desert-grass, named academically by *P. vittata,* was of importance to restore the soil contaminated by multiplicate metals such as Pb, Zn, Cu and As (Zhou et al. 2007). Hereby, *P. vittata* might be an ideal specie applied to restore the polluted soil by multiplicate metals. Moreover, *F. buddlejae* is a species worthy to be noticed. Though Pb contents in the caudexes and leaves of *F. buddlejae* were lower than those in *P. vittata*, but higher obviously than other species. The more importance is, acted as a small arbor kind plant, its biomass is far more than other herbage plants, the total Pb quantity absorbed from the soil of *F. buddlejae* thus should be far more than that of *P. vittata.* Mani et al. (2007) considered arbor kind plant would be more potential used as hyperaccumulator for soil polluted by metal due to its more biomass and flourishing root than those of herbage kind plant. The *F. buddlejae* was found growing well in soils polluted badly by Pb, and its leaves accumulated higher concentration of Pb (305 mg kg^{-1}) and formed a harmonious community environment together with other herbage plants. Hereby, *F. buddlejae* might be a significantly potential hyperaccumulator in dealing with the soil polluted in Pb/Zn mineland. It was discovered that *P. vittata* and *F. buddlejae* were plants existing in nearly all of the sampled subareas with plentiful biomass and well growth, indicating they have powerful diffusivity and adaptability despite growing in the soil polluted seriously by Pb/Zn and could be chosen as pioneers of phytoremediations to restore Pb/Zn mineland.

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