Appendix A. Supplementary data

Leaching of heavy metals from farmland soils in China: The status and ecological risk assessment

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Text S1: Empirical coefficients (1/12 and 1/6)

The origin of the empirical coefficient 1/12:

Cambier et al. (2014) and Filipović et al. (2016) have published data from the QualiAgro field experiment in Feucherolles, near Paris (France), which belongs to the French national SOERE-PRO network. They measured Cd water concentrations and fluxes from nearly six years of lysimeter sampling at a 45 cm depth, from three plots cultivated with maize and wheat. One was a control plot, the other two received either sewage sludge compost or municipal waste compost. The Cd concentration measured in the percolating water (Cambier et al., 2014) was 15 times lower than that predicted by Eq. (1). **The Cd leached ranged between 0.11 and 0.14 g Cd ha^{-1} yr^{-1} (Filipović et al., 2016), i.e. about 12 times lower than that estimated with Eq. (1) .**

The origin of the empirical coefficient 1/6:

Bengtsson et al. (2006) measured Cd concentrations and fluxes in percolating soil water and surface run-off on arable land, in an experimental farm from Northern Sweden. They collected soil water for five years, from four sampling sites differing in soil type and agricultural practice. There were three to four replicated water sampling devices (suction cup lysimeters) collecting water at three depths (20, 50 and 80 cm). Topsoils contained 0.10–0.11 mg Cd kg^{-1}. The Cd concentration measured in the solutions at the different depths was on average 12 times lower than that estimated with Eq. (1) . **However, the authors estimated the average Cd flow through percolation and run-off to be 0.35 g Cd ha^{-1} yr^{-1}, which is about 6 times lower than the 2.56 g Cd ha^{-1} yr^{-1} average leaching calculated at the European level by Six and Smolders (2014) using Eq. (1).**

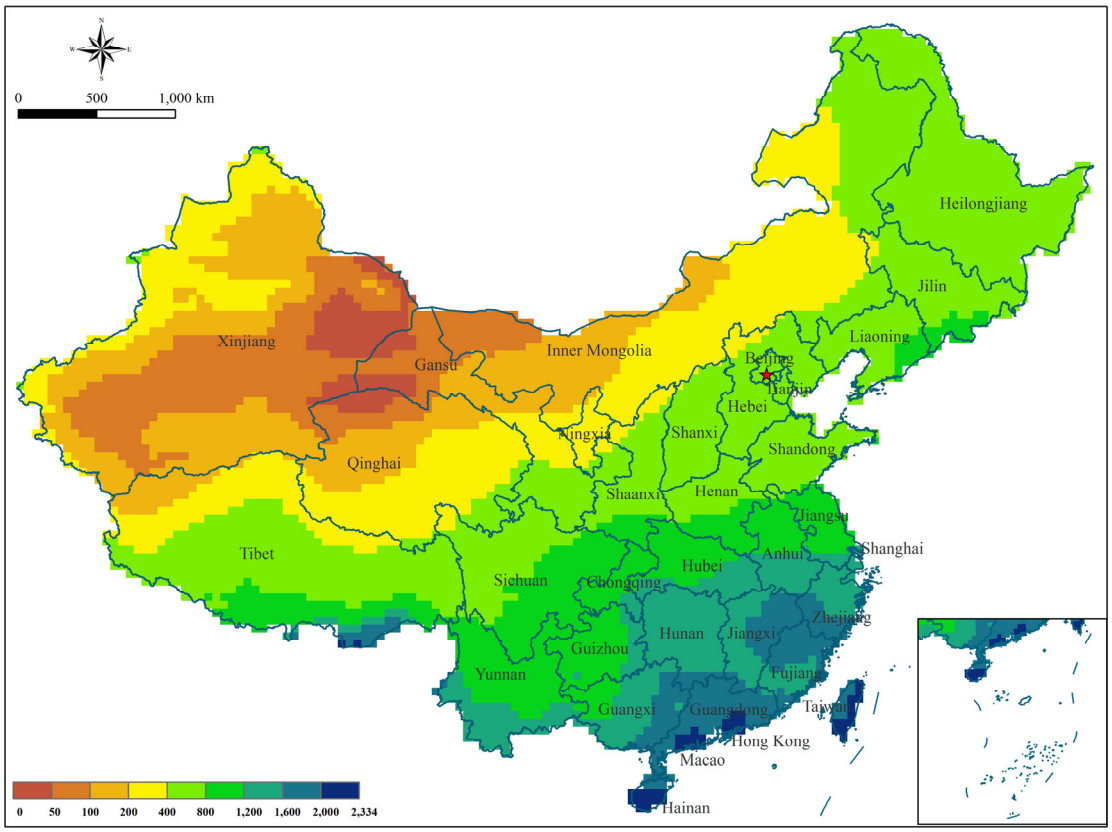


Figure S1. Geographical distribution of average annual precipitation in mm in China.

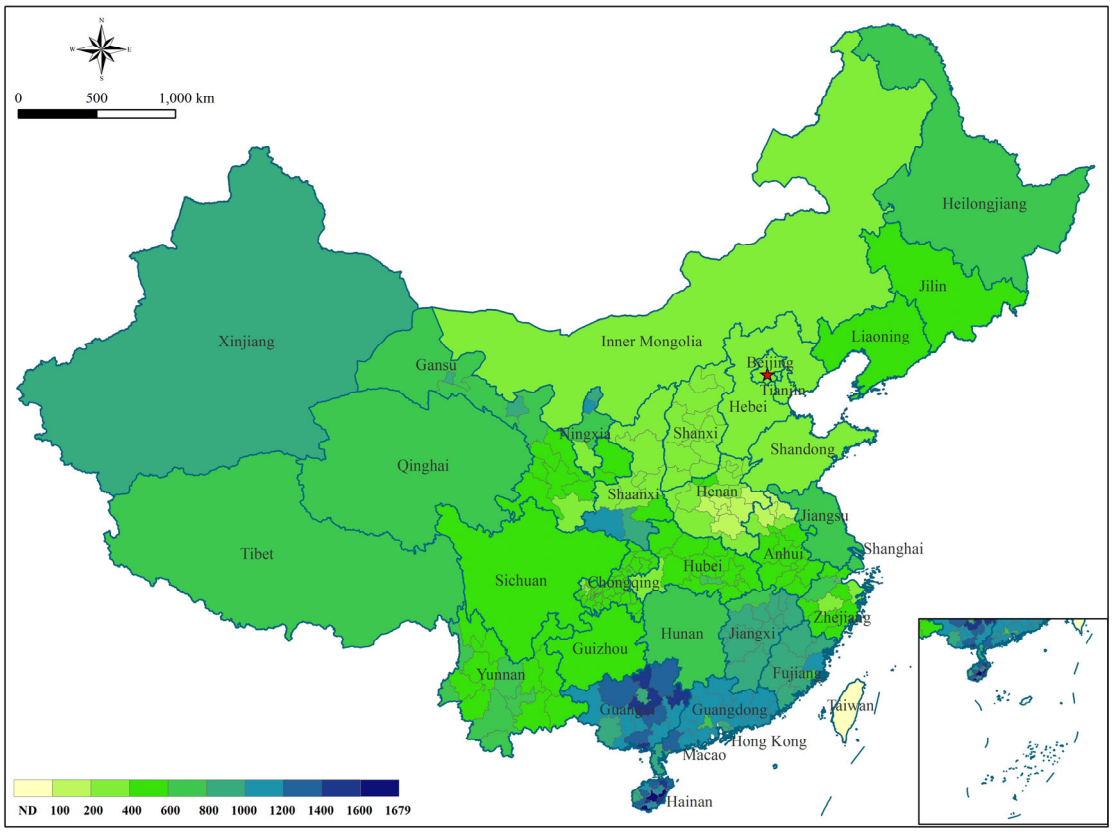


Figure S2. Geographical distribution of average annual irrigation in mm in China.

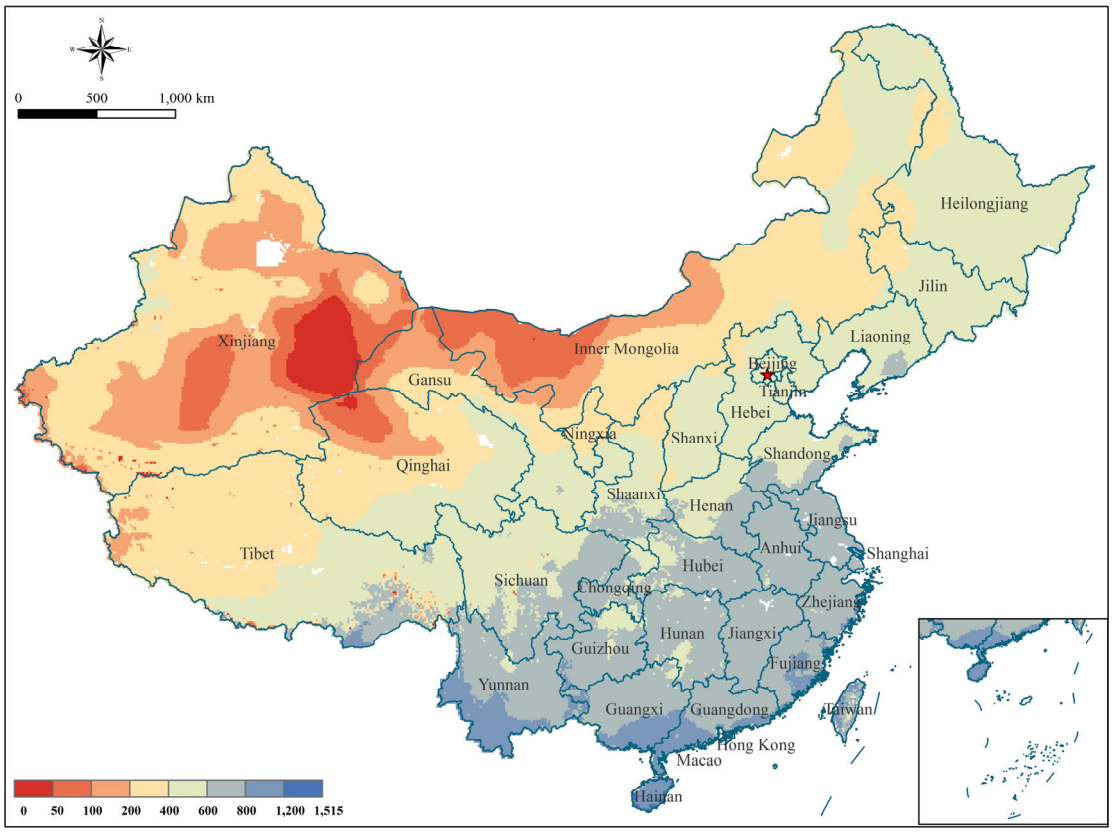


Figure S3. Geographical distribution of average annual evapotranspiration in mm in China

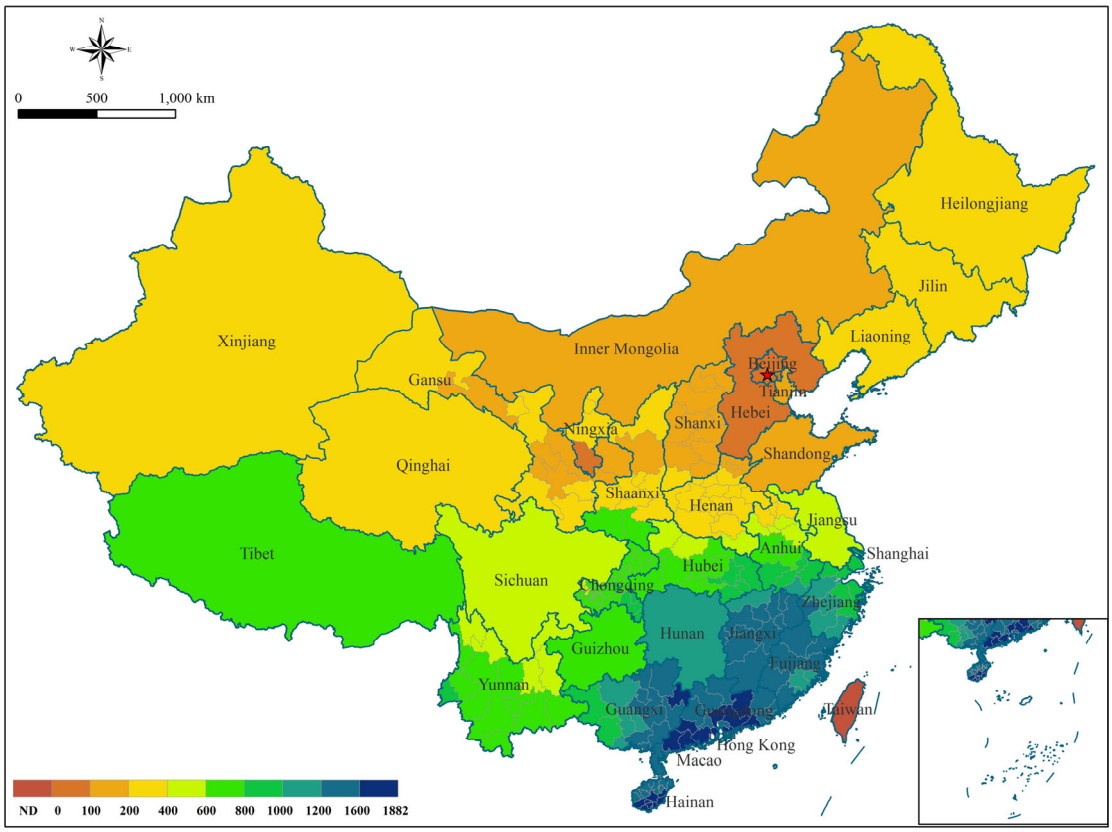


Figure S4. Geographical distribution of average annual infiltration water in mm in China.

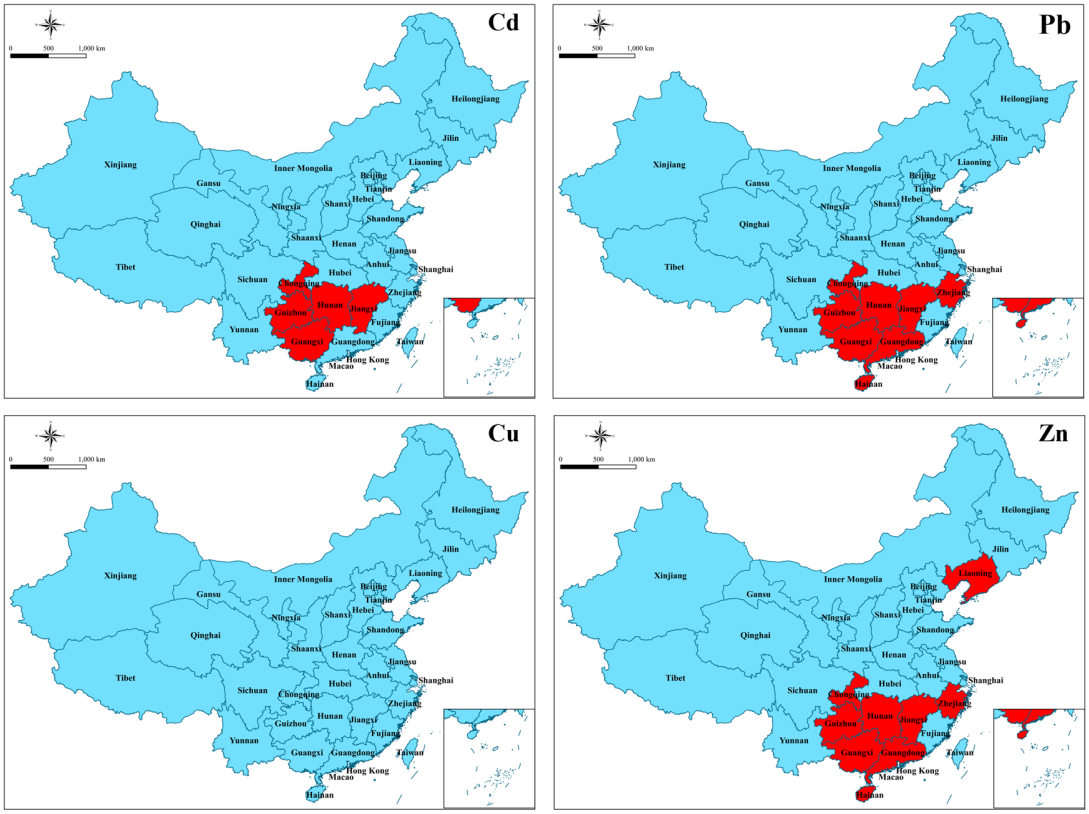


Figure S5. The calculated heavy metals content in the leachate exceeds the drinking water quality (GB 5749-2022). The red area exceeds the standard, and the blue area does not exceed the standard.

Table S1. Mean heavy metal content in soil used in Leaching models.

Province	Total Cd* mg kg^{-1}	Total Pb* mg kg^{-1}	Total Cu* mg kg^{-1}	Total Zn* mg kg^{-1}
Beijing	0.153	23.57	23.9	83.77
Tianjin	0.351	25.99	32.86	82.03
Hebei	0.181	26.42	26.39	81.3
Shanxi	0.18	27.22	31.38	80.15
Inner Mongolia	0.142	23.24	23.87	63.03
Liaoning	0.218	29.27	24.35	76.86
Jilin	0.189	25.91	27.33	77.49
Heilongjiang	0.138	23.71	22.94	64.66
Shanghai	0.138	26.18	29.58	95.35
Jiangsu	0.244	28.73	30.55	83.9
Zhejiang	0.204	40.84	26.9	95.08
Anhui	0.241	39.57	28.75	80.15
Fujian	0.24	52.48	24.18	105.45
Jiangxi	0.22	44.32	29.5	87.9
Shandong	0.198	24.13	25.6	75.51
Henan	0.252	30.03	32.9	84.72
Hubei	0.259	31.75	29.35	86.46
Hunan	0.381	42.78	35.5	104.25
Guangdong	0.195	39.47	25.15	97.43
Guangxi	0.317	34.92	25.22	107.98
Hainan	0.288	33.62	29.64	89.34
Chongqing	0.4	50.46	26.34	93.72
Sichuan	0.268	32.28	30.89	107.58
Guizhou	0.365	38.49	29.35	103.86
Yunnan	0.249	35.74	32.56	93.49
Tibet	/	/	/	/
Shaanxi	0.314	32.63	34.31	75.99
Gansu	0.213	27.52	26.83	86.43
Qinghai	0.171	24.64	26.16	89.62
Ningxia	0.211	23.26	22.7	66.95
Xinjiang	0.274	18.02	32.79	81.55

*Chen et al., 2020

*Table S2. Soil pH and SOM values in various provinces in China.

Province	pH	SOM (g kg^{-1})	References
Beijing	7.71	44.99	Xu et al., 2021a; Jia et al., 2011
Tianjin	8.11	18.23	Zheng et al., 2013; Feng et al., 2016; Zhao et al., 2014
Hebei	8.18	11.68	Jia et al., 2011; Zheng et al., 2013; Feng et al., 2016; Zhao et al., 2014; Zeng et al., 2015
Shanxi	8.22	21.85	Feng et al., 2016; Zhao et al., 2014
Inner Mongolia	8.00	21.70	Jia et al., 2011; Zheng et al., 2013; Feng et al., 2016; Zhao et al., 2014; Wang et al., 2011; Bao et al., 2011
Liaoning	5.84	21.27	Zheng et al., 2013; Feng et al., 2016; Zhao et al., 2014; Zeng et al., 2015
Jilin	7.10	34.30	Xu et al., 2021a; Zheng et al., 2013; Feng et al., 2016; Zhao et al., 2014
Heilongjiang	6.32	38.75	Jia et al., 2011; Feng et al., 2016; Zhao et al., 2014; Zeng et al., 2015; Wang et al., 2011; Bao et al., 2011
Shanghai	7.35	31.10	Gu et al., 2009; Chen et al., 2003
Jiangsu	6.58	32.94	Zheng et al., 2013; Feng et al., 2016; Zhao et al., 2014; Zeng et al., 2015
Zhejiang	5.59	18.92	Jia et al., 2011; Zeng et al., 2015
Anhui	6.40	18.12	Feng et al., 2016; Zhao et al., 2014
Fujian	7.08	25.23	Zheng et al., 2013
Jiangxi	5.55	11.16	heng et al., 2013; Feng et al., 2016; Zhao et al., 2014; Zeng et al., 2015; Bao et al., 2011
Shandong	7.86	14.32	Jia et al., 2011; Feng et al., 2016; Zhao et al., 2014; Zeng et al., 2015; Wang et al., 2011; Bao et al., 2011
Henan	8.07	12.33	Xu et al., 2021a; Jia et al., 2011; Feng et al., 2016; Zhao et al., 2014; Zeng et al., 2015; Wang et al., 2011; Bao et al., 2011
Hubei	6.97	17.00	Zeng et al., 2015; Wang et al., 2011
Hunan	5.75	17.75	Xu et al., 2021a; Jia et al., 2011; Zheng et al., 2013; Feng et al., 2016; Zhao et al., 2014
Guangdong	5.74	35.64	Xu et al., 2021a; Jia et al., Zeng et al., 2015
Guangxi	5.41	20.56	Zheng et al., 2013; Zeng et al., 2015; Wang et al., 2011; Bao et al., 2011
Hainan	5.50	28.95	Jia et al., 2011; Zheng et al., 2013; Zeng et al., 2015
Chongqing	5.92	16.50	Jia et al., 2011; Zheng et al., 2013; Feng et al., 2016; Zhao et al., 2014
Sichuan	7.38	15.44	Zeng et al., 2015; Wang et al., 2011
Guizhou	4.70	48.22	Zheng et al., 2013; Zeng et al., 2015; Wang et al., 2011; Bao et al., 2011
Yunnan	6.61	23.02	Feng et al., 2016; Zhao et al., 2014; Wang et al., 2011; Bao et al., 2011
Tibet	6.72	81.78	Zheng et al., 2013
Shaanxi	7.39	11.09	Jia et al., 2011; Zheng et al., 2013; Feng et al., 2016; Zhao et al., 2014; Wang et al., 2011; Bao et al., 2011
Gansu	8.30	16.36	Jia et al., 2011; Zheng et al., 2013; Feng et al., 2016; Zhao et al., 2014
Qinghai	7.47	93.79	Dao et al., 2019
Ningxia	8.25	8.08	Wang et al., 2011; Bao et al., 2011
Xinjiang	8.11	27.59	Jia et al., 2011; Zheng et al., 2013; Feng et al., 2016; Zhao et al., 2014; Wang et al., 2011; Bao et al., 2011

Table S3. Biological toxicity factor, background value in soil, and standard concentration in groundwater of the target heavy metal.

Province	*Ti**				The background value(mg kg^{-1}) for metals in soils**				Standard concentration (mg kg^{-1}) of the target heavy metal in groundwater***			
	Cd	Pb	Cu	Zn	Cd	Pb	Cu	Zn	Cd	Pb	Cu	Zn
Beijing					0.074	25.4	23.6	102.6				
Tianjin					0.09	21	28.8	79.3				
Hebei					0.094	21.5	21.8	78.4				
Shanxi					0.128	15.8	26.9	75.5				
Inner Mongolia					0.053	17.2	14.1	59.1				
Liaoning					0.108	21.4	19.8	63.5				
Jilin					0.099	28.8	17.1	80.4				
Heilongjiang					0.086	24.2	20	70.7				
Shanghai					0.138	25	27.2	81.3				
Jiangsu					0.126	26.2	22.3	62.6				
Zhejiang					0.07	23.7	17.6	70.6				
Anhui					0.097	26.6	20.4	62				
Fujian					0.074	41.3	22.8	80.1				
Jiangxi					0.108	32.3	20.3	69.4				
Shandong					0.084	25.8	24	63.5				
Henan	30	5	5	1	0.074	19.6	19.7	60.1	0.005	0.01	1	1
Hubei					0.172	26.7	30.7	83.6				
Hunan					0.126	29.7	27.3	94.4				
Guangdong					0.056	36	17	47.3				
Guangxi					0.267	24	27.8	75.6				
Hainan					0.14	34.4	25.3	85.4				
Chongqing					0.28	28.1	24.6	81.9				
Sichuan					0.079	30.9	31.1	86.5				
Guizhou					0.659	35.2	32	99.5				
Yunnan					0.218	40.6	46.3	89.7				
Tibet					0.081	29.1	21.9	74				
Shaanxi					0.094	21.4	21.4	69.4				
Gansu					0.116	18.8	24.1	68.5				
Qinghai					0.137	20.9	22.2	80.3				
Ningxia					0.112	20.6	22.1	58.8				
Xinjiang					0.12	19.4	26.7	68.8				

*(Sharifi et al., 2016; Wen et al., 2019)

**(State Bureau of Environmental Conservation. 1990.; Xu et al., 2021b; Bao et al., 2020)

***State Administration for Market Regulation of the People's Republic of China, 2017

Table S4. Evaluation criteria for potential ecological risk index method.

Grades	***EI***	***ERI***
Low ecological risk	$EI < 40$	$ERI < 150$
Moderate ecological risk	$40 \leq EI < 80$	$80 \leq ERI < 300$
Considerable ecological risk	$80 \leq EI < 160$	$300 \leq ERI < 600$
Very high ecological risk	$160 \leq EI < 320$	$600 \leq ERI < 1200$
Extremely high ecological risk	$EI \geq 320$	$ERI \geq 1200$

Table S5. The annual leaching amounts of Cd, Pb, Cu, and Zn under different leaching models calculated using Eqs. (1)-(3).

Province	W_{lea}	Cd (g ha^{-1} yr^{-1})			Pb (g ha^{-1} yr^{-1})			Cu (g ha^{-1} yr^{-1})			Zn (g ha^{-1} yr^{-1})		
	mm yr^{-1}	L	L_6	L_{12}	L	L_6	L_{12}	L	L_6	L_{12}	L	L_6	L_{12}
Beijing[a]	52.15	0.072	0.012	0.006	0.496	0.083	0.041	8.865	1.478	0.739	22.423	3.737	1.869
Tianjin[a]	103.80	0.419	0.070	0.035	1.177	0.196	0.098	31.140	5.190	2.595	48.785	8.131	4.065
Hebei[a]	89.19	0.244	0.041	0.020	1.208	0.201	0.101	27.647	4.608	2.304	52.619	8.770	4.385
Shanxi[a]	142.49	0.226	0.038	0.019	1.403	0.234	0.117	32.772	5.462	2.731	48.446	8.074	4.037
Inner Mongolia[a]	181.05	0.292	0.049	0.024	1.856	0.309	0.155	38.020	6.337	3.168	66.988	11.165	5.582
Liaoning[a]	357.79	11.471	1.912	0.956	34.393	5.732	2.866	425.775	70.963	35.481	4390.137	731.690	365.845
Jilin[a]	352.59	1.513	0.252	0.126	7.339	1.223	0.612	126.932	21.155	10.578	433.684	72.281	36.140
Heilongjiang[a]	306.13	2.192	0.365	0.183	11.312	1.885	0.943	159.189	26.532	13.266	942.889	157.148	78.574
Shanghai[b]	806.78	2.047	0.341	0.171	14.211	2.369	1.184	282.372	47.062	23.531	911.658	151.943	75.972
Jiangsu[b]	594.37	6.264	1.044	0.522	22.603	3.767	1.884	374.452	62.409	31.204	1806.878	301.146	150.573
Zhejiang[b]	972.64	42.545	7.091	3.545	172.907	28.818	14.409	1672.938	278.823	139.412	23411.402	3901.900	1950.950
Anhui[b]	658.73	13.65	2.275	1.138	55.122	9.187	4.594	665.321	110.887	55.443	4057.800	676.300	338.150
Fujian[b]	1199.93	8.577	1.430	0.715	60.320	10.053	5.027	479.972	79.995	39.998	2663.844	443.974	221.987
Jiangxi[b]	1346.63	101.117	16.853	8.426	351.175	58.529	29.265	3703.238	617.206	308.603	48371.026	8061.838	4030.919
Shandong[a]	193.05	0.713	0.119	0.059	2.888	0.481	0.241	63.708	10.618	5.309	146.721	24.454	12.227
Henan[a]	258.12	1.065	0.178	0.089	4.263	0.711	0.355	103.247	17.208	8.604	180.683	30.114	15.057
Hubei[b]	786.53	9.486	1.581	0.791	32.397	5.400	2.700	542.704	90.451	45.225	2328.120	388.020	194.010
Hunan[b]	1092.84	77.975	12.996	6.498	181.605	30.268	15.134	2284.046	380.674	190.337	23845.877	3974.313	1987.156
Guangdong[b]	1562.18	33.217	5.536	2.768	170.343	28.391	14.195	1484.068	247.345	123.672	18605.526	3100.921	1550.461
Guangxi[b]	1311.43	103.256	17.209	8.605	225.925	37.654	18.827	2308.121	384.687	192.343	44208.386	7368.064	3684.032
Hainan[b]	1540.26	76.081	12.680	6.340	199.062	33.177	16.589	2387.403	397.901	198.950	28802.861	4800.477	2400.238
Chongqing[b]	735.23	48.123	8.021	4.010	128.511	21.419	10.709	1051.385	175.231	87.615	11910.794	1985.132	992.566
Sichuan[b]	511.29	4.229	0.705	0.352	15.330	2.555	1.278	286.324	47.721	23.860	1078.830	179.805	89.903

Guizhou[b]	755.96	80.828	13.471	6.736	180.922	30.154	15.077	1557.279	259.547	129.773	36989.159	6164.860	3082.430
Yunnan[b]	659.53	9.105	1.518	0.759	36.360	6.060	3.030	540.815	90.136	45.068	2835.980	472.663	236.332
Tibet[b]	605.33	/	/	/	/	/	/	/	/	/	/	/	/
Shaanxi[a]	356.37	4.426	0.738	0.369	12.613	2.102	1.051	270.843	45.141	22.570	680.672	113.445	56.723
Gansu[a]	214.27	0.460	0.077	0.038	2.291	0.382	0.191	49.282	8.214	4.107	85.708	14.285	7.142
Qinghai[a]	231.37	0.264	0.044	0.022	1.978	0.330	0.165	32.392	5.399	2.699	85.609	14.268	7.134
Ningxia[a]	266.02	1.040	0.173	0.087	3.565	0.594	0.297	85.126	14.188	7.094	156.951	26.159	13.079
Xinjiang[a]	296.40	0.676	0.113	0.056	1.903	0.317	0.159	68.171	11.362	5.681	100.775	16.796	8.398

[a] belong to the North China

[b] belong to the South China

Table S6. Analysis of significant differences among multiple groups for Cd, Pb, Cu, and Zn.

Group Pair	Cd		Pb		Cu		Zn	
	p-value	Significance	p-value	Significance	p-value	Significance	p-value	Significance
Jiangsu vs Zhejiang	0.8182	No	0.5887	No	0.6991	No	0.3939	No
Jiangsu vs Hunan	0.6756	No	0.0411	Yes	0.6429	No	0.0714	No
Jiangsu vs S-15-L	0.0003	Yes	0.0548	No	0.0000	Yes	0.0000	Yes
Jiangsu vs S-15-L_6	0.0184	Yes	0.5693	No	0.0143	Yes	0.0000	Yes
Jiangsu vs S-15-L_{12}	0.2347	No	0.2347	No	0.1317	No	0.0004	Yes
Zhejiang vs Hunan	0.6514	No	0.0411	Yes	0.6429	No	0.2857	No
Zhejiang vs S-15-L	0.0001	Yes	0.0046	Yes	0.0000	Yes	0.0000	Yes
Zhejiang vs S-15-L_6	0.0084	Yes	0.5693	No	0.0016	Yes	0.0024	Yes
Zhejiang vs S-15-L_{12}	0.0796	No	0.2672	No	0.1537	No	0.0110	Yes
Hunan vs S-15-L	0.0000	Yes	0.0001	Yes	0.0147	Yes	0.0147	Yes
Hunan vs S-15-L_6	0.0001	Yes	0.0143	Yes	0.2941	No	0.1324	No
Hunan vs S-15-L_{12}	0.0038	Yes	0.0663	No	0.7206	No	0.7206	No

Table S7. EI and ERI values of the heavy metals in soil.

Province	EI				ERI
	Cd	Pb	Cu	Zn	
Beijing	62.03	4.64	5.06	0.82	72.55
Tianjin	117.00	6.19	5.70	1.03	129.92
Hebei	57.77	6.14	6.05	1.04	71.00
Shanxi	42.19	8.61	5.83	1.06	57.69
Inner Mongolia	80.38	6.76	8.46	1.07	96.67
Liaoning	60.56	6.84	6.15	1.21	74.76
Jilin	57.27	4.50	7.99	0.96	70.72
Heilongjiang	48.14	4.90	5.74	0.91	59.69
Shanghai	30.00	5.24	5.44	1.17	41.85
Jiangsu	58.10	5.48	6.85	1.34	71.77
Zhejiang	87.43	8.62	7.64	1.35	105.04
Anhui	74.54	7.44	7.05	1.29	90.32
Fujian	97.30	6.35	5.30	1.32	110.27
Jiangxi	61.11	6.86	7.27	1.27	76.51
Shandong	70.71	4.68	5.33	1.19	81.91
Henan	102.16	7.66	8.35	1.41	119.58
Hubei	45.17	5.95	4.78	1.03	56.93
Hunan	90.71	7.20	6.50	1.10	105.51
Guangdong	104.46	5.48	7.40	2.06	119.40
Guangxi	35.62	7.28	4.54	1.43	48.87
Hainan	61.71	4.89	5.86	1.05	73.51
Chongqing	42.86	8.98	5.35	1.14	58.33
Sichuan	101.77	5.22	4.97	1.24	113.20
Guizhou	16.62	5.47	4.59	1.04	27.72
Yunnan	34.27	4.40	3.52	1.04	43.23
Tibet	/	/	/	/	/
Shaanxi	100.21	7.62	8.02	1.09	116.94
Gansu	55.09	7.32	5.57	1.26	69.24
Qinghai	37.45	5.89	5.89	1.12	50.35
Ningxia	56.52	5.65	5.14	1.14	68.45
Xinjiang	68.5	4.64	6.14	1.19	80.47

Table S8. The concentration, EI and ERI value for heavy metals (Cd, Pb, Cu, and Zn) in leachate.

Province	Concentration in leachate (mg/L)				EI				ERI
	Cd	Pb	Cu	Zn	Cd	Pb	Cu	Zn	
Beijing	0.000138	0.000951	0.017	0.043	0.83	0.48	0.09	0.04	1.43
Tianjin	0.000403	0.00113	0.030	0.047	2.42	0.57	0.15	0.05	3.18
Hebei	0.000273	0.00135	0.031	0.059	1.64	0.68	0.16	0.06	2.53
Shanxi	0.000158	0.000985	0.023	0.034	0.95	0.49	0.12	0.03	1.59
Inner Mongolia	0.000161	0.00103	0.021	0.037	0.97	0.51	0.11	0.04	1.62
Liaoning	0.00321	0.00961	0.119	1.227	19.24	4.81	0.60	1.23	25.86
Jilin	0.000429	0.00208	0.036	0.123	2.57	1.04	0.18	0.12	3.92
Heilongjiang	0.000716	0.00370	0.052	0.308	4.30	1.85	0.26	0.31	6.71
Shanghai	0.000254	0.00176	0.035	0.113	1.52	0.88	0.18	0.11	2.69
Jiangsu	0.00105	0.00380	0.063	0.304	6.32	1.90	0.32	0.30	8.84
Zhejiang	0.00437	0.0178	0.172	2.407	26.25	8.89	0.86	2.41	38.40
Anhui	0.00207	0.00837	0.101	0.616	12.43	4.18	0.51	0.62	17.74
Fujian	0.000715	0.00503	0.040	0.222	4.29	2.51	0.20	0.22	7.22
Jiangxi	0.00751	0.0261	0.275	3.592	45.05	13.04	1.38	3.59	63.06
Shandong	0.000369	0.00150	0.033	0.076	2.21	0.75	0.17	0.08	3.20
Henan	0.000413	0.00165	0.040	0.070	2.48	0.83	0.20	0.07	3.57
Hubei	0.00121	0.00412	0.069	0.296	7.24	2.06	0.35	0.30	9.94
Hunan	0.00714	0.0166	0.209	2.182	42.81	8.31	1.05	2.18	54.35
Guangdong	0.00213	0.0109	0.095	1.191	12.76	5.45	0.48	1.19	19.88
Guangxi	0.00787	0.0172	0.176	3.371	47.24	8.61	0.88	3.37	60.11
Hainan	0.00494	0.0129	0.155	1.870	29.64	6.46	0.78	1.87	38.74
Chongqing	0.00655	0.0175	0.143	1.620	39.27	8.74	0.72	1.62	50.35
Sichuan	0.000827	0.00230	0.056	0.211	4.96	1.50	0.28	0.21	6.95
Guizhou	0.0107	0.0239	0.206	4.893	64.15	11.97	1.03	4.89	82.04
Yunnan	0.00138	0.00551	0.082	0.430	8.28	2.76	0.41	0.43	11.88
Tibet	/	/	/	/	/	/	/	/	/
Shaanxi	0.00124	0.00354	0.076	0.191	7.45	1.77	0.38	0.19	9.79
Gansu	0.000214	0.00107	0.023	0.040	1.29	0.53	0.12	0.04	1.98
Qinghai	0.000114	0.000855	0.014	0.037	0.69	0.43	0.07	0.04	1.22
Ningxia	0.000391	0.00134	0.032	0.059	2.35	0.67	0.16	0.06	3.24
Xinjiang	0.000228	0.000642	0.023	0.034	1.37	0.32	0.12	0.03	1.84

The standard limits for Cd, Pb, Cu, and Zn in drinking water (GB 5749-2022) and Class IIIl of gorund water are 0.005, 0.01, 1, and 1 mg L^{-1}, respectively.

Table S9. Heavy metal content in leachate in critical provinces before and after pH control.

Province	pH before adjustment	Cd	Pb	Cu	Zn	pH after adjustment	Cd	Pb	Cu	Zn
Liaoning	5.84	0.00321	0.00961	0.119	1.227	6	0.00264	0.00826	0.105	0.955
Zhejiang	5.59	0.00437	0.0178	0.172	2.407	6	0.00271	0.0122	0.125	1.296
Jiangxi	5.55	0.00751	0.0261	0.275	3.592	6	0.00444	0.0173	0.193	1.818
Hunan	5.75	0.00714	0.0166	0.209	2.182	6	0.00533	0.0132	0.172	1.495
Guangdong	5.74	0.00213	0.0109	0.095	1.191	6	0.00213	0.0109	0.095	1.191
Guangxi	5.41	0.00787	0.0172	0.176	3.371	6	0.00395	0.0100	0.111	1.378
Hainan	5.5	0.00494	0.0129	0.155	1.870	6	0.00494	0.0129	0.155	1.870
Chongqing	5.92	0.00655	0.0175	0.143	1.620	6	0.00592	0.0162	0.134	1.423
Guizhou	4.7	0.0107	0.0239	0.206	4.893	6	0.00232	0.00721	0.074	0.676

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