Supplementary information

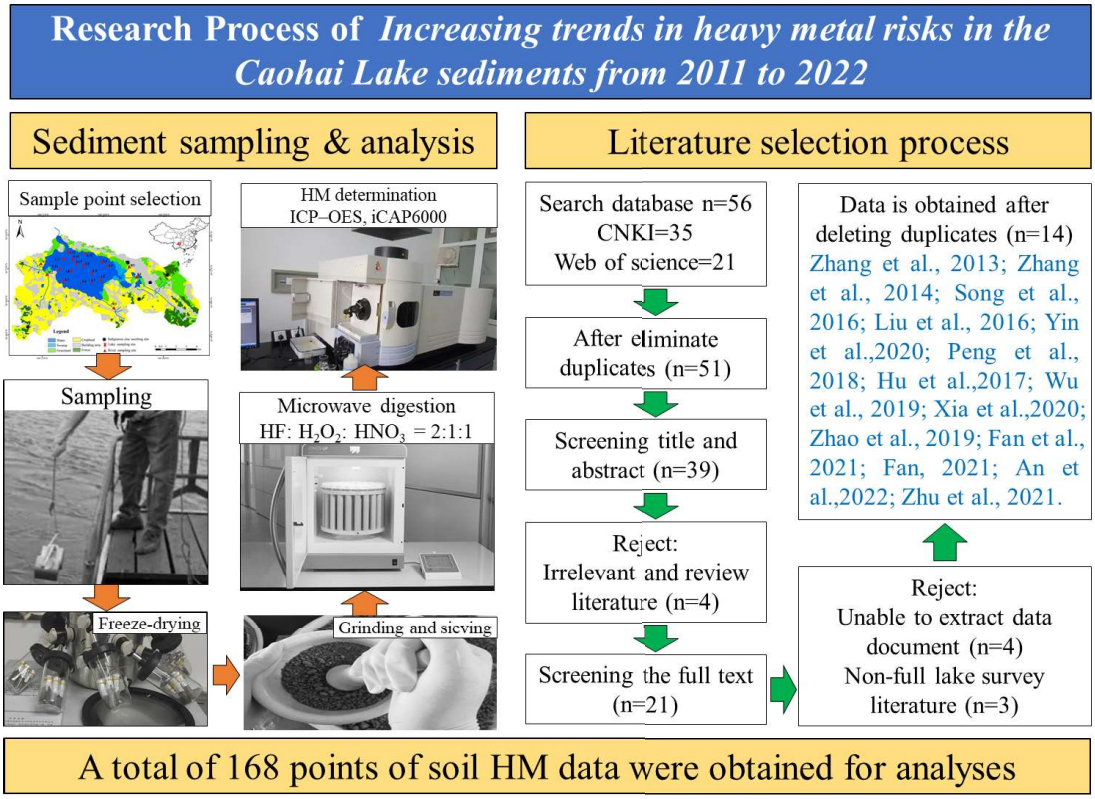

Fig. S1. Sampling Analysis and Literature Collection Process

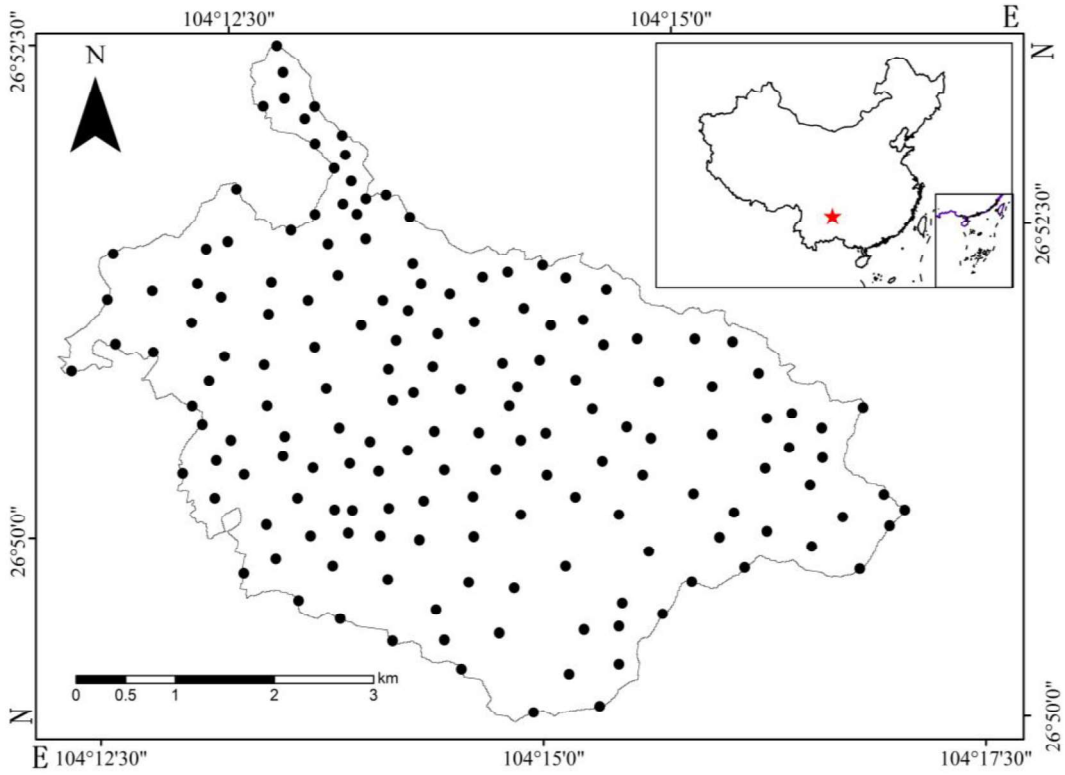

Fig. S2. Sampling sites collected from literatures

Table S1 Summary of data collected

No	Sampling time	Point	Longitude (°)	Latitude (°)	Depth (cm)	Digestion method	Instrument	Heavy metals(mg/kg)								Reference
								As	Cd	Cr	Cu	Hg	Pb	Ni	Zn	
1	2011	N1	104.231415	26.852024	10	HNO_3-H_2O_2	ICP-MS, Agilent 7500a	16	0.84	45.55	18.47	0.38	36.15		356.41	Zhang et al., 2013
		N2	104.229184	26.853862				15	0.68	42.31	18.64	0.35	36.73		347.55	
		N3	104.226953	26.856159				15	0.49	47.41	15.26	0.25	33.13		335.32	
		N4	104.224894	26.859529				15	0.32	40.19	14.39	0.22	24.64		326.45	
		N5	104.224036	26.861061				15	0.29	34.17	14.09	0.11	23.15		305.82	
		S1	104.231415	26.848501				17	0.89	31.25	20.45	0.40	32.31		358.10	
		S2	104.231243	26.844058				16	0.84	27.60	18.67	0.36	31.92		390.24	
		S3	104.231587	26.840688				16	0.70	41.66	19.10	0.35	28.04		394.92	
		S4	104.231072	26.835326				16	0.56	49.48	24.48	0.34	26.33		444.34	
		S5	104.230385	26.830270				15	0.66	31.25	24.11	0.22	25.09		450.04	
		E1	104.235872	26.849261				16	0.87	44.12	20.49	0.37	31.96		298.92	
		E2	104.239819	26.847729				17	1.28	31.33	15.58	0.36	34.27		289.21	
		E3	104.249256	26.847729				19	1.42	40.81	17.52	0.38	55.32		289.79	
		E4	104.255605	26.845278				14	1.42	9.53	14.67	0.39	58.11		345.15	
		E5	104.263670	26.843440				14	2.01	25.51	26.88	0.46	61.87		386.83	
		E6	104.270019	26.844665				18	2.45	25.00	82.25	0.57	82.92		520.97	

2	2011	54	26.845888	104.248238	20	HNO_3-$HCLO_4$-HF	ICP-MS，Agilent 7500a	17	18.09	37.88	18.56	34.46		431.31	Zhang et al., 2014
3	2012	1	104.700185	26.840471	N. D.	HNO_3-HF	ICP-MS，ELANDRC-e		14.83	142.0		108.0	180.7	515.00	Song et al., 2016
		2	104.231550	26.814914					7.42	69.37		51.00	15.01	278.00	
		3	104.316421	26.826027					12.34	50.80		67.00	16.34	294.00	
		4	103.988007	26.843602					3.85	12.45		29.00	14.94	161.00	
		5	104.331181	26.832627					3.12	50.67		26.00	15.70	140.00	
		6	104.545203	26.819809					15.64	30.25		91.00	28.70	510.00	
		7	104.488007	26.840307					14.78	17.80		96.00	13.74	507.00	
		8	104.620849	26.830914					15.23	38.40		92.00	27.40	411.00	
		9	104.059963	26.830479					4.65	38.40		28.00	28..3	187.00	
		10	104.165129	26.823778					9.97	13.70		74.00	14.87	279.00	
		11	104.301661	26.845201					10.68	17.80		84.00	133.6	421.00	
		12	104.552583	26.859543					6.03	50.80		43.00	39.47	243.00	
		13	104.139299	26.845075					6.14	17.80		27.00	27.15	184.00	
		14	104.423432	26.855179					14.25	21.35		117.0	14.84	547.00	
		15	104.080258	26.840378					7.38	30.25		66.00	28.60	276.00	
		16	104.135609	26.829375					5.17	31.46		64.00	37.45	144.00	
		17	104.428967	26.835804					21.67	12.45		86.00	17.90	418.00	
		18	104.583948	26.853172					4.68	13.75		30.00	40.84	141.00	

		19	104.443727	26.815854					23.57	12.41			85.00	31.56	337.00	
4	2013	S1	104.266667	26.850000	30	HCl-HNO_3-HF-$HClO_4$	ICP-MS Thermo X Series II, Cd、Cr、Pb、Cu Zn; AFS-820 Double-channel Atomic Fluorescence Photometer, As, Hg.	21	10.10	75.53	50.26	0.07	75.38		468.19	Liu et al., 2016
		S2	104.266667	26.833333				18	8.96	68.66	44.87	0.06	61.54		305.27	
		S3	104.250000	26.850000				15	1.22	121.0	23.33	0.04	35.38		254.54	
		S4	104.250000	26.833333				18	1.67	79.30	22.43	0.09	75.38		142.90	
		S5	104.233333	26.833333				23	1.82	110.7	43.07	0.08	109.2		226.73	
		S6	104.250000	26.833333				11	1.59	59.76	14.80	0.05	37.69		99.08	
		S7	104.250000	26.833333				16	5.77	79.59	49.36	0.09	106.1		247.05	
		S8	104.250000	26.833333				6	0.91	25.12	18.39	0.05	25.38		74.53	
		S9	104.233333	26.850000				22	1.14	132.0	29.62	0.04	37.69		113.51	
		S10	104.233333	26.850000				18	1.29	134.4	45.77	0.06	67.69		152.48	
		S11	104.233333	26.833333				21	4.10	75.34	39.48	0.05	46.15		210.71	
		S12	104.216667	26.850000				22	0.84	134.6	28.27	0.04	46.15		121.55	
5	2014	L1	104.220083	26.850119	5	HNO_3-HF-	HR-CS-ETAAS, Contra AA 300/700, Analytik Jena AG, Germany		0.48				27.98		91.93	Yin et al.,2020
		L2	104.235875	26.849480					3.90				33.19		206.50	
		L3	104.253030	26.849241					2.57				38.52		163.45	
		L4	104.261819	26.850286					2.93				46.33		152.60	
		L5	104.243544	26.857427					4.86				126.0		282.92	
		L6	104.226847	26.861913					5.75				76.68		226.18	
		L7	104.230272	26.837888					7.96				41.32		180.03	

		L8	104.252247	26.839975					68.98				129.1	1194.91	
		L9	104.259486	26.857427					61.85				235.4	858.94	
		L10	104.249294	26.862840					41.03				171.8	801.86	
		L11	104.220583	26.871963					9.69				44.20	216.03	
		L12	104.224963	26.851261					34.73				151.6	619.15	
		L13	104.209913	26.840963					19.01				95.76	303.59	
		L14	104.234455	26.829825					36.45				134.4	162.79	
		L15	104.233583	26.830166					25.94				155.1	449.68	
		L16	104.261741	26.840569					24.06				117.6	353.33	
6	2014	CL1	104.220083	26.850119	30	HNO_3-HF	Graphite furnace, flame, cold atomic absorption spectroscopy (2400 Z AA, Agilent, USA; contr AA 300; AFS-230 E)		69.30				120.9	1147.17	Peng et al., 2018
		CL2	104.235875	26.849481					56.37				215.9	874.72	
		CL3	104.253031	26.849242					39.09				168.2	803.53	
		CL4	104.261819	26.850286					9.77				43.84	216.19	
		CL5	104.243544	26.857428					34.19				148.1	639.68	
		CL6	104.226847	26.861914					19.05				91.54	293.59	
		CL7	104.230272	26.837889					33.91				145.6	164.37	
		CL8	104.252247	26.839975					25.57				155.6	459.40	
		CL9	104.259486	26.857428					22.35				114.3	376.29	
7	2015	S3	104.247000	26.871000	10	HNO_3-HCl	Atomic Fluorescence	9	18.17	58.62	23.08	1.22	54.31	457.82	Hu et al.,2017
		S4	104.257000	26.864000				20	23.49	43.59	18.28	0.82	40.43	396.52	

		S5	104.263000	26.855000			Spectrometry (AFS, Titian, China); atomic absorption spectrophotometer (FAAS, Perkin Elmer, Graphite furnace, flame, cold atomic absorption spectroscopy (2400 ZAA, Agilent, USA; contr AA 300; AFS-	16	14.38	26.97	23.24	0.49	20.04		381.69	
		S6	104.234000	26.855000				24	6.26	35.65	22.97	0.38	28.16		289.23	
		S8	104.249000	26.838000				17	1.74	24.47	20.32	0.17	21.08		357.48	
		S11	104.248000	26.855000				13	11.43	41.33	23.31	0.21	31.19		435.71	
8	2015	W1	104.267979	26.842517	40	HNO_3-HF		13		53.72	17.68		59.62		291.91	Wu et al., 2019
		W2	104.250298	26.851097				8		19.27	14.45		10.74		24.26	
		W3	104.226952	26.858298				14		35.90	13.44		94.87		551.47	
		W4	104.218712	26.846501				10		38.41	18.43		55.56		191.18	
		W5	104.238625	26.829645				15		75.76	16.89		47.43		177.21	
9	2017	A1	104.274158	26.844519	10	HNO_3-HF-$HClO_4$	Atomic fluorescence spectrophotometer (AFS-933, Titan, China).		3.52	101.6	27.06	0.81	55.29	40.15	165.68	Xia et al.,2020
		A2	104.281368	26.841456					6.05	99.23	28.41	0.99	60.37	52.67	20.45	
		A3	104.286861	26.839158					5.27	105.2	29.59	0.82	59.74	61.89	349.05	
		A4	104.297504	26.839618					3.43	103.6	30.30	0.78	57.36	38.87	183.46	
		B1	104.255619	26.829202					2.56	108.5	26.75	0.58	51.22	49.72	118.85	
		B2	104.256477	26.825679					2.39	107.6	27.13	0.60	54.45	48.67	134.12	
		B3	104.258537	26.823075					3.12	110.2	27.21	0.62	53.26	57.51	186.75	
		B4	104.266605	26.818632					3.63	114.7	28.73	0.67	53.27	56.18	204.69	

		C1	104.231243	26.835942					2.13	123.8	28.96	0.46	45.21	58.97	104.27	
		C2	104.224376	26.834871					1.51	124.9	29.29	0.44	44.85	59.56	113.56	
		C3	104.223691	26.829508					3.05	122.1	27.35	0.43	46.71	54.62	146.85	
		C4	104.226951	26.827977					2.76	119.2	26.58	0.45	48.47	49.78	164.22	
		D1	104.210128	26.855241					1.74	123.6	32.88	0.55	44.16	63.54	127.98	
		D2	104.205322	26.853709					1.66	125.3	34.26	0.60	45.43	64.85	175.24	
		D3	104.203433	26.852331					3.29	117.5	33.14	0.68	46.13	50.19	175.24	
		D4	104.197597	26.851411					3.15	115.1	33.56	0.62	46.78	36.09	159.57	
		E1	104.224376	26.863816					2.03	126.8	30.78	0.44	42.60	58.64	110.41	
		E2	104.219741	26.861213					1.91	125.4	31.24	0.48	42.30	65.31	153.82	
		E3	104.213905	26.863816					4.16	123.4	32.86	0.52	45.81	60.59	159.62	
		E4	104.212017	26.869788					2.75	122.0	33.13	0.50	46.23	56.74	150.93	
10	2017	S1	104.263859	26.844889	10	HNO3-HF-HClO4	ICP-OES, iCAP6000, USA Thermo Fisher	12	1.24	57.30	27.70		46.60	29.00	314.93	Zhao et al., 2019
		S2	104.248066	26.837536				22	0.45	80.10	19.90		57.70	51.10	260.03	
		S3	104.255447	26.831867				17	0.35	78.50	18.70		46.10	38.80	130.02	
		S4	104.242058	26.830488				15	0.42	58.70	19.30		55.80	37.60	103.05	
		S5	104.228668	26.836616				15	0.35	36.10	21.60		56.50	29.80	209.95	
		S6	104.233818	26.846727				17	0.28	68.50	16.60		64.70	36.10	116.53	
		S7	104.246006	26.855459				14	0.68	58.80	18.50		67.90	36.40	261.96	
		S8	104.232788	26.856837				14	1.50	39.60	17.20		84.00	26.40	473.84	

		S9	104.217339	26.857450				19	1.47	48.80	19.20		46.60	29.60	67.42	
		S10	104.216995	26.848719				12	0.63	37.80	26.80		16.40	23.90	255.22	
11	2018	1	104.243259	26.846357	10	HCl-HNO_3-HF-$HClO_4$	AAS-ZEEnit700P(150Z7P 1064);	12	11.05	64.99	17.73	0.67	38.79			Fan et al., 2021
12	2018	25	26.84343620	104.233131	5	HCl-HNO_3-HF-$HClO_4$	AAS-ZEEnit700P(150Z7P 1064);	8	15.23	54.06	22.14	0.55	34.42			Fan, 2021
13	2020	1	104.220000	26.852667	10	HNO_3-HCl-H_2O_2	ICP-MS; Agilent 7900, USA		3.57				9.66	13.28	6.76	An et al.,2022
		2	104.220000	26.852667					0.72				23.19	13.86	60.82	
		3	104.255000	26.859500					3.47				27.05	13.85	96.86	
		4	104.255000	26.859500					5.26				40.10	16.75	96.86	
		5	104.270000	26.846833					8.20				23.67	17.38	126.15	
		6	104.270000	26.846833					6.03				34.78	25.79	128.40	
		7	104.271667	26.841833					12.49				65.70	25.21	202.74	
		8	104.271667	26.841833					17.70				70.53	25.69	186.97	
		9	104.250000	26.825000					19.00				86.96	23.85	211.75	
		10	104.250000	26.825000					22.04				94.20	32.84	466.29	
		11	104.225000	26.833500					23.39				151.2	35.11	486.56	
		12	104.225000	26.833500					23.63				107.7	55.25	479.81	

		13	104.211667	26.841667				34.10		121.7	56.93	790.67		
		14	104.211667	26.841667				35.40		177.7	55.24	797.43		
		15	104.208333	26.851667				40.47		205.3	49.19	772.65		
		16	104.208333	26.851667				46.98		237.2	46.82	1088.01		
		17	104.208333	26.861167				53.25		272.4	52.23	1220.92		
14	2020	S1	104.215475	26.854571	10	HF-$HClO_4$	ICP-MS, Agilent 7900, USA	18.70	30.51	187.0		366.94	Zhu et al., 2021	
		S2	104.222889	26.854571				65.45	28.22	271.4		1371.77		
		S3	104.232421	26.854571				34.36	34.39	205.5		643.55		
		S4	104.241088	26.854667				14.91	24.91	121.2		265.32		
		S5	104.250330	26.854476				22.23	61.85	127.8		372.58		
		S6	104.261595	26.854381				19.45	17.51	126.7		304.84		
		S7	104.215436	26.846571				15.16	24.44	124.6		270.97		
		S8	104.222755	26.846667				54.83	31.63	232.8		993.55		
		S9	104.232384	26.846857				37.91	39.33	215.5		745.16		
		S10	104.241050	26.846952				46.75	25.24	227.5		807.26		
		S11	104.250293	26.846762				41.95	33.97	216.7		767.74		
		S12	104.261173	26.846762				16.42	19.63	128.1		287.90		
		S14	104.223003	26.838190				60.14	28.88	226.6		993.55		
		S15	104.232343	26.838190				40.18	34.79	210.4		716.94		
		S16	104.241488	26.837905				52.31	34.55	225.6		976.61		
		S17	104.250251	26.838000				16.67	30.45	180.2		372.58		
		S18	104.261420	26.838000				17.43	27.65	158.7		400.81		
		S20	104.232309	26.831238				7.07	29.45	115.5		225.81		

		S21	104.240975	26.831333				47.00		38.18	257.2		970.97		
		S22	104.250315	26.831333				23.24		32.56	158.8		462.91		
15	2022	L1	26.847861	104.265286	10	HF-H_2O_2-HNO_3	ICP–OES, iCAP6000, Thermo Fisher, USA	7.47	86.53	22.09	88.55	37.44	379.32	This study	
		L2	26.835461	104.258700				21.31	74.35	29.64	120.5	35.67	953.87		
		L3	26.832653	104.249400				10.99	88.91	24.89	119.3	38.30	519.91		
		L4	26.851517	104.259114				7.46	89.75	23.69	94.53	37.14	739.89		
		L5	26.841486	104.249619				20.61	66.45	29.56	120.3	36.22	1052.65		
		L6	26.831575	104.239492				19.40	52.55	22.02	104.5	25.64	828.56		
		L7	26.834139	104.222389				11.41	85.65	24.57	118.0	37.39	494.06		
		L8	26.843272	104.229972				19.18	55.81	23.22	107.2	25.14	797.64		
		L9	26.848097	104.237306				14.10	96.18	25.61	151.5	31.46	754.72		
		L10	26.858553	104.243525				15.81	73.74	25.40	90.34	29.47	1380.19		
		L11	26.850036	104.230894				17.32	109.1	33.36	181.4	52.65	1821.24		
		L12	26.859978	104.226472				5.73	99.92	23.47	76.32	39.81	425.80		
		L13	26.858553	104.243525				9.36	95.10	25.32	99.23	38.75	1560.46		
		L14	26.853636	104.221994				8.69	99.15	25.03	97.45	41.01	657.62		
		L15	26.849756	104.234119				12.55	80.87	23.10	130.0	32.25	759.55		

Table S2 Contamination categories based on geo-accumulation index (Igeo).

Igeo	Pollution Intensity	TRI	Toxicity Intensity
<0	unpolluted	<5	No toxicity risk
0-1	lightly polluted	5-10	Low toxicity risk
1-2	moderately polluted	10-15	Moderate toxicity risk
2-3	moderately to heavily polluted	15-20	Considerable toxicity risk
3-4	heavily polluted	>20	very high toxicity risk
4-5	heavily to extremely polluted		
>5	extremely polluted		

Table S3 Statistics on fertilizer use and pollutant emissions in the Weining County from 2005 to 2020

Year	2020	2019	2018	2017	2016	2015	2014	2013	2012	2011	2010	2005
Agricultural fertilizer application (million tons)			12.56	13.28	13.19	12.62	11.89	14.87	11.89	11.32	9.84	7.37
Motor vehicle ownership (10,000 vehicles)	15.44	13.60	10.86	10.30	8.62							
Industrial smoke emissions (tons)					594	1388	1666	1285	1061			
Total industrial wastewater discharge (tons)			61	42	23	618	754	371	386	355		
Total domestic wastewater discharge (tons)			1610	1571	977	1629	1501	1447	1313	1313		
Total wastewater discharge (tons)			1671	1613	1001	2249	2257	1819	1700	1668		
Industrial solid waste generation (tons)			8.17	4.26	2.36	35.10	40.22	41.24				
Industrial solid waste emissions (tons)			0.00	4.26	0.00	0.00	0.02	24.84	0.02			
Amount of domestic waste disposed of in an environmentally sound manner (tons)					7.96	5.03	5.03	5.11	5.47	15.00		

Note: These data are derived from the Statistical Yearbook of Winnebago County (Weining County Bureau of Statistics, 2005-2020).

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