Supplementary Data

Figure S1: PCA graph of different chemical forms of all the PTMs for lower soil layer.

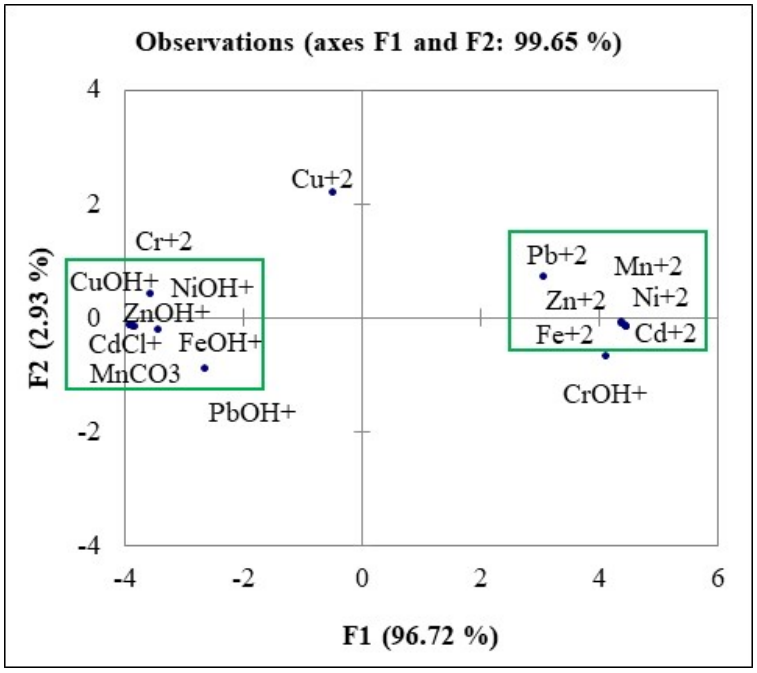

Figure S2: PCA graph of all the agronomic fields (F1-F15) for lower soil layers.

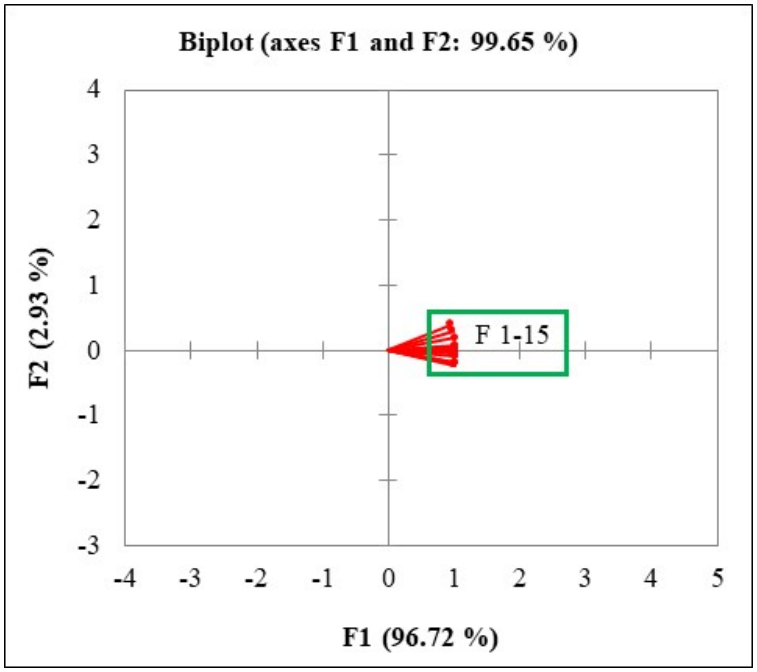

Table S1: Literature data regarding soil contamination by PTMs. Limit values obtained from (Antoniadis et al., 2019; Natasha et al., 2022).

PTM	Area	Soil level	Limit	Times higher of limit	Reference
Lead	Apalachicola Forest, USA	221	60	3.7	(Wu et al., 2022)
Lead	Cyprus	7265	60	121.1	(Christou et al., 2022)
Lead	Northwestern-Iran	390	60	6.5	(Khalilzadeh et al., 2021)
Lead	New York, USA	1150	60	19.2	(Egendorf et al., 2021)
Lead	Punjab, Pakistan	88.6	60	1.5	(Natasha et al., 2022)
Lead	Guangzhou, China	168	60	2.8	(Xiong et al., 2016)
Lead	Loiret, France	236	60	3.9	(Mombo et al., 2016)
Lead	Smelter, France	2000	60	33.3	(Xiong et al., 2014)
Lead	Northern France	1176	60	19.6	(Shahid et al., 2014)
Lead	Toulouse, France	39800	60	663.3	(Foucault et al., 2013)
Lead	Toulouse, France	39250	60	654.2	(Shahid et al., 2012)
Lead	Toulouse, France	1830	60	30.5	(Arshad et al., 2008)
Cadmium	Erbil City, Iraq	23	1	23.0	(Tariq, 2021)
Cadmium	Hamadan, Iran	4.8	1	4.8	(Kharazi et al., 2021)
Cadmium	China	16	1	16.0	(Yang et al., 2021)
Cadmium	Iran	55	1	55.0	(Khalilzadeh et al., 2021)
Cadmium	Shujabad, Pakistan	4.3	1	4.3	(Ahmad et al., 2021)
Cadmium	Pakistan	35	1	35.0	(Anwar et al., 2021)
Cadmium	Punjab-Pakistan	5.6	1	5.6	(Iqbal et al., 2021)
Cadmium	Vehari, Pakistan	3.6	1	3.6	(Sardar et al., 2020)
Cadmium	Guangzhou, China	6.3	1	6.3	(Xiong et al., 2016)
Cadmium	Vehari, Pakistan	4.0	1	4.0	(Shah et al., 2022)
Cadmium	Northern France	300	1	300	(Austruy et al., 2014)
Chromium	Erbil City, Iraq	131	100	1.3	(Tariq, 2021)
Chromium	Punjab, Pakistan	153	100	1.5	(Iqbal et al., 2021)
Chromium	Murrongoze River sediment	367	100	3.7	(Marove et al., 2022)
Chromium	Moatize River sediments	135	100	1.4	(Marove et al., 2022)
Chromium	Guangzhou, China	417	100	4.2	(Xiong et al., 2016)
Copper	Chile	1106	100	11.1	(Yáñez et al., 2022)
Copper	Iran	1485	100	14.9	(Khalilzadeh et al., 2021)
Copper	China	3200	100	32.0	(Zhang et al., 2022)
Copper	Punjab, Pakistan	181.6	100	1.82	(Natasha et al., 2022)
Manganese	Ethiopia	17919	2000	9.0	(Gemeda et al., 2021)
Nickel	Murrongoze River sediment	83	50	1.7	(Marove et al., 2022)
Nickel	Erbil City, Iraq	108	50	2.2	(Tariq, 2021)
Nickel	Hamadan, Iran	84	50	1.7	(Kharazi et al., 2021)
Nickel	Iran	116	50	2.3	(Khalilzadeh et al., 2021)
Nickel	Ethiopia	475	50	9.5	(Gemeda et al., 2021)
Nickel	Multan, Pakistan	67	50	1.3	(Anjum et al., 2021)
Arsenic	Punjab, Pakistan	5.0	5.0	1.0	(Natasha et al., 2022)
Arsenic	Vehari, Pakistan	20	5.0	4.0	(Natasha et al., 2021)
Arsenic	Jiangnan, China	138	5.0	27.6	(Lian et al., 2020)
Arsenic	Beijing. China	288	5.0	57.6	(Irshad et al., 2020)
Arsenic	Global	1540	5.0	308	(Hussain et al., 2021)

Table S2: Soil attributes for upper and lower soil layers.

Parameters	EC dS/m	pH —	Na meq/L	K meq/L	Ca meq/L	Ca+Mg meq/L	Cl meq/L	CO3 meq/L	HCO3 meq/L	OM %
0-15 cm										
Mean	1.1	7.1	109	23	38	7	12	0.4	1.7	1.4
Minimum	0.3	2.3	13	2	3	2	2	0.0	0.0	0.5
Maximum	5.4	8.1	788	326	230	33	50	1.3	3.7	2.8
15-30 cm										
Mean	0.9	7.1	88	19	31	5	7	0.4	1.6	0.9
Minimum	0.2	2.4	13	1	7	1	1	0.0	0.0	0.2
Maximum	4.2	8.1	313	400	87	37	34	1.2	3.2	3.2
% variation	*16*	*0*	*19*	*17*	*17*	*29*	*38*	*-6*	*9*	*36*

Table S3: Chemical speciation of PTMs in different agronomic fields (F5-F15) in upper soil layer.

PTM	Speciation	F-5	F-6	F-7	F-8	F-9	F-10	F-11	F-12	F-13	F-14	F-15
Pb	Pb^{+2}	87.2	61.9	86.4	85.9	99.3	82.7	90.3	88.4	88.1	93.7	97.2
	$PbOH^{+}$	11.9	34.7	12.1	13.4	0.5	16.5	9.1	11.0	11.2	6.0	2.6
Cd	Cd^{+2}	98.6	97.0	97.6	99.1	99.5	99.2	99.2	99.4	99.3	99.5	99.5
	$CdCl^{+}$	1.4	2.9	2.3	0.8	0.5	0.7	0.8	0.6	0.6	0.5	0.5
Zn	Zn^{+2}	99.4	96.8	99.3	99.3	100.0	99.0	99.5	99.4	99.4	99.7	99.9
	$ZnOH^{+}$	0.5	2.2	0.6	0.6	0.0	0.8	0.4	0.5	0.5	0.3	0.0
Mn	Mn^{+2}	100.0	99.9	99.9	100	100	100	100	100	100	100	100
	$MnCO_3$	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Fe	Fe^{+2}	99.8	99.1	99.8	99.7	100	99.7	99.8	99.8	99.8	99.9	100
	$FeOH^{+}$	0.2	0.9	0.2	0.2	0.0	0.3	0.2	0.2	0.2	0.1	0.0
Cu	Cu^{+2}	29.0	10.3	39.6	43.5	99.1	22.2	49.7	54.5	39.2	74.5	97.1
	$CuOH^{+}$	5.0	7.3	7.0	8.5	0.6	5.6	6.3	8.5	6.3	6.0	2.1
Cr	Cr^{+2}	2.2	0.5	2.2	1.9	38.0	1.5	3.0	2.4	2.4	4.6	4.6
	$CrOH^{+}$	97.8	99.5	97.8	98.1	62.0	98.5	97.0	97.6	97.6	95.4	95.4
Ni	Ni^{+2}	99.9	99.7	99.9	99.9	100	99.9	99.9	99.9	99.9	100	99.9
	$NiOH^{+}$	0.1	0.1	0.1	0.1	0.0	0.1	0.1	0.1	0.1	0.0	0.0
Average	*M^{+2}*	*77*	*71*	*78*	*79*	*92*	*76*	*80*	*80*	*79*	*84*	*87*
Average	*Others*	*15*	*18*	*15*	*15*	*8*	*15*	*14*	*15*	*15*	*14*	*13*

Table S4: Chemical speciation of PTMs in different agronomic fields (F5-F15) in lower soil layer.

PTM	Speciation	F-5	F-6	F-7	F-8	F-9	F-10	F-11	F-12	F-13	F-14	F-15
Pb	Pb^{+2}	95.4	91.4	89.0	83.1	90.3	79.0	99.0	92.3	88.2	85.5	93.7
	$PbOH^{+}$	4.1	7.9	9.7	16.3	9.3	19.9	0.8	7.2	11.2	13.9	5.9
Cd	Cd^{+2}	99.2	99.4	98.0	99.6	99.4	99.6	99.6	99.6	99.7	99.4	99.7
	$CdCl^{+}$	0.8	0.6	2.0	0.3	0.5	0.3	0.4	0.4	0.2	0.5	0.3
Zn	Zn^{+2}	99.9	99.7	99.6	99.1	99.5	98.8	100.0	99.6	99.4	99.2	99.7
	$ZnOH^{+}$	0.1	0.1	0.1	0.8	0.4	1.0	0.0	0.3	0.5	0.6	0.3
Mn	Mn^{+2}	100	100	100	100	100	100	100	100	100	100	100
	$MnCO_3$	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Fe	Fe^{+2}	100	100	99.9	99.7	99.8	99.6	100	99.9	99.8	99.7	99.9
	$FeOH^{+}$	0.0	0.0	0.1	0.3	0.2	0.4	0.0	0.1	0.2	0.3	0.1
Cu	Cu^{+2}	87.8	70.1	58.7	41.5	55.1	20.0	98.3	71.2	43.8	33.1	80.3
	$CuOH^{+}$	3.1	4.9	5.1	10.3	7.1	6.4	1.0	7.0	7.0	6.8	6.4
Cr	Cr^{+2}	2.9	1.5	1.2	1.5	2.9	1.2	27.8	3.8	2.4	1.9	4.7
	$CrOH^{+}$	97.1	98.5	98.8	98.5	97.1	98.8	72.2	96.2	97.6	98.1	95.3
Ni	Ni^{+2}	100	100	99.9	99.9	99.9	99.9	100	99.9	99.9	99.9	100
	$NiOH^{+}$	0.0	0.0	0.0	0.1	0.1	0.1	0.0	0.0	0.1	0.1	0.0
Average	*M^{+2}*	*86*	*83*	*81*	*78*	*81*	*75*	*91*	*83*	*79*	*77*	*85*
Average	*Others*	*13*	*14*	*14*	*16*	*14*	*16*	*9*	*14*	*15*	*15*	*14*

Table S5: Pearson correlations among all the agronomic fields (F1-F15) for upper and lower sol layers.

Variables	F-1	F-2	F-3	F-4	F-5	F-6	F-7	F-8	F-9	F-10	F-11	F-12	F-13	F-14
F-2	1.00													
F-3	1.00	1.00												
F-4	0.97	0.98	0.98											
F-5	0.98	0.99	0.99	1.00										
F-6	1.00	1.00	1.00	0.97	0.98									
F-7	0.97	0.98	0.98	1.00	1.00	0.97								
F-8	0.97	0.98	0.98	1.00	1.00	0.97	1.00							
F-9	0.81	0.83	0.83	0.90	0.89	0.81	0.91	0.91						
F-10	0.99	0.99	0.99	1.00	1.00	0.99	1.00	0.99	0.87					
F-11	0.96	0.97	0.97	1.00	0.99	0.96	1.00	1.00	0.93	0.99				
F-12	0.96	0.97	0.97	1.00	0.99	0.95	1.00	1.00	0.93	0.98	1.00			
F-13	0.97	0.98	0.98	1.00	1.00	0.97	1.00	1.00	0.91	1.00	1.00	1.00		
F-14	0.92	0.93	0.93	0.98	0.97	0.92	0.98	0.99	0.96	0.96	0.99	0.99	0.98	
F-15	0.87	0.89	0.89	0.95	0.94	0.87	0.95	0.96	0.97	0.92	0.97	0.97	0.95	0.99

Table S6: Pearson correlations among all the agronomic fields (F1-F15) for upper and lower sol layers.

Variables	F-1	F-2	F-3	F-4	F-5	F-6	F-7	F-8	F-9	F-10	F-11	F-12	F-13	F-14
F-2	1.00													
F-3	1.00	1.00												
F-4	0.97	0.98	0.98											
F-5	0.98	0.99	0.99	1.00										
F-6	1.00	1.00	1.00	0.97	0.98									
F-7	0.97	0.98	0.98	1.00	1.00	0.97								
F-8	0.97	0.98	0.98	1.00	1.00	0.97	1.00							
F-9	0.81	0.83	0.83	0.90	0.89	0.81	0.91	0.91						
F-10	0.99	0.99	0.99	1.00	1.00	0.99	1.00	0.99	0.87					
F-11	0.96	0.97	0.97	1.00	0.99	0.96	1.00	1.00	0.93	0.99				
F-12	0.96	0.97	0.97	1.00	0.99	0.95	1.00	1.00	0.93	0.98	1.00			
F-13	0.97	0.98	0.98	1.00	1.00	0.97	1.00	1.00	0.91	1.00	1.00	1.00		
F-14	0.92	0.93	0.93	0.98	0.97	0.92	0.98	0.99	0.96	0.96	0.99	0.99	0.98	
F-15	0.87	0.89	0.89	0.95	0.94	0.87	0.95	0.96	0.97	0.92	0.97	0.97	0.95	0.99

Table S7: Component factors (CF) of PCA for upper and lower sol layers.

Variables	0-15 cm		15-30 cm	
	CF1	CF2	CF1	CF2
F-1	6.6	10.5	6.6	10.5
F-2	6.7	7.7	6.7	7.7
F-3	6.7	7.6	6.7	7.6
F-4	6.9	0.0	6.9	0.0
F-5	6.9	0.5	6.9	0.5
F-6	6.5	11.0	6.5	11.0
F-7	6.9	0.0	6.9	0.0
F-8	6.9	0.0	6.9	0.0
F-9	5.7	33.3	5.7	33.3
F-10	6.8	2.0	6.8	2.0
F-11	6.9	0.8	6.9	0.8
F-12	6.8	1.1	6.8	1.1
F-13	6.9	0.0	6.9	0.0
F-14	6.7	7.4	6.7	7.4
F-15	6.3	18.0	6.3	18.0
Eigenvalue	14.5	0.4	14.5	0.4
Variability (%)	96.7	2.9	96.7	2.9
Cumulative %	96.7	99.7	96.7	99.7

References

Ahmad, I., Malik, S.A., Saeed, S., Rehman, A.-u., Munir, T.M., 2021. Phytoextraction of Heavy Metals by Various Vegetable Crops Cultivated on Different Textured Soils Irrigated with City Wastewater. Soil Systems 5, 35.

Anjum, M.A., Hussain, S., Arshad, P., Hassan, A., 2021. Irrigation water of different sources affects fruit quality attributes and heavy metals contents of un-grafted and commercial mango cultivars. Journal of Environmental Management 281, 111895.

Antoniadis, V., Shaheen, S.M., Levizou, E., Shahid, M., Niazi, N.K., Vithanage, M., Ok, Y.S., Bolan, N., Rinklebe, J., 2019. A critical prospective analysis of the potential toxicity of trace element regulation limits in soils worldwide: Are they protective concerning health risk assessment?-A review. Environment international 127, 819-847.

Anwar, H., Shahid, M., Niazi, N.K., Khalid, S., Tariq, T.Z., Ahmad, S., Nadeem, M., Abbas, G., 2021. Risk assessment of potentially toxic metal (loid) s in Vigna radiata L. under wastewater and freshwater irrigation. Chemosphere 265, 129124.

Arshad, M., Silvestre, J., Pinelli, E., Kallerhoff, J., Kaemmerer, M., Tarigo, A., Shahid, M., Guiresse, M., Pradère, P., Dumat, C., 2008. A field study of lead phytoextraction by various scented Pelargonium cultivars. Chemosphere 71, 2187-2192.

Austruy, A., Shahid, M., Xiong, T., Castrec, M., Payre, V., Niazi, N.K., Sabir, M., Dumat, C., 2014. Mechanisms of metal-phosphates formation in the rhizosphere soils of pea and tomato: environmental and sanitary consequences. Journal of Soils and Sediments 14, 666-678.

Christou, A., Hadjisterkotis, E., Dalias, P., Demetriou, E., Christofidou, M., Kozakou, S., Michael, N., Charalambous, C., Hatzigeorgiou, M., Christou, E., Stefani, D., Christoforou, E., Neocleous, D., 2022. Lead contamination of soils, sediments, and vegetation in a shooting range and adjacent terrestrial and aquatic ecosystems: A holistic approach for evaluating potential risks. Chemosphere 292, 133424.

Egendorf, S.P., Spliethoff, H.M., Shayler, H.A., Russell-Anelli, J., Cheng, Z., Minsky, A.H., King, T., McBride, M.B., 2021. Soil lead (Pb) and urban grown lettuce: Sources, processes, and implications for gardener best management practices. Journal of environmental management 286, 112211.

Foucault, Y., Lévêque, T., Xiong, T., Schreck, E., Austruy, A., Shahid, M., Dumat, C., 2013. Green manure plants for remediation of soils polluted by metals and metalloids: ecotoxicity and human bioavailability assessment. Chemosphere 93, 1430-1435.

Gemeda, F.T., Guta, D.D., Wakjira, F.S., Gebresenbet, G., 2021. Occurrence of heavy metal in water, soil, and plants in fields irrigated with industrial wastewater in Sabata town, Ethiopia. Environmental Science and Pollution Research 28, 12382-12396.

Hussain, M.M., Bibi, I., Niazi, N.K., Shahid, M., Iqbal, J., Shakoor, M.B., Ahmad, A., Shah, N.S., Bhattacharya, P., Mao, K., Bundschuh, J., Ok, Y.S., Zhang, H., 2021. Arsenic biogeochemical cycling in paddy soil-rice system: Interaction with various factors, amendments and mineral nutrients. Science of The Total Environment 773, 145040.

Iqbal, Z., Abbas, F., Ibrahim, M., Mahmood, A., Gul, M., Qureshi, T.I., 2021. Ecological risk assessment of soils under different wastewater irrigation farming system in Punjab, Pakistan. International Journal of Environmental Science and Technology, 1-12.

Irshad, M.K., Noman, A., Alhaithloul, H.A.S., Adeel, M., Rui, Y., Shah, T., Zhu, S., Shang, J., 2020. Goethite-modified biochar ameliorates the growth of rice (Oryza sativa L.) plants by suppressing Cd and As-induced oxidative stress in Cd and As co-contaminated paddy soil. The Science of the total environment 717, 137086.

Khalilzadeh, R., Pirzad, A., Sepehr, E., Khan, S., Anwar, S., 2021. The Salicornia europaea potential for phytoremediation of heavy metals in the soils under different times of wastewater irrigation in northwestern Iran. Environmental Science and Pollution Research, 1-14.

Kharazi, A., Leili, M., Khazaei, M., Alikhani, M.Y., Shokoohi, R., 2021. Human health risk assessment of heavy metals in agricultural soil and food crops in Hamadan, Iran. Journal of Food Composition and Analysis 100, 103890.
Lian, F., Liu, X., Gao, M., Li, H., Qiu, W., Song, Z., 2020. Effects of Fe-Mn-Ce oxide-modified biochar on As accumulation, morphology, and quality of rice (Oryza sativa L.). Environmental science and pollution research international 27, 18196-18207.
Marove, C.A., Sotozono, R., Tangviroon, P., Tabelin, C.B., Igarashi, T., 2022. Assessment of soil, sediment and water contaminations around open-pit coal mines in Moatize, Tete province, Mozambique. Environmental Advances 8, 100215.
Mombo, S., Foucault, Y., Deola, F., Gaillard, I., Goix, S., Shahid, M., Schreck, E., Pierart, A., Dumat, C., 2016. Management of human health risk in the context of kitchen gardens polluted by lead and cadmium near a lead recycling company. Journal of Soils and Sediments 16, 1214-1224.
Natasha, Bibi, I., Niazi, N.K., Shahid, M., Ali, F., Masood Ul Hasan, I., Rahman, M.M., Younas, F., Hussain, M.M., Mehmood, T., Shaheen, S.M., Naidu, R., Rinklebe, J., 2022. Distribution and ecological risk assessment of trace elements in the paddy soil-rice ecosystem of Punjab, Pakistan. Environmental Pollution 307, 119492.
Natasha, N., Shahid, M., Khalid, S., Niazi, N.K., Murtaza, B., Ahmad, N., Farooq, A., Zakir, A., Imran, M., Abbas, G., 2021. Health risks of arsenic buildup in soil and food crops after wastewater irrigation. Science of The Total Environment 772, 145266.
Sardar, A., Shahid, M., Khalid, S., Anwar, H., Tahir, M., Shah, G.M., Mubeen, M., 2020. Risk assessment of heavy metal (loid) s via Spinacia oleracea ingestion after sewage water irrigation practices in Vehari District. Environmental Science and Pollution Research 27, 39841–39851.
Shah, A.H., Shahid, M., Tahir, M., Natasha, Bibi, I., Tariq, T.Z., Khalid, S., Nadeem, M., Abbas, G., Saeed, M.F., Ansar, S., Dumat, C., 2022. Risk assessment of trace element accumulation in soil and Brassica oleracea after wastewater irrigation. Environmental geochemistry and health doi.org/10.1007/s10653-022-01351-4.
Shahid, M., Arshad, M., Kaemmerer, M., Pinelli, E., Probst, A., Baque, D., Pradere, P., Dumat, C., 2012. Long-term field metal extraction by pelargonium: phytoextraction efficiency in relation to plant maturity. International Journal of Phytoremediation 14, 493-505.
Shahid, M., Xiong, T., Masood, N., Leveque, T., Quenea, K., Austruy, A., Foucault, Y., Dumat, C., 2014. Influence of plant species and phosphorus amendments on metal speciation and bioavailability in a smelter impacted soil: a case study of food-chain contamination. Journal of Soils and Sediments 14, 655-665.
Tariq, F., 2021. Heavy metals concentration in vegetables irrigated with municipal wastewater and their human daily intake in Erbil city. Environmental Nanotechnology, Monitoring & Management 16, 100475.
Wu, Y., Qi, L., Wang, B., Medley, P., Drake, J., Vernon, J., Ibeanusi, V., Chen, G., 2022. Assess long-term As, Pb and Cr contamination and uptake by Eriocaulon decangulare in the Apalachicola National Forest. Science of The Total Environment 838, 156040.
Xiong, T., Dumat, C., Pierart, A., Shahid, M., Kang, Y., Li, N., Bertoni, G., Laplanche, C., 2016. Measurement of metal bioaccessibility in vegetables to improve human exposure assessments: field study of soil–plant–atmosphere transfers in urban areas, South China. Environmental geochemistry and health 38, 1283-1301.
Xiong, T., Leveque, T., Shahid, M., Foucault, Y., Mombo, S., Dumat, C., 2014. Lead and cadmium phytoavailability and human bioaccessibility for vegetables exposed to soil or atmospheric pollution by process ultrafine particles. Journal of environmental quality 43, 1593-1600.
Yáñez, C., Verdejo, J., Moya, H., Donoso, P., Rojas, C., Dovletyarova, E.A., Shapoval, O.A., Krutyakov, Y.A., Neaman, A., 2022. Microbial responses are unreliable indicators of copper ecotoxicity in soils contaminated by mining activities. Chemosphere 300, 134517.

Yang, S., Feng, W., Wang, S., Chen, L., Zheng, X., Li, X., Zhou, D., 2021. Farmland heavy metals can migrate to deep soil at a regional scale: A case study on a wastewater-irrigated area in China. Environmental Pollution 281, 116977.
Zhang, X., Fu, G., Xing, S., Fu, W., Liu, X., Wu, H., Zhou, X., Ma, Y., Zhang, X., Chen, B., 2022. Structure and diversity of fungal communities in long-term copper-contaminated agricultural soil. Science of The Total Environment 806, 151302.