Ecological load and migration of heavy metals in soil profile in wheat-corn rotation system

Yi Zhang[1], Kunling Zheng[1], Yinxian Song[1,*], Tengjie Cui[1], Zhongyao Chen[1], Chunjun Tao[2,*]

[1] Department of Geosciences, Faculty of Land Resource Engineering, Kunming University of Science and Technology, Kunming 650093, Yunnan Province, China; zhangyi00606@outlook.com (Y.Z.); kunlingzheng1@gmail.com (K.Z.); realbobohu04@stu.kust.edu.cn (T.C.); 18971898831@163.com (Z.C.)

[2] Geological Survey of Anhui Province (Anhui Institute of Geological Sciences), Hefei, 230001, Anhui Province, China;

* Correspondence: 20200135@kust.edu.cn or songyinxian@gmail.com (Y.S.); tcj9903@sina.com (C.T.)

Table S1. Parameters for PROFILE models

parameter	unit	minimum	maximum	average value
pH		7.13	8.85	8.27
DOC	g/kg	0.007	0.148	0.046
Feo (Oxalate-extractable Fe)	g/kg	2.99	3.62	3.25
Fed (Citrate-Bicarbonate-Dithionite extractable Fe)	g/kg	25.47	73.24	40.06
Al[a]	cmol/kg	0.00	0.00	0.00
BC[a]	cmol/kg	10.60	35.40	25.77
θ	%	0.26	0.33	0.29
XClay[a]	%	18.90	58.90	42.20
XSilt[a]	%	25.50	47.10	34.60
XSand[a]	%	2.40	45.60	23.30
Aw (10^6)	m2/m3	3.11	8.69	5.74
ϱ (Soil bulk density) [a]	kg/m3	1200	1560	1358.46
Average Month Temperature[b]	℃	28.7	3.3	16.58
Average Month Precipitation[b]	mm	296.9	13.6	887.4

a. Data from the Soil Science Database (http://vdb3.soil.csdb.cn/);

b. Data from the National Bureau of Statistics (http://www.stats.gov.cn/tjsj/ndsj/2020/indexch.htm).

Table S2. Soil Characteristics of the 16 individual farmland sampling sites.

Sample	Chinese Soil Classification	pH	TFe_2O_3 %	TOC g/kg	Bulk Density (g/cm^3)
PM01	Shajiang Black Soil	8.03	5.06	4.28	1.36
PM02	Shajiang Black Soil	8.23	5.24	3.41	1.36
PM03	Shajiang Black Soil	8.58	4.59	4.98	1.36
PM04	Shajiang Black Soil	8.46	3.89	4.19	1.36
PM05	Shajiang Black Soil	8.40	4.26	3.92	1.36
PM06	Shajiang Black Soil	8.72	3.44	2.86	1.36
PM07	Shajiang Black Soil	8.54	3.72	2.67	1.36
PM08	Shajiang Black Soil	8.38	4.37	5.84	1.36
PM09	Shajiang Black Soil	8.37	6.16	6.30	1.36
PM10	Shajiang Black Soil	8.25	4.76	5.51	1.36
PM11	Shajiang Black Soil	8.39	5.24	6.02	1.36
PM12	Shajiang Black Soil	8.43	4.20	3.75	1.36
PM13	Shajiang Black Soil	7.75	5.56	4.35	1.36
PM14	Shajiang Black Soil	7.87	5.43	5.51	1.36
PM15	Shajiang Black Soil	8.13	6.48	4.79	1.42
PM16	Shajiang Black Soil	7.75	4.70	5.42	1.36

Table S3. Average weathering rate (Rw) and base cation release rate (R_{BC}) in soil.

Release rate	unit	Surface soil （0-20cm）	subsoil （20-200cm）
K^+	keq/(ha·a)	0.03	0.11
Na^+	keq/(ha·a)	0.16	0.52
Ca^{2+}	keq/(ha·a)	0.94	3.17
Mg^{2+}	keq/(ha·a)	0.31	1.36
R_W	keq/(ha·a)	1.27	4.35
R_{BC}	keq/(ha·a)	1.44	5.16

Table S4. Correlation of heavy metal release rates with soil properties.

		RmAs	RmHg	RmCu	RmPb	RmZn	RmCd	RmCr
wheat	Feo/Fed	-0.155	-0.169	-0.253*	-0.252*	-0.128	-0.21	-0.078
	Feo	0.172	0.182	0.277*	0.29**	0.149	0.24*	0.111
	Fed	0.19	0.198	0.306**	0.343**	0.177	0.283*	0.157
	PH	0.201	0.183	0.13	0.086	0.245*	0.218	0.224*
	Clay	0.379**	0.318**	0.339**	0.367**	0.396**	0.295**	0.453**
	Silt	0.163	0.223*	0.171	0.13	0.186	0.193	0.158
	Sand	-0.372**	-0.342**	0.342**	-0.353**	-0.395**	-0.311**	-0.433**
	TOC	-0.395**	-0.181	-0.376**	-0.413**	0.382**	-0.244*	-0.494**
corn	Feo/Fed	-0.129	-0.143	-0.227*	-0.222	-0.099	-0.186	-0.045
	Feo	0.145	0.155	0.25*	0.261*	0.121	0.216	0.078
	Fed	0.163	0.171	0.279*	0.314**	0.149	0.259*	0.124
	pH	0.226*	0.207	0.156	0.115	0.27*	0.239*	0.253*
	Clay	0.39**	0.325**	0.351**	0.38**	0.407**	0.304**	0.465**
	Silt	0.166	0.227*	0.175	0.134	0.188	0.197	0.159
	Sand	-0.382**	-0.348**	-0.352**	-0.366**	-0.404**	-0.32**	-0.444**
	TOC	-0.403**	-0.179	-0.383**	-0.421**	-0.388**	-0.247*	-0.501**

Table S5. Average Input and Output Flux of Heavy Metals in surface Soil ($g \cdot ha^{-1} \cdot a^{-1}$).

	Wheat Output fluxes			**Corn Output fluxes**			**Input fluxes**		
	U_g	**U_s**	**R_m**	**U_g**	**U_s**	**R_m**	**Q_a**	**Q_i**	**Q_f**
As	0.09	3.19	8.39	0.001	0.04	8.41	9.12	31.72	2.82
Hg	0.01	0.04	0.03	0.0001	0.001	0.03	0.13	0.12	0.01
Cu	11.76	55.77	18.74	0.115	1.13	18.75	31.84	26.19	2.76
Pb	0.15	8.84	19.23	0.002	0.09	19.25	49.78	16.44	1.75
Zn	67.17	122.55	51.03	1.147	1.77	51.07	139.21	36.68	9.08
Cd	0.04	1.07	0.13	0.0002	0.006	0.13	1.12	0.17	0.05
Cr	0.31	3.04	48.44	0.007	1.21	48.44	57.63	0	4.16

U_g-Grain Absorption Flux; U_s-Straw absorption flux; R_m-Heavy metal leaching; Q_a-atmospheric deposition flux; Q_i-irrigation flux; Q_f-fertilizer flux.

Table S6. Multiple Regression Prediction Results.

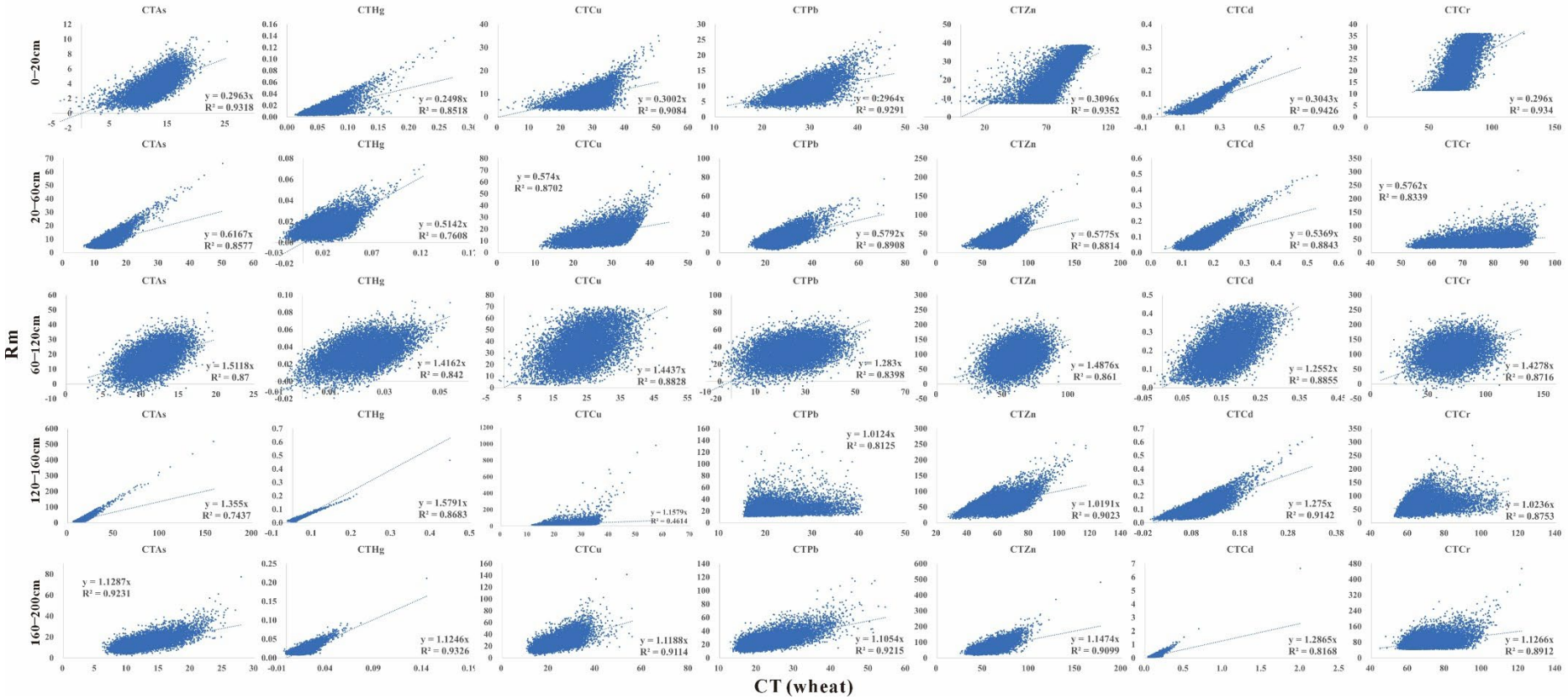
	wheat			corn		
0-20cm	**R^2**	**p**	**Regression equation**	**R^2**	**p**	**Regression equation**
As	0.863	<0.001	CT=-123.071+42.257Feo-0.256Fed-0.037Clay+0.217TOC+0.758PH+0.3Rm	0.861	<0.001	CT=-122.413+41.767Feo-0.243Fed-0.036Clay+0.220TOC+0.817PH+0.200Rm
Hg	0.868	<0.001	CT=1.665-0.573Feo+0.008Fed-0.000231Clay+0.002TOC-0.012PH+1.764Rm	0.864	<0.001	CT=1.749-0.603Feo+0.008Fed-0.000226Clay+0.002TOC-0.012PH+1.348Rm
Cu	0.873	<0.001	CT=-155.475+77.419Feo-0.716Fed+0.049Clay+0.338TOC-6.476PH+0.839Rm	0.874	<0.001	CT=-152.356+76.810Feo-0.699Fed+0.054Clay+0.334TOC-6.691PH+0.641Rm
Pb	0.708	<0.001	CT=-27.590+30.024Feo-0.195Fed+0.047Clay+0.454TOC-5.862PH+0.898Rm	0.698	<0.001	CT=-20.496+27.775Feo-0.154Fed+0.055Clay+0.457TOC-6.026PH+0.669Rm
Zn	0.747	<0.001	CT=-726.471+304.676Feo-3.510Fed-0.052Clay+1.076TOC-10.911PH+1.281Rm	0.747	<0.001	CT=-707.855+299.655Feo-3.408Fed-0.037Clay+1.063TOC-11.670PH+0.982Rm
Cd	0.889	<0.001	CT=-1.068+0.489Feo-0.006Fed-0.001Clay+0.006TOC-0.026PH+1.752Rm	0.888	<0.001	CT=-0.962+0.456Feo-0.006Fed-0.001Clay+0.007TOC-0.028PH+1.346Rm
Cr	0.895	<0.001	CT=-200.058+73.694Feo+0.053Fed-0.107Clay+0.524TOC+2.873PH+0.246Rm	0.895	<0.001	CT=-194.306+72.028Feo+0.080Fed-0.105Clay+0.520TOC+2.708PH+0.191Rm

	wheat			corn		
20-60cm	**R^2**	**p**	**Regression equation**	**R^2**	**p**	**Regression equation**
As	0.780	<0.001	CT=148.949-43.148Feo+0.591Fed-0.084XClay+0.596TOC-2.835PH+0.665Rm	0.784	<0.001	CT=162.746-47.363Feo+0.651Fed-0.086XClay+0.612TOC-3.153PH+0.541RmAs
Hg	0.794	<0.001	CT=0.547-0.139Feo+0.000495Fed-0.001XClay+0.004TOC-0.011PH+1.575Rm	0.802	<0.001	CT=0.620-0.163Feo+0.001Fed-0.001XClay+0.004TOC-0.013PH+1.266RmHg
Cu	0.977	<0.001	CT=-65.786+23.257Feo+0.179Fed-0.052XClay+0.674TOC+0.524PH+0.209Rm	0.977	<0.001	CT=-52.360+19.215Feo+0.230Fed-0.056XClay+0.687TOC+0.235PH+0.174RmCu
Pb	0.883	<0.001	CT=297.447-69.556Feo+0.704Fed-0.257XClay+1.575TOC-9.835PH+0.799Rm	0.882	<0.001	CT=329.466-79.361Feo+0.852Fed-0.258XClay+1.592TOC-10.588PH+0.643RmPb
Zn	0.841	<0.001	CT=-378.822+120.114Feo-1.506Fed-0.491XClay+2.808TOC+12.293PH+0.547Rm	0.847	<0.001	CT=-300.299+96.444Feo-1.187Fed-0.502XClay+2.901TOC+10.485PH+0.450RmZn
Cd	0.803	<0.001	CT=0.030-0.043Feo-0.001Fed-0.001XClay+0.014TOC+0.027PH+0.867Rm	0.808	<0.001	CT=0.214-0.097Feo-0.000398Fed-0.001XClay+0.014TOC+0.023PH+0.706RmCd
Cr	0.977	<0.001	CT=-74.315+9.442Feo+1.284Fed-0.063XClay-0.695TOC+8.146PH+0.048Rm	0.977	<0.001	CT=-67.241+7.416Feo+1.309Fed-0.065XClay-0.681TOC+7.958PH+0.040RmCr

	wheat			corn		
60-120cm	**R^2**	**p**	**Regression equation**	**R^2**	**p**	**Regression equation**
As	0.910	<0.001	CT=-160.893+56.145Feo-0.692Fed-0.079XClay+0.506TOC+2.013PH+0.193Rm	0.908	<0.001	CT=-165.522+57.916Feo-0.711Fed-0.080XClay+0.493TOC+1.979PH+0.153RmAs
Hg	0.451	<0.001	CT=0.556-0.106Feo+0.001Fed-0.000304XClay+0.002TOC-0.029PH+0.520Rm	0.444	<0.001	CT=0.530-0.097Feo+0.001Fed-0.000299XClay+0.002TOC-0.028PH+0.406RmHg
Cu	0.980	<0.001	CT=-159.683+56.972Feo-0.312Fed-0.102XClay+0.185TOC+1.375PH+0.098Rm	0.979	<0.001	CT=-163.871+58.516Feo-0.327Fed-0.101XClay+0.171TOC+1.355PH+0.077RmCu
Pb	0.965	<0.001	CT=424.284-130.783Feo+2.272Fed-0.056XClay+0.438TOC-8.119PH+0.043Rm	0.965	<0.001	CT=423.202-130.392Feo+2.270Fed-0.056XClay+0.431TOC-8.128PH+0.033RmPb
Zn	0.914	<0.001	CT=-302.041+96.955Feo-0.532Fed-0.299XClay+0.484TOC+8.014PH+0.161Rm	0.914	<0.001	CT=-308.167+100.443Feo-0.566Fed-0.304XClay+0.440TOC+7.576PH+0.129RmZn
Cd	0.759	<0.001	CT=3.194-0.940Feo+0.013Fed-0.001XClay+0.009TOC-0.069PH+0.418Rm	0.756	<0.001	CT=3.167-0.929Feo+0.013Fed-0.001XClay+0.009TOC-0.070PH+0.332RmCd
Cr	0.993	<0.001	CT=141.789-46.558Feo+1.897Fed-0.021XClay-0.108TOC+0.266PH+0.033Rm	0.993	<0.001	CT=139.186-45.409Feo+1.886Fed-0.021XClay-0.123TOC+0.189PH+0.026RmCr

	wheat			corn		
120-160cm	R^2	p	Regression equation	R^2	p	Regression equation
As	0.948	<0.001	CT=-12.759-0.834Feo+0.222Fed-0.191XClay-0.041TOC+3.007PH+0.286Rm	0.948	<0.001	CT=-8.085-2.374Feo+0.251Fed-0.189XClay-0.038TOC+2.895PH+0.232RmAs
Hg	0.834	<0.001	CT=-0.222+0.103Feo-0.002Fed-0.000329XClay-0.001TOC-0.003PH+0.892Rm	0.848	<0.001	CT=-0.191+0.092Feo-0.001Fed-0.000323XClay-0.001TOC-0.003PH+0.713RmHg
Cu	0.958	<0.001	CT=-131.512+35.899Feo+0.063Fed-0.091XClay+0.645TOC+4.595PH+0.026Rm	0.957	<0.001	CT=-130.712+35.652Feo+0.067Fed-0.092XClay+0.645TOC+4.575PH+0.021RmCu
Pb	0.980	<0.001	CT=86.769-33.903Feo+1.086Fed-0.002XClay+0.132TOC+0.404PH-0.017Rm	0.980	<0.001	CT=87.417-34.418Feo+1.098Fed+0.001XClay+0.146TOC+0.467PH-0.019RmPb
Zn	0.859	<0.001	CT=-430.343+134.692Feo-1.367Fed-0.380XClay+2.738TOC+12.674PH+0.229Rm	0.860	<0.001	CT=-411.063+129.049Feo-1.269Fed-0.379XClay+2.724TOC+12.044PH+0.189RmZn
Cd	0.926	<0.001	CT=-0.838+0.216Feo-0.003Fed-0.002XClay+0.011TOC+0.044PH+0.419Rm	0.927	<0.001	CT=-0.778+0.198Feo-0.003Fed-0.002XClay+0.010TOC+0.042PH+0.345RmCd
Cr	0.950	<0.001	CT=309.553-93.603Feo+2.186Fed-0.122XClay+1.575TOC-2.950PH+0.068Rm	0.951	<0.001	CT=311.219-93.639Feo+2.189Fed-0.128XClay+1.547TOC-3.145PH+0.059RmCr

	wheat			corn		
160-200cm	**R^2**	**p**	**Regression equation**	**R^2**	**p**	**Regression equation**
As	0.981	<0.001	CT=69.750-16.046Feo+0.389Fed-0.070XClay+0.155TOC-2.501PH+0.213Rm	0.982	<0.001	CT=71.320-16.898Feo+0.409Fed-0.073XClay+0.138TOC-2.445PH+0.177RmAs
Hg	0.844	<0.001	CT=0.215-0.029Feo+0.00002429Fed-0.000273XClay+0.000188TOC-0.013PH+0.737Rm	0.863	<0.001	CT=0.222-0.034Feo+0.000136Fed-0.000279XClay+0.000078TOC-0.012PH+0.600RmHg
Cu	0.988	<0.001	CT=15.344+3.545Feo+0.360Fed-0.103XClay+0.666TOC-2.213PH+0.125Rm	0.989	<0.001	CT=20.792+1.911Feo+0.383Fed-0.107XClay+0.642TOC-2.338PH+0.108RmCu
Pb	0.948	<0.001	CT=203.262-40.335Feo+0.712Fed-0.085XClay+0.574TOC-9.583PH+0.172Rm	0.945	<0.001	CT=201.646-41.689Feo+0.760Fed-0.090XClay+0.532TOC-9.059PH+0.138RmPb
Zn	0.924	<0.001	CT=-255.784+85.503Feo-0.459Fed-0.345XClay+0.599TOC+6.223PH+0.271Rm	0.929	<0.001	CT=-199.901+76.405Feo-0.417Fed-0.361XClay+0.552TOC+2.753PH+0.239RmZn
Cd	0.932	<0.001	CT=-1.279+0.347Feo-0.003Fed-0.000488XClay+0.002TOC+0.044PH+0.285Rm	0.931	<0.001	CT=-1.078+0.323Feo-0.003Fed-0.001XClay+0.002TOC+0.030PH+0.267RmCd
Cr	0.984	<0.001	CT=297.991-96.290Feo+2.356Fed-0.180XClay-0.506TOC-1.037PH+0.118Rm	0.984	<0.001	CT=300.454-97.412Feo+2.390Fed-0.192XClay-0.560TOC-1.043PH+0.100Rm


CTAs
CTHg
CTCu
CTPb
CTZn
CTCd
CTCr
0–20cm
20–60cm
60–120cm
120–160cm
160–200cm
y = 0.2963x
R² = 0.9318
y = 0.2498x
R² = 0.8518
y = 0.3002x
R² = 0.9084
y = 0.2964x
R² = 0.9291
y = 0.3096x
R² = 0.9352
y = 0.3043x
R² = 0.9426
y = 0.296x
R² = 0.934
y = 0.6167x
R² = 0.8577
y = 0.5142x
R² = 0.7608
y = 0.574x
R² = 0.8702
y = 0.5792x
R² = 0.8908
y = 0.5775x
R² = 0.8814
y = 0.5369x
R² = 0.8843
y = 0.5762x
R² = 0.8339
y = 1.5118x
R² = 0.87
y = 1.4162x
R² = 0.842
y = 1.4437x
R² = 0.8828
y= 1.283x
R² = 0.8398
y = 1.4876x
R² = 0.861
y = 1.2552x
R² = 0.8855
y = 1.4278x
R² = 0.8716
y = 1.355x
R² = 0.7437
y = 1.5791x
R² = 0.8683
y = 1.1579x
R² = 0.4614
y = 1.0124x
R² = 0.8125
y = 1.0191x
R² = 0.9023
y = 1.275x
R² = 0.9142
y = 1.0236x
R² = 0.8753
y = 1.1287x
R² = 0.9231
y = 1.1246x
R² = 0.9326
y = 1.1188x
R² = 0.9114
y = 1.1054x
R² = 0.9215
y = 1.1474x
R² = 0.9099
y = 1.2865x
R² = 0.8168
y = 1.1266x
R² = 0.8912
Rm
CT (wheat)

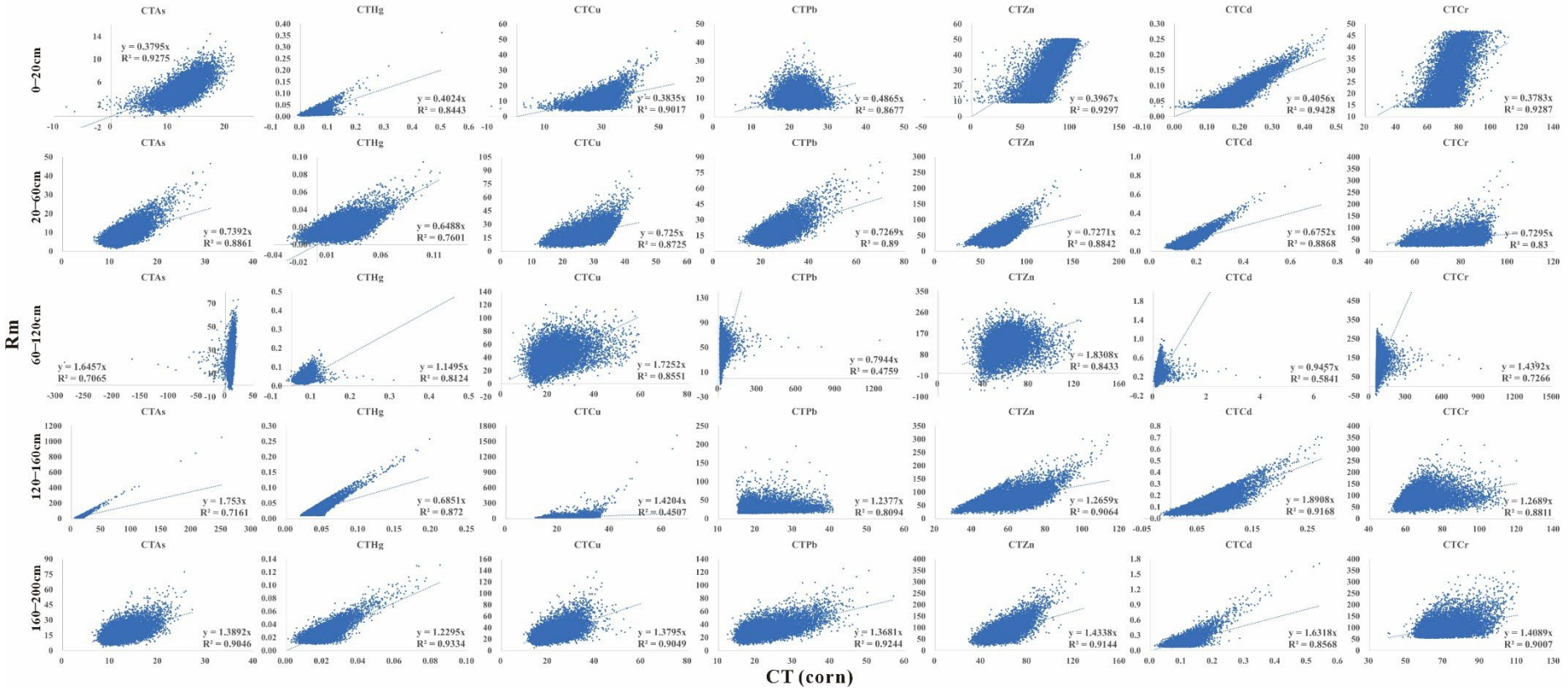
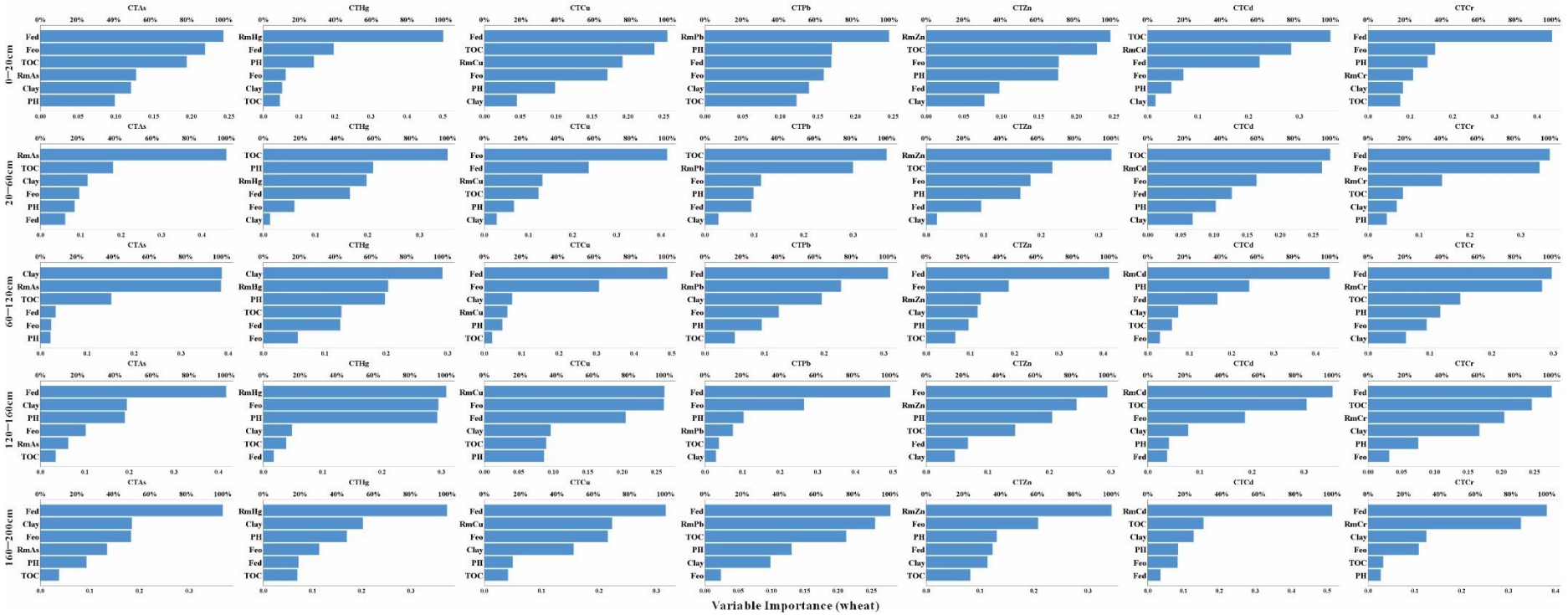


Figure S1. Monte Carlo's prediction of heavy metal concentrations


CTAs
CTHg
CTCu
CTPb
CTZn
CTCd
CTCr
0%
20%
40%
60%
80%
100%
0–20cm
Fed
Feo
TOC
RmAs
Clay
PH
0.00
0.05
0.10
0.15
0.20
0.25
RmHg
PH
Feo
Clay
TOC
0.0
0.1
0.2
0.3
0.4
0.5
RmCu
RmPb
RmZn
RmCd
RmCr
20–60cm
60–120cm
120–160cm
160–200cm
Variable Importance (wheat)

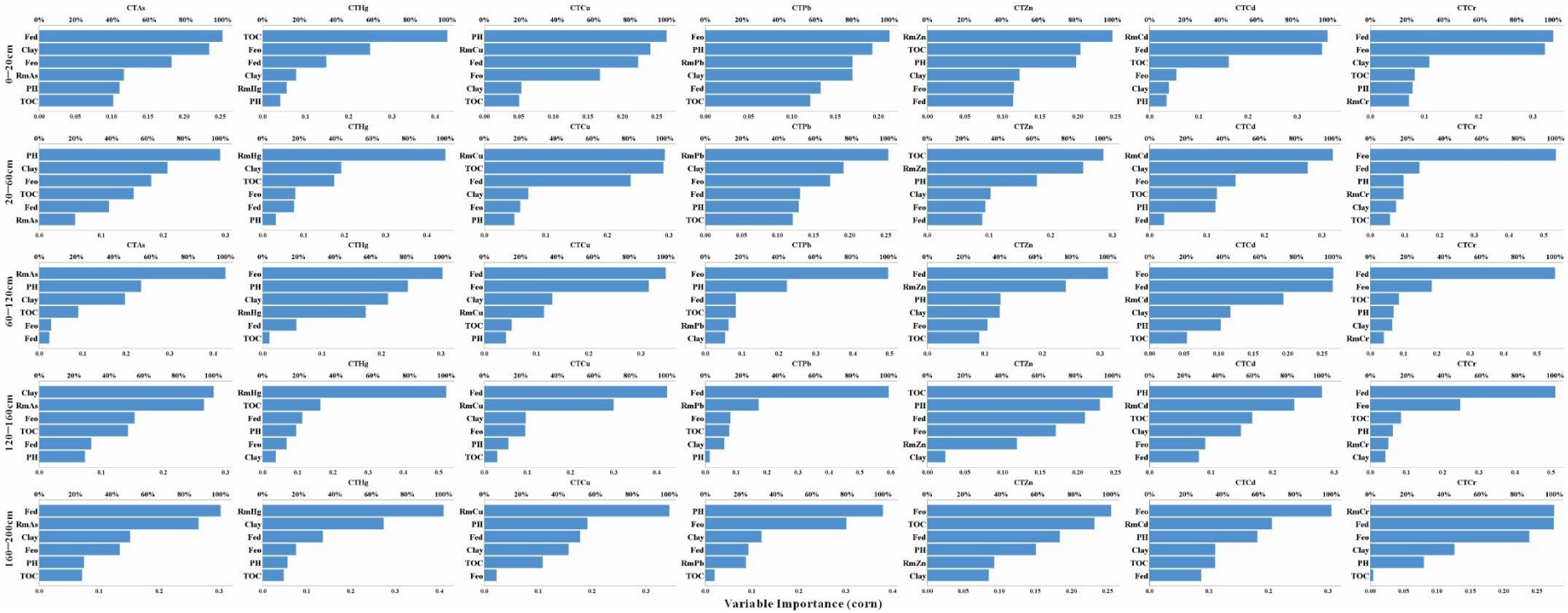
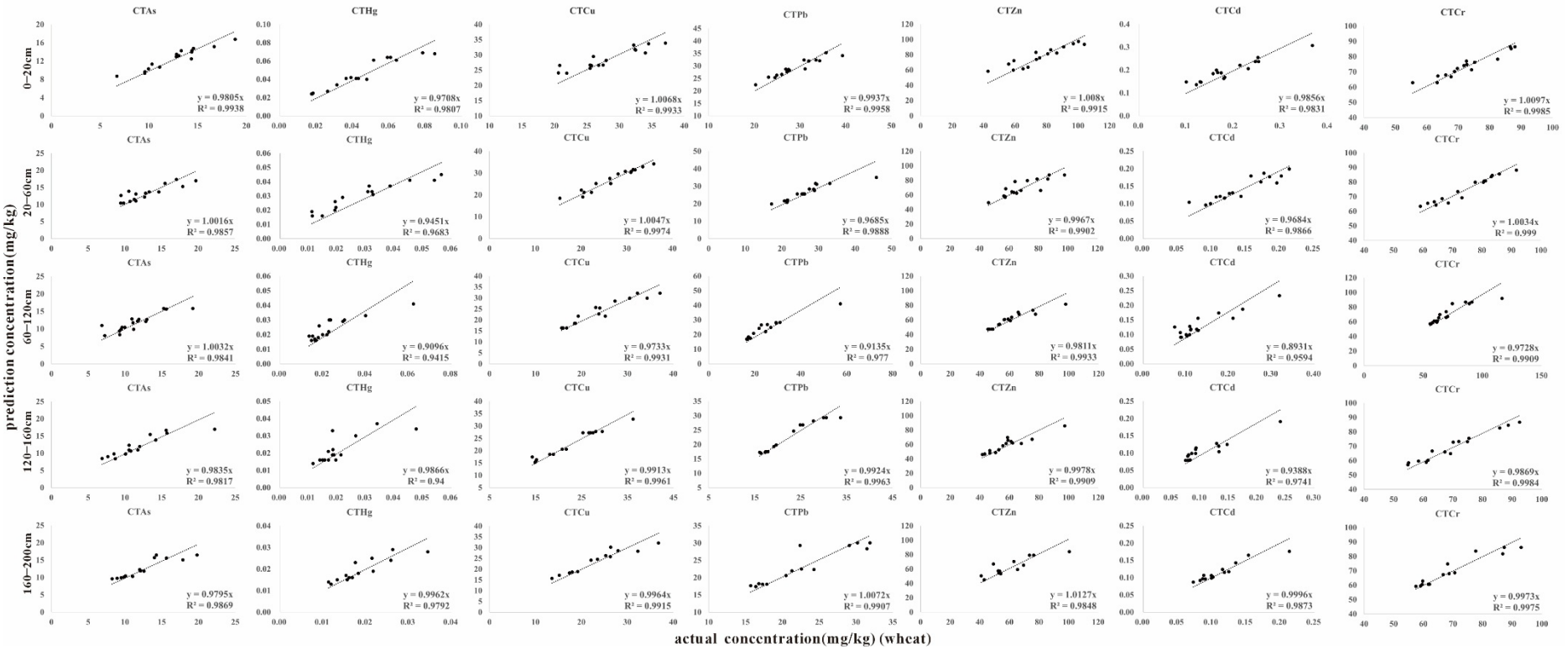


Figure S2. Normalized Importance of Neural Network Variables.


CTAs
CTHg
CTCu
CTPb
CTZn
CTCd
CTCr
0–20cm
20–60cm
60–120cm
120–160cm
160–200cm
y = 0.9805x R² = 0.9938
y = 0.9708x R² = 0.9807
y = 1.0068x R² = 0.9933
y = 0.9937x R² = 0.9958
y = 1.008x R² = 0.9915
y = 0.9856x R² = 0.9831
y = 1.0097x R² = 0.9985
y = 1.0016x R² = 0.9857
y = 0.9451x R² = 0.9683
y = 1.0047x R² = 0.9974
y = 0.9685x R² = 0.9888
y = 0.9967x R² = 0.9902
y = 0.9684x R² = 0.9866
y = 1.0034x R² = 0.999
y = 1.0032x R² = 0.9841
y = 0.9096x R² = 0.9415
y = 0.9733x R² = 0.9931
y = 0.9135x R² = 0.977
y = 0.9811x R² = 0.9933
y = 0.8931x R² = 0.9594
y = 0.9728x R² = 0.9909
y = 0.9835x R² = 0.9817
y = 0.9866x R² = 0.94
y = 0.9913x R² = 0.9961
y = 0.9924x R² = 0.9963
y = 0.9978x R² = 0.9909
y = 0.9388x R² = 0.9741
y = 0.9869x R² = 0.9984
y = 0.9795x R² = 0.9869
y = 0.9962x R² = 0.9792
y = 0.9964x R² = 0.9915
y = 1.0072x R² = 0.9907
y = 1.0127x R² = 0.9848
y = 0.9996x R² = 0.9873
y = 0.9973x R² = 0.9975
prediction concentration(mg/kg)
actual concentration(mg/kg) (wheat)

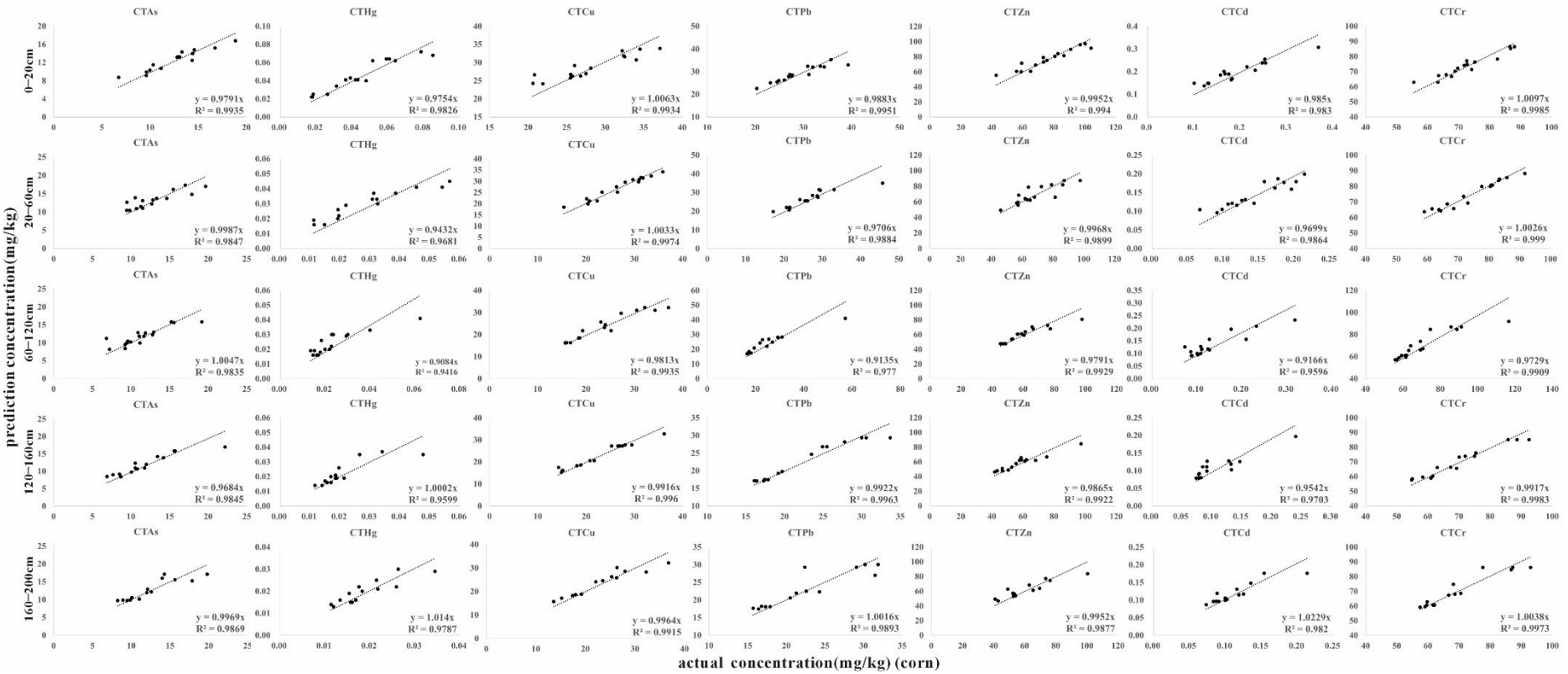


Figure S3. Random Forest Predictions of Heavy Metal Concentrations.