

Supplementary Materials

Accumulation and Origin of Phosphorus and Heavy Metals in Citrus Orchard Soils in Jeju Island, South Korea: Potential Ecological Risks and Bioavailability

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1. Physicochemical analysis

Among soil physicochemical properties, soil pH (H₂O), soil organic matter (SOM), and cation exchange capacity (CEC) were analyzed with 1:5 ratio, Walkley-Black wet digestion method and 1 M ammonium acetate (pH 7.0) extraction (Kjeldahl method), according to the Methods of Soil Chemical Analysis suggested by the National Institute of Agricultural Science of Rural Development Administration, Republic of Korea [1]. Soil pH (NaF), phosphorus retention capacity (P_{RC}), and selective extraction were analyzed following the Soil Survey Laboratory Method Manual [2]. Soil pH (H₂O, NaF) was measured using a pH meter (Orion star A211, Thermo Scientific, USA); phosphorus of P_{RC} was measured using a UV/Vis spectrometer (Lambda 25, PerkinElmer, USA); and selective extraction (Al, Fe) were measured using an inductively coupled plasma optical emission spectrometer (JY138 Ultrace, Jobin Yvon, France).

Table S1. Summary of physicochemical properties in the topsoil and subsoil samples collected from citrus orchard and forestland sites in Jeju Island, South Korea

Site			pH		SOM ^a	CEC ^b	P _{RC} ^c	Al _o +1/2Fe _o
			H ₂ O	NaF	(g/kg)	(cmolc/kg)	(%)	(%)
Citrus orchard	Topsoil	Mean	5.02	9.31	96.3	6.55	69.4	3.18
		SD	0.65	0.56	43.8	0.18	20.8	1.13
		Max	6.68	10.2	185	6.95	95.4	4.85
		Min	4.04	8.28	20.7	6.18	25.6	0.92
	Subsoil	Mean	4.84	9.75	83.3	6.55	80.9	3.75
		SD	0.66	0.95	46.1	0.19	19.8	1.60
		Max	6.54	11.2	197	6.85	99.3	7.36
		Min	4.02	7.94	11.9	6.19	30.1	0.95
Forestland	Topsoil	Mean	4.73	10.2	182	6.53	95.0	3.96
		SD	0.26	0.7	31.3	0.14	4.0	0.98
		Max	5.08	11.2	219	6.85	98.6	5.50
		Min	4.23	8.84	124	6.38	86.2	2.93
	Subsoil	Mean	4.83	10.9	154	6.61	99.4	5.46
		SD	0.20	0.5	41.7	0.21	0.9	1.90
		Max	5.14	11.5	204	6.97	100.0	8.90
		Min	4.37	9.93	79.4	6.39	97.5	3.31

^a Soil organic matter, ^b Cation exchange capacity, ^c Phosphorus retention capacity.

Table S2. Concentration of total phosphorus (TP) and heavy metals in the topsoil and subsoil collected from citrus orchard and forestland sites in Jeju Island, South Korea

Site	TP (%)			Pb (mg/kg)			Zn (mg/kg)			Cu (mg/kg)			Cd (mg/kg)			Cr (mg/kg)			Ni (mg/kg)			Al (%)		
	A	B	C	A	B	C	A	B	C	A	B	C	A	B	C	A	B	C	A	B	C	A	B	C
Citrus orchard soils																								
1	1.14	0.19	6.2	23.9	19.2	1.2	269.3	117.8	2.3	145.9	50.8	2.9	1.3	0.8	1.6	144.4	97.5	1.5	62.2	80.4	0.8	4.70	5.36	0.9
2	0.77	0.08	9.4	24.4	17.6	1.4	144.9	143.4	1.0	116.8	41.8	2.8	1.0	0.8	1.3	127.3	95.0	1.3	71.9	82.5	0.9	6.40	7.58	0.8
3	0.31	0.12	2.6	18.6	15.1	1.2	145.7	118.9	1.2	54.5	42.2	1.3	0.8	0.8	1.0	129.0	119.3	1.1	101.8	105.9	1.0	6.48	6.75	1.0
4	0.24	0.05	4.7	20.2	14.2	1.4	135.9	116.0	1.2	84.9	54.8	1.5	0.9	0.9	1.0	165.3	155.4	1.1	155.8	158.8	1.0	7.00	6.62	1.1
5	0.55	0.22	2.5	20.0	12.4	1.6	142.5	141.1	1.0	111.1	60.0	1.9	0.9	1.0	0.9	244.9	149.8	1.6	75.9	96.0	0.8	6.04	8.32	0.7
6	0.64	0.21	3.0	23.6	21.8	1.1	182.5	149.7	1.2	106.9	54.8	2.0	1.2	1.0	1.2	127.3	118.5	1.1	85.3	100.1	0.9	6.49	7.43	0.9
7	0.14	0.07	2.0	15.1	12.7	1.2	97.2	68.0	1.4	47.9	25.3	1.9	0.5	0.4	1.3	72.6	59.9	1.2	40.5	36.5	1.1	2.97	2.89	1.0
8	0.38	0.14	2.7	14.5	13.8	1.1	92.0	79.0	1.2	35.2	30.2	1.2	0.7	0.6	1.2	86.8	76.3	1.1	56.6	59.2	1.0	3.77	4.29	0.9
9	0.40	0.15	2.6	35.0	17.1	2.0	192.6	127.2	1.5	173.7	85.2	2.0	1.3	1.0	1.3	184.4	122.3	1.5	71.6	81.0	0.9	5.92	6.81	0.9
10	1.36	0.97	1.4	37.0	27.6	1.3	255.3	176.9	1.4	246.7	109.6	2.3	1.1	0.8	1.4	240.7	90.7	2.7	59.5	47.0	1.3	4.79	5.85	0.8
11	0.39	0.23	1.7	14.9	12.4	1.2	147.1	138.5	1.1	111.4	61.9	1.8	0.9	0.8	1.1	102.3	70.1	1.5	37.7	38.9	1.0	5.86	6.44	0.9
12	0.27	0.09	3.0	19.4	17.8	1.1	153.0	110.4	1.4	95.0	45.8	2.1	0.7	0.9	0.8	150.6	102.4	1.5	87.0	79.7	1.1	5.85	6.08	1.0
13	0.83	0.28	2.9	16.9	16.5	1.0	113.7	98.2	1.2	83.8	53.6	1.6	0.9	0.6	1.5	92.1	65.0	1.4	57.4	54.3	1.1	6.26	4.96	1.3
14	0.31	0.24	1.3	22.6	16.7	1.4	212.9	136.9	1.6	168.6	82.9	2.0	1.1	0.9	1.2	112.1	103.0	1.1	72.3	83.9	0.9	6.05	4.48	1.4
15	0.34	0.10	3.3	26.7	19.3	1.4	189.6	114.5	1.7	173.8	72.1	2.4	1.0	1.0	1.0	237.0	114.9	2.1	71.8	69.0	1.0	5.40	5.56	1.0
16	0.50	0.18	2.8	23.6	15.7	1.5	184.2	97.6	1.9	67.5	49.6	1.4	1.2	0.8	1.5	141.4	103.4	1.4	79.4	90.5	0.9	4.91	6.23	0.8
17	0.76	0.34	2.2	22.4	17.2	1.3	250.5	135.7	1.8	130.5	75.5	1.7	1.2	0.7	1.7	147.4	106.9	1.4	120.1	106.2	1.1	5.70	5.35	1.1
18	0.21	0.14	1.5	12.5	12.1	1.0	147.0	135.7	1.1	38.3	22.9	1.7	0.8	0.9	0.9	37.4	35.8	1.0	23.1	23.6	1.0	3.41	3.73	0.9
19	0.33	0.18	1.8	20.2	13.4	1.5	163.6	128.0	1.3	48.6	33.7	1.4	0.7	0.7	1.0	50.1	44.9	1.1	32.2	31.3	1.0	3.70	3.42	1.1
20	0.96	0.40	2.4	21.4	13.9	1.5	192.2	106.6	1.8	76.4	40.5	1.9	1.0	0.9	1.1	147.2	141.3	1.0	71.5	78.5	0.9	3.85	4.73	0.8
21	0.24	0.19	1.3	15.5	16.4	0.9	119.6	117.2	1.0	52.1	44.6	1.2	0.7	0.8	0.9	109.4	106.8	1.0	65.5	70.7	0.9	4.08	4.08	1.0
22	0.44	0.45	1.0	23.7	19.7	1.2	193.3	181.9	1.1	64.0	60.2	1.1	1.1	1.0	1.1	130.6	128.2	1.0	97.0	101.6	1.0	5.58	5.04	1.1
23	0.37	0.18	2.1	16.3	9.8	1.7	144.7	114.3	1.3	109.9	62.4	1.8	0.9	0.6	1.5	73.9	70.7	1.0	59.1	55.2	1.1	4.58	5.29	0.9
24	0.25	0.08	3.1	18.0	17.0	1.1	114.4	88.1	1.3	52.5	38.9	1.3	0.7	0.7	1.0	91.1	86.8	1.0	82.5	76.6	1.1	4.62	4.85	1.0
25	0.22	0.11	2.0	29.2	17.0	1.7	112.6	93.0	1.2	50.8	36.0	1.4	0.9	0.5	1.8	102.1	78.9	1.3	50.3	52.8	1.0	4.36	5.17	0.8
26	0.39	0.19	2.0	14.6	7.9	1.8	136.6	97.6	1.4	60.3	48.8	1.2	1.0	0.6	1.7	89.2	71.1	1.3	53.2	44.9	1.2	4.96	5.89	0.8
27	1.23	0.68	1.8	35.6	14.3	2.5	191.0	159.1	1.2	108.7	63.5	1.7	1.7	1.0	1.7	161.2	94.8	1.7	58.8	52.6	1.1	3.64	4.19	0.9
28	0.33	0.35	1.0	14.0	4.9	2.9	144.7	89.0	1.6	73.1	57.2	1.3	0.9	0.9	1.0	79.5	58.0	1.4	55.0	43.5	1.3	4.15	6.84	0.6
29	0.24	0.09	2.6	18.3	14.1	1.3	103.7	93.7	1.1	40.8	33.5	1.2	0.8	0.7	1.1	89.0	82.7	1.1	70.5	74.2	1.0	5.52	5.72	1.0

30	0.21	0.09	2.4	21.8	17.8	1.2	129.9	92.7	1.4	48.7	27.5	1.8	0.8	0.6	1.3	80.0	70.5	1.1	53.2	40.9	1.3	5.53	5.70	1.0
31	1.65	1.27	1.3	39.8	31.2	1.3	192.0	159.4	1.2	106.4	87.9	1.2	1.5	1.1	1.4	171.1	149.9	1.1	105.4	97.5	1.1	6.15	6.68	0.9
32	0.29	0.21	1.4	17.2	16.2	1.1	173.5	97.4	1.8	108.8	60.1	1.8	0.8	0.6	1.3	86.9	90.0	1.0	69.5	70.1	1.0	5.96	5.30	1.1
33	0.56	0.21	2.7	22.7	24.1	0.9	110.5	98.0	1.1	71.6	41.8	1.7	0.7	0.4	1.8	103.1	85.1	1.2	66.9	66.4	1.0	4.98	5.46	0.9
34	0.18	0.13	1.4	27.2	33.2	0.8	128.7	93.1	1.4	44.0	36.3	1.2	0.5	0.6	0.8	66.8	70.4	0.9	54.7	57.9	0.9	6.15	4.48	1.4
35	0.45	0.23	2.0	15.7	14.7	1.1	153.7	118.8	1.3	58.8	43.7	1.3	0.6	0.5	1.2	82.1	87.9	0.9	58.5	74.2	0.8	4.16	5.07	0.8
Mean	0.51	0.25	2.5	21.8	16.7	1.4	158.9	118.1	1.4	90.5	52.4	1.7	0.9	0.8	1.2	121.6	94.4	1.3	69.5	70.9	1.0	5.14	5.50	0.9
SD	0.37	0.25	1.6	6.9	5.7	0.4	44.1	27.2	0.3	47.8	19.4	0.4	0.3	0.2	0.3	51.0	29.3	0.3	25.2	27.0	0.1	1.04	1.20	0.2
Max	1.65	1.27	9.4	39.8	33.2	1.2	269.3	181.9	1.5	246.7	109.6	2.3	1.7	1.1	1.5	244.9	155.4	1.6	155.8	158.8	1.0	7.00	8.32	0.8
Min	0.14	0.05	1.0	12.5	4.9	2.6	92.0	68.0	1.4	35.2	22.9	1.5	0.5	0.4	1.3	37.4	35.8	1.0	23.1	23.6	1.0	2.97	2.89	1.0
Forestland soils																								
1	0.08	0.06	1.3	27.9	20.6	1.4	102.9	95.8	1.1	25.1	24.6	1.0	1.0	0.6	1.7	48.3	55.2	0.9	29.9	34.7	0.9	5.24	6.09	0.9
2	0.10	0.13	0.8	28.4	22.1	1.3	105.2	102.5	1.0	26.4	23.7	1.1	1.0	0.9	1.1	33.4	33.7	1.0	21.2	22.2	1.0	5.64	6.25	0.9
3	0.15	0.11	1.3	39.9	20.1	2.0	109.7	97.8	1.1	42.6	38.1	1.1	1.2	0.9	1.3	85.0	127.3	0.7	49.9	61.6	0.8	4.58	5.27	0.9
4	0.10	0.06	1.8	25.2	25.3	1.0	98.0	105.6	0.9	20.5	23.6	0.9	0.8	0.6	1.3	40.6	43.5	0.9	20.4	22.8	0.9	3.70	5.36	0.7
5	0.06	0.06	1.0	22.1	16.9	1.3	89.8	99.2	0.9	31.0	37.3	0.8	0.7	0.7	1.0	91.6	108.6	0.8	52.0	83.2	0.6	5.49	7.02	0.8
6	0.13	0.09	1.4	38.8	16.2	2.4	100.1	87.6	1.1	32.8	30.0	1.1	1.0	0.8	1.3	64.6	70.5	0.9	34.0	43.7	0.8	4.13	5.18	0.8
7	0.15	0.09	1.6	17.5	12.2	1.4	97.0	93.2	1.0	34.0	30.7	1.1	1.0	1.0	1.0	77.9	82.5	0.9	35.3	36.7	1.0	5.62	4.85	1.2
8	0.12	0.11	1.1	45.2	19.9	2.3	99.8	99.6	1.0	23.4	23.1	1.0	1.2	0.7	1.7	35.1	31.7	1.1	17.8	18.1	1.0	3.71	6.35	0.6
9	0.07	0.07	1.0	38.3	12.3	3.1	70.5	66.7	1.1	25.1	90.7	0.3	0.8	0.7	1.1	84.8	89.9	0.9	23.7	40.8	0.6	5.68	7.15	0.8
10	0.08	0.09	0.8	13.9	13.2	1.1	68.0	74.0	0.9	31.5	38.3	0.8	0.8	0.7	1.1	83.5	86.9	1.0	39.4	54.3	0.7	5.26	5.92	0.9
Mean	0.10	0.09	1.2	29.7	17.9	1.7	94.1	92.2	1.0	29.2	36.0	0.9	1.0	0.8	1.3	64.5	73.0	0.9	32.4	41.8	0.8	4.90	5.94	0.8
SD	0.03	0.03	0.3	10.4	4.5	0.7	14.1	12.6	0.1	6.5	20.2	0.3	0.2	0.1	0.3	23.0	32.0	0.1	12.1	20.1	0.1	0.81	0.78	0.2
Max	0.15	0.13	1.8	45.2	25.3	1.8	109.7	105.6	1.0	42.6	90.7	0.5	1.2	1.0	1.2	91.6	127.3	0.7	52.0	83.2	0.6	5.68	7.15	0.8
Min	0.06	0.06	0.8	13.9	12.2	1.1	68.0	66.7	1.0	20.5	23.1	0.9	0.7	0.6	1.2	33.4	31.7	1.1	17.8	18.1	1.0	3.70	4.85	0.8

A: Topsoil, B: Subsoil, C: Topsoil/Subsoil ratio (bold).

Table S3. Principal component analysis (PCA) results of phosphorus and heavy metals in the topsoil collected from citrus orchard sites in Jeju Island, South Korea

Component	PC 1	PC 2
Cd	0.848	0.119
TP	0.841	-0.045
Zn	0.833	0.091
Pb	0.793	0.164
Cu	0.787	0.226
Cr	0.695	0.465
Al	0.065	0.891
Ni	0.153	0.866
Total variance (%)	53.3	18.5
Cumulative variance (%)	53.3	71.8

Extraction method: Principal component analysis

Rotation method: Varimax with Kaiser Normalization

(PC1–principal component 1, PC2–principal component 2)

References

1. NAAS (National Institute Academy of Agricultural Science). *Methods of Soil Chemical Analysis*. Rural Development Administration, Wanjju, Korea. 2010. (in Korean).
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