Supplementary material

Remediation of agricultural soils with long-term contamination of arsenic and copper in two Chilean Mediterranean areas

Pedro Mondaca, Patricio Valenzuela, Nicole Roldán, Waldo Quiroz, Mónika Valdenegro, Juan L. Celis-Diez

Table S1. Chemical characterization of compost.

Parameter*	Unit	Measured value
pH	-	7.6
Electrical conductivity	dS m^{-1}	0.87
Total N	%	2.0
Ammonia nitrogen N-NH$_4^+$	mg kg^{-1}	4.6
Nitric nitrogen N-NO$_3^-$	mg kg^{-1}	45
NH$_4$/NO$_3$ rate	-	0.10
Organic matter content	%	62
Carbon	%	34
C/N rate	-	18
Total P	%	0.25
Total K	%	0.47

* Routinary laboratory analyses performed [1].

Table S2. Soil contamination factors (C_f) of the experimental plots before starting the experiment (n=3).

Trace element	Background concentration (mg kg^{-1})	C_f *			
		Sandy soil plots		Loamy soil plots	
		U-SS	C-SS	U-LS	C-LS
As	13	0.85	1.5	1.0	2.2
Cu	68	0.90	3.6	1.1	9.0

* C_f < 1 = Low contamination; $1 \leq C_f \leq 3$ = moderate contamination; $3 \leq C_f \leq 6$ = considerable contamination; and $6 < C_f$ = very high contamination [2]. Background concentration are based on national report [3]. C-SS = Contaminated sandy soil plot; U-SS = uncontaminated sandy soil plot; C-LS = contaminated loamy soil plot; U-LS = uncontaminated loamy soil plot.

Table S3. Average amendment performance in terms of food safety and investment (n=5).

Parameter	C-SS						C-LS					
	C+	DP	FP	IS	OM	CP	C+	DP	FP	IS	OM	CP
DI of arsenic, mg kg^{-1} bw day^{-1}	7.8E-5	5.6E-5	5.9E-5	6.1E-5	6.6E-5	4.2E-5	9.1E-5	6.3E-5	5.5E-5	8.8E-5	1.1E-4	8.0E-5
HQ through lettuce consumption	0.26	0.19	0.20	0.20	0.22	0.14	0.30	0.21	0.18	0.29	0.35	0.27
Δ HQ (HQ treated – HQ C+)	-	- 0.071	- 0.063	- 0.055	+ 0.037	- 0.12	-	- 0.092	- 0.11	- 0.009	- 0.052	- 0.034
Efficacy, (- (Δ HQ / HQ C+))	-	27%	24%	21%	- 14%	45%	-	31%	39%	3%	17%	11%
Investment, USD/ha	-	≈ 225	≈650	≈ 350	≈ 1000	≈2500	-	≈ 225	≈650	≈ 350	≈ 1000	≈2500
Efficiency, USD/ha per each 1% reduced (Investment/efficacy in percentage)	-	8.3	27	17	-	56		7.2	17	116	59	227

DI = daily intake; HQ = hazard quotient; C-SS = contaminated sandy soil; C-LS = contaminated loamy soil; C+ = contaminated soil without amendment; DP = diammonium phosphate; FP = ferrous phosphate; IS = iron sulfate; OM = organic matter addition; CP= calcium peroxide.

Table S4. Average concentrations ± SD of As and Cu in soils surrounding roots of the sampled lettuces (n=5).

| Treatment | Trace element concentration in soils (mg kg^{-1}) | | | |
| | As | | Cu | |
	C-SS	C-LS	C-SS	C-LS
C+	20.8 ± 1.6 a	28.9 ± 4.2 a	204 ± 11 a	565 ± 9.5 bc
DP	18.9 ± 2.0 a	28.7 ± 2.9 a	197 ± 9.7 a	600 ± 3.0 a
IS	19.2 ± 1.5 a	29.0 ± 0.85 a	210 ± 7.5 a	559 ± 12 bc
FP	21.6 ± 1.3 a	25.4 ± 4.9 a	219 ± 20 a	576 ± 17 abc
CP	20.0 ± 2.3 a	29.7 ± 1.2 a	204 ± 13 a	553 ± 17 c
OM	20.0 ± 1.2 a	28.7 ± 1.2 a	208 ± 17 a	585 ± 17 ab

Different letters indicate significant differences between treatments based on Tukey multiple comparison test (p ≤ 0.05). C+ = contaminated soil without amendment; DP = diammonium phosphate; FP = ferrous phosphate; IS = iron sulphate; OM = organic matter addition; CP= calcium peroxide. C-SS = Contaminated sandy soil plot; U-SS = uncontaminated sandy soil plot; C-LS = contaminated loamy soil plot; U-LS = uncontaminated loamy soil plot.

Table S5. Mantel test applied to soil As and Cu concentration in function of the spatial distance of the contaminated plots.

Trace element	Background concentration (mg kg^{-1})	Distance				
		C-SS		C-LS		
		r	p-value	r	p-value	
As	13	0.0099	0.42	-0.024	0.62	
Cu	68	-0.015	0.57	-0.0058	0.51	

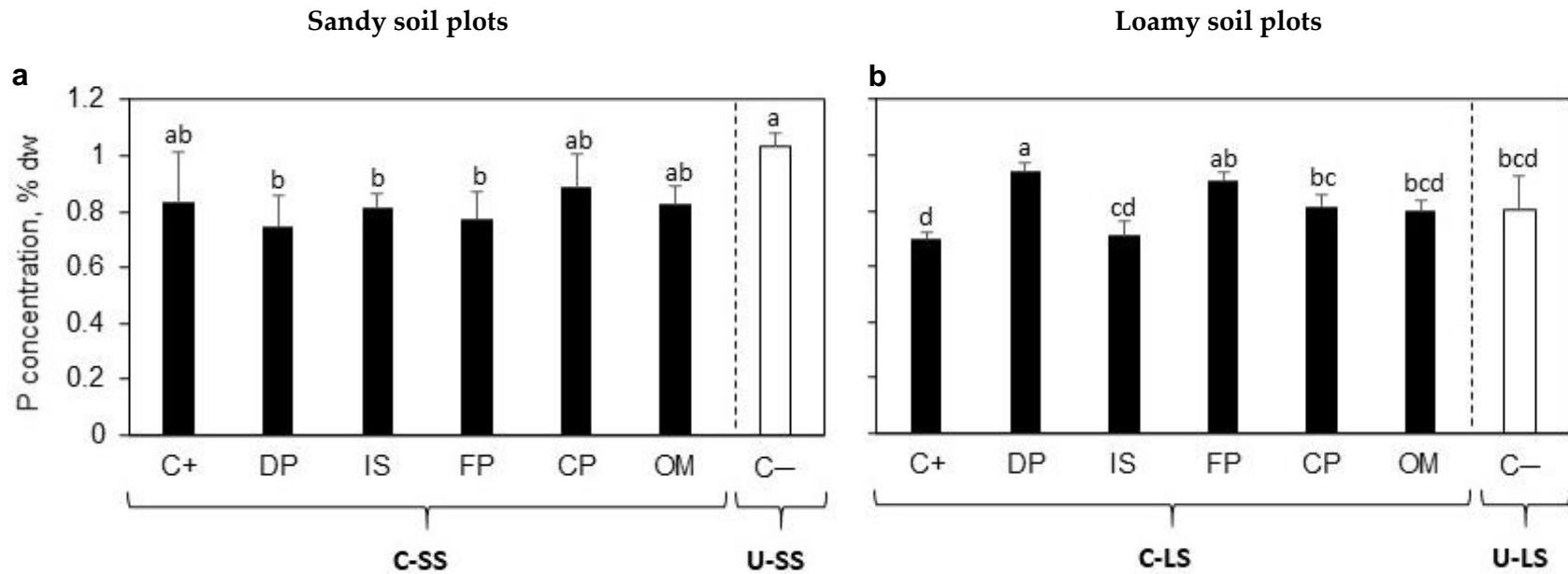**Equation S1.** Calculation of Concentration factor.

$$C_f = \frac{\text{Trace element concentration in the studied soils}}{\text{Trace element background concentration}}$$

Figure S1. Experimental plots. C-SS = Contaminated sandy soil plot; U-SS = uncontaminated sandy soil plot; C-LS = contaminated loamy soil plot; and U-LS = uncontaminated loamy soil plot. Pictures were taken by Christopher Ulriksen.

Sandy soil plots   **Loamy soil plots**
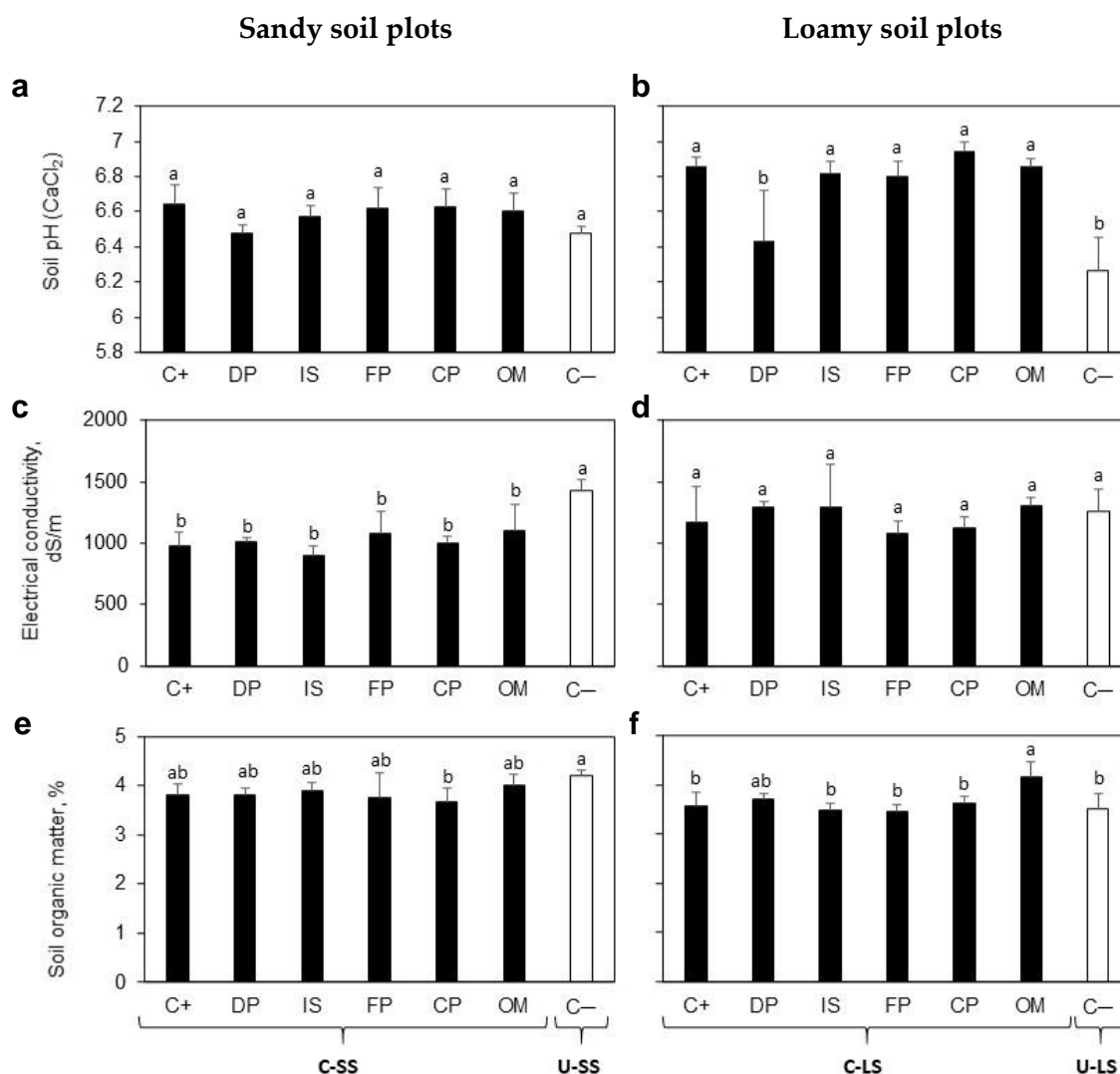
Figure S2. Chemical parameters of soils surrounding roots of lettuces. (a), (c), and (e) are the results of lettuces grown on sandy soils, while (b), (d), and (f) are those for lettuce grown on loamy soils. Bars are means ± SD (n = 5). Different letters above the means indicate significant differences between treatments based on Tukey multiple comparison test (p ≤ 0.05). Black bars represent contaminated soil under different treatments: C+ = contaminated soil without amendment; DP = diammonium phosphate; FP = ferrous phosphate; IS = iron sulphate; OM = organic matter addition; CP= calcium peroxide. White bar represents uncontaminated soil: C- = uncontaminated soil without amendment. C-SS = Contaminated sandy soil plot; U-SS = uncontaminated sandy soil plot; C-LS = contaminated loamy soil plot; U-LS = uncontaminated loamy soil plot.

Figure S3. Phosphorus concentration in the edible tissues of lettuces (on a dry weight basis) grown on (a) sandy soils and (b) loamy soils. Bars are means ± SD (n = 5). Different letters above the means indicate significant differences between treatments based on Tukey multiple comparison test (p ≤ 0.05). Black bars represent contaminated soil under different treatments: C+ = contaminated soil without amendment; DP = diammonium phosphate; FP = ferrous phosphate; IS = iron sulfate; OM = organic matter addition; CP= calcium peroxide. White bar represents uncontaminated soil: C- = uncontaminated soil without amendment. C-SS = contaminated sandy soil plot; U-SS = uncontaminated sandy soil plot; C-LS = contaminated loamy soil plot; and U-LS = uncontaminated loamy soil plot.
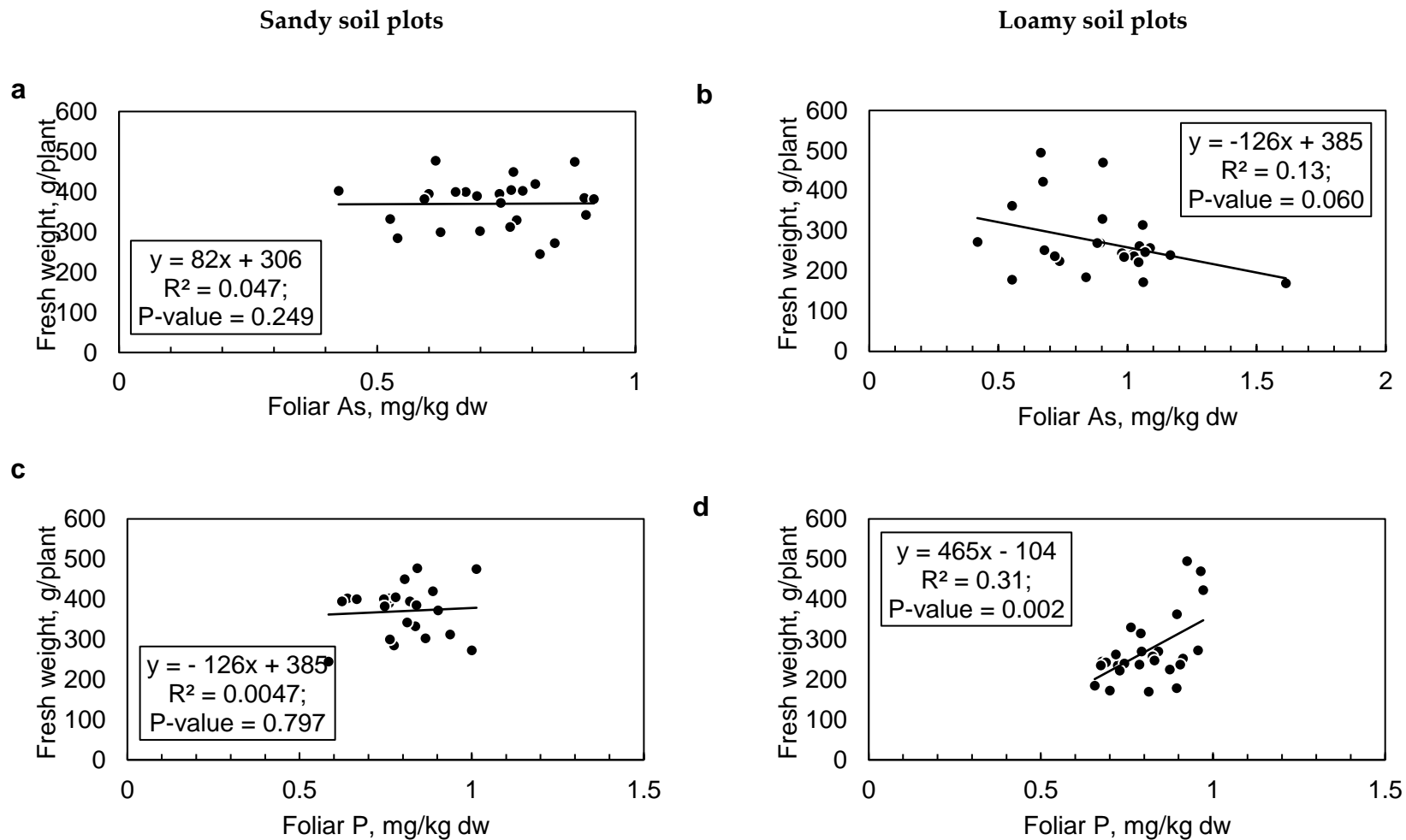
Figure S4. Scatterplot between the fresh weight of lettuces and the As and P concentration in their leaves. (a) and (c) are the results of lettuces grown on sandy soils, while (b), and (d) are those for lettuce grown on loamy soils.

Supplementary references

1. Sadzawka, A.; Carrasco, M.A.; Grez, R.; Mora, M. Métodos de análisis de compost; Instituto de Investigaciones Agropecuarias: Santiago, Chile, 2005.

2. Hakanson, L. An ecological risk index for aquatic pollution control. A sedimentological approach. Water Research 1980, 14, 975-1001, doi:10.1016/0043-1354(80)90143-8.

3. PGS. Muestreo de suelos para las comunas de Quintero y Puchuncaví, Región de Valparaíso; PGS: Santiago, Chile, 2015.