

# Assessing soil contamination and temporal trends of heavy metal contents in greenhouses on semiarid land

Carlos Gil<sup>1</sup>  | Rafael Boluda<sup>2</sup>  | José Antonio Rodríguez Martín<sup>3</sup>  | Miguel Guzmán<sup>1</sup>  | Fernando del Moral<sup>1</sup>  | Joaquín Ramos-Miras<sup>1</sup> 

<sup>1</sup>Departamento Agronomía: cei A3; CIEMBITAL, Escuela Superior de Ingeniería, Universidad de Almería, La Cañada 04120, Spain

<sup>2</sup>Departamento Biología Vegetal, Facultat de Farmacia, Universitat de Valencia, Burjassot 46100, Spain

<sup>3</sup>Departamento Medio Ambiente, Instituto Nacional de Investigación y Tecnología Agraria y Alimentaria (I.N.I.A), Madrid 28040, Spain

## Correspondence

Carlos Gil, Área de Edafología y Química Agrícola, Escuela Superior de Ingeniería, Univ. de Almería, cei A3. Ctra. de Sacramento s/n, 04120 La Cañada (Almería), Spain.  
Email: cgil@ual.es

## Funding information

Spanish Ministry of Science and Innovation, Grant/Award Numbers: AGL2011-29382 and CGL2013-43675-P

## Abstract

Information about the behavior and temporal evolution of heavy metals in agricultural soils is limited, particularly about greenhouse soils on semiarid lands, indeed it nonexistent. Western Almería (Southern Spain) is a semiarid land where some 30,000 ha are occupied by greenhouses with high productivity. As these greenhouses are fundamental to the socioeconomic development of this area, they should be maintained and well conserved. However, there are indications that long-term intensive agriculture with considerable agrochemicals use can deteriorate soil quality, which in turn, would reduce productivity and food quality. This study was conducted to investigate soil contamination and the temporal trends of heavy metal concentrations in greenhouse soils of western Almería. Contamination level, availability, and sources of metals were evaluated by the extractable fraction percentage, by indices zinc equivalent, geoaccumulation, enrichment factor and pollution load, and by a correlation analysis between soil properties and metal contents. The results showed that the total contents of Cd, Cu, Ni, and Pb, and the available concentration of Cd, Cu, Pb, and Zn, were significantly higher than background levels. Temporal patterns indicated that these elements accumulate in greenhouse soils. After more than 20 years of intensive agriculture, the available concentration of elements, and contamination, had clearly increased.

## KEYWORDS

agrochemicals, contamination indices, heavy metal enriched, Horticultural greenhouse impact, trace elements

## 1 | INTRODUCTION

Soil heavy metal (HM) pollution are related to different sources of pollution, such as such as mining (Mirzaei, Ghorbani, HafeziMoghaddas, & Martín, 2014; Odumo et al., 2014; Rodríguez Martín et al., 2014; Sakizadeh, Mirzaei, & Ghorbani, 2015), industry, and energy production (Gutiérrez et al., 2016; Hou, O'Connor, Nathanail, Tian, & Ma, 2017; Rodríguez Martín & Nanos, 2016), which results in very high concentrations in the soil. However, the contamination by HMs takes greater importance in cultivated soils, although the contents in soils are smaller than in industrial soils (Shao, Zhan, Zhou, & Zhu, 2016; Tianlik, Norulaini, Shahadat, Yoonsing, & Omar,

2016). Soil is one of the most important reservoirs of HMs and one of the principal routes of metals uptake in crops. High-intensity cropping can increase the HM concentration in soil and water, which could affect the content of HM in plants (Li, Shi, Jin, Wu, & Sheng, 2017; Zhang et al., 2017). It can potentially affect human health; metals such as Pb and Cd can enter soil via the food chain, exceed normal limits, and have harmful effects (Roy & McDonald, 2015).

The intensification of certain agricultural practices, the continued or excessive use of fertilizers and pesticides, as well as machinery, all increase HM contents in agricultural soils (Ding et al., 2017; Khaledian, Pereira, Brevik, Pundyte, & Paliulis, 2017; Long, Wang, & Da, 2013; Ramos-Miras, Roca-Pérez, Guzmán-Palomino, Boluda, & Gil, 2011).

Several studies have revealed that greenhouse intensive-production practices increase the accumulation of trace metals in soils, particularly Cd, Cu, Hg, and Zn, which has been related with agrochemical inputs (Gil, Ramos-Miras, Roca-Pérez, & Boluda, 2010; Ramos-Miras, Roca-Pérez, Boluda, & Gil, 2012; Rodríguez-Martín, Ramos-Miras, Boluda, & Gil, 2013). Others have shown evidences about the behaviour of HM over time (Álvarez-Ayuso, García-Sánchez, Querol, & Moyano, 2008; Chen, Tian, Gao, & Tian, 2016; Kochem-Mallmann et al., 2012; Long et al., 2013; Ordoñez, Alvarez, De Miguel, & Charlesworth, 2015), but information on this topic in greenhouse soils (GSs) is scarce, and no other information except for Ramos-Miras et al. (2012) on Hg have been found about GS in semiarid lands in the Mediterranean region.

The western Almería region is a very important semiarid land of south Spain from both socioeconomic and agricultural viewpoints. The zone is intensively cultivated but not industrialized. Agriculture is based on greenhouses, wherein soils have been vastly altered by the intense agricultural practices to which they are submitted. The average yearly consumption of fertilizers in western Almería is about 2,200 kg ha<sup>-1</sup> and can even reach 3,400 kg ha<sup>-1</sup> for some crops, whereas the yearly consumption of pesticides is about 150 kg or 150 L ha<sup>-1</sup> (Gil, Boluda, & Ramos, 2004; Plaza-Bolaños, Padilla-Sánchez, Garrido-Freniche, Romero-González, & Martínez-Vidal, 2012), and there is evidence that this intense agricultural activity is having a very strong impact on soils and on the environment (Plaza-Bolaños et al., 2012).

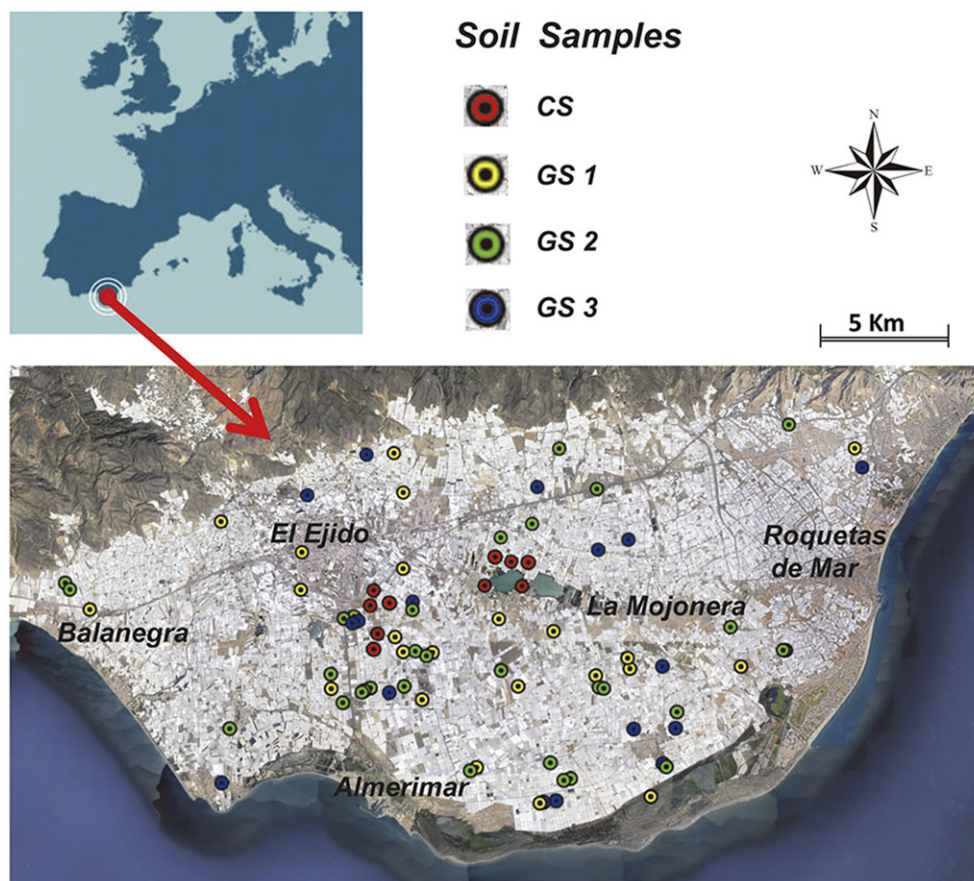
For these reasons, this work aimed to (a) determine soil properties, total contents, and the extractable concentrations with

ethylenediaminetetraacetic acid (EDTA) of Cd, Co, Cu, Fe, Ni, Pb, and Zn, in 71 GS from a traditionally Mediterranean agricultural area of semiarid land (western Almería, South Spain); (b) assess soil contamination; (c) establish relationships between metal contents and soil properties; and (d) assess temporal trends after three periods (<10, 10–20, and >20 years) of intensive cultivation. The results were compared with the background levels obtained for soils in the same area. Different indices have been used to assess soil pollution. Some provide a simple, comparative means for assessing the level of HM pollution in soils. Indices are proposed by Andreu and Boluda (1995), Ramos-Miras et al. (2011), and Tamim et al. (2016), among others. Thus, metal available fraction percentage (A), zinc equivalent (ZnEq), geoaccumulation index (Igeo), enrichment factor (EF), contamination factor (CF), and pollution load index (PLI) were used as methods to assess soil contamination.

## 2 | MATERIALS AND METHODS

### 2.1 | Study area and sampling

The study area (330 km<sup>2</sup> in a coastal lying area with about 30,000 ha occupied by greenhouses) is located to the southeast of Spain (Figure 1). The predominant crops in the area are fruits and vegetables: pepper, tomato, zucchini, cucumber, melon, eggplant, and watermelon. The soil of most greenhouses could be classified as Technosols (IUSS Working Group WRB, 2015) and could be



**FIGURE 1** Location of area and for sampling soil points. CS: control soil; GS: greenhouse soil [Colour figure can be viewed at [wileyonlinelibrary.com](https://onlinelibrary.wiley.com)]

described as an artificially layered soil, with commonly three layers over the natural substrate (Figure 2): sand layer (10–15 cm), organic matter layer (manure, 5–10 cm), and a clay layer (CL), brought in origin from quarries and commonly known as “Tierra de Cañada” (25–50 cm). CL is important because it is involved in water storage, nutrient retention, and contaminant fixing, on which fertilizers were supplied to the GS and therefore where toxic elements could be accumulated. For these reasons, CL was sampled and analyzed. Its features are calcareous nature, colluvium–alluvium origin, red to red-dish brown, and loam to clayed.

Seventy-one GS samples were selected to study soils. To obtain true reference values (control soils [CS] samples), 11 quarry samples were collected too as CS. These quarry soils have similar edaphic characteristics, and all the GS studied were built up with these similar sediments. The background level is calculated as the mean value of these 11 CS. Figure 1 illustrates the location of the 82 sampling points. Random subsampling was carried out at each site to obtain five subsamples, which were mixed and homogenized to form a single compound sample, which was analyzed.

Compound samples were then grouped according to their cropping age as follows: 24 samples from GS used for less than 10 years (GS1), 29 GS used for 10–20 years (GS2), and 18 samples from GS used for more than 20 years (GS3).

## 2.2 | Analytical methods

All the samples were air-dried, sieved with a 2-mm grid sieve, and stored in hermetically sealed polyethylene bags until analyzed. Standard soil analyses were carried out according to the methods referenced by Gil et al. (2004), Ramos-Miras et al. (2011), and Rodríguez-Martín et al. (2013). Three soil granulometric fractions (sand, silt, and clay) were determined by the pipette method. Soil pH was measured in a 1:5 (soil:distilled water) extract. Soil organic matter (SOM) was analyzed by the Walkley–Black method and carbonate concentration by gas volumetric method. Available P was established following the Olsen method. Determination of HM total content was performed using hydrofluoric and nitric acid digestion in a microwave

oven Milestone 1200 (Milestone Inc, CT USA) following the EPA 3052 procedure reported by Kingston and Walter (1995) using a sample of 0.5 g. A reference material was used (BCR-141, calcareous loamy soil, sample no. 00051; BCR, Brussels, Belgium) to assess the method's accuracy. EDTA-extractable content was carried out by the Quevauviller et al. (1994) procedure, which estimates the available fraction for plants in calcareous soils (Ramos-Miras et al., 2011; Zeng et al., 2011). Five grams of soil was extracted with 50 ml of EDTA 0.05 M, pH = 7.

The metal concentrations in digested and EDTA extracts were measured in atomic absorption spectrophotometer equipment GBC-906AA (GBC, Hampshire, IL, USA). Matrix interferences were checked by standard addition techniques; no matrix interferences were observed for Cd, Co, Cu, Fe, Ni, Pb, and Zn. The blank sample and the certified reference material were extracted and analyzed with each batch of 15 samples. Good recoveries were obtained: 96% Cd, 103% Co, 92% Cu, 97% Fe, 94% Ni, 110% Pb, and 102% Zn. Three replicates were analyzed per sample to check the accuracy of the results. The determined analytical variations were lower than 10% for all the metals. The limit of quantification ranged between 0.07 (Cd) and 32 mg kg<sup>-1</sup> (Fe). All the analyzed metals were above the limit of quantification. The concentrations of elements are presented as mg kg<sup>-1</sup> dry matter.

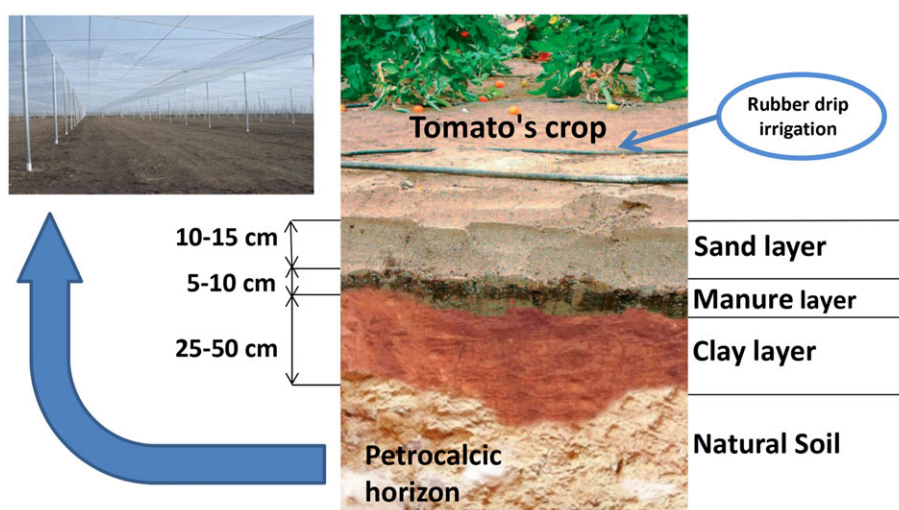
## 2.3 | Quantification of soil contamination

The metal available A, ZnEq, Igeo, EF, CF, and PLI were used as methods to assess GS contamination.

Following Andreu and Boluda (1995); Gimeno-García, Andreu, and Boluda (1996); Gil et al. (2004); and Ramos-Miras et al. (2011), A and ZnEq indices were calculated and applied to provide a basis to compare the potential HM mobility and toxicity in soils in accordance with expressions (1) and (2):

$$A = (MA/MT) \times 100, \quad (1)$$

where MA is the EDTA-extractable metal concentration, MT is the total metal content.



**FIGURE 2** Schematic representation showing soil profile of a greenhouse in Almería [Colour figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

$$\text{ZnEq} = [\text{Zn}] + 2 \times [\text{Cu} + 8 \times [\text{Ni}]], \quad (2)$$

where [Zn], [Cu], and [Ni] are the respective total concentrations of metals in soils.

Igeo, EF, CF, and PLI to CS and GS were calculated and interpreted according to Müller (1969); Bloundi, Duplay, and Quaranta (2009); Zhang, Tang, Zhang, and Wu (2009); and Tamim et al. (2016). In our study, we used the metal abundances of the CS, as the background data, and iron, as the commonest reference element. Expressions (3), (4), (5), and (6) were used, respectively, in each case:

$$\text{Igeo} = \text{Log}_2[\text{C}_{\text{GS}} / (1.5 \times \text{C}_{\text{CS}})], \quad (3)$$

$$\text{EF} = (\text{C}_{\text{GS}} / \text{C}_{\text{CS}}) / (\text{Fe}_{\text{GS}} / \text{Fe}_{\text{CS}}), \quad (4)$$

$$\text{CF} = \text{C}_{\text{GS}} / \text{C}_{\text{CS}}, \quad (5)$$

$$\text{PLI} = (\text{CF}_1 \times \text{CF}_2 \times \text{CF}_3 \times \dots \times \text{CF}_n)^{1/n}, \quad (6)$$

where  $\text{C}_{\text{GS}}$  is the metal content in the tested soil (GS),  $\text{C}_{\text{CS}}$  is the metal concentration in the control soils (CS, background level),  $\text{Fe}_{\text{GS}}$  and  $\text{Fe}_{\text{CS}}$  are the iron concentrations in the GS and the CS, respectively,  $\text{CF}_1$  to  $\text{CF}_n$  represent the contamination factors for the specific toxic metals, and  $n$  is the total number of considered contamination factors; in our case,  $n = 6$  (excludes Fe).

## 2.4 | Statistical analyses

All the statistical analyses were performed using the SPSS 15.0 software. The results showed a mean value, ranges (min–max), standard deviation, and Spearman's correlation analysis. Mann–Whitney U test and Kruskal–Wallis test, followed by Scheffe's post hoc test, were used to test for any significant differences in the HM between the CS and the considered GS. The confidence interval for the student's  $t$  test was calculated at  $\alpha = 0.05$ .

## 3 | RESULTS AND DISCUSSION

Soil characteristics and metal concentrations, together as the main descriptive statistics for the CL of GS in western Almería, are listed in Tables 1 and 2. In general, the GS had a high clay content, slightly basic, moderate fertility (lower SOM and cationic exchange capacity),

were highly calcareous with some salinity problems, which is in accordance with that previously reported by Gil et al. (2004).

### 3.1 | Total and available metal contents in soils

Table 2 summarizes the main descriptive statistics related to the total and available metal contents in both the CS (background values) and GS. Table 2 also includes the background levels obtained by Gil et al. (2004), Sierra et al. (2007), and Ramos-Miras et al. (2011) and the baselines proposed for Almería soils by Aguilar et al. (2005). Our background levels were like those obtained by Gil et al. (2004) and Ramos-Miras et al. (2011), and all, except Zn, were lower than the baseline values proposed by Aguilar et al. (2005) for soils in Almería. The levels in GS of Cd, Cu, Pb, and Ni were significantly higher than the CS, whereas the levels of Zn and Co were similar among CS and GS. When comparing the mean total concentration values, they were generally 1.5- to 2.0-fold higher CS than GS. The levels obtained for the GS were higher than those described by Rodríguez-Martín, López-Arias, and Grau-Corbí (2009) for the whole Spanish territory and higher than those described for farming soil (Andreu & Boluda, 1995; Gimeno-García et al., 1996), whereas the CS were lower than the background levels proposed for Almería (Sierra et al., 2007) and Andalusia (Aguilar et al., 2005), except for Cd and Zn levels, which were higher in the CS. The concentrations of the HM in the CS were significantly lower than in the GS, except for Zn and Co, which were similar (Table 2A). The origin of these two elements could be endogenous; this fact explained that the levels of Zn and Co were similar in both GS and CS (Magrisso, Belkin, & Errel, 2009).

Our results revealed a significant increase in the available concentrations of Cd, Cu, Pb, Ni, and Zn in GS compared with the CS, which were 3-, 8-, 4-, 2-, and 5-fold higher, respectively (Table 2). The available levels were in the same order as or higher than, those reported for Spanish agricultural soils (Andreu & Boluda, 1995; Gimeno-García et al., 1996; Ramos-Miras et al., 2011), and were similar to those previously described by Ramos-Miras et al. (2011).

Table 2B provides the extractable fraction percentage values for the studied elements and confirms increased availability (A), where Cd, Pb, Zn, and Cu are 1.6-, 3-, 6-, and 6-fold higher in the GS compared with the CS, respectively. The available fraction percentage (A) is an indicator of its comparative mobility (Boluda, Andreu, Gilabert, & Sobrino, 1993; Ramos-Miras et al., 2011) and is an appropriate indicator for recent soil pollution history (Massas, Ehaliotis, Gerontidis, & Sarris, 2009). The A values are Cd, 11.0–62.0; Co, 3.8–25.0; Cu, 4.0–79.5; Ni, 0.8–12.0; Pb, 10.6–81.5; and Zn, 1.1–20.2. The order

**TABLE 1** Main descriptive parameters for GS soils ( $n = 71$ )

	Clay (%)	SOM (%)	CEC (cmol <sub>c</sub> kg <sup>-1</sup> )	pH	ESP (%)	EC (dSm <sup>-1</sup> )	P <sub>2</sub> O <sub>5</sub> (mg 100 g <sup>-1</sup> )	CaCO <sub>3</sub> (%)
min	13.6	0.3	4.6	7.6	1.0	0.7	4.9	5.6
Mean	32.7	1.0	9.9	8.3	12.0	2.0	25.3	31.6
max	57.1	3.0	15.5	8.9	40.0	5.2	79.3	64.9
SD	9.15	0.5	6.6	0.3	6.0	1.0	20.6	15.3

Note. SOM: soil organic matter; CEC: cationic exchange capacity; ESP: exchangeable sodium percentage; EC: electric conductivity; P<sub>2</sub>O<sub>5</sub>: available phosphorus; min: minimum value; Mean: mean value; max: maximum value; SD: standard deviation.

**TABLE 2** Main descriptive statistics for metals in CS ( $n = 11$ ) and GS ( $n = 71$ ), mg kg<sup>-1</sup> dry weight

A	CS				GS				C	BA
	min	Mean	max	SD	min	Mean	max	SD		
CdT	0.46	0.63 <sub>a</sub>	0.80	0.10	0.71	1.27 <sub>b</sub>	1.89	0.27	0.6 <sup>a</sup>	0.3
CdA	0.11	0.13 <sub>a</sub>	0.16	0.02	0.15	0.41 <sub>b</sub>	0.80	0.12	0.4 <sup>c</sup>	Na
CuT	9.9	16.4 <sub>a</sub>	27.1	5.7	9.5	28.7 <sub>b</sub>	67.1	9.7	21.2 <sup>b</sup>	47.2
CuA	0.2	0.8 <sub>a</sub>	1.2	0.3	0.9	6.8 <sub>b</sub>	41.9	7.4	9.5 <sup>c</sup>	Na
PbT	15.3	49.3 <sub>a</sub>	74.2	20.4	18.0	75.4 <sub>b</sub>	404.9	52.1	46.2 <sup>a</sup>	93.9
PbA	1.2	6.0 <sub>a</sub>	12.2	3.8	5.8	24.3 <sub>b</sub>	171.4	25.5	26.3 <sup>c</sup>	Na
ZnT	60.1	158.5 <sub>a</sub>	213.4	58.1	63.4	156.6 <sub>a</sub>	374.7	72.2	135.7 <sup>b</sup>	129
ZnA	0.2	2.1 <sub>a</sub>	4.4	1.4	1.6	11.5 <sub>b</sub>	53.8	9.4	11.8 <sup>c</sup>	Na
NiT	17.6	26.5 <sub>a</sub>	37.7	6.2	19.7	39.6 <sub>b</sub>	53.6	8.1	23.4 <sup>a</sup>	70.9
NiA	0.1	0.8 <sub>a</sub>	1.4	0.5	0.4	1.7 <sub>b</sub>	3.5	0.5	1.7 <sup>c</sup>	Na
CoT	9.8	14.4 <sub>a</sub>	18.1	3.0	12.1	17.1 <sub>a</sub>	23.3	2.3	13.7 <sup>b</sup>	26.7
CoA	0.2	1.0 <sub>a</sub>	2.6	1.0	0.6	1.6 <sub>a</sub>	4.6	0.7	1.7 <sup>c</sup>	Na

B	CS				GS			
	min	Mean	max	SD	min	Mean	max	SD
Cd	15.7	20.8 <sub>a</sub>	25.8	3.9	11.0	33.7 <sub>b</sub>	62.0	11.7
Cu	2.2	3.9 <sub>a</sub>	5.2	1.1	4.0	21.9 <sub>b</sub>	79.5	15.7
Pb	7.6	10.4 <sub>a</sub>	16.5	10.8	10.0	30.5 <sub>b</sub>	81.5	10.8
Zn	0.3	1.3 <sub>a</sub>	2.5	0.8	1.1	7.6 <sub>b</sub>	20.2	4.8
Ni	1.9	3.2 <sub>a</sub>	5.0	2.2	0.8	4.6 <sub>a</sub>	12.0	2.0
Co	1.4	8.2 <sub>a</sub>	22.7	8.5	3.8	9.5 <sub>a</sub>	25.0	4.0

Note. Panel A: total content (-T) and available content (-A). Panel B: available fraction percentage. Different letter indicates statistically significant differences ( $p < 0.05$ ) after Mann-Whitney U test. CS: control soil; GS: greenhouse soil; min: minimum value; Mean: mean value; max: maximum value; SD: standard deviation; C: comparison with background levels; BA: baselines proposed for Almería by Aguilar et al. (2005); Na: not available.

<sup>a</sup>Comparison with background levels by Gil et al. (2004).

<sup>b</sup>Comparison with background levels by Sierra, Martínez, and Aguilar (2007).

<sup>c</sup>Comparison with background levels by Ramos-Miras et al. (2011).

of available fraction percentage among the analyzed samples was Cd > Pb > Cu > Co > Zn > Ni. This result indicated that the most toxic elements were also the most mobile ones and also revealed that except for Co and Ni, they were significantly higher in the GS than in the CS. Moreover, the value of these indices for the GS were in the same order as those described by several authors for Mediterranean farm soils, whereas the CS gave lower values, save Co and Ni (Andreu & Boluda, 1995; Ramos-Miras et al., 2011). According to Tarvainen and Kallio (2002), levels of the available fraction index below 5% indicate noncontaminated soils. Our results revealed that all save Co, the mean A values in the GS were over 5%, which was very possibly due to the excessive use of agrochemicals, the irrigation system, and its high frequency.

The above described facts suggest that intensive agriculture that greenhouses undergo favored the accumulation and availability of HM in soils, which has been previously described by Gil et al. (2004), Ramos-Miras et al. (2011), and Rodríguez-Martín et al. (2013). Long et al. (2013) assessed the spatial-temporal variations of HM in farmland soils in Shanghai (China) to find that Cd, Zn, and Hg contributed the most pollution. Recently, Zhang et al. (2017) found that Cd, Cu, Hg, and Zn had clearly accumulated in GS in southwest China, which favors their enrichment in vegetable crops due to high-intensity

cropping and management practices. Li et al. (2017) found that greenhouse cultivation greatly enhanced the bioconcentration of most metals from soil to plant roots.

### 3.2 | Assessing soil contamination

The concentrations of Cd, Cu, and Pb exceed our background levels. The high concentrations of Cd and Cu may be due to abusive use of fertilizers; the high Pb contents observed in GS must be similarly related to the greenhouse activities carried out by means of farming machinery and emissions deriving from burning fuel within a confined area (Rodríguez-Martín et al., 2013). Moreover, when using the mean CS value criterion plus twice the standard deviation as a reference level, 66 GS (93%) exceeded this value for CdT, as did 47 GS (66%) for CuT, whereas three (4%) and 60 (85%) GS, respectively, exceeded these values when using the baselines proposed by Aguilar et al. (2005) and the criterion of Sierra et al. (2007). This demonstrates the importance of using the obtained geochemical levels, within the same area, to more suitably assess contamination status in soil.

Indices ZnEq, Igeo, EF, and PLI (Tables 3 and 4) confirmed the potential risk of contamination. The Igeo, EF, and PLI values were higher for Cd, Cu, and Pb. Major differences were found when

**TABLE 3** Contamination indexes

Index	Element	min	Mean	max	SD
ZnEq	(Zn, Cu, Ni) T	266	531	851	116
	(Zn, Cu, Ni) E	14	38	111	20
Igeo	CdT	-0.4	0.4	1.0	0.3
	CdE	-0.4	1.0	2.0	0.5
	CoT	-0.8	-0.3	0.1	0.2
	CoE	-0.9	0.5	2.1	0.6
	CuT	-1.2	0.3	1.6	0.5
	CuE	-0.2	2.3	5.4	1.1
	NiT	-1.0	0.0	0.5	0.3
	NiE	-1.1	0.8	2.0	0.5
	PbT	-1.9	-0.1	2.6	0.7
	PbE	-0.4	1.3	4.5	0.9
	ZnT	-1.8	-0.7	0.7	0.6
	ZnE	-1.0	1.5	4.1	1.0
	MULT EL T	-1.1	-0.1	0.5	0.3
	MULT EL E	0.5	1.2	2.2	0.4
EF	CdT	0.9	2.2	5.1	0.9
	CdE	0.2	3.4	14.9	2.7
	CoT	0.6	1.3	2.4	0.5
	CoE	0.1	2.3	10.1	1.8
	CuT	0.5	2.2	5.3	1.0
	CuE	1.1	8.0	38.0	7.0
	NiT	0.4	1.7	3.4	0.7
	NiE	0.5	2.8	10.9	2.0
	PbT	0.3	1.9	16.7	2.0
	PbE	0.3	4.7	38.5	5.5
	ZnT	0.2	1.2	3.7	0.8
	ZnE	0.2	4.5	18.0	3.2
	MULT EL T	0.6	1.8	5.5	0.8
	MULT EL E	0.5	4.3	12.3	2.5
PLI	CdT	1.1	2.0	3.0	0.4
	CdE	1.1	3.2	6.1	0.9
	CoT	0.8	1.2	1.6	0.2
	CoE	0.8	2.3	6.6	1.0
	CuT	0.6	2.0	4.6	0.7
	CuE	1.3	10.1	62.6	10.4
	NiT	0.8	1.5	2.1	0.3
	NiE	0.7	2.8	5.9	0.9
	PbT	0.4	1.7	8.9	1.1
	PbE	1.1	4.8	33.6	5.0
	ZnT	0.4	1.0	2.5	0.5
	ZnE	0.7	5.2	24.5	4.3
	MULT EL T	0.7	1.5	2.2	0.3
	MULT EL E	2.1	3.7	7.1	1.1

Note. min: minimum value; Mean: mean value; max: maximum value; SD: standard deviation; Igeo: geoaccumulation index; EF: enrichment factor; PLI: potential load index.

comparing the values obtained from the total (T) concentration or the available fraction (E: EDTA-extractable content), and the indices gave higher values when the available concentration is employed to calculate them. According to ZnEq-T only 35% of the GS (25 samples) have toxicity problems; however, ZnEq-E showed that 97% of the GS (69 samples) may have toxicity problems (Table 4).

Based on previously adopted criteria (Tamim et al., 2016) and on the available concentration for Igeo calculations, when total concentration was applied, 56% GS were considered practically uncontaminated (Class 0), and 44% were uncontaminated to moderately contaminated (Class 1).

The EF index related the geochemical origin of the element, its mobilization and the anthropogenic nature of its abundance (Tamim et al., 2016). The highest EF values were the same as in the previous case for available metals: for CuE (EF = 8.0), PbE (EF = 4.7), ZnE (EF = 4.5), and CdE (EF = 3.4). Five GS groups were established according to degree of modification: nonmodification (10%), minor

**TABLE 4** Results after soil contamination indices interpretation (n, numbers of GS affected; %, percentage of GS affected)

Index	Interpretation	Total metal (T)		Extractable metal (E)	
		n	%	n	%
ZnEq <sup>a</sup>	No contaminated soil	46	65	2	3
	Soil with toxicity problems	25	35	69	97
Igeo	Class 0 (practically uncontaminated)	40	56	0	0
	Class 1 (uncontaminated to moderately contaminated)	31	44	24	34
	Class 2 (moderately contaminated)	0	0	45	63
	Class 3 (moderately to heavily contaminated)	0	0	2	3
EF	Nonmodification (<1.5)	28	40	7	10
	Minor modification (1.5–3.0)	40	56	17	24
	Moderate modification (3.0–5.0)	2	4	21	30
	Severe modification (5.0–10)	0	0	23	32
	Very severe modification (>10)	0	0	3	4
PLI	No deterioration (PLI ≤ 1)	3	4	0	0
	Progressive deterioration (PLI > 1)	68	96	71	100

Note. Igeo: geoaccumulation index; EF: enrichment factor; PLI: potential load index; ZnEq: zinc equivalent; SD: standard deviation.

<sup>a</sup>ZnEq-T: no contaminated soil (ZnEq-T ≤ 580), soil toxicity (ZnEq-T > 580); ZnEq-E: no contaminated soil (ZnEq-E ≤ 17), soil toxicity (ZnEq-E > 17). Reference value calculated as background level + 2 × SD.

modification (24%), moderate modification (30%), severe modification (32%), and very severe modification (4%). A higher EF value than the unit meant that the element was anthropogenic in nature (Tamim et al., 2016). When total concentration was used, 14 GS obtained an EF ≤ 1, whereas only two GS obtained this same value using the available concentration. For EF-E, 23 GS presented severe modification, whereas no GS was classified as such when total concentration was used. The PLI indices suggested that more than 97% of GS showed progressive deterioration.

All these results confirmed that the total concentration of an element or using a single index was not sufficiently reliable to properly assess soil pollution. Differences were found in interpretations depending on the adopted criterion. In any case, all these results confirmed that a large proportion of GSs in the study area could have soils that present adverse effects. The potential risk contemplated jointly by Cd, Cu, and Pb was confirmed by the indices employed herein to generally assess soil pollution status, which agrees with several other authors (Bloundi et al., 2009; Tamim et al., 2016; Zhang et al., 2009; Zhang et al., 2017) and demonstrates their effectiveness.

### 3.3 | Assessing relationship with soil parameters

Table 5 showed the correlation analyses to determine the extent of the relationship between the parameters investigated. SOM and carbonate contents are the main properties that show a higher affinity

**TABLE 5** Correlation coefficients among elemental concentrations and soil properties in GS ( $n = 71$ )

	Clay	SOM	CEC	ESP	pH	P <sub>2</sub> O <sub>5</sub>	CaCO <sub>3</sub>	CdT	CdE										
Clay	1																		
SOM	n.s.	1																	
CEC	0.421**	n.s.	1																
ESP	0.304**	n.s.	n.s.	1															
pH	n.s.	n.s.	n.s.	n.s.	1														
P <sub>2</sub> O <sub>5</sub>	n.s.	0.576**	n.s.	-0.301**	n.s.	1													
CaCO <sub>3</sub>	0.299**	n.s.	n.s.	n.s.	n.s.	n.s.	1												
CdT	n.s.	0.449**	n.s.	-0.408**	n.s.	0.458**	0.250*	1											
CdE	n.s.	0.386**	n.s.	n.s.	n.s.	n.s.	0.435**	0.363**	1										
CuT	n.s.	0.313**	n.s.	n.s.	n.s.	0.441**	0.228*	0.484**	0.323**	1									
CuE	-0.317**	0.475**	n.s.	-0.242*	-0.261*	0.357**	n.s.	0.413**	0.320**		1								
PbT	0.398**	n.s.	n.s.	n.s.	n.s.	n.s.	0.556**	0.283**	0.318**			1							
PbE	n.s.	0.444**	n.s.	n.s.	n.s.	0.352**	0.512**	0.395**	0.471**				1						
ZnT	0.450**	n.s.	0.303**	n.s.	n.s.	n.s.	0.462**	n.s.	0.345**					1					
ZnE	n.s.	0.509**	n.s.	n.s.	n.s.	0.467**	0.271*	0.474**	0.564**						1				
NiT	n.s.	0.297**	0.321**	-0.226*	n.s.	0.431**	n.s.	0.489**	n.s.							1			
NiE	n.s.	0.351**	-0.313**	n.s.	n.s.	n.s.	n.s.	n.s.	0.237*								1		
CoT	0.323**	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	0.239*	n.s.									1	
CoE	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.	n.s.										1
ZnEq	0.293*	0.277*	0.322**	n.s.	n.s.	0.342**	0.259*	0.471**	0.378**										
	CuT	CuE	PbT	PbE	ZnT	ZnE	NiT	NiE	CoE	CoE									
CuT	1																		
CuE	0.590**	1																	
PbT	0.372**	n.s.	1																
PbE	0.263*	0.276*	0.769**	1															
ZnT	0.368**	n.s.	0.649**	0.413**	1														
ZnE	0.652**	0.689**	0.320**	0.540**	0.333**	1													
NiT	0.572**	0.248*	0.311**	0.316**	n.s.	0.263*	1												
NiE	n.s.	0.386**	n.s.	n.s.	n.s.	n.s.	0.240*	1											
CoT	0.300**	n.s.	n.s.	n.s.	n.s.	n.s.	0.484**	n.s.	1										
CoE	n.s.	n.s.	n.s.	n.s.	-0.312**	n.s.	n.s.	n.s.	n.s.	1									
ZnEq	0.747**	n.s.	0.581**	0.460**	0.660**	0.471**	0.798**	n.s.	0.333**	n.s.									

Note. SOM: soil organic matter; CEC: cation exchange capacity; ESP: exchangeable sodium percentage; -T: concentration of total metal; -E: concentration of available metal; ZnEq; Zn equivalent; n.s.: not significant.  $n = 71$ .

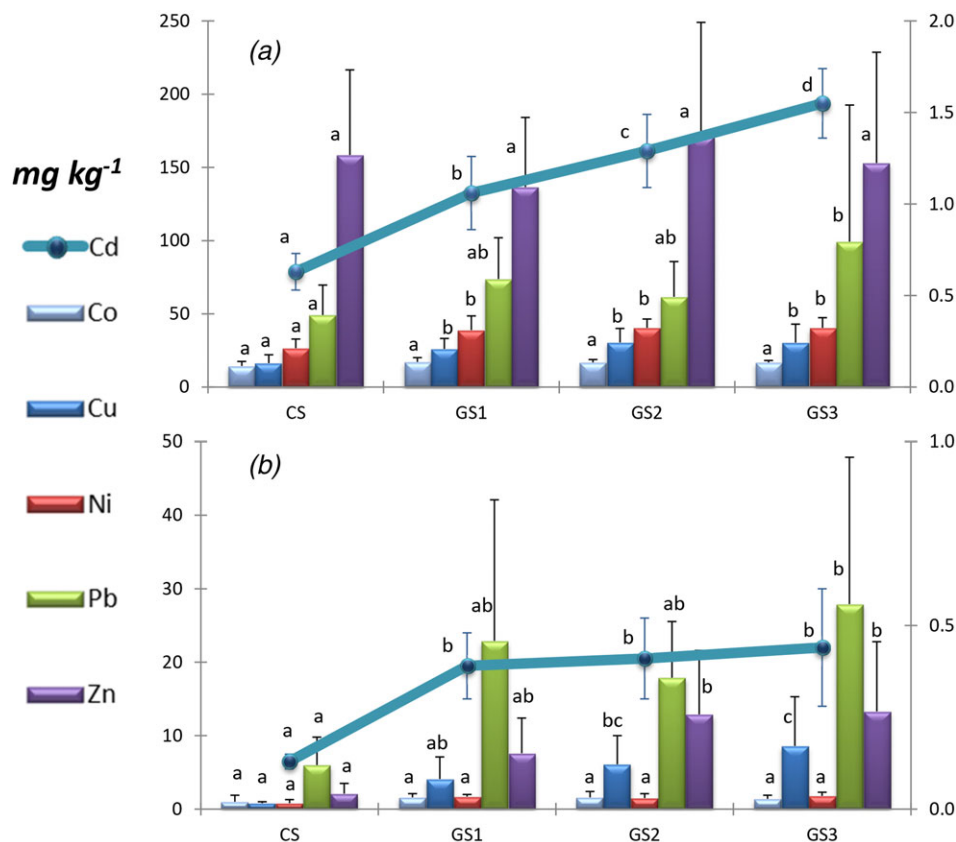
\* $p < 0.05$ . \*\* $p < 0.01$ .

to HM. Different authors have demonstrated the affinity of SOM to distinct HM in soils (Gil et al., 2004; Rodríguez Martín, López Arias, & Grau Corbí, 2006; Tume et al., 2006; Massas, Ehaliotis, Kalivas, & Panagopoulou, 2010; Roca-Pérez et al., 2010; Businelli, Onofri, & Massaccesi, 2011; among others). Our results confirmed the role that soil carbonates play in retaining HM, just as several authors have already reported (Businelli et al., 2011). One noteworthy aspect was the correlations obtained between available phosphorus and Cd, Cu, Ni, Pb, Zn, and ZnEq as this fact was related to the use of fertilizers in the agrosystem, which could be due to presence of HM in phosphorous fertilizers (Gil et al., 2004; Gimeno-García et al., 1996). Moreover, the relation between HM could indicate that these elements in the GS could share a common origin that could be related with anthropic activity (Gil et al., 2004). The positive and highly significant correlations found between MT and MA were logical; this fact has been associated with an enriched exogenous origin, which diminished the

retention force between the HM and soil colloids that favored the mobility of HM, which could pass to other environmental compartments (Massas et al., 2010; Ramos-Miras et al., 2011; Tarvainen & Kallio, 2002).

### 3.4 | Temporal trends of HMs in GS

No historic data about the concentration of HM in the GS are available. Using the background levels obtained in this study, and based on knowledge about the cropping ages of the GS, we could establish temporal distribution patterns for the studied elements. Figure 3 shows the changes in the temporal trends of HM. The total contents in HM showed a significant increase for the Cd, Cu, Pb, and Ni contents in GS compared with the CS, whereas no variations were observed for Co and Zn (Figure 3a). The same behavior pattern was observed for available concentration, but in this case, Zn also



**FIGURE 3** Temporal trends of heavy metal contents in greenhouse soils (GS) of western Almería, Spain (mean  $\pm$  standard deviation). (a) Total content and (b) available content. CS: control soil (0 year); GS1: <10 years; GS2: 10 to 20 years; GS3: >20 years. Right scale is only for Cd values. Different letter indicates statistically significant differences ( $p < 0.05$ ) after Krustall-Wallis and Scheffe's post hoc tests [Colour figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

presented major variations between the contemplated soil groups (Figure 3b). This fact revealed that progressive enrichment and the availability of Cd, Cu, Pb, and Zn increased in the GS with cropping age. This scenario confirmed that soil management and the use of agrochemicals was the main source of HM (Gonçalves, Nacke, Schwantes, Nava, & Strey, 2011; Molina, Aburto, Calderón, Cazanga, & Escudey, 2009; Uprety, Hejzman, Száková, Kunzová, & Tlustoš, 2009; Zaccone, Di Caterina, Rotunno, & Quinto, 2010). In the case of Cd, the main source would be phosphorus fertilizers (Gimeno-García et al., 1996). It has also been observed that nitrogen fertilizers favor Cd accumulation in plants (Wångstrand, Eriksson, & Oborn, 2007). Regarding Cu, apart from it being present in agrochemicals, the irrigation waters of the greenhouses in Almería are continuously treated with copper salts (SO<sub>4</sub>Cu) for biocide purposes (Casas et al., 2011), which contributes to soil enrichment. For ZnT and CoT, no accumulation was observed; thus, the original matter must be the main source for soils (Magrisso et al., 2009).

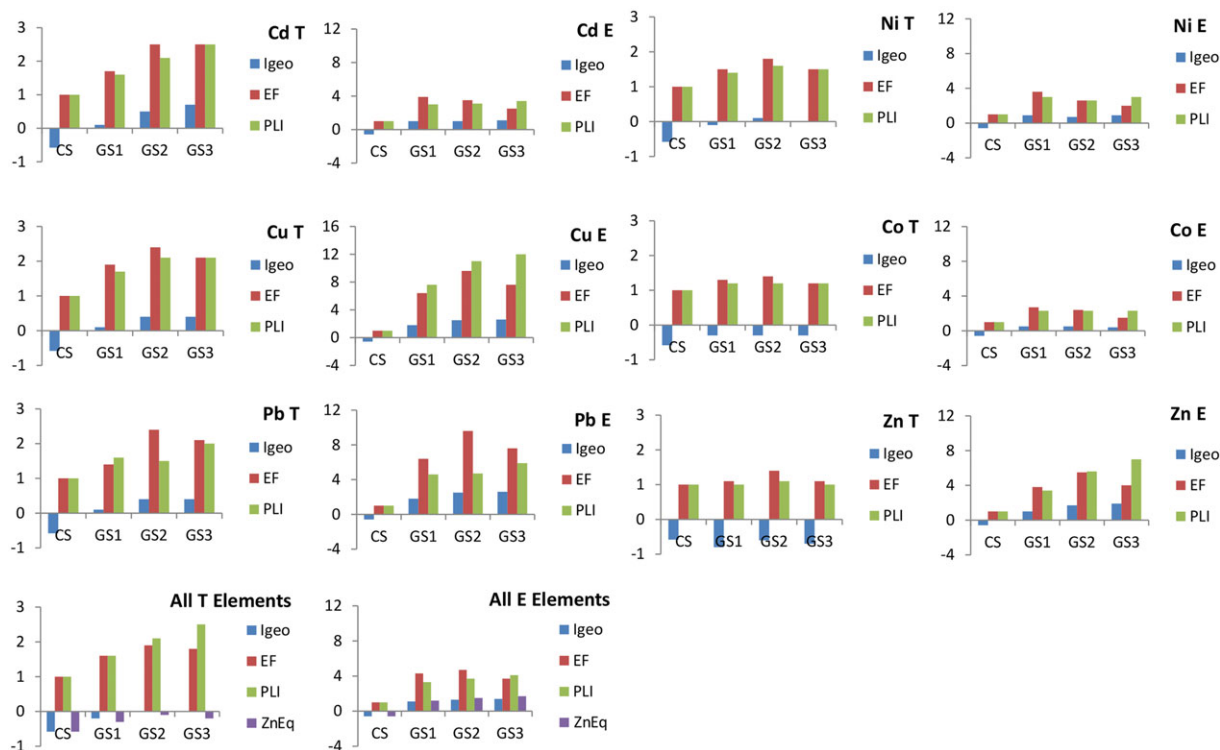
The increase observed in the available concentrations of Cd, Cu, Ni, Pb, and Zn (Figure 3b), especially for GS2 and GS3, could be related with the fact that HM added to soil exogenously favors the mobility of HM (Ghaderi, Abdul, Karbassi, Nasrabadi, & Khajeh, 2012; Massas et al., 2009). This might represent an important health risk (Roy & McDonald, 2015; Zhang et al., 2017).

Regarding the contamination indices, a significant increase in ZnEq, Igeo, EF, and PLI was observed (Figure 4), which was also

observed with HM as indices ZnEq, Igeo, EF, and PLI showed the same behavior patterns. Thus, cultivation led to rapidly increasing index values in the first years after setting up greenhouses (GS1, 0–10 years). After 20 years of cultivation, all the index values exceeded those of the background values (CS), except for CoT, CoE, and ZnT.

In relation to temporal trends, some studies have specifically shown that the concentrations of Hg, Cd, Cu, and Zn tend to rise in GS with cropping ages (Chen et al., 2016; Ramos-Miras et al., 2012). Li, Stagnitti, Xiong, and Peterson (2009) observed temporal distribution patterns for Cd, Cr, Cu, Ni, Pb, and Zn in soil layers at 0–20 cm, following exponential regression equations, caused by sewage irrigation after more than one century. Long et al. (2013) assessed the spatial-temporal variations of HM in farmland soils of Shanghai (China) and found that Cd, Zn, and Hg contributed the most pollution. When use of fertilizers was reduced, among other human activities, the impact of agricultural activities on the temporal variation of farmland soil quality weakened. Although some studies have shown that the reduced application of nutrients and pesticides reduces metal contamination in soil (Narimanidze & Brückner, 1999), using fewer fertilizers has been reported to relieve the impact of agriculture on temporal farmland soil quality variation (Long et al., 2013). This work demonstrates for the first time the HM concentration trend in GS in a semiarid area and warns about the risks that stem from intensive agriculture.





**FIGURE 4** Temporal trends of contamination indices values. Igeo: geoaccumulation index; EF: enrichment factor; PLI: potential load index; ZnEq: zinc equivalent; T: total content; E: ethylenediaminetetraacetic acid-extractable content; CS: control soils (0 year); GS1: <10 years; GS2: 10 to 20 years; GS3: >20 years [Colour figure can be viewed at [wileyonlinelibrary.com](http://wileyonlinelibrary.com)]

## 4 | CONCLUSIONS

A true trend pattern for temporal accumulation of Cd, Cu, and Pb is demonstrated for the GS of western Almería (Spain), which is associated with the intensive agriculture practices. In addition to metal accumulation, increasing the available concentration implies enhanced Cd, Cu, Pb, Zn, and Ni mobility. The values of the pollution indices (ZnEq, Igeo, EF, and PLI) employed in this work reveal the same behavior pattern, which confirms the above-indicated aspects. Thus, they are good tools to assess soil contamination and to address monitoring studies.

The disproportionate volume of agrochemical substances constantly used in GS is linked to increase of HMs. All the above indications suggest that this situation could reduce soil quality in the long term and could affect the crop yields and nutritional quality of the obtained vegetables. This study also demonstrates the importance of soil monitoring and assessment in the subjected GS to find the possible problems caused by intensive agriculture practices and to prevent pollution risks. SOM, soil carbonates, and greenhouse management can help reduce the mobility and bioavailability of these metals in the GS. Sustainable management practices (e.g., use of quality compost instead of manure amendments) should be assessed, and monitoring studies should continue in the future. The Cd, Cu, Ni, Pb, and Zn inputs in the GS should also be controlled to minimize pollution effects and to maintain soil quality. Therefore, future studies about plant uptake and ecological risk assessments should be conducted.

## ACKNOWLEDGMENT

This research was sponsored by the Spanish Ministry of Science and Innovation, projects AGL2011-29382 and CGL2013-43675-P.

## ORCID

Carlos Gil <http://orcid.org/0000-0002-9903-1303>

Rafael Boluda <http://orcid.org/0000-0001-7682-758X>

José Antonio Rodríguez Martín <http://orcid.org/0000-0002-9158-9564>

Miguel Guzmán <http://orcid.org/0000-0002-5648-7554>

Fernando del Moral <http://orcid.org/0000-0002-8323-0850>

Joaquín Ramos-Miras <http://orcid.org/0000-0001-6194-7191>

## REFERENCES

- Aguilar, J., Dorronsoro, C., Fernández, J. A., Bellver, R., Fernández, E., García, I., ... Izarra, R. (2005). *Estudio de elementos traza en suelos de Andalucía*. Sevilla: Consejería de Medio Ambiente Junta de Andalucía.
- Álvarez-Ayuso, E., García-Sánchez, A., Querol, X., & Moyano, A. (2008). Trace element mobility in soils seven years after the Aznalcóllar mine spill. *Chemosphere*, 73, 1240–1246. <https://doi.org/10.1016/j.chemosphere.2008.07.028>
- Andreu, V., & Boluda, R. (1995). Application of contamination indexes on different farming soils. *Bulletin of Environmental Contamination and Toxicology*, 54, 228–236. <https://doi.org/10.1007/BF00197435>
- Bloundi, M. K., Duplay, J., & Quaranta, G. (2009). Heavy metal contamination of coastal lagoon sediments by anthropogenic activities: The case of Nador (East Morocco). *Environmental Geology*, 56, 833–843. <https://doi.org/10.1007/s00254-007-1184-x>

- Boluda, R., Andreu, V., Gilabert, M. A., & Sobrino, P. (1993). Relation between reflectance of rice crop and indices of pollution by heavy metals in soils of Albufera Natural Park (Valencia, Spain). *Soil Technology*, 6, 351–363. [https://doi.org/10.1016/0933-3630\(93\)90025-A](https://doi.org/10.1016/0933-3630(93)90025-A)
- Businelli, D., Onofri, A., & Massaccesi, L. (2011). Factors involved in uptake of lead by some edible crops grown in agricultural soils of Central Italy. *Soil Science*, 176, 472–478. <https://doi.org/10.1097/SS.0b013e31822769fa>
- Casas, J. J., Sánchez-Oliver, J. S., Sanz, A., Furné, M., Trenzado, C., Juan, M., ... Ramos-Miras, J. J. (2011). The paradox of the conservation of an endangered fish species in a Mediterranean region under agricultural intensification. *Biological Conservation*, 144, 253–262. <https://doi.org/10.1016/j.biocon.2010.08.023>
- Chen, Z., Tian, T., Gao, L., & Tian, Y. (2016). Nutrients, heavy metals and phthalate acid esters in solar greenhouse soils in Round-Bohai Bay-Region, China: Impacts of cultivation year and biogeography. *Environmental Science and Pollution Research*, 23, 13076–13087. <https://doi.org/10.1007/s11356-016-6462-2>
- Ding, F., He, Z., Liu, S., Zhang, S., Zhao, F., Li, Q., & Stoffella, P. J. (2017). Heavy metals in composts of China: Historical changes, regional variation, and potential impact on soil quality. *Environmental Science and Pollution Research*, 24, 3194–3209. <https://doi.org/10.1007/s11356-016-8057-3>
- Ghaderi, A. A., Abdul, M. A., Karbassi, A. R., Nasrabadi, T., & Khajeh, M. (2012). Evaluating the effects of fertilizers on bioavailable metallic pollution of soils, case study of Sistan farms, Iran. *International Journal of Environmental Research*, 6, 565–570. <https://doi.org/10.22059/ijer.2012.526>
- Gil, C., Boluda, R., & Ramos, J. (2004). Determination and evaluation of cadmium lead and nickel in greenhouses soils of Almería (Spain). *Chem*, 55, 1027–1034. <https://doi.org/10.1016/j.chemosphere.2004.01.013>
- Gil, C., Ramos-Miras, J., Roca-Pérez, L., & Boluda, R. (2010). Determination and assessment of mercury content in calcareous soils. *Chemosphere*, 78, 409–415. <https://doi.org/10.1016/j.chemosphere.2009.11.001>
- Gimeno-García, E., Andreu, V., & Boluda, R. (1996). Heavy metals incidence in the application of inorganic fertilizers and pesticides to rice farming soils. *Environmental Pollution*, 92, 19–25. [https://doi.org/10.1016/0269-7491\(95\)00090-9](https://doi.org/10.1016/0269-7491(95)00090-9)
- Gonçalves, A. C., Nacke, H., Schwantes, D., Nava, I. A., & Strey, L. (2011). Phytoavailability of toxic heavy metals and productivity in wheat cultivated under residual effect of fertilization in soybean culture. *Water, Air, & Soil Pollution*, 220, 205–211. <https://doi.org/10.1007/s11270-011-0747-3>
- Gutiérrez, C., Fernández, C., Escuer, M., Campos-Herrera, R., Beltrán Rodríguez, M. E., Carbonell, G., & Rodríguez Martín, J. A. (2016). Effect of soil properties, heavy metals and emerging contaminants in the soil nematodes diversity. *Environmental Pollution*, 213, 184–194. <https://doi.org/10.1016/j.envpol.2016.02.012>
- Hou, D., O'Connor, D., Nathanail, P., Tian, L., & Ma, Y. (2017). Integrated GIS and multivariate statistical analysis for regional scale assessment of heavy metal soil contamination: A critical review. *Environmental Pollution*, 231, 1188–1200. <https://doi.org/10.1016/j.envpol.2017.07.021>
- IUSS Working Group WRB 2015. World Reference Base for Soil Resources. International soil classification system for naming soils and creating legends for soil maps. World Soil Resources Reports No. 106. FAO, Rome.
- Khaledian, Y., Pereira, P., Brevik, E. C., Pundyte, N., & Paliulis, D. (2017). The influence of organic carbon and pH on heavy metals, potassium, and magnesium levels in Lithuanian Podzols. *Land Degradation & Development*, 28, 345–354. <https://doi.org/10.1002/ldr.2638>
- Kingston HM, Walter PJ. 1995. USEPA method 3052: Microwave assisted digestion of siliceous and organically based matrices. USEPA.
- Kochem-Mallmann, F. J., Rheinheimer dos Santos, D. R., Cambier, P., Labanowski, J., Lamy, I., Santanna, M. A., ... van Oort, F. (2012). Using a two site-reactive model for simulating one century changes of Zn and Pb concentration profiles in soils affected by metallurgical fallout. *Environmental Pollution*, 162, 294–302. <https://doi.org/10.1016/j.envpol.2011.11.039>
- Li, F., Shi, W., Jin, Z., Wu, H., & Sheng, G. D. (2017). Excessive uptake of heavy metals by greenhouse vegetable. *Journal of Geochemical Exploration*, 173, 76–84. <https://doi.org/10.1016/j.gexplo.2016.12.002>
- Li, P. J., Stagnitti, F., Xiong, X., & Peterson, J. (2009). Temporal and spatial distribution patterns of heavy metals in soil at a long-standing sewage farm. *Environmental Monitoring Assessment*, 49, 275–282. <https://doi.org/10.1007/s10661-008-0201-1>
- Long, Q., Wang, J. Y., & Da, L. J. (2013). Assessing the spatial-temporal variations of heavy metals in farmland soil of Shanghai, China. *Fresenius Environmental Bulletin*, 22, 928–938.
- Magrisso, S., Belkin, S., & Errel, Y. (2009). Lead bioavailability in soil and soil components. *Water, Air and Soil Pollution*, 20, 315–323. <https://doi.org/10.1007/s11270-009-9978-y>
- Massas, I., Ehaliotis, C., Gerontidis, S., & Sarris, E. (2009). Elevated heavy metal concentrations in top soils of an Aegean island town (Greece): Total and available forms, origin and distribution. *Environmental Monitoring and Assessment*, 151, 105–116. <https://doi.org/10.1007/s10661-008-0253-2>
- Massas, I., Ehaliotis, C., Kalivas, D., & Panagopoulou, G. (2010). Concentrations and availability indicators of soil heavy metals; the case of children's playgrounds in the city of Athens (Greece). *Water, Air and Soil Pollution*, 212, 51–63. <https://doi.org/10.1007/s11270-009-0321-4>
- Mirzaei, R., Ghorbani, H., HafeziMoghaddas, N., & Martín, J. A. R. (2014). Ecological risk of heavy metal hotspots in topsoils in the province of Golestan, Iran. *Journal of Geochemical Exploration*, 147, 268–276. <https://doi.org/10.1016/j.gexplo.2014.06.011>
- Molina, M., Aburto, F., Calderón, R., Cazanga, M., & Escudey, M. (2009). Trace element composition of selected fertilizers used in Chile: Phosphorus fertilizers as a source of long-term soil contamination. *Soil and Sediment Contamination*, 18, 497–511. <https://doi.org/10.1080/15320380902962320>
- Müller, G. (1969). Index of geoaccumulation in sediments of the Rhine River. *Geo Journal*, 2(3), 108–118.
- Narimanidze, E., & Brückner, H. (1999). Survey on the metal contamination of agricultural soils in Georgia. *Land Degradation & Development*, 10, 467–488. [https://doi.org/10.1002/\(SICI\)1099-145X\(199909/10\)10:5<467::AID-LDR344>3.0.CO;2-6](https://doi.org/10.1002/(SICI)1099-145X(199909/10)10:5<467::AID-LDR344>3.0.CO;2-6)
- Odumo, B., Carbonell, G., Angeyo, H., Patel, J., Torrijos, M., & Rodríguez Martín, J. (2014). Impact of gold mining associated with mercury contamination in soil, biota sediments and tailings in Kenya. *Environmental Science and Pollution Research*, 21, 12426–12435. <https://doi.org/10.1007/s11356-014-3190-3>
- Ordoñez, A., Alvarez, R., De Miguel, E., & Charlesworth, S. (2015). Spatial and temporal variations of trace element distribution in soils and street dust of an industrial town in NW Spain: 15 years of study. *Science of the Total Environment*, 524, 93–103. <https://doi.org/10.1016/j.scitotenv.2015.04.024>
- Plaza-Bolaños, P., Padilla-Sánchez, J. A., Garrido-Freniche, A., Romero-González, R., & Martínez-Vidal, J. L. (2012). Evaluation of soil contamination in intensive agricultural areas by pesticides and organic pollutants: South-eastern Spain as a case study. *Journal of Environmental Monitoring*, 14, 1182–1189. <https://doi.org/10.1039/C2EM10993J>
- Quevauviller Ph, L. M., Barahona, E., Rauret, G., Ure, A., Gómez, A., & Muntau, H. (1994). Interlaboratory comparison of EDTA and DTPA procedures prior to certification of extractable trace elements in calcareous soil. *Science of the Total Environment*, 178, 127–132. [https://doi.org/10.1016/0048-9697\(95\)04804-9](https://doi.org/10.1016/0048-9697(95)04804-9)
- Ramos-Miras, J. J., Roca-Pérez, L., Boluda, R., & Gil, C. (2012). Content and evolution of mercury in greenhouse soils of Almería, Spain. *Acta Horticulturae*, (927), 821–826. <https://doi.org/10.17660/ActaHortic.2012.927.102>
- Ramos-Miras, J. J., Roca-Pérez, L., Guzmán-Palomino, M., Boluda, R., & Gil, C. (2011). Background levels and baseline values of available heavy metals in Mediterranean greenhouse soils (Spain). *Journal of*

- Geochemical Exploration*, 110, 186–192. <https://doi.org/10.1016/j.gexplo.2011.05.009>
- Roca-Pérez, L., Gil, C., Cervera, M. L., González, A., Ramos-Miras, J., Pons, V., ... Boluda, R. (2010). Selenium and heavy metals content in some Mediterranean soils. *Journal of Geochemical Exploration*, 107, 110–116. <https://doi.org/10.1016/j.gexplo.2010.08.004>
- Rodríguez Martín, J. A., Gutiérrez, C., Escuer, M., García-González, M. T., Campos-Herrera, R., & Águila, N. (2014). Effect of mine tailing on the spatial variability of soil nematodes from lead pollution in La Union (Spain). *Science of the Total Environment*, 473–474, 518–529. <https://doi.org/10.1016/j.scitotenv.2013.12.075>
- Rodríguez Martín, J. A., & Nanos, N. (2016). Soil as an archive of coal-fired power plant mercury deposition. *Journal of Hazardous Materials*, 308, 131–138. <https://doi.org/10.1016/j.jhazmat.2016.01.026>
- Rodríguez Martín, J. A., López Arias, M., & Grau Corbí, J. M. (2006). Heavy metal contents in agricultural topsoils in the Ebro basin (Spain). Application of multivariate geostatistical methods to study spatial variations. *Environmental Pollution*, 144, 1001–1012. <https://doi.org/10.1016/j.envpol.2006.01.045>
- Rodríguez-Martín, J. A., López-Arias, M., & Grau-Corbí, J. M. (2009). *Metales pesados, materia orgánica y otros parámetros de los suelos agrícolas y pastos de España*. Madrid: Ministerio de Medio Ambiente Rural y Marino.
- Rodríguez-Martín, J. A., Ramos-Miras, J., Boluda, R., & Gil, C. (2013). Spatial relations of heavy metals in arable and greenhouse soils of a Mediterranean environment region (Spain). *Geoderma*, 200–201, 180–188. <https://doi.org/10.1016/j.geoderma.2013.02.014>
- Roy, M., & McDonald, L. M. (2015). Metal uptake in plants and health risk assessments in metal-contaminated smelter soils. *Land Degradation & Development*, 26, 785–792. <https://doi.org/10.1002/ldr.2237>
- Sakizadeh, M., Mirzaei, R., & Ghorbani, H. (2015). The extent and prediction of heavy metal pollution in soils of Shahrood and Damghan, Iran. *Bulletin of Environmental Contamination and Toxicology*, 95, 770–776. <https://doi.org/10.1007/s00128-015-1632-3>
- Shao, D., Zhan, Y., Zhou, W., & Zhu, L. (2016). Current status and temporal trend of heavy metals in farmland soil of the Yangtze River Delta Region: Field survey and meta-analysis. *Environmental Pollution*, 219, 329–336. <https://doi.org/10.1016/j.envpol.2016.10.023>
- Sierra, M., Martínez, F. J., & Aguilar, J. (2007). Baselines for trace elements and evaluation of environmental risk in soils of Almería (SE Spain). *Geoderma*, 139, 209–219. <https://doi.org/10.1016/j.geoderma.2007.02.003>
- Tamim, U., Khan, R., Jolly, Y. N., Fatema, K., Das, S., Naher, K., ... Hossain, S. M. (2016). Elemental distribution of metals in urban river sediments near an industrial effluent source. *Chemosphere*, 155, 509–518. <https://doi.org/10.1016/j.chemosphere.2016.04.099>
- Tarvainen, T., & Kallio, E. (2002). Baselines of certain bioavailable and total heavy concentrations in Finland. *Applied Geochemistry*, 17, 975–980. [https://doi.org/10.1016/S0883-2927\(02\)00003-3](https://doi.org/10.1016/S0883-2927(02)00003-3)
- Tianlik, T., Norulaini, N. A. R. N., Shahadat, M., Yoonsing, W., & Omar, A. K. M. (2016). Risk assessment of metal contamination in soil and groundwater in Asia: A review of recent trends as well as existing environmental laws and regulations. *Pedosphere*, 26, 431–450. [https://doi.org/10.1016/S1002-0160\(15\)60055-8](https://doi.org/10.1016/S1002-0160(15)60055-8)
- Tume, P., Bech, J., Longan, L., Tume, L., Reverter, F., & Sepulveda, B. (2006). Trace elements in natural surface soils in Sant Climent (Catalonia, Spain). *Ecological Engineering*, 27, 145–152. <https://doi.org/10.1016/j.ecoleng.2006.01.004>
- Uprety, D., Hejzman, M., Száková, J., Kunzová, E., & Tlustoš, P. (2009). Concentration of trace elements in arable soil after long-term application of organic and inorganic fertilizers. *Nutrient Cycling in Agroecosystems*, 85, 241–252. <https://doi.org/10.1007/s10705-009-9263-x>
- Wångstrand, H., Eriksson, J., & Oborn, I. (2007). Cadmium concentration in winter wheat as affected by nitrogen fertilization. *European Journal of Agronomy*, 26, 209–214. <https://doi.org/10.1016/j.eja.2006.09.010>
- Zaccone, C., Di Caterina, R., Rotunno, T., & Quinto, M. (2010). Soil-farming system-food-health: Effect of conventional and organic fertilizers on heavy metal (Cd, Cr, Cu, Ni, Pb, Zn) content in semolina samples. *Soil and Tillage Research*, 107, 97–105. <https://doi.org/10.1016/j.still.2010.02.004>
- Zeng, F., Ali, S., Zhang, H., Ouyang, Y., Qiu, B., Wu, F., & Zhang, G. (2011). The influence of pH and organic matter content in paddy soil on heavy metal availability and their uptake by rice plants. *Environmental Pollution*, 159, 84–91. <https://doi.org/10.1016/j.envpol.2010.09.019>
- Zhang, H., Huang, B., Dong, L., Hu, W., Akhtar, M. S., & Qu, M. (2017). Accumulation, sources and health risks of trace metals in elevated geochemical background soils used for greenhouse vegetable production in southwestern China. *Ecotoxicology and Environmental Safety*, 137, 233–239. <https://doi.org/10.1016/j.ecoenv.2016.12.010>
- Zhang, X. Y., Tang, L. S., Zhang, G., & Wu, H. D. (2009). Heavy metal contamination in a typical mining town of a minority and mountain area, South China. *Bulletin of Environmental Contamination and Toxicology*, 82, 31–38. <https://doi.org/10.1007/s00128-008-9569-4>

**How to cite this article:** Gil C, Boluda R, Rodríguez Martín JA, Guzmán M, del Moral F, Ramos-Miras J. Assessing soil contamination and temporal trends of heavy metal contents in greenhouses on semiarid land. *Land Degrad Dev*. 2018;29:3344–3354. <https://doi.org/10.1002/ldr.3094>