Impact assessment of agricultural activities on heavy metal accumulation in soil

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Abstract. Use of chemical fertilizers, pesticides and sewage sludge in agriculture lands caused the increase of heavy metals concentration in the soils in many parts of the world. The purpose of this article is studying the impacts of agricultural activities on the accumulation of Cd, Co, Cr, Cu, Ni and Pb in the soil. The soil samples were taken by systematic method from agriculture fields with cultivation period of 65, 35 and 20 years in Hamadan province situated in the west of Iran. The results showed a significant difference between the concentrations of heavy metals, except for Pb, in the soil under different cultivation periods. The trend of heavy metal accumulation for Cd, Co, Cr and Cu was exponential, whereas for Ni and Pb, it was quadratic and linear, respectively. Also, based on calculated contamination factor, the soils considered as none and none to medium pollution classes were generally found in the lands with more length of cultivation.
Key Words: accumulation trend, agricultural activities, contamination factor, heavy metal.

Introduction. The most important soil contaminants are heavy metals, acid precipitation and organic materials. In recent years, soil pollution by heavy metals has received much attention due to their properties (Yalcin et al 2007). Spatial variations of heavy metals contents in agricultural topsoil may be affected by soil parent material and anthropogenic sources (De Temmerman et al 2003). Agricultural activities cause enrichment of elements in agricultural ecosystems. Heavy metal can enter soil due to the use of solid and liquid inorganic and organic fertilizers (Kashem & Singh 2001; Mantovi et al 2003). The application of fertilizers is an important source of heavy metals especially Cd, Cu and Zn (Nicholson et al 2003) and causes the accumulation of these metals in soil (Yalcin et al 2007). Application of phosphate fertilizers in agricultural soils can lead to increasing the levels of Cd, Ar, Cr and Pb in soil and decreasing soil pH, and consequently the mobility of heavy metals from the soil matrix (Alloway 1995; Sofianska et al 2013). Alloway (1990) reported that soil contamination by heavy metals resulting from application of phosphate fertilizers is a cause for concern in some countries.

Nowadays, due to limited access to fresh water for irrigation, wastewater, especially sewage water is being used for irrigation of agricultural lands and several researches have confirmed that the use of sewage water for agricultural irrigation can be useful (Mehrdadi et al 2007; Palese et al 2009). Application of sewage water leads to improvement of the physico-chemical properties and nutrient status of the soil and increase in crop production (Panicker 1995). On the other hand, the use of sewage water in agriculture is associated with some risks, because it increases the presence of disease microorganisms (Toze 2006), metal contaminations such as Cu, Ni, Cd, Cr, Zn (Misra & Mani 1991) and poly chloride (Bansal 1998). McGrath et al (1995) declared that after 25 years, more than 80 percent of the added toxic metals to the surface layer of soil was due to application of sewage sludge.

Agriculture soils, both directly and indirectly, affect on public health by food production; therefore, conservating this source and ensuring its constancy could be very important. Rapid industrial developing and high release of chemical materials used in agriculture cause concerns about accumulation of heavy metals in agricultural soils (Alloway 1990; Wong et al 2002). Heavy metals can accumulate in crops or plants and cause the damage and changes in humans and animals' physiological functions through the food chain (Otte et al 1993; Dudka et al 1994; Kong 2014). Previous studies have shown human exposure to the risk of accumulation of heavy metals and their accumulation in the fatty tissues of the human body (Abdelhafez et al 2012). It may impact the central nervous system or be settled in circulatory system and disrupt the normal function of internal organs (Waisberg et al 2003; Bocca et al 2004).

Sun et al (2006) studied the spatial distribution and temporal variation of cadmium contamination using kriging methods in Shenyang Zhangshi irrigation area (SZIA) in China. The Shenyang Zhangshi irrigation area in China is a representative area of heavy metal contamination of soils resulting from sewage irrigation for about 30 years. The soil samples were collected from topsoils (0-20 cm) and subsoils (20-40 cm) from the SZIA in 1990, 2004. The kriging map showed that long-term sewage irrigation had caused serious Cd contamination in topsoil and subsoil. Compared with that in 1990, the mean and maximum concentration of Cd as well as the area with Cd more than 1.5 mg kg^{-1} in the soil was increased in 2004, both in topsoils and subsoils.

Chen et al (2007) have studied spatial-temporal variations of heavy metals contamination of sediments using GIS 3D spatial analysis methods in Dexing mines, Jiangxi province, China. The geo-accumulation index (Igeo) was used to assess the environment quality. Spatial-temporal contrast was performed using GIS 3D spatial analysis from original testing data and geo-accumulation index. The results of the maximum increases in As, Hg, Cd, Cr, Zn, Cu and Pb concentration in 2004 and in sediments they were up to 9, 4, 4.6, 1.5, 5.9, 6.3 and 5.6 times higher than those in 1989, respectively. All the contrast results indicated that the extent and scope of heavy metals contamination of sediments in 2004 were higher than those in 1989.

Overuse of chemical fertilizers in Iran (placed among the first 12 countries of the world's fertilizer consumers until 2006 (Khodakarami 2009)), its rapid growing consumption (21.5% increase in fertilizer consumption in 2003 compared to 2002, the direct use of wastewater for agricultural purposes due to lack of irrigation water and lack of sewage treatment systems in many cities (Motasadi Zarandi & Babran 2009), have caused the transfer of heavy metals to agricultural soils. Due to concerns about the effects of heavy metals in soil fertility and their potential transfer to the human's diets, we need to quantify the metal inputs to agricultural soils and evaluate the soil contamination. The accumulation of heavy metals in agricultural soils has been investigated in few studies in Iran (Amini et al 2007). The aim of this study is to determine the impact of agriculture activities on the accumulation of Cd, Co, Cr, Cu, Ni and Pb in the soil under different cultivation conditions.

Material and Method

Study area. Hamadan province is one of the western provinces of Iran with a population of 1700000 and area of 19547 km^2. It is located between 33°58′ and 35°44′ northern latitude and 47°48′ and 49°28′ eastern longitude (Figure 1). Dominant geological formations are: alluvial fans from quaternary, orbitolina limestone, sheil and marn from Keratase, catabolized sandstone from Jurassic, and andesite lava, limestone from early Neozhen & late Paleozhen. Soil depth is shallow to semi deep, gravelly, with light to medium texture and lime accumulated layer. Soil texture is clay-loam and loam (Khodakarami 2009). Total agricultural lands have 1,005,000 hectares area with 719,000 hectares being under cultivation and the rest is fallow. In the study area, 43.3% of cultivated lands are irrigated and the rest of them are dry farm, gardens and nurseries. The major crops include wheat, barley, alfalfa, potato & maize (Khodakarami 2009).

Soil sampling and chemical analysis. The satellite images of different years were used to determine the cultivated lands having cultivation period. Referring to selected sites and a questionnaire, the cultivation period in each site was determined. Soil sampling was done in six sites having a cultivation duration of 65 (37 samples), 35 (31 samples) and 20 (29 samples) years as well as 12 samples from non-agriculture lands as control points (Figure 1). The surface soil samples (0–20 cm) were collected by systematic methods in late September 2010 after crop harvesting. All sites were located in the same bedrocks (alluvial).

Soil samples were air dried, grinded and sieved. Solution of aqua regia (HNO_3, HCL, H_2O_2) was used for digestion of soil samples. The concentration of Co, Cr, Cu, Ni and Pb were determined using atomic absorption spectrophotometer (model AAnalyst 700 Perkian Elmer). HNO_3 and H_2O_2 were used for digestion of Cd because HCl interferes with the analysis of Cd by graphite furnace (McGrath & Cunliffe 1985).

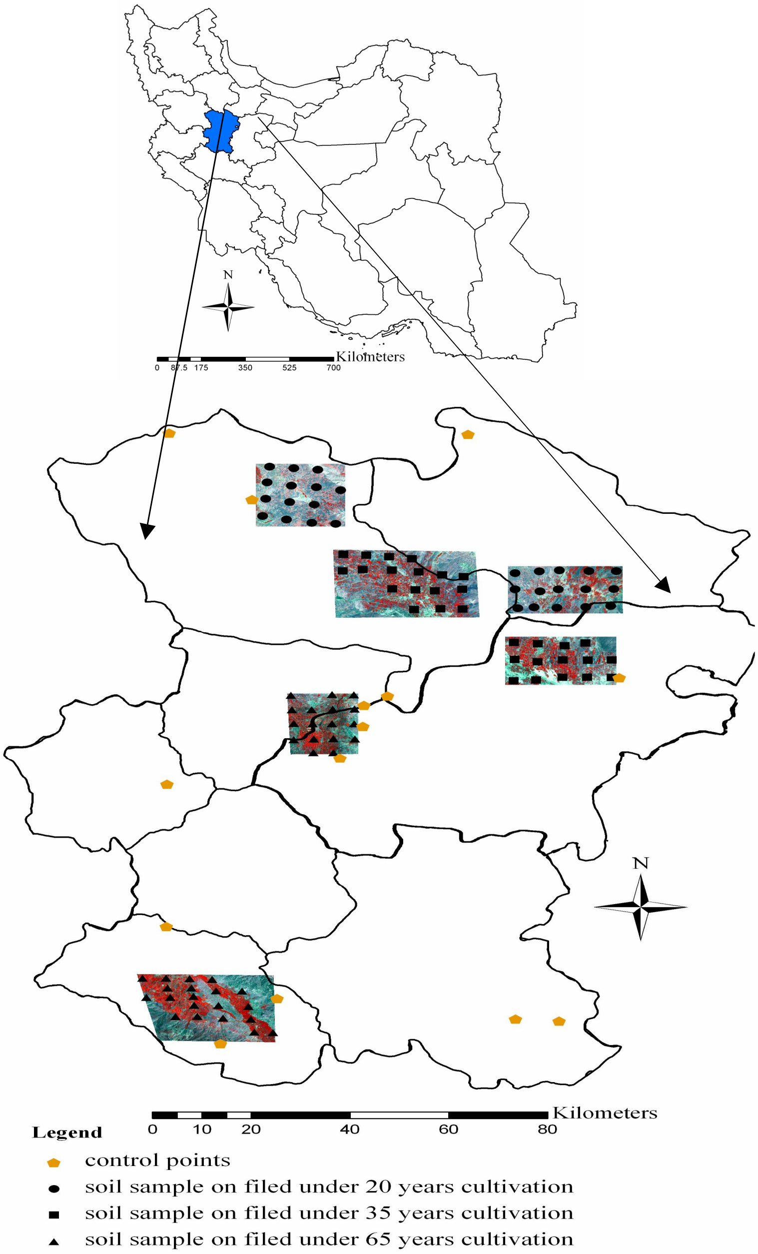

Figure 1. Location of Hamadan province in Iran and sampling sites.

Descriptive statistic analysis and heavy metal accumulation trend. The normality of data was verified by Kolmogorov-Smirnov test. ANOVA was used to compare the concentration of heavy metals. To determine trend in the metals concentration, Kendall and Spearman tests were used. The statistic analysis was performed using SPSS 15.0 software and Minitab 15.

Soil contamination factor. The soil contamination risk was evaluated by contamination factor (equation 1) (Abrahim & Parker 2008). According to this factor, soil contamination was classified in seven classes (Table1):

CF= [Concentration] heavy metal/[Concentration] background (1)

Table 1

Classification levels of contamination factor (Bhuiyan et al 2010)

Pollution	*Contamination factor*
None	0
None to medium	1
Moderate	2
Moderately to strong	3
Strongly polluted	4
Strong to very strong	5
Very strong	6

Results and Discussion

Descriptive statistic. Descriptive statistics are shown in Table 2. The concentration of Cd, Co, Cr, Cu, Ni and Pb in the agriculture soils were more than control points. In general, the mean concentration of heavy metals in the soils was increased with the cultivation period. The concentration of all elements in the soil had normal distribution. However, due to non homogeneity of variance for Cd, Cr and Ni, a logarithmic transformation was used for Cr and Ni, and Box-Cox transformation for Cd. However, there are variations between the elements. For Cd, Co, Cr, Cu and Ni, the mean concentration in the soil under 65 years-cultivation was significantly higher than control points. Whereas for Pb, there is no significant difference between the mean concentration in the soil (Table 3). This can indicate the effect of cultivation longevity on the heavy metals accumulation in the soils. The presence of these metals in chemical fertilizers and sewage samples used in the study area can be the reason for their increase in the soil (Khodakarami 2009).

Table 2

Descriptive statistics of heavy metals concentration (mg kg^{-1}) in agricultural soils

Site		*Cd*	*Co*	*Cr*	*Cu*	*Ni*	*Pb*
Control points	Mean	0.13	16.22	49.95	23.94	60.1	20.7
	Min	0.04	10	20	14.98	29	10.62
	Max	0.34	20	110	33.16	82	23
	Median	0.09	17	32.17	14.15	65.06	22
	K-S	0.11	0.84	0.16	0.99	0.71	0.84
20 years	Mean	0.14	17.94	58.89	27.53	61.93	23.97
	Min	0.09	8.10	24.08	15	26	12.21
	Max	0.33	26.54	110	41	102.01	52.43
	Median	0.11	17.6	60	29.3	54	22.4
	K-S	0.07	0.96	0.99	0.64	0.27	0.28
35 years	Mean	0.16	18.05	72.99	23.94	61.63	25.41
	Min	0.09	8.90	28.49	19.75	27	15.10
	Max	0.33	22	110	47	88	50.18
	Median	0.14	18	75	30.5	62	24
	K-S	0.11	0.33	0.52	0.83	0.97	0.61
65 years	Mean	0.19	20.79	88.09	34.96	73.85	26.13
	Min	0.09	11	43.64	19	39	14.78
	Max	0.51	32.05	160	52	122.28	81.14
	Median	0.17	20	78	34.5	73	24
	K-S	0.19	0.66	0.29	0.90	0.80	0.17

K-S: Kolmogorov-Smirnov Test.

Table 3

ANOVA test results in agricultural regions

	Cd	*Co*	*Cr*	*Cu*	*Ni*	*Pb*
65 years	0.19[a]	20.79[a]	88.09[a]	34.96[a]	73.85[a]	26.13[a]
35 years	0.16a[b]	18.05[b]	72.99[b]	31.98[a]	61.63[b]	25.41[a]
20 years	0.14[b]	17.94[b]	58.89[c]	27.53[b]	61.93[b]	23.97[a]
Control point	0.13[b]	16.22[b]	49.95[c]	23.94[b]	60.10[b]	20.70[a]

Different letters in each column indicate significant differences in mean level of 0.05.

Probably, increased concentration of heavy metals in agriculture soils can be a result of presence of heavy metals in irrigation sewage (Al-Nakshabandi et al 1997) as well as the use of chemical/organic fertilizers (Huang & Jin 2008). Khodakarami (2009) investigated the concentration of 14 heavy metals in different land uses in a part of Hamadan province. He indicated that the main factor controlling the concentration of Cd, Co, Cr, Cu, Ni and Pb in study area was geological formation, but excessive use of chemical fertilizers can contribute to increasing the concentration of these heavy metals in soil.

Heavy metal accumulation trend. The results of Kendall and Spearman tests showed a trend in the concentrations of all metals. To understand heavy metals accumulation trend in 65 year old fields, it was necessary to have information about the concentration variations of heavy metals in the soil. The current metal concentration in the soils of 35 and 20 year cultivation period can be simulated as the reference concentration in the soils of 65 year old fields. Because there is a similarity between the lands both geologically and in terms of farming management. The curve of heavy metal accumulation trend in the soil under 65 year cultivation has been drawn using current concentration of metals (Figure 2).

Heavy metals accumulation trend in 65 year old field soils was shown in Table 4. The heavy metal accumulation trend of Cd, Co, Cr and Cu was exponential, but this pattern was quadratic for Ni and linear for Pb.

Table 4

Equations for accumulation trend of heavy metal in the soil under 65 year cultivation

Metal	*Equation*	*R^2*
Cd	$y = 0.13e^{0.005x}$	0.98
Co	$y = 16.29e^{0.003x}$	0.96
Cr	$y = 50.44e^{0.008x}$	0.97
Cu	$y = 24.53e^{0.005x}$	0.94
Ni	$y = 0.004x^2-0.09x+60.52$	0.82
Pb	$y = 0.08x+21.64$	0.83

Li et al (2009) studied three regions irrigated with sewage in different temporal periods. In this study, 41 soil samples were taken from 0-20 and 20-40 cm of soil depth, 17 samples in regions irrigated with sewage since 1893 (LFA_{1893}), 10 samples from regions irrigated with sewage since 1920 (LFA_{1920}), 8 samples in regions irrigated with sewage since 1944 (LFA_{1944}) and 6 samples in regions not irrigated with sewage, as control points. The results showed that heavy metals concentration in LFA_{1893} was more than LFA_{1920}, that of LFA_{1920} was more than LFA_{1944} and those of all three regions were more than control points. Moreover, in order to understand temporal accumulation pattern in LFA_{1893} region, the current heavy metal concentration in LFA_{1920} and LFA_{1944} was used in the absence of related archive data. The results showed that distribution pattern of heavy metals were exponential in 0-20 cm depth and linear in 20-40 cm depth.

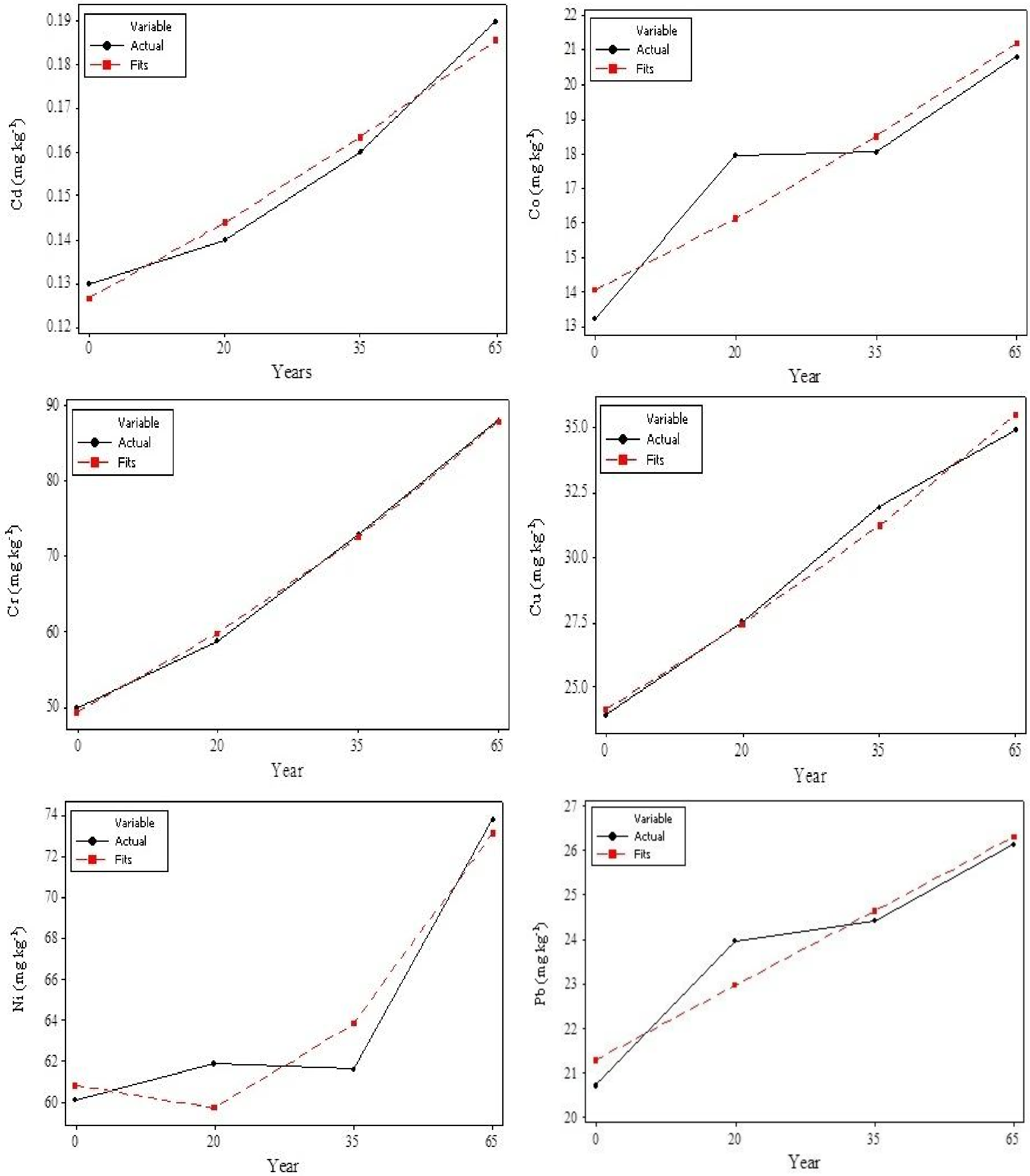

Figure 2. The heavy metals accumulation trend in the soil under 65 year cultivation.

Soil contamination factor. To calculate background concentration, the mean concentration of 12 soil samples from uncultivated lands on alluvial bed was used. The "Geochemical background" can be described as: normal abundant of an element in an empty and barren land. In other words, it is the supply of rare elements which are in soil without human activities (McGrath et al 1995). The results of calculated contamination factor show that the majority of sample can be classified as none polluted and none to medium polluted classes (Figure 3). For both metal Cd and Cr, 2.7 percent of the samples cultivated in 65 year were classified as moderate to strong contamination class, 24.3 and 29.7 percent of samples as moderate contamination class, respectively. For Cu and Ni, 10.8 and 2.1 percent of the samples in 65 year cultivated lands were as moderate contamination class, respectively. Co had the lowest contamination factor. The soil contamination index in 65 year cultivated land was more than 20 and 35 year cultivated lands (Figure 3).

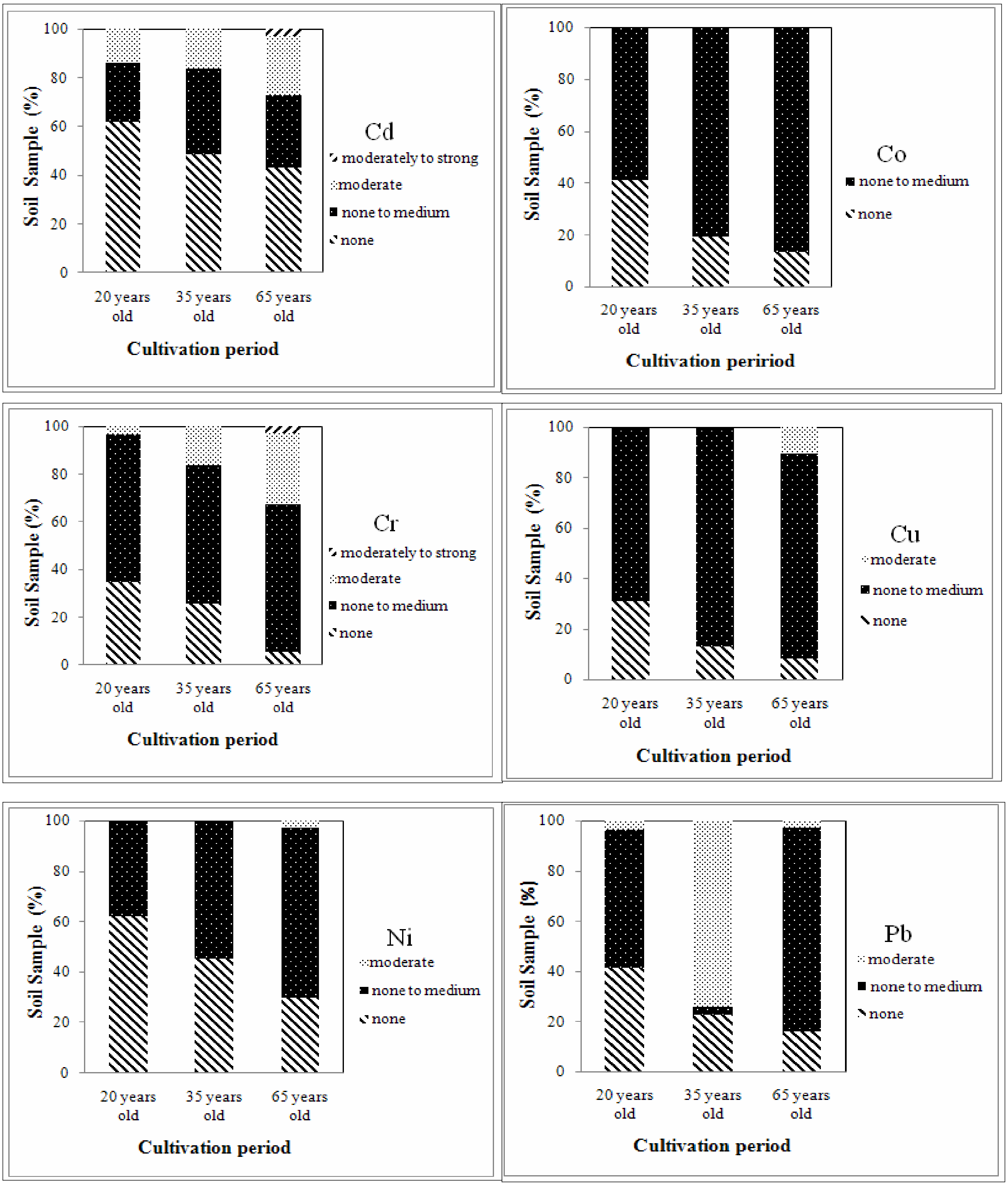

Figure 3. Contamination factor for heavy metals in agriculture lands.

Liu et al (2005) studied the effects of irrigation with sewage on pollution and heavy metals distribution in Beijing, the changes of Cd, Cr, Cu, Zn and Pb concentration in the three kinds of soils including cultivated soil, industrial park and a region not irrigated with sewage. They also calculated the amount of pollution load index (PLI), enrichment factor (EF) and contamination factor (CF). The results of EF indicated that the concentration of each metal in soil has an increasing trend (EF was more than 1). The contamination factor (CF) as well as pollution load index (PLI) values indicated the accumulation of these metals during the past 20 years.

Conclusions. The results indicated that the mean concentration of heavy metals in the fields with 65 year cultivation period has a significant difference from background concentration. The increasing trend of the metal concentration with the cultivation duration was obvious. The soil contamination factor in the lands under 65 years of cultivation was maximum. The increase of heavy metals concentration in the soil was followed by the increase in the use of fertilizers in the region. The cultivated soils are important ways for entrance of trace elements in the human's food chain. Therefore, monitoring contaminated soils is necessary to control and prevent the risks from contamination soils.

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