Mobility and Phytoavailability of Arsenic and Cadmium at the Soil-corn Interface in Sewage Irrigation Polluted Farmlands Northwest, China

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Abstract

The activity of heavy metals in farmland soil and the safety of food crops after long-term sewage irrigation are the premise of sustainable development of agriculture. The major pollutants of arsenic (As) and cadmium (Cd) in farmland-corn system affected by sewage irrigation for 40 years were studied. The results showed that although the content of total As in soil was high, the absorption and transport of total As by corn were relatively small, and As could not migrate down the soil profile. On the contrary, Cd mainly exists in the form of exchangeable and carbonate-bound which has strong fluidity. By optimizing the method of investigating the bioavailability of pollutants and systematically and comprehensively studying the migration law of pollutants in various parts of maize tissues, it is concluded that Cd still has high activity after stopping sewage irrigation, and the development process of maize roots has a significant relationship between the contact of heavy metals in the surface soil and the enrichment of heavy metals in the upper part of corn tissues ($p < 0.01$). Therefore, this study can increase the understanding of the migration law of pollutants in corn, and provide a basis for remediation of contaminated soil in local and other areas.

1. Introduction

Heavy metals pollution in agricultural soil has attracted worldwide attention due to its negative impact on food security and soil environmental quality. Among pollutants, As and Cd are toxic, universal and persistent (Xia, Peng et al. 2016). The sources of these pollutants has natural causes, such as parent material differentiation, but most of them come from agricultural activities such as industrial production, mining and transportation (Li, Liu et al. 2013).

Baiyin City is located on the northwest edge of the Loess Plateau, east of Qilian Mountain and the transitional zone to Tengger Desert. It is the largest industrial base of non-ferrous metals with various varieties in China. With abundant resources, the industrial economy is developing rapidly. During the process of mining, smelting and processing copper, lead, zinc and their associated minerals like Cd, As and other associated mineral resources are discarded at will in the form of waste residue or sewage (Du, Zhou et al. 2020). Once these wastes are disposed improperly, the surrounding environment will be polluted.

Dongdagou is a drainage ditch. It receives industrial wastewater and the domestic sewage from residents of Baiyin east district along the way. Eventually, it became the largest sewage channel in Baiyin and the largest source of heavy metals in the Yellow River basin. The original wastewater from the residents in the west area of the city is directly discharged into Xidagou, and finally flows into the Yellow River through Shuichuan town. The soil in this area is considered as an important farmland, because many crops including corn grow here.

Due to the local government and enterprises did not have enough sewage treatment funds, and the natural conditions of Baiyin City were limited by drought and little rain, the sewage from Dongdagou and Xidagou began to irrigate the farmland from 1960, and stopped in 2000, so the sewage irrigation lasted at less 40 years (Nan Zhongren 2002). Heavy metals in contaminated farmland and irrigation water can enter the plant through their root, causing phytotoxicity to some crops (Palansooriya, Shaheen et al. 2020). Concentrations of As and Cd in the edible part of the crop depend on the availability of As and Cd in the soil and the ability of the crop to absorb and transfer them to target organs (Balkhair and Ashraf 2016, Rai, Lee et al. 2019). The transfer of As and Cd from soil to edible plant is a key step for them to enter the human food chain (Ismael, Elyamine et al. 2019). Corn is a widely planted in this area. Corn is considered to be a digestible carbohydrate, which is rich in protein. In many rural areas, this grain provides nearly 70% of calories and 50% of intake protein. In addition, corn was commonly used as livestock feed (Nuss and Tanumihardjo 2010). After decades of sewage irrigation, it is unclear whether the local corn can be safely consumed by people and livestock.

Generally speaking, the migration and enrichment rule of heavy metals in crops is mainly through correlation analysis to analyze the relationship between heavy metals content and soil physical-chemical properties, as well as heavy metals content in crop roots, stems, leaves and fruits (Yang, Zhu et al. 2018). However, for corn with large biomass, the tissues are roughly divided into four parts, which can not accurately explain the migration characteristics of heavy metals from soil to tissues. Furthermore the study on the availability of heavy metals in soil is also facing challenges, and there are different opinions on how to improve the accurate expression availability of heavy metals. In this study, a field investigation with soil-corn system was conducted to research the accumulation and bioavailability of As and Cd in a long-time sewage irrigation farmland. A multiple linear regression model was established to predict the accumulation of As and Cd in grain, which provided reference for the prediction of As and Cd content in grain. Therefore this study will provide theoretical basis and data support for the safe use of agricultural land and the establishment of revised standards.

2. Materials And Method

2.1 Study area

The study area is located in the Dongdagou and Xidagou streams of Baiyin City, Gansu Province, the arid area in Northwest China (Shu and Deng-Rong 2004). The Dongdagou and Xidagou streams (average temperature of 9.2°C, annual precipitation is 244 mm, and the annual evaporation was 1871 mm) belong to the arid and semi-arid continental climate (Cao, Chen et al. 2016). This area is near the upper reaches of the Yellow River, and it is a major agricultural region. The Dongdagou and Xidagou streams grow corn, wheat and vegetables to provide food for residents (Liu, Ai et al. 2017). In recent years, with the start of ecological restoration projects of Dongdagou and Xidagou streams, the proportion of sewage irrigation has obviously decreased, and the Yellow River water has been used instead of irrigation.

2.2 Collection of soil and corn samples

Mature corn in Dongdagou and Xidagou farmland in Baiyin suburbs at the end of September 2019. 3 ~ 4 corn samples were collected at one point by the grid method (1×1m). A total of 45 samples were collected in Dongdagou and 34 from Xidagou. In principle, one sampling point was selected every 1km along the streams. The soil sample was located 0 ~ 20cm on the surface of corn field soil. After stirring evenly, take out 1.00 kg according to the quarter method to form a soil sample. 10 soil profiles and 8 soil profiles were collected every 2 kilometers along Dongdagou and Xidagou. The sampling depths were 1 ~ 10 cm, 10 ~ 20 cm, 20 ~ 30 cm, 30 ~ 40 cm and 40 ~ 50cm. A total of 95 soil profile samples were collected in plastic bag (Fig. 1). From bottom to top, corn samples were divided into ten parts: root, stem1, stem2 (decomposed on location of corncob), leaf1, leaf2, male inflorescence, grain, husk, corncob and corn stigma (Fig. 6).

2.3 Pretreatment of samples and determination of physical and chemical index

Brought the soil samples back to the laboratory, placed them in a cool place indoors, and dried them naturally. Gravel, plant and animal impurities in the soil sample were selected and then mixed evenly. Grinded it with a mortar and passed through 2 mm, 0.8 mm, and 0.149 mm nylon sieve. Put the soil in plastic bag and storaged in a cool place for future use. 0.3000 g of soil sample was weighed and put into the digestion tube, and then 10 mL of Aqua Regia (Hydrochloric acid: Nitric acid = 3: 1 v/v) was added for microwave digestion (Anton Paar, Multiwave PRO 3000). After digestion, the acid was driven by electric heating plate at 140°C for 2 h, filtered and fixed to 50 mL volumetric flask with ultrapure water. The As content in the soil sample was analyzed by atomic fluorescence spectrometer (AFS-2880) and used absorption flame spectrophotometer (iCE 3000 SERIES) to determine Cd content.

The plant samples were washed three times with distilled water to eliminate the influence of dust on the experimental results, and then placed in marked envelope. After natural air drying. Crushed the dried thickness samples with a crusher, and put in the marked valve bag for the next test. Weighed 0.5000 g of plant sample into the digestion tube, add 10 mL of Aqua Regia, digest with microwave digestion instrument, drive acid at 140°C for 1 h, filter, dilute to 25 mL with ultrapure water, and determine As and Cd content. All digestion procedures are 20 min to increase the temperature to 180°C, hold on 45 min, and cool down for 30 min.

The pH and EC were measured following the soil-water ratio of 1:2.5, using a pH meter (PHS-3E) and a conductivity meter (HI9811) (Zang, Wang et al. 2017). The content of organic matter was determined by the potassium dichromate oxidation-volumetric method, and the content of carbonate was determined by hydrochloric acid titration (Li, Wang et al. 2019). The soil particle size measured with a laser particle size analyzer (Mastersize 2000). The total salt content of soil solution was measured by the soil-water ratio of 1: 5 and a conductivity meter (HI9811) (Xu, Huang et al. 2013). Tessier five-step method was used to extract sequentially. There are five chemical constituents in soil, such as the exchangeable fraction (F1), the carbonate-bound fraction (F2), the Fe-Mn oxides-bound fraction (F3), the organic-bound fraction (F4) and the residual fraction (F5). (Tessier, Campbell et al. 1979). The bioavailability of heavy metals was evaluated by using DTPA single extraction method (pH = 7.3) (Feng, Shan et al. 2005).

2.4 Quality assurance and quality control

The loess GSS-8 (GBW 07408) reference material and Henan wheat GSB-24 (GBW 10046) reference material provided by China Academy of Metrology were used to evaluate each batch of samples (every 20 samples correspond to one blank and one standard). The recovery rate of element: As (99%±2.6%), Cd (95.5%±2.8%), which is repeated every 20 samples to ensure that the standard deviation is within 5%. All the glassware used in the experiment should be soaked in 10% nitric acid solution for over 24 h, and then washed with tap water and deionized water. The experimental reagents are all guarantee reagent.

2.5 Bioaccumulation and translocation factors

By calculating the migration of heavy metals from the soil to the plant, we can better understand the accumulation and migration capabilities of heavy metals in corn. (Adamczyk-Szabela, Lisowska et al. 2020).

$$W = a/b \quad (1)$$

$$D = c/d \quad (2)$$

W is the bioaccumulation factors, a is the heavy metals content in the plant (mg kg^{-1}), b is the heavy metals content in the soil (mg kg^{-1}); D is the translocation factors, and c is the heavy metals content in the above-ground of plants (mg kg^{-1}), d is the heavy metals content in underground of plants (mg kg^{-1}).

2.6 Statistical analysis

Microsoft Office2013 software was used to record the data, calculate the mean value, standard deviation, and coefficient of variation. SPSS22 was used to analyze the correlation of the data, and one-way analyses of variance (ANOVA).

3. Results And Discussion

3.1 Effect of sewage irrigation on physical-chemical properties of topsoil

Table 1 showed the physical-chemical properties and the concentration of As and Cd of topsoil. The content of As and Cd in Dongdagou were 4.93 ~ 557.55 mg kg^{-1} and 0.18 ~ 107.58 mg kg^{-1} with an average of 74.94 mg kg^{-1} and 20.18 mg kg^{-1}, compared with the risk screening values for soil contamination of agricultural land (CEPA 2018) there were 2.36 times and 67.27 times higher than the standard. Among them, As content exceeded the standard in 24 sampling points, with the exceeding standard rate of 52.17%. There were 44 sampling points where Cd exceeded the standard, and the over-standard rate was 97.72%. The content of As and Cd in the Xidagou were 6.11 ~ 19.44 mg kg^{-1} and 2.99 ~ 20.51 mg kg^{-1}, respectively, with an average of 14.09 mg kg^{-1} and 7.08 mg kg^{-1}. The Cd content exceeded the standard rate of 100%. Although As did not exceed the screening value, some samples exceed the soil background value of Gansu Province, which was still worthy of attention. In general, the pollution degree of Dongdagou was more serious than that of Xidagou.

Table 1
The physical-chemical properties of topsoil and the average concentration of As and Cd.

	Dongdagou(n = 45)				BV[a]	RSV[b]	*Xidagou*(n = 34)			
	MIN	MAX	AVG	CV(%)			MIN	MAX	AVG	CV(%)
As($mg \cdot kg^{-1}$)	4.93	557.55	74.94	124	12.60	30.00	6.11	19.44	14.09	22.06
Cd($mg \cdot kg^{-1}$)	0.18	107.58	20.18	124	0.12	0.30	1.58	13.96	4.88	50.32
DTPA-Cd($mg \cdot kg^{-1}$)	0.26	26.50	5.30	112.53	-	-	0.06	7.57	2.01	86.80
DTPA-As($mg \cdot kg^{-1}$)	0.01	12.37	2.13	142.69	-	-	0.01	0.49	0.17	63.84
Clay < 0.002mm(%)Slit0.002mm-0.02mm（%）	7.38	9.70	8.21	8.83	-	-	2.35	11.79	6.496	38.59
	27.61	43.39	34.689	16.40	-	-	9.49	49.75	25.52	41.09
Sand > 0.02mm(%)	49.18	64.95	57.103	10.27	-	-	38.46	88.16	67.98	19.06
pH	7.12	8.55	7.98	3.53	8.40	-	7.56	8.07	7.85	2
EC($\mu s \cdot cm^{-1}$)	326.00	4170.00	960	83.23	-	-	213.00	927.50	431.99	35
TS($g \cdot kg^{-1}$)	0.44	5.23	1.34	89.81	-	-	0.53	2.43	0.84	45.18
$CaCO_3$(%)	5.89	14.29	11.26	16.39	11.8	-	3.42	12.67	9.07	23
SOM($g \cdot kg^{-1}$)	2.44	42.31	21.55	35.78	9.40	-	1.03	37.64	15.74	50

[a] Refer to the background value of soil elements in *Gansu* province.

[b] Refer to the risk screening value for soil contamination of agricultural land ($6.5 < pH \leq 7.5$) (Trial implementation). '-'Represents did not mention in the passage; 'TS' represents the total salinity of the soil solution; 'SOM' represents soil organic matter; 'EC' represents electrical conductivity; 'CV' represents coefficient of variation; 'MIN' represents minimal value; 'MAX' represents maximal value; AVG represents mean value.

The pH value of Dongdagou was between 7.12 ~ 8.55, the average value was 7.98. In Xidagou was between 7.56 ~ 8.07, the average value was 7.85. Background value was 8.40, and the soil pH value has dropped. This may be a long-term acid wastewater irrigation and fertilization (Li, Wang et al. 2019). According to reports, after chemical fertilizers were applied to farmlands all over the world, the pH value of the soil dropped severely, and acidic wastewater irrigation was a key factor (Kunhikrishnan, Bolan et al. 2012). Soil acidification will lead to the dissolution of carbonate and the release of pollutants so the carbonate content of Dongdagou and Xidagou is 11.26% and 9.07%, respectively, which is lower than the background value of 11.8%.

The content of soil organic matter was higher than the background value. This was due to local agricultural customs. Every winter, before fertilization, fertilizers was accumulated in farmland. Application of organic fertilizer can increase soil fertility, while organic matter can retain more heavy metals due to its powerful supplementary ability (Violante, Cozzolino et al. 2010). The EC in the soil shows its salt content. The high salinity in soil directly leads to the inability of crops to grow (Shrivastava and Kumar 2015). Under the conditions of less rainfall and large evaporation in northwest China, the accumulation of salt on the soil surface will increase the salt content and pH value, and in some areas will cause salinization. The highest value of total salinity in Dongdagou reaches 5.23 g kg^{-1} belonging to strong saline soil, and the average value of 1.34 g kg^{-1} belonged to mildly saline soil. The average value of total salinity in Xidagou was 0.84 g kg^{-1} belonged to non-saline soil, but the maximum value of 2.43 g kg^{-1} belonged to moderate saline soil, indicating that some land has begun to be salinized. Therefore, the salinity of Dongdagou is more serious than that of Xidagou. This may be due to the high salinity of sewage irrigation water, resulting in the accumulation of topsoil in farmland (Angin, Yaganoglu et al. 2005). This will bring adverse consequences, such as soil compaction and fertility decline, as well as adverse effects, such as crops are difficult to absorb nutrients, hindering crop growth (Chojnacka, Witek-Krowiak et al. 2020).

According to the grain size distribution of Dongdagou and Xidagou, the soil structure is mainly composed of mineral loam and loam. The distribution of soil is affected by its service life and spatial location. Related studies show that the longer the service life, the finer the soil composition (Shepherd, Saggar et al. 2001). In addition, farmland vegetation coverage is different, and it is located in different areas affected by main wind direction all year round, so the responses to wind erosion is different (Zuazo and Pleguezuelo 2008). Compared with farmland, the volume content of clay and slit particles in the upper reaches of Dongdagou wasteland was reduced by 8.32%~23.38% and 15.19%~30.24% also confirmed this point, so ensuring long-term and reasonable farming management methods will help stabilize and improve soil grain size distribution.

3.2 Bioavailability evaluation of As and Cd in topsoil

Studies have shown that the content of Cd in grains is mainly related to the bioavailability of Cd in the soil, but not to the total amount of Cd in the soil (Kim, Kim et al. 2016). Therefore, we need to know the distribution of Cd in various sewage irrigation area. In the (Fig. 2A-B) Cd mainly existed in carbonate-bound fraction (F2) and exchangeable fraction (F1). The total amount of these two forms exceeded 50% in Dongdagou and approached 60% in Xidagou. Exchangeable Cd is mainly composed of Cd^{2+}, $CdCl^{+}$, $CdSO_4$ and $CdHCO_3^{+}$ (Filipović, Romić et al. 2018). These ions are easily absorbed and enriched by plants. The next fractions were Fe-Mn oxides-bound fraction (F3) representing the reduced form, organic-bound fraction (F4), and residual fraction (F5), which all were not easily dissolved under natural conditions and were sealed in the crystal lattice structure of soil minerals (Tessier, Campbell et al. 1979). Cd content in F5 of some sampling points in Dongdagou is low. In this study area, the exchangeable fraction and the carbonate-bound fraction accounted for a relatively high proportion, so there were higher bioavailability and ecological safety risks (Zhao, He et al. 2020).

Arsenic in the soil mainly exists as residual fraction (F5), accounting for 84.32% ~ 96.98% (Fig. 2C-D), which could explain why the total As content in soil is high, while the As content in the corn plants is low (Fig. 5). Exchangeable parts can be released and transported by dissolution, ion exchange and desorption, which has great influence on soil and plants. In this study, the exchangeable fraction account for the smallest proportion of total As content in the soil, but they could migrate with surface water and interstitial water. Poisoning occurs after drinking or being absorbed by plants and animals (Ruíz-Huerta, Varela et al. 2017). The reduced state is mainly in the form of Fe-Mn oxides-bound fraction. The proportion of Xidagou is around 5%, and Dongdagou is close to 10%. Under the influence of soil pH, Eh and some iron-reducing microorganisms, this part can be transformed into exchangeable As (Masscheleyn, Delaune et al. 1991).

The amount of DTPA extracted from metal elements is regarded as an effective forms of heavy metals (Lee, Park et al. 2011). The DTPA extraction from Dongdagou and Xidagou have a good linear relationship between summation of exchangeable and carbonate-bound fraction (Fig. 3). Some studies have concluded that the sum of exchangeable and carbonate-bound heavy metals can be regarded as an effective state for plants to absorb and utilize (Nie, Yang et al. 2018). We have found a simpler and more efficient method, that is, to judge the bioavailability and toxicity of heavy metals in this area by single-step extraction.

3.3 Distribution of As and Cd in profile

Under the effects of farming, leaching, etc., the heavy metals accumulated in the soil will migrate horizontally and vertically, making it show different accumulation characteristics in the profile soil (Bongoua-Devisme, Balland-Bolou-Bi et al. 2018). The research shows that heavy metals accumulation in soil profile is affected by pollution sources. For example, there was a significant bottom-layer enrichment of heavy metals in the soil near the Urad Houqi mining area (Song, Li et al. 2018). Profile distribution can reflect the accumulation of heavy metals pollution in farmland. Comparing Fig. 4A-B, although Cd pollution in Xidagou is lighter than that in Dongdagou, it still exceeds the screening value of soil risk at 50cm of soil profile. However, in Dongdagou, the soil at the surface layer of 20 cm exceeds the risk control value, but it is lower than the risk screening value at 50 cm, which is clean soil. Therefore, when investigating the pollution of farmland in long-term polluted irrigation areas, only collected the soil at the surface layer of 20cm can not represent the pollution degree, and it is necessary to collect the profile soil over 50cm for further study.

Xidagou profile of As (Fig. 4C) basically within the safe range, there is no pollution and there is no obvious correlation with depth. On the contrary, Dongdagou (Fig. 4D) (PE1, 2, 5, 6, 7) exceeds the risk screening value of 30 mg kg^{-1} (solid blue line) at 0 ~ 30cm, and PE5 and PE7 surface soil 0 ~ 20cm exceeded the risk control value of 120 mg kg^{-1} (solid red line), indicating that the point source pollution at the middle and upper reaches profile of Dongdagou has surface accumulation phenomenon, and the pollution was serious. At the same time, pollution was concentrated in the topsoil (10 ~ 30cm), which indicates that the mobility of As is not high.

The soil in this study area was sierozem, with mineral loam and loam structure, so the soil was loose and sandy. When the concentration of Cd was less than 3 mg kg^{-1}, the migration of Cd in the soil was more intense (Fig. 4A). On the contrary, because of the competition and exchange of ions (Table 1), the migration becomes smaller with the increase of soil depth (Fig. 4B). The results showed that the improvement of Cd availability will not improve the migration capacity of Cd in the soil if it did not exceed the adsorption capacity of Cd in the soil (Filipović, Romić et al. 2018). When the As concentration is less than 10 mg kg^{-1}, there is no obvious change in As concentration with the increase of soil depth (Fig. 4C). The main reason is that the concentration is within the range of soil background value (Table 1). On the contrary, arsenic is mainly concentrated in the soil with a depth of 10 ~ 30cm (Fig. 4D). With special environmental conditions conductive to the attenuation of As. Therefore, ionic As exists in a stable solid residual state (Wang and Mulligan 2006). At the same time, during irrigation process, the concentrations of As and Cd mainly migrate in the form of suspended solids, and the purification effect of heavy metals in the upper middle reaches is shorter than that in the lower reaches. Therefore, they accumulate on the surface in the middle and upper reaches of the sewage irrigation. Due to self-purification, the concentrations of As and Cd in sewage decrease, and so do the downstream (de Santiago-Martín, Meffe et al. 2020).

3.4 The accumulation of As and Cd in different parts of corn

The range of As and Cd contents in Dongdagou grain was 0 ~ 0.265 mg kg^{-1} and 0.001 ~ 0.503 mg kg^{-1}, the average value was 0.038 mg kg^{-1} and 0.095 mg kg^{-1} which were lower than the national food safety standard maximum levels of contaminants in food limit of As 0.5 mg kg^{-1} and Cd 0.1 mg kg^{-1} (CEPA 2017). However, the Cd of some points in Dongdagou exceeded the standard, with an over-standard rate of 32.35% but As is safe. The content of As and Cd in Xidagou grain ranged from 0.013 to 0.097 mg kg^{-1} and 0.01 to 0.138 mg kg^{-1}, with the average values of 0.033 mg kg^{-1} and 0.051 mg kg^{-1}, respectively. 12.90% of samples exceeded the standard, and As was also safe.

In related research, the migration coefficient from roots to grain in sandy loam was 3.2%~4.7% (this study was 4%), which is not high (Table 2) (Lottermoser 2012, Yadav, Minhas et al. 2015, Meng, Wang et al. 2016, Farahat, Galal et al. 2017, Xue, Zhao et al. 2019). It also showed that the self-protection mechanism of corn tissue makes it difficult to accumulate in the grains, so the edible part of contaminated soil is within a safe range (Fig. 5). Some pollutants can enter plants through stomata of leaves. Biological monitoring of plants has been used to monitor chromium, nickel, copper, zinc, arsenic, cadmium and lead in the atmosphere (Suvarapu and Baek 2016). Contaminants enter the leaves through soil-root-leaf, air-leaf and stem-leaf, which can explain why As and Cd content in leaves are higher than other organs (Fig. 5) (He, Wang et al. 2021). But in this study, there is no direct evidence that the enrichment of heavy metals in leaves mainly comes from air pollutants. The content of As and Cd in corn inflorescence was higher than that in other parts (except roots and leaves). The main reason was that the growth period of male inflorescence of corn is short (10-15d), and a lot of nutrients are needed in a short time during the growth process. At the same time, pollutants can also enter the male inflorescences with nutrients (Li, Wang et al. 2019). By the transpiration, the metal ions in the plant were transported upwards, and eventually reached the male inflorescence and accumulated. Pollen development of corn male inflorescence was important, and any abnormality will affect the yield of corn. Previous studies have shown that heavy metals can cause nutrient deficiency during the development of microspores, so reducing As and Cd transport in corn plays an important role in corn yield (Mohsenzadeh, Chehregani Rad et al. 2011).

Table 2
Comparison of As and Cd concentrations in sewage irrigated soil of different regions.

Site	As concentration range (mg·kg^{-1})	Cd concentration range (mg·kg^{-1})	pH	SOM (%)	TF	BCF	Soil types	Sewage irrigation time (a)	Data source
Berlin, Germany	0.7–4.1	0.14–31.5	3.93–6.92	-	-	-	Sandy soil	110	(Lottermoser 2012)
Faridabad, India	-	1.38–3.24	8.2 ± 0.3	2.4 ± 0.4	Cd < 1	-	Sandy loam	25	(Yadav, Minhas et al. 2015)
Hebei, China	8.83–19.7	0.21–1.19	7.91–8.33	-	-	Cd < 1 As < 1	-	50	(Xue, Zhao et al. 2019)
Cairo, Egypt	-	0.22 ± 0.01	7.21 ± 0.02	-	Cd < 1	Cd > 1	-	20	(Farahat, Galal et al. 2017)
Tianjin, China	5.14–19.7	0.05–1.17	8.44 ± 0.44	2.66 ± 0.32	-	Cd < 1 As < 1	-	35	(Meng, Wang et al. 2016)
Gansu, China	4.93-557.75	0.18-107.38	7.12–8.55	1.87 ± 0.35	Cd > 1 As < 1	Cd < 1 As < 1	mineral loam and loam	40	This study

By comparing the transfer coefficient of aboveground parts of plants (Table 3), it can be seen that the Cd (TF > 1) is easier to transfer in tissues than As. According to the bioaccumulation factors (BCF < 1), As and Cd in roots can not accumulate from soil (Table 2). This shows that the interaction between the root system and metals is decisive factor. The average content of Cd in different parts of the corn is in order of: root > leaf > male inflorescence > husk > stem > corn stigma > corncob > grain and the average As content in order: root > leaf > male inflorescence > husk > stem > corn stigma > grain > corncob. Generally speaking, two kinds of pollutants are easy to migrate and accumulate in leaves and male inflorescences.

Table 3
Bioaccumulation and translocation factors of As and Cd in different maize organs.

		BCF/TF (Average ± S.D.)									
		R/Soil	S 1/R	S 2/R	L 1/R	L 2/R	MI/R	H/R	C/R	CS/R	G/R
W	As	0.08 ± 0.09	0.22 ± 0.19	0.12 ± 0.10	0.83 ± 0.60	0.66 ± 0.54	0.40 ± 0.56	0.25 ± 0.25	0.01 ± 0.04	0.11 ± 0.26	0.04 ± 0.19
	Cd	0.11 ± 0.13	0.48 ± 1.30	0.47 ± 0.56	0.83 ± 1.00	1.11 ± 1.73	1.19 ± 1.71	0.62 ± 1.58	0.24 ± 0.29	0.32 ± 0.33	0.13 ± 0.20
E	As	0.10 ± 0.08	0.24 ± 0.29	0.16 ± 0.18	1.14 ± 0.97	0.88 ± 0.69	1.02 ± 0.84	0.33 ± 0.59	0.01 ± 0.01	0.27 ± 0.37	0.04 ± 0.04
	Cd	0.09 ± 0.06	0.31 ± 0.27	0.57 ± 0.41	2.92 ± 2.72	2.61 ± 2.40	2.28 ± 2.68	0.74 ± 0.70	0.37 ± 0.42	1.20 ± 1.39	0.30 ± 0.30

BCF: bioaccumulation factors; TF: translocation factors.

W: *Xidagou*; E: *Dongdagou*.

R: Roots; S 1: Stem 1; S 2: Stem 2; L 1: Leaf 1; L 2: Leaf 2; MI: Male Inflorescence; H: Husk; CS: Corn Stigma; C: Corncob; G: Grain.

Stems and leafs are not directly eaten by human beings, but used as corn silage, which can be eaten by livestock (Li, Wang et al. 2019). According to the feed hygiene standards (SAQSIQ 2017), the limits of As and Cd in plant feed ingredients are 4 mg kg^{-1} and 1 mg kg^{-1}, respectively. In the corn tissues of Dongdagou and Xidagou, the Cd in feed exceeded the standard in turn, and the Cd exceeding standard rate of leaf 1 and leaf 2 of Xidagou corn were 17.65% and 14.70% respectively (Fig. 5). The over-standard rates of stem 1, stem 2, leaf 1, leaf 2, male inflorescence and husk corn in Dongdagou were 10.26%, 7.69%, 15.38%, 30.77%, 30.56% and 7.69%, respectively, and the highest exceeded the standard by 3 times. Cd pollution is serious in Dongdagou, which poses a threat to human health through the food chain. Therefore, it is necessary to strengthen the safe utilization of land and ensure the safety of feed (Zhou, Chen et al. 2018, Li, Wang et al. 2019).

3.5 Relationships between As or Cd with different maize organs.

The above research shows that the content of As and Cd in corn tissues is related to the physical -chemical properties of soil. From Table 4 we know that soil pH, SOM and others factors has a bad pearson correlation coefficient with maize organs. But the correlation between grain As or Cd and SOM showed significant positive correlation at $P< 0.05$. Because organic matter can provide organic chemicals substances for soil solution, and can also be used as chelating agent to improve the utilization rate of metals by plants (Vega, Covelo et al. 2004).

Table 4
Pearson correlation coefficients for metal concentrations in different corn tissues, soil metal contents and soil properties.

	R	S1	S2	L1	L2	M	H	G	B
pH	NS	NS	NS	**-0.567***	**-0.497***	NS	NS	NS	NS
SOM	NS	NS	0.55*	NS	NS	NS	NS	**0.564*** (0.479*)	NS (0.58**)
DTPA-As	**0.502*** (0.689**)	**0.613**** (0.781**)	**0.761**** (0.598**)	(0.495*)	**0.554*** (0.754**)	**0.709****	**0.563*** (0.62**)	(0.524*)	**0.495*** (0.666**)
DTPA-Cd	**0.738**** (0.678**)	NS	**0.66****	(0.503*)	**0.572*** (0.751**)	**0.71****	NS	(0.5*)	NS
As	(0.55*)	**0.595**** (0.509*)	**0.81****	NS	(0.641**)	NS	NS	**0.493*** (0.757**)	**0.458*** (0.929**)
Cd	**0.472*** (0.589**)	**0.538*** (0.755**)	**0.521*** (0.611**)	(0.593**)	**0.623**** (0.655**)	**0.716****	**0.674**** (0.77**)	NS	NS
P1	(0.489*)	NS	**0.71****	NS	(0.694**)	NS	NS	**0.848**** (0.616**)	**0.593**** (0.554*)
P2	NS	NS	**0.543***	NS	(0.626**)	NS	NS	**0.633**** (0.5*)	NS
P3	NS	NS	**0.474***	NS	(0.535*)	NS	NS	(0.508*)	NS

Bold represents the correlation coefficients of Cd, and the other stands for As. P1, P2 and P3 is heavy metals content in soil profile at 0-10cm, 10-20cm and 20-30cm.

** $P< 0.01$.

* $P< 0.05$.

NS: not significant.

Although the content of total As, total Cd and DTPA extracted from corn surface soil (0-20cm) are significantly related to corn root, stem 1, stem 2 and leaf 2 ($P< 0.05$), it only show that the migration of heavy metals from soil to corn is in the final stage from germination to maturity, and its migration rule can not be accurately expressed. Heavy metals were absorbed by maize and accumulated in the body during the whole development process. We introduced the profile soil around corn (distance = 30cm) as a control to simulate corn absorption characteristics of As and Cd during returning green stage, jointing stage, heading stage and mature stage (total 180d) (Fig. 6). We found that As and Cd in the soil layer were significantly correlated with the upper tissues such as stem 2 and leaf 2 of corn ($P < 0.01$) (Table 4). There is a significant correlation between 0–10 cm topsoil and the heavy metals in grains ($P < 0.01$), which indicates that during the growth of corn, with the development of roots system, a large number of nutrient elements and harmful heavy metals will be absorbed, and finally accumulated in the upper tissues of maize due to transportation and transpiration in plants. Therefore, the content of heavy metals in the topsoil is the key to its enrichment in corn, and the topsoil comes into contact with and absorbs elements at the earliest time during the development of maize roots.

The relationship between the content of heavy metals in grain and the content of heavy metals in soil and their physical-chemical parameters was studied by multiple regression analysis (Table 5). The results of correlation analysis (Table 4) are different from those of multiple regression analysis (Table 5). The results indicated that the absorption of As and Cd by corn grains under field conditions was mainly affected by the physiological characteristics of crops (Zang, Wang et al. 2017). However, the effects of As and Cd absorption and transportation from soil to grains are still worth further study and exploration.

Table 5
Multiple regressions between the concentration of metals in corn grains, the concentration of metals and their physico-chemical parameters in the soil.

	Multiple regressions equation	R^2
Grain	$As = 0.086 - 0.06[pH_S]-2.801[EC_S]-0.104[CaCO_{3S}] + 0.148[SOM_S] + 2.732[TS_S]$	0.937**
	$+ 1.168[DTPA\text{-}As_S] + 0.281[DTPA\text{-}Cd_S]-0.47[As_S]-1.13[Cd_S] + 0.619[P1_S]-0.611[P2_S]$	
	$+ 0.82[P3_S]-0.59[P4_S] + 0.193[P5_S]$	
Grain	$Cd=-0.861 + 0.799[pH_S] + 1.176[EC_S]-0.008[CaCO_{3S}]-0.438[SOM_S]-1.114[TS_S]-1.567[DTPA\text{-}As_S]$	0.942**
	$+ 0.742[DTPA\text{-}Cd_S] + 2.551[As_S] + 0.977[Cd_S] + 0.057[P1_S] + 1.349[P2_S]-2.293[P3_S]$	
	$+ 2.667[P4_S]-1.41[P5_S]$	
$[X_S]$ denotes material content in soils.		
** $P < 0.01$.		

4. Conclusion

Long-term sewage irrigation leads to the intensification of soil salinization. At the same time, the average value of As and Cd in the soil of the study area is higher than the national risk screening value, and some soils exceed the risk control value of the quality and safety of edible agricultural products. Arsenic in soil mainly exists in residual state, while Cd exists in the exchange state and carbonate-bound state. The bioavailability of As and Cd in soil can be predicted by single-step extraction DTPA method. The mobility of Cd in profile soil is stronger than that of As. Arsenic is not contaminated in corn tissues, while Cd is contaminated in leaves, stems, male inflorescence and grains. The correlation between the enrichment and migration characteristics of As and Cd in maize tissues with soil physical-chemical properties and heavy metals content shows that soil physical-chemical properties have little effect on the enrichment of heavy metals in corn. The transfer ability of Cd in corn tissue is greater than that of As, and heavy metals can not accumulate in roots. The content of heavy metals in topsoil is the key to entering into upper tissues during the growth and development of corn. After remediation of heavy metals polluted soil, reasonable irrigation and fertilization can reduce the absorption of heavy metals by corn, thus ensuring the safety of food and feed.

Declarations

Ethics approval and consent to participate

Not applicable

Competing interests

The authors declare that they have no competing interests.

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Consent for publication

Not applicable

Availability of data and materials

The datasets used or analysed during the current study are available from the corresponding author on reasonable request.

Authors' contributions

Xiang Ning: Investigation, formal analysis, measurement, graph drawing, writing-original draft. Shengli Wang: Conceptualization, Methodology, validation, Resources, Writing-Original Draft, Writing- Review & Editing, Supervision. Yi Wu: Investigation, formal analysis. Mengbo Liu: Pretreatment, Measurement. Meng Yang: Pretreatment, Measurement. Yuqing Wang: Measurement, Graph Drawing. Zhongren Nan: Writing- Review & Editing, validation.

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Figures

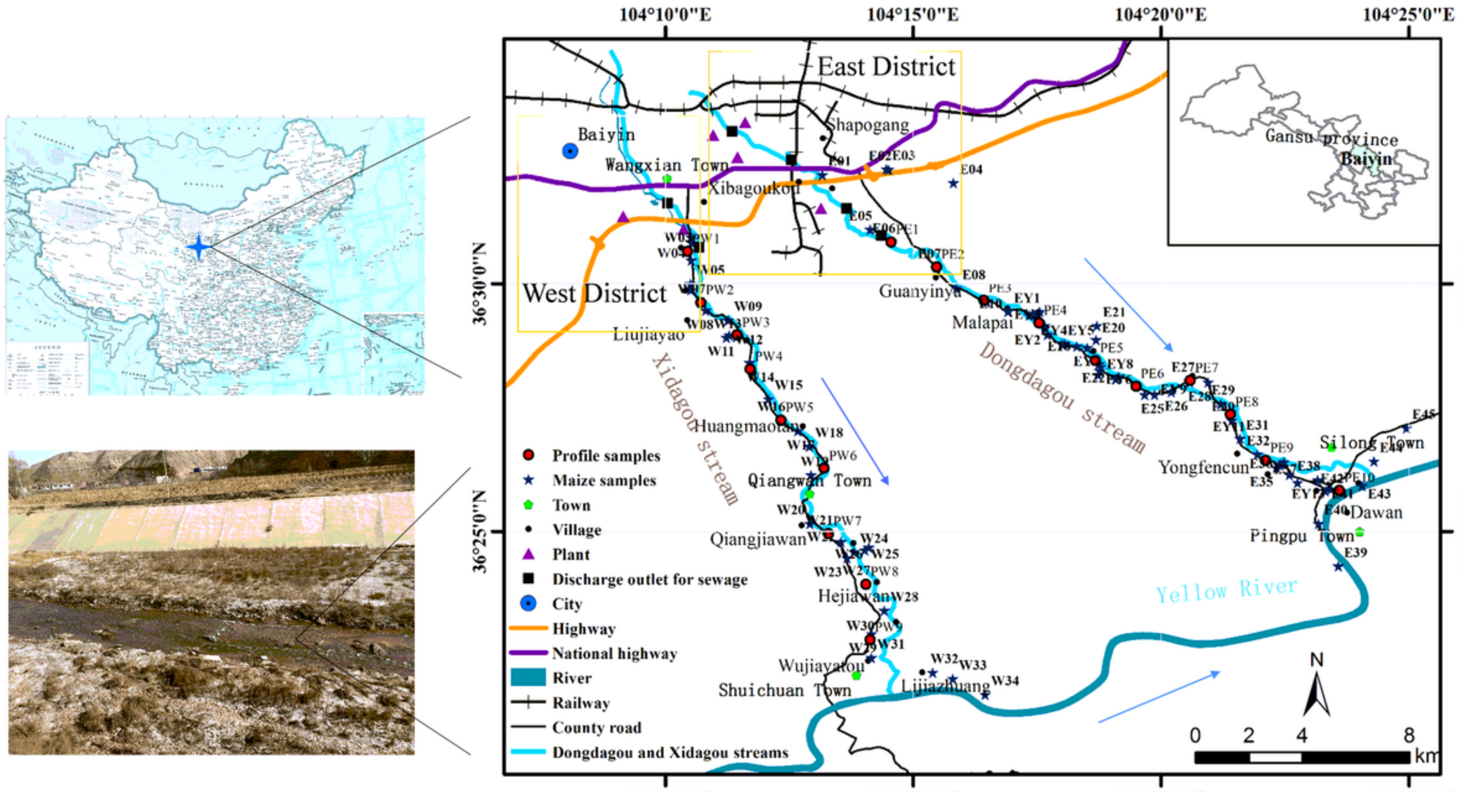

Figure 1

Location of the sampling site.

Figure 2

As and Cd fractionation in the surface layers of Xidagou and Dongdagou corn field. (A) is Cd fractionation in Xidagou soil (n=33), (B) is Cd fractionation in Dongdagou soil (n=45), (C) is As fractionation in Xidagou soil (n=33), (D) is As fractionation in Dongdagou soil (n=45). F1 is the exchangeable fraction, F2 is the carbonate-bound fraction, F3 is the Fe-Mn oxides-bound fraction, F4 is the organic-bound fraction, F5 is the residual fraction. Red line represents extraction by DTPA method (mg kg-1).

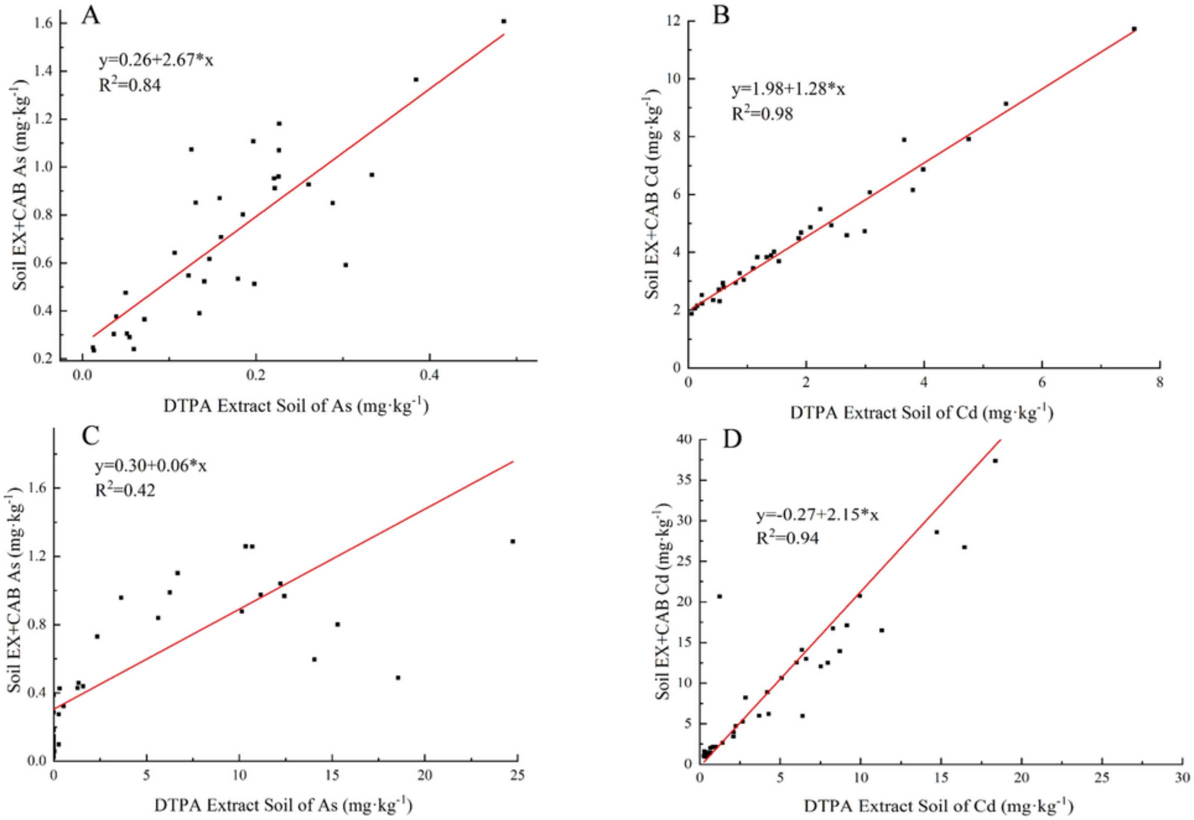

Figure 3

Comparing analyses of four kinds of linear fitting models for multi-group data. A and B is soil in Xidagou, C and D is soil in Dongdagou, EX is Exchangeable fraction in soil, CAB is Carbonate-Bound fraction in soil.

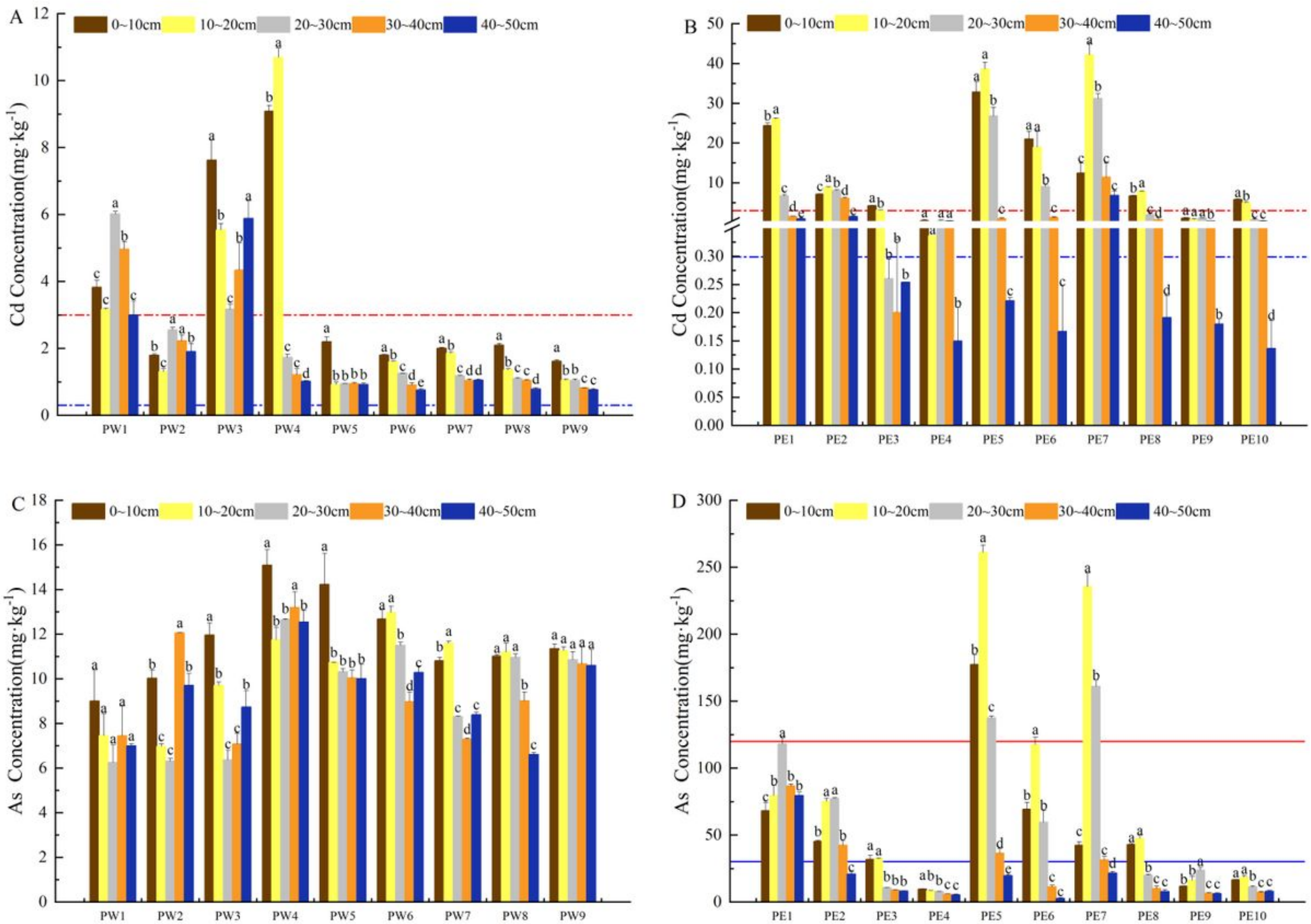

Figure 4

The profile distribution of As and Cd. A and B the profile distribution of Cd in Xidagou and Dongdagou, C and D the profile distribution of As in Xidagou and Dongdagou. A and B: blue dotted line is risk screening values for soil contamination of agricultural land of Cd 0.3 mg kg-1, red dotted line is risk intervention values for soil contamination of agricultural land of Cd 3 mg kg-1. D: The blue lines is risk screening values for soil contamination of agricultural land of As 30 mg kg-1, the red lines is risk intervention values for soil contamination of agricultural land of As 120 mg kg-1. Different lowercase letters indicate significant differences in depth and heavy metal concentration at $p < 0.05$. Error bars represented standard deviation (n = 3).

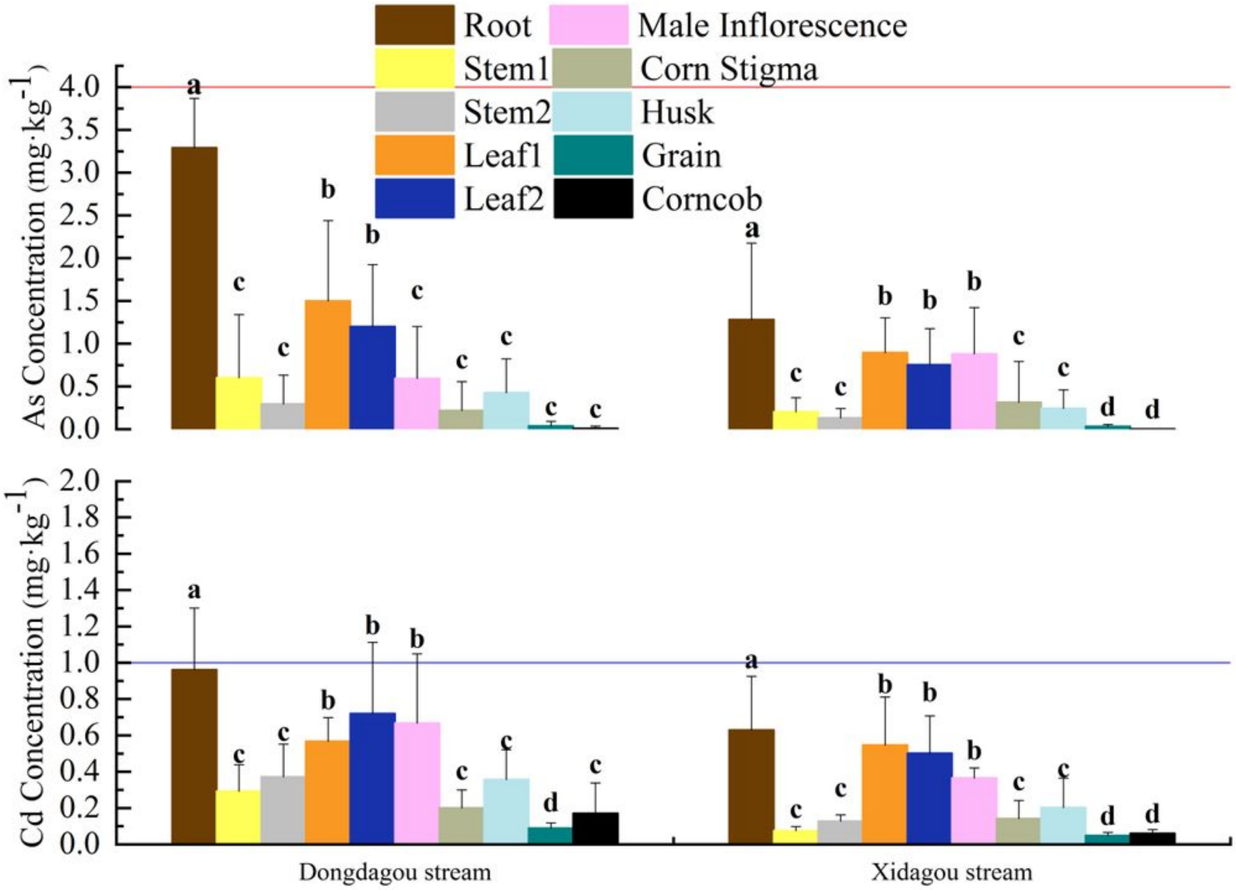

Figure 5

Average contents of As and Cd in corn tissue. Different letters on the columns indicated significant statistically differences among different parts of corn tissue at $p < 0.05$. Error bars represented standard deviation (n =45 in Dongdagou and n = 34 in Xidagou). Red line is hygienical standard for feeds limit value of As and blue line is limit value of Cd.

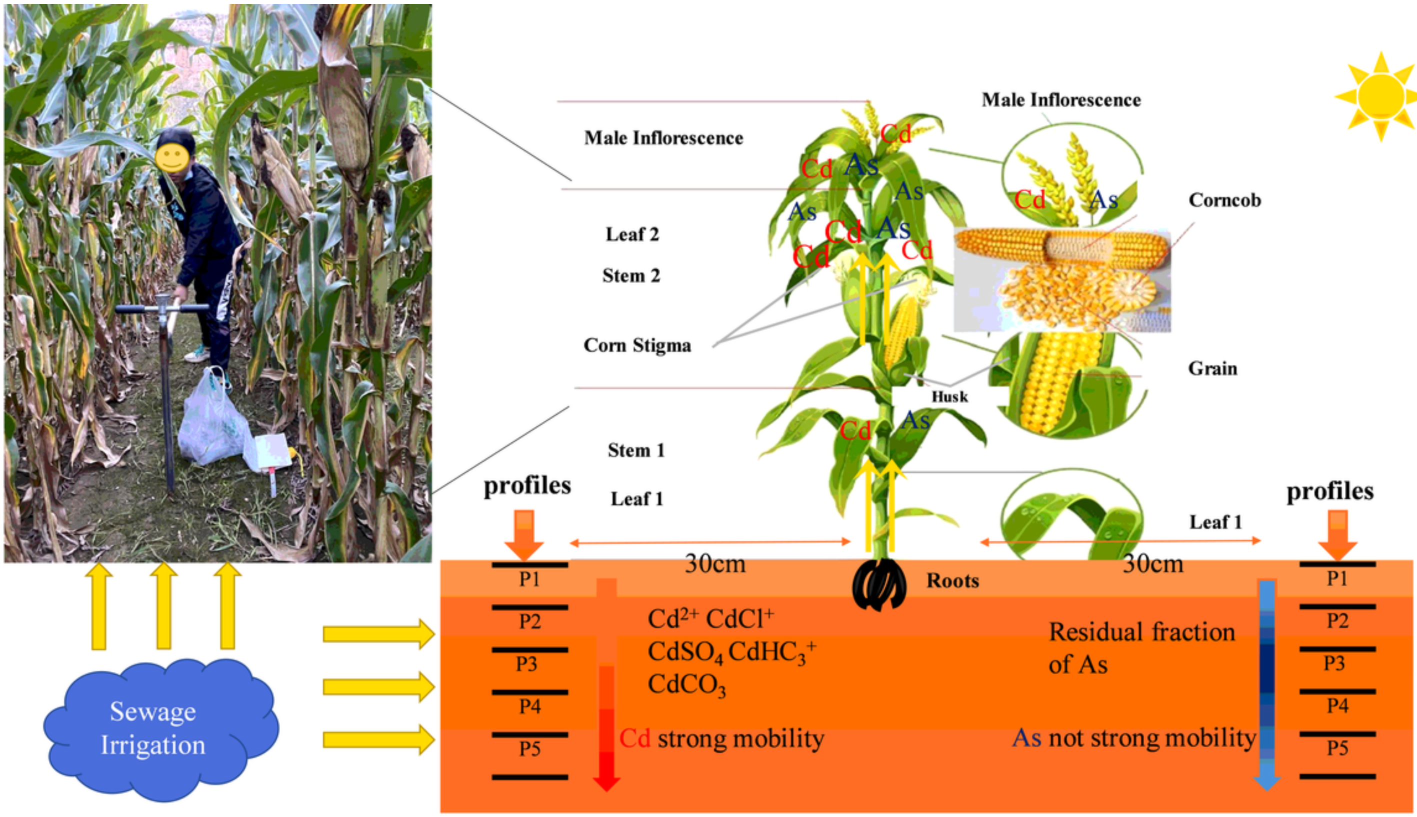

Figure 6

Migration characteristics of As and Cd in corn and soil.

Supplementary Files

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