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Distribution Characteristics of Heavy Metal Pollution in Soil Under Recycling Mode of Direct Straw Manuring

Leiming Fu, Chunxia He† and Yifei Chen
Nanjing Agricultural University, Nanjing, Jiangsu, 210031, China
†Corresponding author: Chunxia He

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ABSTRACT

A large number of straw resources are inefficiently used in China. Direct straw manuring is the most common method for efficiently utilizing these resources. The migration of nutrients and the transfer of hazardous elements such as heavy metals during the cycle, endanger the growth of crops, thereby decreasing food production and adversely affecting human health through the food chains. The mechanism of heavy metal-polluted soil under the recycling mode of direct straw manuring is analysed to determine its distribution characteristics. The concentration distribution characteristics of heavy metals such as Pb, Cr, Cd and As in the soil are identified through experiments. Empirical study shows that the contents of the four heavy metals in the soil of the wheat-maize rotating farmland under direct straw manuring mode do not exceed the values listed in the Environmental Quality Standards for Soils (GB15618-1995). The contents of Cr and Cd in the roots of wheat and maize are significantly higher than those in the soil, suggesting the accumulation of these metals in the roots, while those of Pb and As do not have this phenomenon. The contents of the heavy metals in the roots, stems, leaves and seeds of the crops decrease in order. The pollution of the heavy metals including Pb, Cr, Cd, As under the direct straw manuring is explored, and their migration rules in the farmland ecosystem are systematically explored. The results provide a basis for regulating and blocking heavy metals, preventing heavy metal pollution and safeguarding food safety.

INTRODUCTION

In the past 20 years, with the rapid improvements in reform and opening up processes and the advancement in urbanization, industrialization and modernization, the pollution in rivers, suburbs and mining areas has intensified; the number of food production areas affected by heavy metal pollution has increased annually. Grain output continues to increase, as shown in Fig. 1, but heavy metal pollution products are also increasing. Heavy metals enter the soil through atmospheric precipitation, discharge of industrial wastewaters and slag and other means, and are absorbed by soil colloids through complexation or precipitation. These metals are not easily degraded or eliminated, thereby accumulating in soil. Heavy metals in the soil are absorbed by agricultural products, penetrate the human body through the food chain, and endanger human health. In the agricultural cycle, heavy metals in soils are absorbed by plants through respiration and ion exchange in the roots, which remain in the soil and return to the earth after the plants mature. The straws are used to cultivate edible fungi or as fodder to feed livestock. Fungal slag and the livestock waste are thrown into the farmland as fertilizers. In this study, the heavy metals tested, belong to the agricultural cycle part. The crop seeds will be consumed by humans, and the meat and milk produced by mature edible mushrooms and livestock will be a part of the food for humans. In the accumulation of heavy metals in the roots, stems, leaves and seeds of crops, whether the residual content exceeds the standard of food hygiene has become a research hotspot. The migration of heavy metals in soil and the relevant pollution restoration has also gained increasing attention. To protect the agricultural ecological environment and maximize the use of high-end and modern technologies, scholars recommend the following to achieve the virtuous circle of ecology and the sustainable development of agricultural economy: Adjustment and optimization of the internal and industrial structures of the agricultural ecological system, improvement of the multistage cyclic utilization of matter and energy of agricultural system, exert strict control over the input of external hazardous substances and production of agricultural wastes, minimize the pollution of agricultural production and life to the environment, and fully integrate clean production technology and efficient matter and energy cycle into agricultural ecological system cycle.

In this study, the distribution characteristics of heavy metal pollution in the soil under the mode of direct straw manuring (straws of old crops in the wheat-maize rotating field is returned as manures for new crops) are analysed.

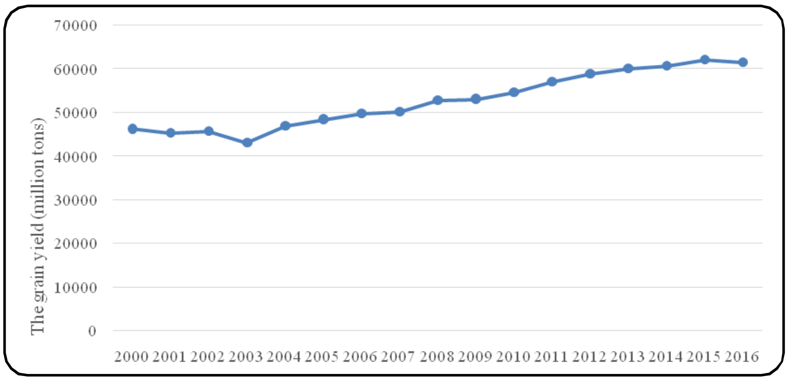

Fig. 1: Grain output in China in 2000-2016.

Measures for controlling straw-polluted soil are then proposed.

Scholars, all over the world, have investigated the distribution characteristics of heavy metal pollution in soil under the mode of direct straw manuring. The water, manure, gas and thermal environments of the soil and the microclimate environment of the farmland change after the straws are returned to the farmland, thereby significantly affecting the growth and output of the crops. For example, Daum analysed the absorption characteristics of soil for ammonium nitrogen and phosphate radicals after adding straws processed through different methods to the soil (Daum et al. 1998). Wang suggested that an organic repair agent has direct absorption on the heavy metal ions in the soil solution, thereby changing the surface activity of the soil matter and affects the chemical behaviour of heavy metals (Wang et al. 2000). Schmidt concluded that after incomplete combustion, the straws will become black charcoal, which exhibits chemical and microbiological inertia (Schmidt et al. 2000). Dong supposed that approximately 10,000 m^2 of farmland was polluted by Cd in China, and that straw manuring may help reduce Cd pollution (Dong et al. 2001). Dumat reported that the turnover rate of rotten straws in the soil is significantly affected by heavy metal content; that is, high contents of heavy metals lead to slow turnover rate (Dumat et al. 2006). Rhodes found that the black charcoal produced by straws has a strong absorption on the heavy metals in the soil and can be used to restore heavy metal-polluted soil because the black charcoal is slowly mineralized in the soil (Rhodes et al. 2008). Zhou concluded that adding black charcoal formed by rice and cotton straws to the Cd-polluted soil can increase the crop yield, significantly reduce the biological effectiveness of Cd, and decrease the crop absorption of Cd in the soil (Zhou et al. 2008). Kuzyakov indicated that black charcoal formed after the straws are combusted can remain in the soil extensively and that the black charcoal made of ryegrass straws has an annual degradation rate of 0.5% only under the best conditions of the soil (Kuzyakov et al. 2009). Ussiri studied the four kinds of absorption of straws for ammonium nitrogen in the water solution and found that the polymolecular layer absorption on the surface of straws may be correlated with the possible cellulose in the straws (Ussiri et al. 2009). Jia identified that the content of effective Cd in the Cd-polluted soil increases after the straw manuring is implemented and claimed that the increase is mainly due to the activation of Cd by the soluble organic carbon released through straw decomposing (Jia et al. 2010). Wang (2015) deduced that the soil organic carbon is increased, and the soil quality is improved when the straws are buried in the soil at 20 cm under the ground (Wang et al. 2015). Wu explored the soil organic matter, enzyme activity, and output in the Yangtze River Delta in China and found the characteristics of rice straws buried underground and the most appropriate depth (Wu et al. 2015). Yang considered that the losses caused by rice-wheat rotation may be reduced when a proper amount of wheat straws are buried underground at an appropriate depth (Yang et al. 2016). Chen indicated that straw returning is an effective measure for improving soil fertility and maintaining the crop productivity of agricultural ecological system (Chen et al. 2017). The crop straws contain a significant amount of nutrients (such as carbon, nitrogen, phosphorus and potassium) required by plants as a kind of renewable resource in a special form. The efficient use of crop straws will help in saving agricultural resources and reducing the agricultural nonpoint pollution; therefore, the efficient use of crop straws can be used as an effective measure for energy conservation and emission reduction in the rural areas. However, if the crop straws are directly returned to the farmland without any pretreatment, then the three major components of the crop straws (cellulose, hemicellulose and lignin) will not be easily and quickly decomposed by microorganisms, which may affect the straw-returning effect. Consequently, they cannot be used as manures for the current crops. If the crop straws exposed to plant diseases and insect pests are directly returned to the farmland, then these crop straws will provide a habitat for these diseases and pests, reduce the crop failure, and even lead to total loss of crop yield. Therefore, the distribution characteristics of heavy metal pollution in the soil under the mode of direct straw manuring (the straws of old crops in the wheat-maize rotating field are returned as manures for new crops) are analysed, and the measures for controlling the straw-polluted soil are proposed.

MECHANISM OF HEAVY METAL POLLUTION IN SOIL UNDER THE DIRECT STRAW MANURING MODE

Heavy metals are part of the earth's crust and are distributed in the topsoil in different forms. The soil-plant system is located in the transition zone of the four huge circles of the earth and constitutes the hub of matter and energy recycling in the terrestrial ecological system and the bridge that connects two major production groups, that is, plants and animals. Thus, heavy metals not only influence the growth of plants, but also affect the health of human beings through the food chain via their accumulation in plants. The heavy metal pollution in soil may originate from three major sources: first, transportation pollution caused by dust from oil combustion and tire wearing; second, the extensive use of heavy metal materials in the industrial production; third, the agricultural pollution caused by the vigorous use of fertilizer, pesticides, and mulch. Their pollutants possess poor mobility, extensive retention time, and poor microbial degradation.

Lead (Pb): Pb is a kind of steel gray heavy metal. Pb dust is harmful to human bodies when accumulates to a certain concentration in the air. The public is exposed to Pb through a variety of means: first, lead pigments, especially for several old brands of pigments, which contain a high content of Pb; lead salt, massicotite, white lead, and red lead are the major components of decorative materials, paints, coating, and wallpaper; second, leaded petrol and the tetraethyl lead in the automobile emissions are extremely toxic substances; third, the lead pollution caused by water pipes. Plastic pipes have become indispensable materials for water supply and drainage system of real estate development and home decoration. Considerable lead salts have been added to PVC pipes and pipe fittings as a stabilizer. With the aging and corrosion of pipes, lead dissolution will lead to the pollution of drinking water. Even having stayed in the environment for a long time, Pb still cannot be degraded into harmless final products, so its toxicity will persist for a long time, once discharged into the environment. Pb pollution cannot only remain indefinitely, but also can cause potential toxicity on many organs. For example, Pb may damage the bone marrow haematopoietic and nervous systems, as well as the gonad of males. People affected by Pb may suffer headache, dizziness, fatigue, memory loss, and insomnia accompanied by loss of appetite, constipation, and other digestive system symptoms. In the worst case, people may become insensible until death.

Chromium (Cr): Cr is a silvery metal commonly used in the industrial production. Cr is widely used for ore smelting, electroplating, leather-making, paint and chemical production, and Cr pollution also mainly originates from these sources. Cr mainly exists in the soil in the form of Cr^{3+}. After the Cr^{3+} combined forms enter the soil, 90% of them will be absorbed by soil, so it is difficult for them to migrate. However, when the accumulated amount exceeds the soil's self-purification capacity, it will cause some harm. Cr is toxic when it is in the form of Cr^{6+}, which can strongly stimulate the mucous membrane of the human body and produce the carcinogenic effect. Cr^{6+} mainly exists in a free state after entering the soil solution, and only a small amount of them can be absorbed by the soil. The soil's absorption of Cr^{6+} is closely related to the nature of the soil.

Cadmium (Cd): Cd has strong toxicity and biological migration. With the extensive use of Cd in the industrial production and the increasing irrigation of Cd-contained sewage, the content of Cd in the environment rises year by year. Cd is easily absorbed by organisms and accumulated in their bodies. When entering the human body, it will be easily accumulated in kidney and bones, causing kidney failure and bone shattering and softening. Cd's main target is renal cortex. If an excessive amount of Cd is accumulated in the renal cortex, then Cd will induce diseases, such as renal lesions. Cd is a non-essential element for human bodies. Cd in the human body is absorbed from the external environment after being born, and enters into the human bodies mainly through food, water and air. The smoke and dust that contain Cd can be taken into the liver and kidney through the respiratory tract. The Cd content accumulated in the liver and kidney constitute 60% of the total amount of Cd in the human body. The Cd content in human organs may largely differ in the different regions with the different environmental pollutions and increase with time. Cd in the human body is mainly discharged through urine via the kidney, but a large amount of Cd is also discharged through droppings via the liver. The exhaust velocity of Cd is slow, and the biological half-time of Cd in the renal cortex of human beings is 10-30 years.

Arsenic (As): As is a kind of nonmetallic element and listed as a major heavy metal pollutant, because its physical and chemical properties are similar to those of the heavy metals.

As-contained sewage, exhaust gas, and waste residue may occur in the mining and smelting of As and As-containing metals in producing glass, paints, medicines, and paper with As or As compound as the raw materials and in the combustion process of coal, which causes pollution to the environment. The As pollution in the atmosphere mainly originates from natural sources, such as rock weathering and volcanic eruption, and from the industrial production, the use of As-contained pesticides, and the combustion of coal. The As-bearing sewage, pesticides, smoke and dust will cause pollution of the soil, and the heavy metal pollutants in the soil further lead to the crop failure in many regions. As is accumulated in the soil and then enters the crops; As and As compounds can enter human bodies through water, air and food and cause harm. The simple substance As has a low toxicity, but its compounds are all toxic, especially the As^{3+} compounds. The toxic symptoms may appear after being exposed to As or decades of years after consuming As-bearing drinking water.

EXPERIMENT DESIGN AND METHOD

Experiment design: The study was conducted in 2016 in the randomly selected villages in five cities (Nanjing, Huaian, Yancheng, Yangzhou and Zhenjiang) of Jiangsu Province, China. The research group consisted of randomly selected farmers to directly crush the straws into 5 acres of farmland soil in each village to provide nutrition for the new crops. Under the wheat-maize rotation system, at the maturity period of each season of crops, by adopting five-point sampling method, the farmland soil from 0-20 cm underground and their plant samples were taken at 20 randomly selected farmlands of the five cities for further determination.

Pretreatment of test samples: Quartering method was utilized to treat the soil samples in the laboratory. After the soil samples were air-dried, a disintegrator (RT-02A, hereinafter the same) was used to crush the soil samples, which were later filtered with a screen mesh of 0.4 mm and saved for future use. The plant samples were separated in the laboratory into different parts and then flushed with tap water, distilled water and deionized water sequentially; then, the soil samples were placed into the electro-thermostatic blast oven for fixation at a temperature of 105°C and dried to a constant weight at 80°C, crushed with a disintegrator, filtered with a screen mesh of 0.4 mm and saved for future use. The fungus material, fungus residue, fodder and cow dung samples were dried in an electro-thermostatic blast oven at a temperature of 80°C to dehydrate them, crushed into pieces with a disintegrator, filtered with a screen mesh of 0.4 mm and saved for future use. An electronic balance (Ohaus Shanghai, with an accuracy of 0.0001 g) was used to weigh the samples to obtain 1 g for each share; this step was repeated three times; the soil samples were placed into the 50 mL conical flask and other kinds of samples into the 25 mL conical flasks. Then, a 25 mL digestion mixture (nitric acid: perchloric acid = 4:1, the reagent purity adopted in the experiment is AR) was added to each share of soil sample. The prepared digestion mixture of 15 mL was added to each share of the other kinds of samples, and then the curved funnels were placed in the mouths of all conical flasks. The samples were placed in the cooking stove (KXL-1010 temperature control cooking stove) for heating and digestion. First, low-temperature digestion was conducted. The temperature of the cooking stove was set at 75°C; when the solution becomes clear and bright yellow, the temperature was increased to 240°C. The samples were continuously digested until white smoke disappears and only the white solid matters were left. A minimal amount of deionized water was used after the digested samples were cooled to dissolve the digestive products and flush the mouths of the conical flasks and the curved funnels to reduce losses. In the process, if the dissolution is slow, then the conical flask is placed into the numerical control ultrasonic cleaner (KQ-100DE) for oscillation for 3-5 s to accelerate its dissolution. The dissolved liquid was transferred to 25 mL volumetric flask and flushed several times to fully recover the heavy metals. Then, the deionized water was used to stabilize the volume at 25 mL and finally filtered into the 50 mL centrifuge tube for determination.

Determination of the content of heavy metal: The standard reserving solutions of Pb, Cr, As and Cd, all having concentrations of 1,000 mg/L, used in the experiment were

Table 1: Limits for heavy metal pollutants in the soil.

Sample Type	Pb Concentration (mg/kg)	Cr Concentration (mg/kg)	Cd Concentration (mg/kg)	As Concentration (mg/kg)
Soil	300.0	300.0	0.3	30.0
Wheat seeds	0.5	1.0	0.1	0.5
Maize seeds	0.5	1.0	0.1	0.5
Beef	0.5	1.0	0.1	0.5
Edible mushrooms	1.0	1.0	0.2	0.5

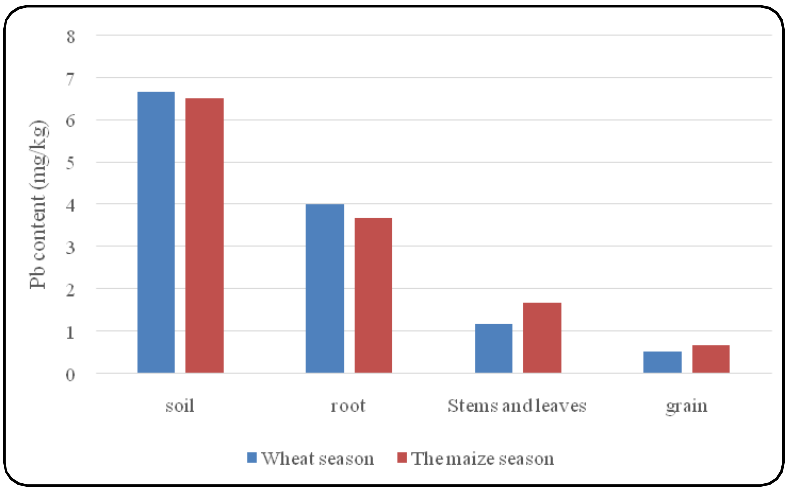

Fig. 2: Concentration of heavy metal Pb under the mode of direct straw manuring.

purchased from TMRM. The standard solution information comes from the National Standard Material Information Center. The standard reserving solutions were diluted into standard metal solutions of 10, 5, 2, 1 and 0.5 mg/kg with the deionized water. Then, the US EIAN DRC-e Inductively Coupled Plasma Mass Spectrometer was used for determining the content of heavy metals. First, the standard heavy metal solutions of the different concentrations were determined separately, and the standard curves were drawn with a fitting degree of more than 99.99%. Second, the absorbance of the samples was determined repeatedly. The difference between the two absorbance values shall not exceed 2% or the instrument shall be adjusted for redetermination. The blank reagent was the deionized water. The deionized water used for flushing was replaced in a timely manner to avoid excessive influence on the measuring results.

Method for measuring heavy metals: Heavy metal content was calculated as follows:

$$C = \frac{c_1 \times v \times 1000}{1000 \times m} \quad ...(1)$$

Where C is the content (unit: mg/kg) of heavy metal, C_1 is the measured content (unit: mg/L) of heavy metal, V is the volume (unit: mL), and m is the weight (unit: g) of the sample. The limits for heavy metals in the soil are listed in Table 1.

EMPIRICAL STUDY

Distribution characteristics of heavy metal Pb under the mode of direct straw manuring: In Fig. 2, the concentration of heavy metal Pb in the soil of wheat season under the mode of direct straw manuring is the highest, reaching 6.68 mg/kg, but is still within the limits required by the Environmental Quality Standards for Soils. The content of heavy metal Pb from the soil to the wheat roots decreases significantly, and the value continues to decrease in the stems, leaves and seeds. The content of Pb in the wheat seeds reaches 0.52 mg/kg, which exceeds the limits required by the national standard. The content of Pb in the maize season also follows the same rules: the content of Pb in the soil reaches the highest value of 6.52 mg/kg, which decreases from the bottom to the top of the plants and reaches 0.68 mg/kg in the maize seeds, significantly exceeding the national standard. Horizontally, the content of heavy metal Pb in the soil decreases with the planting season and the addition of straws, while the content of Pb in the seeds increases minimally. Although the content of Pb in the soil is within the safety range, it is worth noting that the Pb contents in these two kinds of grain seeds both exceed the national food hygiene standards, but are not so significant.

Distribution characteristics of heavy metal Cr under the mode of direct straw manuring: In Fig. 3, the content of heavy metal Cr in the wheat season increases first and then decreases from the soil, roots, stems, and leaves to the seeds. The Cr content in the soil was 5.33 mg/kg, which is lower than the standard set in the Environmental Quality Standards for Soils; the Cr concentration in the seeds reaches 0.17 mg/kg, which is also lower than the national food hygiene standard. The content of heavy metal Cr in the maize season increases greatly from the soil to the maize roots, which then experiences a decline sequentially from the roots, stems and leaves to the seeds. The Cr content in the maize seeds is 0.19 mg/kg, which is lower than the national standards and is within the safety range. Seeing from the whole cycle, the content of heavy metal Cr in the soil does not change significantly, but in terms of seeds, the Cr content in maize

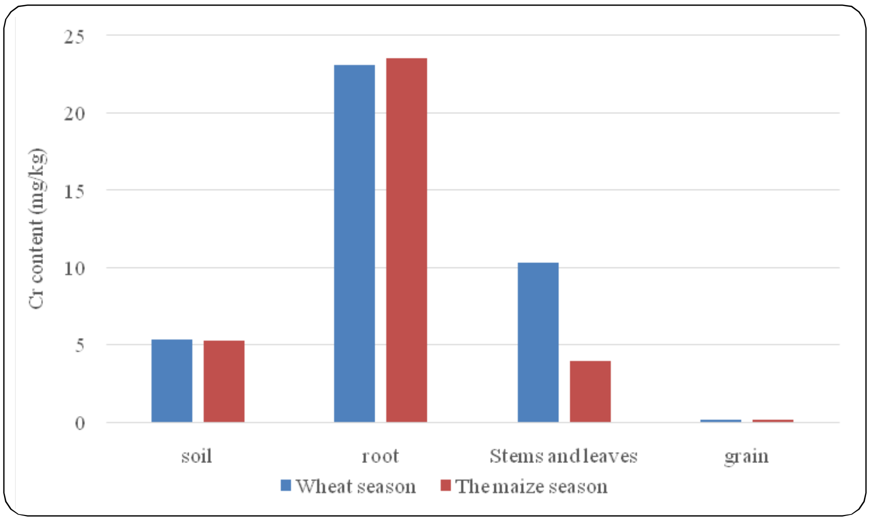

Fig. 3. Concentration of heavy metal Cr under the mode of direct straw manuring.

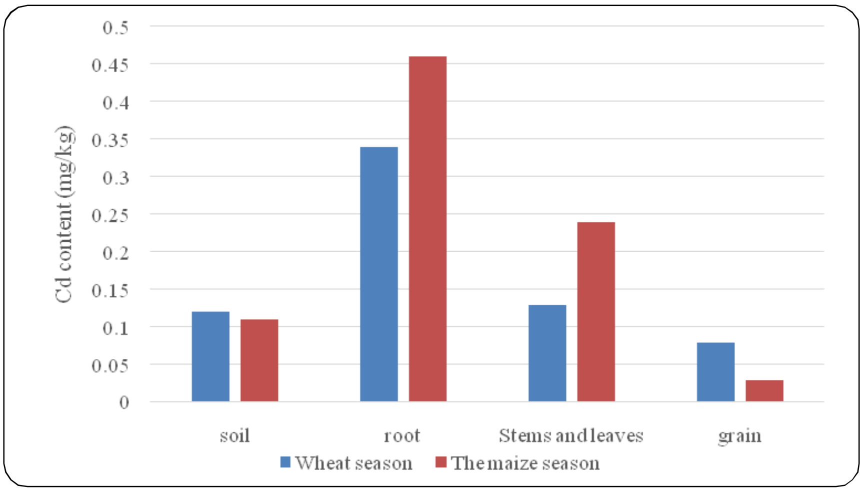

Fig. 4: Concentration of heavy metal Cd under the mode of direct straw manuring.

seeds decreases to 11% compared with that in wheat seeds.

Distribution characteristics of heavy metal Cd under the mode of direct straw manuring: Cd is highly toxic. In accordance with the Environmental Quality Standards for Soils and the National Food Hygiene Standard, the Cd content in the soil in the wheat season was 0.12 mg/kg, which is lower than the national standard, as illustrated in Fig. 4; the value represents a huge increase in the wheat roots, a decrease in the stems and leaves, and the lowest level in the seeds (0.08 mg/kg), which is slightly lower than the national standard. The Cd contents at various key nodes in the maize season are different from those in the wheat season, but the trends are similar, increase first and then decrease. The Cd content in the maize seeds is 0.03 mg/kg. Horizontally, the Cd content in the soil declines slightly, and the maize plants absorb more Cd than the wheat plants.

Distribution characteristics of heavy metal as under the mode of direct straw manuring: In Fig. 5, the highest content of heavy metal As in the wheat soil is 6.25 mg/kg; the amount of As that enters from the soil to the plants is minimal; the As content in the wheat roots reaches 0.96 mg/kg, suggesting a huge decline. The As content increases a bit from the wheat roots to the stems and leaves, reaching 1.73 mg/kg; the As content in the wheat seeds is 0.28 mg/kg, which is lower than the national food hygiene standard (0.70 mg/kg). For each recycling node of the maize season, the As content in the soil is the highest at 5.43 mg/kg and is lower than the Environmental Quality Standards for Soils; this value

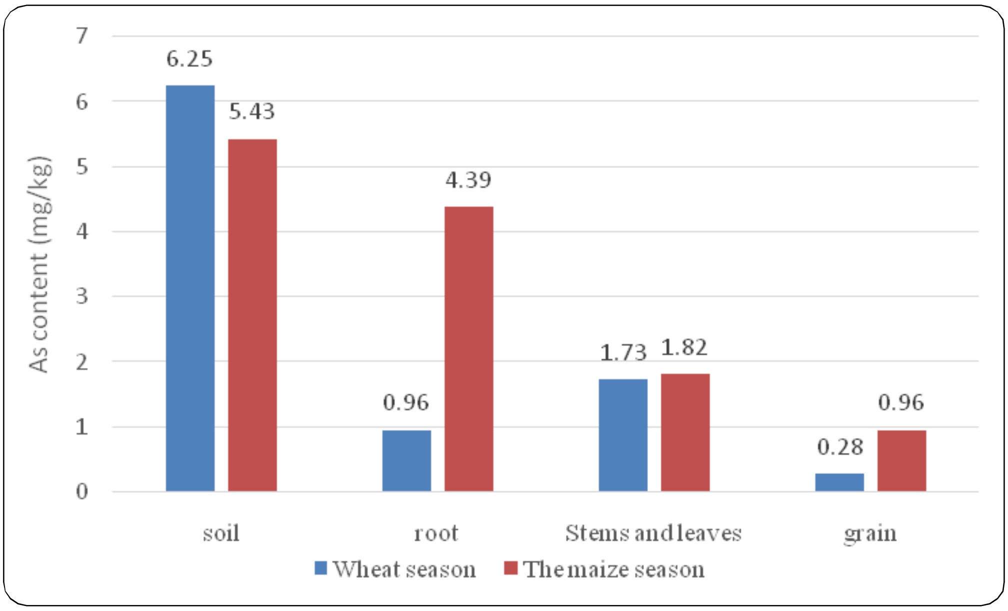

Fig. 5: Concentration of heavy metal As under the mode of direct straw manuring.

is also lower than that in the soil of wheat season. Then, the As content in the roots, stems, leaves and seeds of the maize declines sequentially, and the As content in the maize seeds is 0.96 mg/kg, which is higher than that in the wheat seeds.

CONCLUSIONS

A process occurs for heavy metal to enter the human body from the environment before it can harm humans. This process is from the environment to the farmland, to the agricultural products, and to the food chain of human beings. The migration of heavy metals in this process is evaluated, and the migration rules in the cyclic chain are summarized, thereby promoting the prevention and avoidance of heavy metal harm. We first analysed the effects of heavy metals on soil pollution and then measure the concentration distribution characteristics of heavy metals, namely, Pb, Cr, Cd and As, of the polluted soil through experiments to further analyse the distribution characteristics of heavy metal pollution of soil under the mode of direct straw manuring. The empirical study shows that the contents of the four heavy metals in the soil do not exceed the upper limit specified in the national standards in the wheat-maize rotating farmland under the mode of direct straw manuring; a certain degree of concentration of heavy metals Cr and Cd is found in the roots of wheat and maize, whereas no Pb and As concentrations are found; the content of heavy metals decreases sequentially from the roots, stems and leaves to the seeds. Further research is recommended on the absorption of heavy metals, the agricultural cycle mode of heavy metals, the influence of the soils of different natures and climates on heavy metal migration, and the blocking mechanism of heavy metal pollution under the agricultural bacteria recycling and the agricultural and pastoral recycling modes.

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