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Original Research

Leaching of Heavy Metals from Rice Fields with Different Irrigation Management

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Abstract

To reveal the impact of irrigation management on the release and leaching of soil metals (Cu, Zn, Pb, Cr, and Cd), deep percolation rate and metals contents in soil solutions were observed in rice fields with non-flooding controlled irrigation (NFI) and flooding irrigation (FI) treatments. The contents of Cu and Cr in the deep solutions were safe according to the environmental quality standard for groundwater, but contents of other metals might lead to groundwater contamination, especially for Cd. The release of metals in surface soil was increased for NFI because the wetting-drying cycles in NFI fields resulted in less reluctant and high decomposition and mineralization of soil organic matter in surface soil, and consequently enhanced the release of soil metals into solutions. Seasonal metals leaching losses in NFI fields were 44.9-53.8% lower than in FI, due to the large reduction in both deep seepage rates and metals concentrations in deep soil solutions. Higher release of metals in NFI surface soils might lead to higher bioavailability of micronutrients (Cu and Zn) to crops, but higher risks in toxic metals (Pb, Cr, and Cd) uptakes.

Keywords: heavy metal, release, leaching, water-saving irrigation, rice field

Introduction

Heavy metals such as copper (Cu), zinc (Zn), lead (Pb), chromium (Cr), and cadmium (Cd) can accumulate in soil and dissolve into groundwater in the process of leaching, which result in contamination of both groundwater and surface water. Leaching of soluble metals from surface soil to deep soil or groundwater was sometimes monitored in landfills, compost sites, or contaminated soils [1-3]. But the leaching of heavy metals from arable agricultural soils was always overlooked and regarded as a ‘forgotten’ pollution source [4], although it was reported as an important source of water contamination [5, 6].

Agricultural soils with intensive cultivation are always have high potentials for leaching losses of water and nutrients from soil into groundwater. Rice paddy soil, an important anthropogenic soil widely distributed in monsoon Asia, is always under flooding conditions and commonly characterized by intensive cultivation and high percolation rate. Large percolation leads to high leaching risks of nutrients and contaminants, and a great deal attention has been paid to leaching of nutrients (nitrogen and phosphorus) in rice fields [7-11], but leaching of metals was always overlooked.

The metals in soil are presented in different binding forms that determine the mobility, bioavailability, and toxicity of metals in soil [12, 13]. Only metals dissolved in soil solutions are leachable. The dissolution of these metals is based on the chemistry of the water and the soil. Factors that affect metal mobility include soil pH, dissolved oxygen, oxidation-reduction potential (ORP), specific conductivity, soil organic matter (SOM) content, and Fe and Mn oxide content [13-17].

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Table 1. Heavy metal contents in paddy soil at different depths and in irrigation water.

Items		Metal contents				
		Cu	Zn	Pb	Cr	Cd
Soil (mg·kg^{-1})	0-20cm	28.04	49.46	15.69	36.45	0.799
	20-40 cm	27.54	62.90	15.18	23.30	0.665
	40-60 cm	24.41	48.23	14.34	17.05	0.560
	Limits of 1st grade for soil pollution	35	100	35	90	0.2 (0.3 and 1.0)*
Irrigation water (mg·L^{-1})	July	7.3	47.9	6.4	1.5	3.2
	August	7.0	61.7	7.5	1.3	2.7
	September	10.1	44.9	6.9	1.0	3.6
	Standards for Irrigation Water Quality	1000	2000	100	100	5.0
	Standards for Groundwater Quality**	10-50-1000	50-500-100	5-10-50	5-10-50	0.1-1-5

*The number in bracket is the limits of the second and third grades for soil pollution according to Environmental Quality Standard for Soils.
**The numbers linked by the dash line are the limits of the first, second, and third grades for ground water pollution according to the Quality Standards for Groundwater.

Agricultural practices (such as manure application, tillage, sewage irrigation, etc.) impact the soil characteristics, and hence the mobility, bioavailability, and toxicity of metals in soils [18-22]. Irrigation is one of the most important management practices in rice fields, which was changing from the traditional flooding irrigation (FI) to water-saving irrigation (WSI) due to increasing water scarcity. Several WSI techniques, such as non-flooding controlled irrigation (NFI), alternate dry-wet irrigation (ADWI), and the system of rice intensification (SRI) [23-26] were applied widely in recent decades. The practice of WSI led to huge changes in percolation patterns and soil properties, and hence the soil biological and chemical processes [27-31]. That will indirectly alter the transformation and redistribution of heavy metals in soil, and many researchers have indicated that unsaturated water conditions and the drying process always lead to enhanced mobility and bioavailability of metals in soil [13, 32-37]. However, minimal information is available on risk of metals leaching from paddy fields with different irrigation management systems.

In the current study, field experiments were conducted to determine concentrations of Cu, Zn, Pb, Cr, and Cd in soil solutions and metals leaching risks from rice paddies with FI and NFI irrigation managements.

Materials and Methods

Site Description and Experimental Design

The study was conducted in 2011 at Kunshan irrigation and drainage experiment station (31º15′ 15″ N, 120º57′ 43″ E) in the Tai-Lake region of East China. This area has a subtropical monsoon climate. The groundwater depth ranged from 0.2 m to 0.6 m during rice season. The soil was Stagnic Anthrosol, developed from alluvial deposits. The soil texture in the plowed layer (0-18 cm depth topsoil) is clay. Soil samples were collected at 5 cm, 15 cm, 25 cm, and 35 cm depths at soil profiles by using the soil sample ring kits (100 cm^3), and were saturated by immersing them in distilled water for 24 hours, then those were dried for 12 hours at 105ºC to determine the saturated soil water contents by weighing method. The saturated soil water content (v/v) for the layers 0-20, 0-30, and 0-40 cm are calculated as 54.4, 49.7, and 47.8%, respectively. The contents of soil organic carbon, total nitrogen, and total phosphorus are 12.9 g·Kg^{-1}, 1.03 g·Kg^{-1}, and 1.35 g·Kg^{-1}. Soil pH is 6.84 (soil:water=1:2.5). Total metals contents in irrigation water and soil at different depths (extracted by HNO_3-HF-$HCLO_4$ digestion method) were listed in Table 1. The Cu, Zn, Pb, and Cr contents were much lower than the limits of grade I according to Environmental Quality Standard for Soils (GB 15618-1995) in China [38]. But the Cd contents were higher than the limits of the grade II of soil pollution. There was a certain risk for Cd contamination in rice fields in this region. In irrigation water, the Cu, Zn, Pb, Cr contents were much lower than the limits for irrigation water quality (GB 5084-2005), and Cd contents were slightly lower [39].

There were two irrigation treatments with three replicates: FI and NFI. A randomized complete block design was established in six plots (35 m^2). In FI rice fields, a depth of 3-5 cm standing water was always maintained after transplantation except during the later tillering period and yellow maturity period. When it comes to NFI rice fields, pond water depth was kept between 5 and 25 mm during the first 7 or 8 days after transplantation during the regreening period, then standing water depth was avoided in other stages except during the periods of pesticide and fertilizer application. Standing water was kept during the fertilization period to hydrolyze the broadcasted urea to ammonium, and during the pesticide period to enhance the function of pesticide or herbicide. Irrigation was applied only when soil moisture was approaching the lower thresholds as listed in Table 2 in different stages and to keep the soil moist.

Table 2. Soil moisture limits in different growth stages for non-flooding controlled irrigation.

Limits	R [a]	T			J/B	H/F	M	R
		Initial	Middle	Late				
Upper limit [c]	25 mm [b]	100%θ_{s1}	100%θ_{s1}	100%θ_{s1}	100%θ_{s2}	100%θ_{s3}	100%θ_{s3}	Naturally drying
Lower limit	5 mm [b]	70%θ_{s1}	65%θ_{s1}	60%θ_{s1}	75%θ_{s2}	80%θ_{s3}	70%θ_{s3}	
Observed root zone depth (cm)	–	0-20	0-20	0-20	0-30	0-40	0-40	

[a] R, T, J/B, H/F, M, and R represent regreening stage, tillering stage, jointing/booting stage, heading/flowering stage, milking stage, and ripening stage, respectively.

[b] Data show water depth during green stage. θ_{s1}, θ_{s2}, and θ_{s3} represent average volumetric soil moisture for the layers of 0-20 cm, 0-30 cm, and 0-40 cm, respectively.

[c] In the case of pesticide and fertilizer applications, standing irrigation water up to 5 cm depth will be maintained less than five days.

Standing irrigation water up to 5 cm depth was maintained for less than 5 days when pesticide or fertilizer was applied. The variety of rice was Japonica Rice NJ46. It was transplanted with 13 cm×25 cm hill spacing on June 28 and harvested on October 25. The same doses of fertilizers for each split were applied to each plot, in agreement with the local conventional fertilizer application method.

Field Measurements and Sampling

Irrigation water volumes were recorded using water meters installed on the pipes. The soil moisture in the rice fields was monitored every 2-3 days in three replicates using time-domain reflectometry equipment (TDR, Soil Moisture, USA) and 20 cm waveguides installed vertically at 0-20 and 20-40 cm depths. Water layer was monitored daily using a vertical ruler fixed in the field. The daily rainfall was recorded using an automatic weather station (ICT, Australia).

Soil solutions were collected by clay suction cups (2 cm in inner diameter, 7cm in length) installed vertically at different depths (7-14, 27-34, and 47-54cm). The water samples were acidified with 2-3 drops nitric acid, and kept in polyethylene bottles at 4ºC. The water samples were filtered using the 0.45 μm Millipore filter before the metals concentrations were measured using ICP-OES (ICAP 6000 duo, Thermo Scientific).

Leaching losses were calculated based on the average metals contents in 27-34 and 47-54 cm depth soil solutions and deep seepage rates calculated by water balance equation in 0-40 cm soil depth:

$$S_t=W_{t-1}-W_t+I_t+R_t-D_t-ET_t \qquad (1)$$

...where S is the deep seepage rate. W is the daily water depth for flooding, and soil water storage in 0-40 cm soil calculated based on soil moisture at 0-20 and 20-40 cm depths for non-flooding. I, R, and D is the volume of irrigation, rainfall, and surface drainage. Drainage volume was determined by the water layers before and after drainage. ET is the evapotranspiration, which was measured by bottom sealed lysimeter with the same irrigation management [11].

Results and Discussion

Water Cycles and Water Consumption

The typical flooding water depths and soil moistures in the FI and NFI fields are shown in Fig. 1. There are 12 drying processes in NFI fields, and 2 drying processes in FI fields. The days under non-flooding conditions were about 65 days in NFI fields. As a result, water consumption was reduced greatly in NFI fields (Table 3). The evapotranspiration and deep percolation were 581.7 and 291.1 mm in NFI fields, reduced by 17.6 and 30.9% compared with FI treatment. Consequently, the irrigation volume in NFI fields was reduced by 60.3%. Irrigation volume, evapotranspiration, deep seepage, and water consumption in NFI paddies are all significantly ($p<0.05$) lower than in FI paddies (Table 3). At the same time, rice yields were 8,103.1 kg·ha^{-1} for NFI treatment, increasing by 5.2% compared with FI treatment.

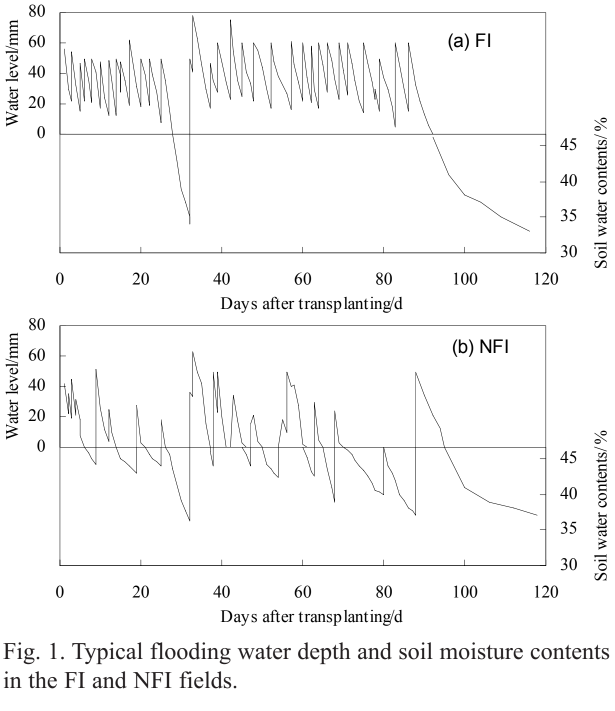

Fig. 1. Typical flooding water depth and soil moisture contents in the FI and NFI fields.

Table 3. Rice yields and water consumption with different irrigation.

Treatment	Yield	Irrigation	Total rainfall	Drainage	Evapotranspiration	Water consumption	Deep seepage
	$kg\cdot ha^{-1}$	mm	mm	mm	mm	mm	mm
NFI	8103.1a	340.4a	403.8	8.5 a	581.7a	872.8 a	291.1a
FI	7705.9a	634.4b	403.8	12.3 a	706.1b	1127.3 b	421.2b

Different letters in each column represent significant differences between the treatments at p=0.05.

Metal Contents in Soil Solutions

Contents of Cu, Zn, Pb, Cr, and Cd in soil solutions fell in the ranges of 2.3-11.2, 14.1-111.4, 0.9-6.8, 1.0-2.6, and 0.4-1.55 $\mu g\cdot L^{-1}$ (Fig. 2). Those ranges of metals concentrations were almost the same ranges as in irrigation water. But sometimes the metals concentrations in soil solutions were higher than the maximum concentrations of metals in irrigation water. The ranges of metals concentrations in 47-54 cm depth were 2.5-8, 17.2-59.2, 2.0-6.1, 1.4-2.2, and 0.4-1.55 $\mu g\cdot L^{-1}$. According to the environmental quality standard for groundwater (Table 1) [40], the contents of Cu and Cr in soil solution was safe. But the contents of Zn and Pb in soil solutions were slightly higher than the pollution limits for grade I (50 and 5 $\mu g\cdot L^{-1}$) occasionally, and the Cd contents in soil solutions were occasionally higher than the pollution limits for grade I (1.0 $\mu g\cdot L^{-1}$). Thus leaching of paddy soil solutions will lead to risks in metals pollution of groundwater, especially for Cd, which was relative high in the paddy soil.

The contents of Cu and Zn in soil solutions were reduced over time in both NIF and FI fields, but the contents of Cd in soil solutions were increased (Fig. 2). This implies that inputs or release of Cu and Zn into the soil solutions were less than the outputs by crop uptakes and leaching, and it was the reverse for metal of Cd. The contents of metals in 7-14 cm depth soil solutions were mostly higher in NFI soil solutions than in FI, except for Cd in the later season. But the contents of metals in 47-54 cm depth soil solutions were mostly lower in NFI soil solutions than in FI. This indicates that wetting-drying cycles in NFI fields resulted in higher release of metals in surface soil, but less soluble metals move downward to deep soil.

The metals contents in 7-14 cm soil solutions in NFI soils were mostly slightly higher than that in FI soils, but metals contents in 47-54 cm soil solutions in NFI soils were more frequently lower than in FI soils (Fig. 2), although the difference between NFI and FI was not significant. The increased metals contents in surface soil solutions in NFI fields confirmed that the drying process and oxidizing condition in paddy fields always led to high metal solubility [13, 41-45]. Metals in soil may exist as ions or complexes with ligands, and inorganic and organic colloidal species. The mobility or solubility of metals in soil was determined by its existing forms. When the rice field was exposed to the drying-wetting cycles condition, there were less reluctant (such as Fe^{2+} and S^{2-}), and fewer metals would precipitate with S^{2-}, or coprecipitate with FeS and MnS [46, 47]. That partially accounted for the increase of metals contents in soil solutions in NFI fields. At the same time, the soil dissolve organic carbon (DOC) contents were highly positively related to the transformation and mobility of metals in soils [14, 37, 41, 42, 48-50]. And the decomposition and mineralization rates of SOM were enhanced, and soil DOC contents increased under the drying-wetting cycles condition [37, 51, 52]. That might be another important reason for high metals contents in surface soil solutions in NFI fields. But the Cd contents in soil solutions in NFI fields were mostly lower than in FI fields in the later season. That may be ascribed to high crop uptakes of Cd in NFI fields, because rice was a hyper-accumulation plant of Cd [53] and the root absorbency was always high in NFI conditions [35, 36].

Since metals of Cu and Zn are essential micronutrients for crops [54, 55] and are in safe concentrations in both the paddy soil and soil solutions, the high solubility of Cu and Zn in NFI soil solutions means high bioavailability of the micronutrients to crops. The metals of Pb, Cr, and Cd are toxic to crops; higher contents of those toxic metals in NFI soil solutions would lead to high risk in pollution of rice. At the same time, all the metals are pollutants to both surface and groundwater. Lower contents of metals in deep soil solutions in NFI fields mean less contamination to groundwater.

Seasonal Leaching Losses of Metals

Seasonal leaching losses of metals Cu, Zn, Pb, Cr, and Cd were calculated as 9.22, 84.5, 8.99, 4.16, and 2.28 $g\cdot ha^{-1}$ in NFI fields (Fig. 3). Those were reduced significantly by 44.9-53.8%, compared to those in FI fields. The differences between the FI and NFI treatments are all significant at $p<0.05$, for all the five heavy metals (Fig. 3). Since both deep seepage rates and metals concentrations in deep soil solutions were reduced in the NFI field, the reduction rates of metals leaching losses are larger than the reduction in deep seepage rates (30.9%). In the flooded paddy soil a large amount of percolation led to high leaching risks of nutrients and contaminants. Nutrients of nitrogen and phosphorus have been thoroughly discussed [7-11]; in current research leaching of heavy metals from paddy soil was confirmed as a potential source of groundwater contamination, especially for the flooding paddy soil. Water-saving irrigation techniques were tested to be efficient in reducing water percolation and the leaching losses of nutrients of nitrogen and phosphorus in rice paddy [11], in current research large reduced percolation in NFI paddy also resulted in lower heavy metals pollution risks to groundwater.

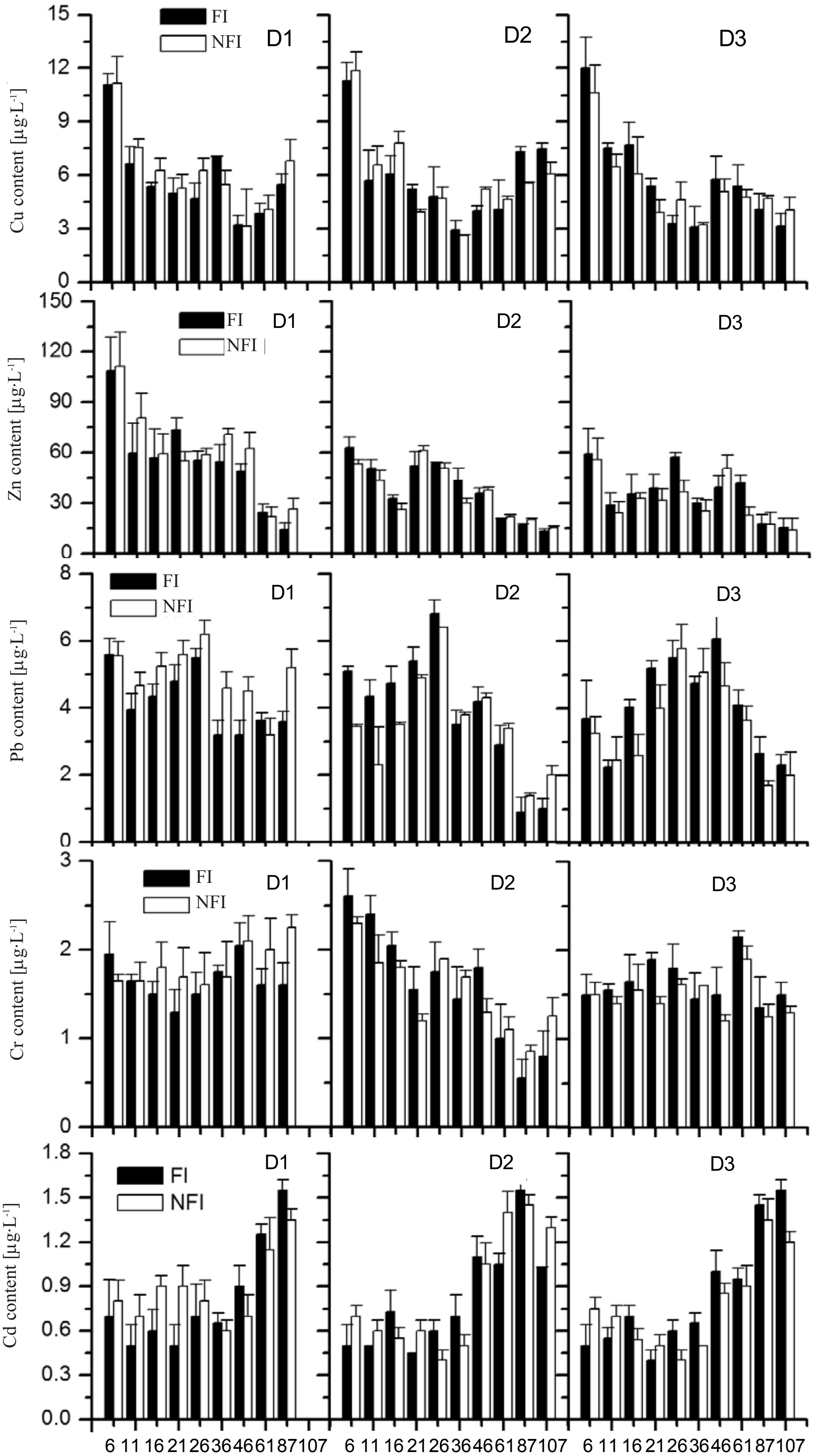

Fig. 2. Metals contents in soil solutions at 7-14 cm (D1), 27-34 cm (D2), and 47-54 cm (D3) depths collected in FI and NFI paddies.

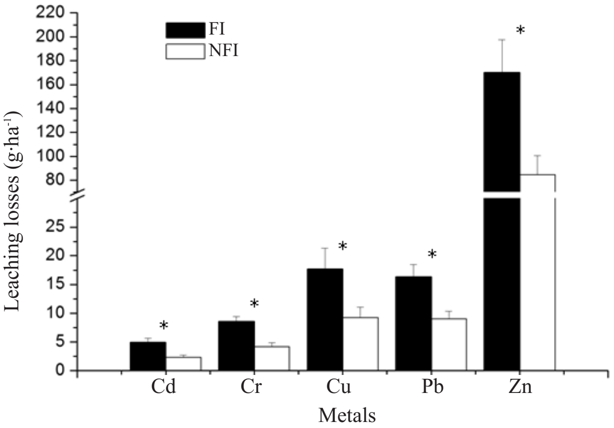

Fig. 3. Seasonal leaching losses of metals from soils in FI and NFI paddies (asterisk on each volume denotes the different is significant at $p<0.05$).

Conclusion

In rice fields in East China there is not a high risk of heavy metals contamination by Cu, Zn, Pb, and Cr, but Cd contamination is a risk. Deep seepage rates and concentrations of Cu, Zn, Pb, Cr, and Cd in soil solutions were observed, and its leaching losses were calculated in rice fields with different irrigation management. The contents of Cu and Cr in the deep soil solution were safe according to the environmental quality standards for groundwater, but contents of other metals might lead to groundwater contamination, especially for the metal of Cd. The wetting-drying cycles in NFI fields resulted in less reluctant and high decomposition and mineralization rates of SOM, and consequently enhanced the release of soil metals into solutions. Seasonal leaching losses of metals in NFI fields were reduced 44.9-53.8% over FI fields due to reduction in both deep seepage rates and metals concentrations in deep soil solutions. That implied NFI resulted in lower risk in leaching of metals into groundwater than FI, although the solubility of metals in surface soil increased. Higher metals solubility in NFI surface soils also meant higher micronutrient (Cu and Zn) bioavailability for crops, but higher toxic metal (Pb, Cr, and Cd) uptakes and risks in food safety.

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