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Long-term combined effects of tillage and rice cultivation with phosphogypsum or farmyard manure on the concentration of salts, minerals, and heavy metals of saline-sodic paddy fields in Northeast China

Lihua Huang [a,b,*], Ying Liu [c,**], Jorge F.S. Ferreira [d], Mingming Wang [a,b], Jia Na [a], Jinxin Huang [a], Zhengwei Liang [a,b]

[a] Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences (CAS), 4888 Shengbei Street, Changchun 130102, China
[b] Da'an Sodic Land Experiment Station, Chinese Academy of Sciences (CAS), Da'an, Jilin 131317, China
[c] School of Management Science and Information Engineering, Jilin University of Finance and Economics, 3699 Jingyue Street, Changchun 130117, China
[d] US Salinity Laboratory (USDA-ARS), 450 W. Big Springs Road, Riverside, CA 92507-4617, USA

A R T I C L E I N F O

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A B S T R A C T

Soil sodicity is a major ecological problem in the western Songnen Plain of Northeast China and rice cultivation is the main approach used to mitigate saline-sodic soils. However, rice cultivation alone may not be the most effective practice. This study aimed to investigate the combined effects of annual tillage and rice cultivation with either phosphogypsum or farmyard manure on soil salinity, mineral status, and concentration of heavy metals in saline-sodic paddy fields. Treatments were: 1) untreated (no amendments), untilled, and uncultivated (no rice) saline-sodic native grasslands (UG); 2) untreated, tilled, rice-cultivated paddy fields (PFU); 3) tilled, rice-cultivated, amended paddy fields with phosphogypsum (PFPG); and 4) tilled, rice-cultivated, amended paddy fields with farmyard manure (PFFM). The effectiveness of these treatments on soil improvement was evaluated after a 10-year field experiment. Compared to the UG control, the 0–20 cm topsoil layer of PFU, PFPG, and PFFM had respective decreases in Na^+ concentrations of 42.9%, 61.5%, and 60.9%; in CO_3^{2-} + HCO_3^- concentrations of 18.9%, 63.2%, and 57.9%; in Cl^- concentration of 64.6%, 75.7%, and 79.9%; in pH units of 0.57, 1.05, and 1.30; in soil electrical conductivity ($EC_{1:5}$) of 18.3%, 49.1%, and 48.3%; and in exchange sodium percentage (ESP) of 47.2%, 66.9%, and 72.5%. Also, the 0–20 cm topsoil layer of PFPG and PFFM had its concentrations of soil organic matter (SOM), available nitrogen (AN), and available phosphorus (AP) significantly ($P < 0.05$) increased compared to the UG control. However, the concentrations of five heavy metals (As, Pb, Cd, Cr, and Hg) were kept within a safe range in saline-sodic paddy fields amended with phosphogypsum or farmyard manure and were far below the environmental quality standard held for Chinese soils. Therefore, phosphogypsum and farmyard manure significantly decreased soil salinity and sodicity while increased soil fertility and SOM. Because these amendments are locally available and affordable to farmers, their use is deemed suitable for large-scale soil reclamation and the mitigation of salinity and sodicity in soils destined for rice cultivation in Northeast China.

1. Introduction

The Songnen Plain of Northeast China occupies a total area of 17.0 million hectares, from which 3.42 million hectares are saline-sodic, making soil salinization and alkalization a major ecological problem in Northeast China (Song et al., 2003; Wang et al., 2003). Over the years,

Abbreviations: UG, uncultivated saline-sodic grassland; PFU, paddy fields untreated; PFPG, paddy fields with phosphogypsum; PFFM, paddy fields with farmyard manure; $EC_{1:5}$, soil electrical conductivity with soil: water ratio of 1:5; ESP, exchangeable sodium percentage; Na_{ex}^+, exchange sodium; CEC, cation exchange capacity; SAR, sodium adsorption ratio; TA, total alkalinity; RSC, residual sodium carbonate; SOM, soil organic matter; AN, available nitrogen; AP, available phosphorus; AK, available potassium; DASLES, Da'an Sodic Land Experiment Station.

* Corresponding author at: Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences (CAS), 4888 Shengbei Street, Changchun 130102, China.
** Corresponding author.
E-mail addresses: huanglihua@iga.ac.cn (L. Huang), lyaihua1995@163.com (Y. Liu).

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overgrazing by livestock, population growth, and improper soil management have been the main anthropogenic factors responsible for increased soil salinization in the Songnen Plain and leading to the decline of grasslands previously colonized by *Leymus chinensis* (Poaceae), a perennial forage grass (Wang et al., 2009). Currently, the Songnen Plain is more explored for agriculture than for livestock herding but low-quality water and improper irrigation-resource management continue to be main factors in increasing soil salinity, sodicity, and alkalinization in recent years. Saline/sodic soils are barren and unfit for crop production but can become a valuable land resource if appropriately managed (Yang and Yao, 2015; Huang et al., 2016). Over the years, numerous researchers devoted themselves to try to recover these soils (Zhao et al., 2001; Chi et al., 2012; Zhang et al., 2016). However, it is necessary to adopt locally produced, low-cost, and effective resources as treatment strategies to reduce soil salinization while improving soil chemical and physical properties.

It is a fact that washing or leaching saline-sodic soils with good-quality (low-salinity) water is the most accepted method for mitigating the problem (Qadir et al., 2000). However, freshwater is increasingly becoming scarce in arid/semiarid areas afflicted by saline-sodic soils (Qadir et al., 2003; Helmreich and Horn, 2009). These soils have high concentrations of montmorillonite clay, which is high in negative charges and adsorb Na^+ very efficiently, leading to clay-particle dispersion into pores, decreasing soil permeability and drainage (Rafiq, 1990). Although low-salinity water sources are few and expensive in arid and semiarid areas, leaching these soils with insufficient water volumes makes their desalination difficult. Furthermore, long-term irrigation with high-salinity groundwater will also increase soil salinity, mainly in superficial layers (Zhang et al., 2012; Huang et al., 2016). Therefore, it is necessary to use tested and proven methods to mitigate soil salinity and sodicity through the lixiviation of salts away from the crop root zone.

Traditional methods of improving saline-sodic soils usually include amendments such as gypsum, sulfuric acid, and organic matter (Sadiq et al., 2007; Yazdanpanah et al., 2011); use of cultural practices such as the addition of sand, deep plowing (inversion of the topsoil layer), leaching with freshwater, drainage systems to remove salts, field covers with crop residues (Zhang, 1995; Mace and Amrhein, 2001; Li et al., 2006; Wang et al., 2010, 2014); and biological remediation with crops known as salt accumulators (Wang et al., 1994, 2014). Generally, the combined application of these methods has produced better results than using a single method (Eynard et al., 2006). Sadiq et al. (2007) discovered that disc plowing combined with sulfuric acid resulted in higher rice (*Oryza sativa*) and wheat (*Triticum aestivum*) yields than those achieved without sulfuric acid application and promoted a faster improvement of saline-sodic soils. Shaaban et al. (2013a,b) observed that the combined application of organic and inorganic amendments played a significant role in improving the properties of salt affected soils under a rice paddy system. Rice cultivation has a long history of 70 years of cultivation in saline-sodic soils in the western Songnen Plain region of Northeast China (Huang et al., 2016). Rice cropping could potentially reduce the negative effects of degraded soil structure by irrigation, progressively removing salinity and sodicity from the soil surface (Huang et al., 2017). Therefore, rice cultivation has become the preferred method of bioremediation of saline-sodic soils, strongly advocated by the Chinese local government in recent years.

Past work done in the Songnen Plain region indicated that the application of chemical amendments such as desulfurized gypsum improved rice yield and soil physicochemical properties during rice cultivation in saline-sodic soils (Chi et al., 2012). The application of phosphogypsum and sheep manure also improved rice yield in saline-sodic soils of Northeast China (Liu et al., 2010a,b). These chemical and organic amendments are byproducts of industrial, agricultural, and livestock operations that are locally available. For instance, phosphogypsum is a byproduct of the manufacturing process of phosphate fertilizer from phosphate ores or phosphoric acid, while farmyard manure comes from livestock production systems such as poultry, swine, and sheep. However, attention should be given to whether these amendments can contaminate the soil with heavy metals such as Ni, Cu, Pb, Cd, and Cr, and decrease rice safety for consumers if these chemical contaminants are absorbed and accumulated by the plant. In Tunisia, soils treated with phosphogypsum had Cd and Cr levels that exceeded Chinese, FAO/WHO, and European allowable standard limits (Ben Chabchoubi et al., 2021). Therefore, a 10-year experiment on rice paddy fields was conducted to evaluate the effects of tillage and soil amendments on soil improvement.

The objectives of the study were (1) to demonstrate the spatial changes (across soil layers) in soil salinity, sodicity, and soil nutrients, (2) to evaluate soil contamination by heavy metals after 10 years of application of phosphogypsum or farmyard manure to saline-sodic soil, and (3) to evaluate the feasibility of using phosphogypsum, farmyard manure, and tillage with rice cultivation to improve saline-sodic soils in Northeast China.

2. Materials and methods

2.1. Experimental site

A 10-year field experiment was conducted at the Da'an Sodic Land Experiment Station (DASLES), between the coordinates 45°35′58″–45°36′28″N and 123°50′27″–123°51′31″E, and situated in the city of Da'an, Jilin Province, China (Fig. 1). The DASLES is affiliated with the Northeast Institute of Geography and Agroecology, Chinese Academy of Sciences. The DASLES site encompasses an area that has a representation of typical saline-sodic soils of the hinterland of the Songnen Plain region (Fig. 1), with elevation ranging between 150 and 200 m above the sea level. The climate type in the Western Jilin Province is semi-arid continental, classified as *Dwa* with short hot summers and dry and long cold winters, according to Köppen's Classification system (Wang et al., 2020). The DASLES has a mean annual temperature of 4.3 °C, varying from −20 °C in January to 26 °C in July, with mean evaporation of 1750 mm (Huang et al., 2015) and a yearly average (data from 2006 to 2016) precipitation of 475.92 mm per year, 80% of which falls between May and September (Wang et al., 2020). According to the World Reference Base for Soil Resources (WRB), the area has saline-sodic meadows that contain an average of 46.9% sand (ranging from 32.34% to 61.77%) and an average of 42.4% silt (ranging from 30.82% to 52.97%). Thus, the general soil type in the area is classified as "sandy" or "sandy loam". The soil salinity, in electrical conductivity (EC), for the experimental area was measured from a ratio of soil:water of 1:5, and denoted as $EC_{1:5}$. The conversion from $EC_{1:5}$ to the widely accepted ECe (EC of soil saturated paste extracts) can be obtained by multiplying $EC_{1:5}$ value by a conversion factor of 10.88, based on the soil type found in the Songnen Plain (Chi and Wang, 2010). Before the experiment, analysis of the 0–20 cm topsoil layer determined that the soil salinity had an $EC_{1:5}$ of 0.72 dS m^{-1} (Table 1), which is equivalent to an ECe of 7.83 dS m^{-1}. Thus, our paddy soils can be classified as moderately saline to highly saline (https://www.agric.wa.gov.au/soil-salinity/measuring-soil-salinity?page=0%2C0#smartpaging_toc_p0_s8_h2).

The vegetation present before paddy field reclamation was the original grassland naturally degraded by soil sodicity and salinization and dominated by *Leymus chinensis* (Trin.) Tzvel., *Phragmites australis* (Poaceae), *Puccinellia tenuiflora* (Griseb.) Scribn. & Merr. (Poaceae), *Chloris virgata* Sw. (Poaceae), *Suaeda corniculata* (C.A.Mey) (Amaranthaceae), and other species adapted to saline-sodic and alkaline soils. In 2006, a piece of grassland with an area of 6 ha was reclaimed into 60 paddy field plots with an area of 1000 m^2 (40 m × 25 m) each. Out of these 60 plots, 12 experimental plots of 1000 m^2 (40 m × 25 m) each were randomly distributed in the 6 ha area of cultivated paddy fields (Fig. 1). These 12 plots were used to apply the treatments (amendments and tillage) and were cultivated with rice yearly. Another 2 ha area of

Fig. 1. The map on the left shows the study site (gray circle) in the city of Da'an, Jilin Province, Northeast China (based on Map data ©2021 Microsoft Corporation-GS [2021]1731, ©2021TomTom). On the upper right picture, the dotted red lines outline a 6 ha area of reclaimed paddy fields, into which 12 experimental plots (1000 m^2 each) were randomly distributed to accommodate the treatments PFU (paddy fields without amendments + tillage), PFPG (paddy fields with phosphogypsum + tillage), and PFFM (paddy fields with farmyard manure + tillage). Below the paddy fields, a 2 ha area of uncultivated grasslands accommodated 4 plots (1000 m^2 each) for the control treatment UG (uncultivated saline-sodic grassland without amendments or tillage) (based on Satellite Map ©2021 Baidu-GS [2019] 5218). On the bottom right, pictures show the steps involved in rotary tillage, harrowing and leveling, seedling transplant, and mechanical harvest. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1
The main chemical characteristics of saline-sodic soil, phosphogypsum, and farmyard manure that were used as saline-sodic soil amendments in this study.

Soil	Soil type	pH	$EC_{1:5}$	Na^+	K^+	$Ca^{2+} + Mg^{2+}$	Cl^-	SO_4^{2-}	$CO_3^{2-} + HCO_3^-$	SOM	AN	AP	AK
			dS m^{-1}			mmol$_c$ L^{-1}				g kg^{-1}		mg kg^{-1}	
Saline-sodic soil	Meadow soil	10.54	0.72	8.23	0.043	0.69	2.93	0.82	5.35	3.08	41.0	27.8	153.8
Amendment	Basic component	pH	$EC_{1:5}$	Na^+	CaO	AN	AP	AK	As	Pb	Cd	Cr	Hg
			dS m^{-1}	mmol$_c$ L^{-1}	%				mg kg^{-1}				
Phosphogypsum	$CaSO_4 \cdot H_2O$	3.63	0.22	1.07	19.93	31.9	134.9	43.2	2.04	14.81	0.02	6.24	1.86*
Farmyard manure	Sheep manure	8.65	0.21	1.78	/	17.2	13.1	20.6	1.46	12.45	1.31*	8.09	0.19*

Notes: SOM, soil organic matter; AN, available nitrogen; AP, available phosphorus; AK, available potassium; pH and $EC_{1:5}$ were measured at the ratio of soil to water of 1:5, the concentrations of five heavy metals in two amendments are in line with the fertilizer standard of the People's Republic of China (GB/T 23349-2009), the items marked with * indicates that the heavy metal concentration exceeds the soil quality standard of the People's Republic of China (GB 15618-2018).

naturally degraded grasslands, caused by soil salinization, was reserved as the control field (Fig. 1). All the plots were irrigated with groundwater from an 80 m-deep well. This water had an EC ranging from 0.51 to 0.93 dS m^{-1} and pH of 7.0–7.5.

2.2. Experimental design and treatments

In this study, the field experiments were carried out in a randomized complete block design with four replicates. The experimental treatments were as follows: (1) uncultivated, untreated, and untilled saline-sodic grassland (UG) as control plots. This area was covered by the original degraded vegetation (4 plots of 1000 m^2 or 40 m × 25 m each) and randomly distributed in the area of 2 ha described above; (2) paddy fields with untreated soil, tilled yearly, and cultivated with rice (PFU); (3) paddy fields amended only once with 25 t ha^{-1} (full recommended dose for this area) of phosphogypsum (PFPG), tilled yearly and cultivated with rice; and (4) paddy fields amended only once with 25 t ha^{-1} of cured farmyard manure (PFFM), usually from sheep, tilled yearly and cultivated with rice. Among the treatments, the areas assigned to PFU, PFPG, and PFFM were tilled. Tillage was carried out by rotary tilling the 0–20 cm topsoil layer using a rotary cultivator to break the topsoil and rice stubble. Then, the soil was left to dry for 2–3 weeks to increase soil temperature in the spring. Then, paddy fields were irrigated with groundwater enough to cover the soil with a water film of 5–10 cm, and left soaking for one week. Next, paddy fields were leveled using a homogenizer or water harrow wheel, left to precipitate the mixed mud for 3–5 days, drained, and re-soaked with new water. Rice seedlings were transplanted mechanically in late May and mature rice plants were reaped with a harvester in mid-October every year (Fig. 1). These plots of paddy fields had individual (separated) irrigation and drainage systems and, during the growing period, were continuously flooded with local groundwater to maintain a water film of 5–10 cm in depth, depending on the rainfall and evapotranspiration during rice growth.

In 2006, soil amendments were applied to the PFPG and the PFFM at once and not reapplied during the 10-year experimental period. The main chemical characteristics of phosphogypsum and farmyard (sheep) manure are listed in Table 1. These amendments were individually and uniformly mixed into the soil 15 cm top layer. The cultivated rice varieties were Changbai 9 (2006–2010) and Dongdao 4 (2011–2015), which are the main local japonica rice varieties (*Oryza sativa* L.) with good salt tolerance and a growth period of 135–138 days. Rice cultivation was started after washing salts and draining soil 2–3 times. In the three tilled paddy field treatments (PFU, PFPG, and PFFM), the same amount of NPK fertilizer was applied every year. A compound fertilizer ($N\text{-}P_2O_5\text{-}K_2O$ = 18–18–18, Sakefu®, Jilin Longyuan Agricultural Materials Co., Ltd) was incorporated into the 10-cm topsoil layer at 500 kg ha^{-1} one week before transplanting rice. Urea (46.5% N, Changshan®, Jilin Changshan chemical fertilizer Group Co. Ltd) was broadcasted onto the soil-water surface 15 days after transplanting, at 100 kg ha^{-1}, to support tillering. Changshan® urea (100 kg ha^{-1}) and potassium sulfate (Hongniu®, 50% K_2O, German Potash Company) were applied at 20 kg ha^{-1} onto the soil-water surface 50 days after transplanting to support panicle formation. The total amounts of N, P, and K fertilizer were 183 kg ha^{-1}, 90 kg ha^{-1}, and 100 kg ha^{-1}, respectively. This conventional rice cultivation and management were consistent with rice recommendations for the region.

2.3. Soil sampling and analysis

The experimental pads had saline-sodic meadow soil (according to WRB) with the specific soil characteristics listed in Table 1. Soil samples were collected from all four treatment plots at the end of October 2015. Samples were composites of 5 soil cores (core diameter of 5 cm) from each plot of 1000 m^2 area. The 60 cm deep sample cores were divided into 0–20, 20–40, 40–60 cm depth increments. Samples were air-dried and passed through a 2 mm mesh sieve for analyses of soil salinity, main nutrients, and heavy metal concentrations.

Soil pH (soil:water ratio of 1:5) was measured by using a CyberScan pH510 pH meter (Eutech Instruments Pte. Ltd., Singapore, USA), soil $EC_{1:5}$ was determined by using a DDS 11AW conductivity meter (Hangzhou Huier Instrument Equipment Co. Ltd., Hangzhou, China). The concentrations of Ca^{2+} and Mg^{2+} in soil solutions were analyzed with an AA-6300 Atomic Absorption Spectrophotometer (Shimadzu, Shanghai, Japan). The concentrations of K^+ and Na^+ in soil solutions were analyzed with a FP6410 Flame photometer (Shanghai Precision & Scientific Instrument Co. Ltd., Shanghai, China). The concentrations of Cl^-, SO_4^{2-}, CO_3^{2-} and HCO_3^- in soil solutions were analyzed with a WDDY-2008 Potentiometric Titrator (Datang Analysis Instrument Co. Ltd., Taizhou, China). Exchange sodium and other cations adsorbed on soil colloids were extracted using 1.0 mol L^{-1} ammonium acetate (NH_4OAc)-alcohol (C_2H_6O) and the extract was analyzed for exchange sodium concentration and soil cation exchange capacity using a FP6410 Flame photometer.

The main parameters of soil salinization include pH, electrical conductivity ($EC_{1:5}$), exchange sodium percentage (ESP), sodium adsorption ratio (SAR), total alkalinity (TA), and residual sodium carbonate (RSC). The ESP, SAR, TA, and RSC were calculated by different ions concentrations, and the formulas are as follows:

$$ESP(\%) = \frac{[Na_{ex}^{+}]}{CEC} \times 100$$

$$SAR = \frac{[Na^{+}]}{\sqrt{\frac{[Ca^{2+}]+[Mg^{2+}]}{2}}}$$

$$TA\ (mmol\ L^{-1}) = [CO_3^{2-}] + [HCO_3^{-}]$$

$$RSC\ (mmol\ L^{-1}) = ([CO_3^{2-}] + [HCO_3^{-}]) - ([Ca^{2+}] + [Mg^{2+}])$$

where $[Na_{ex}^{+}]$ is the exchangeable sodium concentration adsorbed on soil colloids, CEC is the soil cation exchange capacity, $[Na^+]$ is the sodium ion concentration in soil solution, ($[Ca^{2+}]$ + $[Mg^{2+}]$) is the total concentration of calcium and magnesium ions in soil solution, ($[CO_3^{2-}]$ + $[HCO_3^-]$) is the total concentration of carbonate and bicarbonate in soil solution.

Soil organic matter (SOM), available nitrogen (AN), available phosphorus (AP), and available potassium (AK) were measured using conventional analytical methods according to the Handbook for Soil Analysis (Bao, 2000). The concentrations of chromium (Cr), cadmium (Cd), lead (Pb), arsenic (As), and mercury (Hg) in soil were also measured by atomic absorption spectrometry or by atomic fluorescence spectrometry according to the National Standards of the People's Republic of China, 1997, 2008 (GB/T 17137-1997, GB/T 17140-1997 and GB/T 22105-2008).

2.4. Statistical analysis

Statistical analysis was performed using the SPSS 21.0 for Windows software package (SPSS Inc., Chicago, IL, USA). All data (including, but not limited to, soil pH, $EC_{1:5}$, ESP, concentrations of salt ions, concentrations of nutrients, and heavy metals) were collected with four replicates and subjected to a one-way analysis of variance (ANOVA) with the means separated using Duncan's multiple range test at the 5% level of significance ($P < 0.05$). The data were reported as mean ± standard error of the mean (SEM). Correlation analysis was carried out among the main parameters of soil salinization, and the Pearson correlation coefficient was reported with 95% or 99% confidence ($\alpha = 0.05$ or 0.01). Redundancy analysis (RDA) for the effects of soil salt ions on soil nutrients concentrations was performed using the vegan package (Oksanen et al., 2019) in the software R version 3.5.2 (R Core Team, 2019) and RDA graphic was drawn with ggplot2 package software (Wickham, 2016). The other graphs were generated using SigmaPlot Version 11.0 (Systat Software, Inc. SigmaPlot for Windows, San Jose, CA, USA), and the standard error of the means (SEM) were calculated and presented in the graphs as error bars.

3. Results

3.1. Long-term effect of tillage combined with rice cultivation and amendments on soil salt ions, salinization, and alkalization parameters

Both tillage and amendments, applied before rice cultivation, for 10 years significantly decreased total salt ion concentrations under different soil layers, although salinity was always higher in deeper layers than in the topsoil layer (Fig. 2). Tillage and rice cultivation alone (PFU) had a significant effect on the reduction of soil Na^+ and Cl^-, but PFPG and PFFM treatments produced significant individual effects in reducing the concentrations of Na^+, CO_3^{2-} + HCO_3^- and Cl^- ($P < 0.05$), although the reduction of Cl^- was not significantly better than tillage alone (PFU) (Fig. 2). At the 0–20 cm topsoil layer, the concentrations of Na^+ in PFU, PFPG, and PFFM decreased by 42.9%, 61.5%, and 60.9%, respectively, compared with that in UG, and those in PFPG and PFFM decreased by 32.5%, and 31.5%, respectively, compared with that in PFU. Similarly, the concentrations of CO_3^{2-} + HCO_3^- in PFU, PFPG, and PFFM decreased by 18.9%, 63.2%, and 57.9%, respectively, compared with that in UG, and those in PFPG and PFFM decreased by 54.6%, and 48.2%, respectively, compared with that in PFU. The concentrations of Cl^- in PFU, PFPG, and PFFM decreased by 64.6%, 75.7%, and 79.9%, respectively, compared with that in UG. The concentration changes of the above four soil salt ions showed a significant ($P < 0.05$) downward trend in response to annual tillage and to both amendments, the latter applied only once to rice paddy fields at the beginning of the 10-year experimental period.

The concentrations of Ca^{2+} + Mg^{2+}, and SO_4^{2-} in PFPG and PFFM were slightly, but not significantly, increased compared with those in UG or PFU, and the concentration of Ca^{2+}+Mg^{2+} in PFFM was

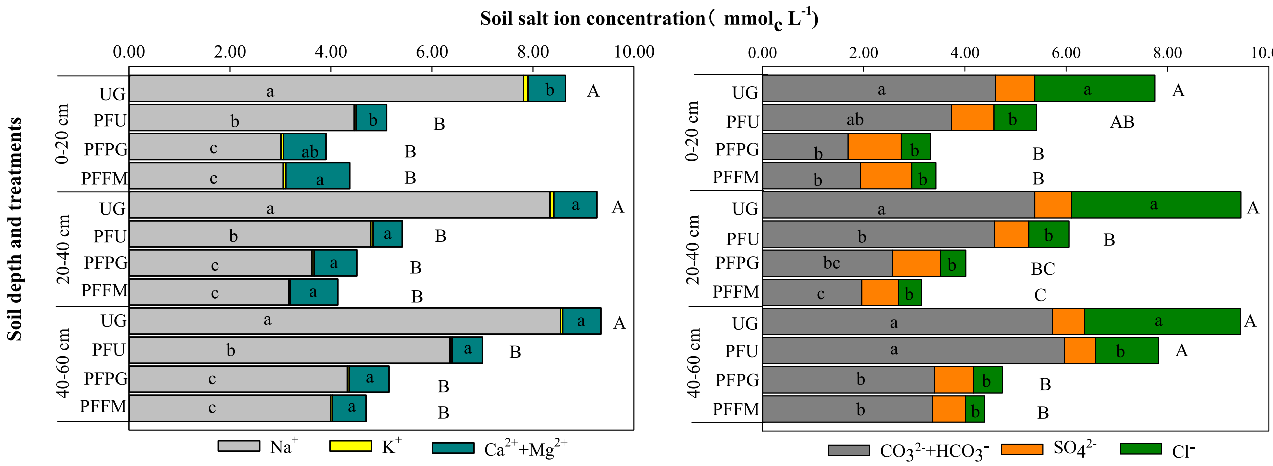

Fig. 2. Changes on the concentration of main salt ions in paddy soils after 10 years under the respective treatments, as follows: UG, uncultivated saline-sodic grassland; PFU, paddy fields without amendments + tillage; PFPG, paddy fields with phosphogypsum + tillage; PFFM, paddy fields with farmyard manure + tillage. Capital letters on the outside of the bar indicate a significant difference of total salt-ion concentration among the different treatments ($P < 0.05$); lowercase letters on the same color bar indicate a significant difference in the concentration of specific ions among the different treatments ($P < 0.05$); no letters on the same color bar indicate no significant difference in the specific ion concentration among the different treatments. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

significantly increased compared with that in UG ($P < 0.05$). There was no significant change in the concentration of K^+ in the four different treatments. With the increase in soil depth, the change trends of various salt ion concentrations were similar to those observed for the topsoil layer. Under the same treatment, the concentration of $Ca^{2+} + Mg^{2+}$ increased with increasing soil depth, but there was no significant difference (Fig. 2).

For the four treatments, the trends in changes in soil pH, salinity $EC_{1:5}$, ESP, SAR, total alkalinity, and RSC in different depths were almost the same, meaning that all soil salinization and alkalization parameters increased with increasing soil depth for each treatment and that they were reduced by tillage or application of soil amendments before planting rice at the same soil depth (Fig. 3). In the 0–20 cm topsoil layer, soil pH of PFU, PFPG, and PFFM significantly ($P < 0.05$) decreased in 0.57, 1.05, and 1.30 units, respectively, compared with that of UG. Similarly, salinity $EC_{1:5}$, ESP, SAR, total alkalinity, and RSC of PFU decreased by 18.3%, 47.2%, 43.3%, 18.9%, and 26.7%, respectively, compared with those of UG. Salinity $EC_{1:5}$, ESP, SAR, total alkalinity, and RSC of PFPG decreased significantly ($P < 0.05$) by 49.1%, 66.9%, 62.2%, 63.3%, and 71.2%, respectively, compared with those of UG. Salinity $EC_{1:5}$, ESP, SAR, total alkalinity, and RSC of PFFM decreased significantly ($P < 0.05$) by 48.3%, 72.5%, 64.4%, 58.0%, and 62.9%, respectively, compared with those of UG.

With increasing soil depth, the parameters of soil pH, $EC_{1:5}$, ESP, SAR, total alkalinity, and RSC all increased gradually in the same treatment, but there was no significant difference ($P \geq 0.05$) for different depths (Fig. 3). For the same soil depth, these parameters in PFPG and PFFM were significantly smaller ($P < 0.05$) than the corresponding values in UG, and those in PFU were slightly less than the corresponding values in UG. There were no significant differences for most parameters between PFU and UG, and for all parameters between PFPG and PFFM ($P \geq 0.05$).

Soil pH and salinity $EC_{1:5}$ were measured directly by the appropriate instruments, while ESP, SAR, and RSC were calculated using the concentration of relevant salt ions. The correlation analysis among these parameters indicated that soil pH, $EC_{1:5}$, ESP, SAR, and RSC were highly, and positively, correlated with each other ($P < 0.01$) in saline-sodic soils (Table 2). These salinization and alkalization parameters were significantly and positively correlated with the concentrations of CO_3^{2-} + HCO_3^-, Na^+ (especially Na_{ex}^+) and Cl^-, and were significantly and negatively correlated with the concentrations of SO_4^{2-} ($P < 0.01$). These salinization and alkalization parameters were not significantly correlated with the concentrations of $Ca^{2+} + Mg^{2+}$ (except for soil pH) or K^+ in saline-sodic soils (Table 2). Among various soil salt ions, the significant correlations were also found among CO_3^{2-} + HCO_3^-, Na^+ (including Na_{ex}^+) and Cl^- ($P < 0.01$), between Na^+ and K^+ ($P < 0.05$), and between $Ca^{2+} + Mg^{2+}$ and SO_4^{2-} ($P < 0.05$).

3.2. Long-term effects of tillage combined with rice cultivation and amendments on soil fertility

Tillage combined with rice cultivation and the application of amendments before planting rice increased the concentrations of SOM, AN, AP, and AK at different soil depths (Fig. 4). With increasing soil depth, the concentrations of these nutrients remained unchanged in the same treatment, and there was no significant difference among the different soil depths for the same nutrient concentration ($P \geq 0.05$). In the 0–20 cm topsoil layer, the concentrations of SOM, AN, AP, and AK in the PFU increased 42.6%, 16.9%, 32.9%, and 16.2% compared with those in the UG, respectively. The concentration of AP increased significantly ($P < 0.05$), but the other three nutrients had no significant difference. The concentrations of SOM, AN, AP, and AK increased in 116.5%, 74.3%, 32.4%, and 15.2%, respectively, in PFPG, and in 165.6%, 103.6%, 39.9%, and 45.4%, respectively, in PFFM compared with those in UG. Except for AK, the concentrations of SOM, AN, and AP in PFPG and PFFM were significantly higher than those in UG ($P < 0.05$).

The trend changes of soil nutrient concentrations were roughly contrary to the concentration changes of main salt ions (Fig. 2) and the parameters of soil salinization/alkalization (Fig. 3) in the four different treatments. The concentrations of SOM, AN, AP, and AK were all affected by the concentrations of various salt ions. The redundancy analysis indicated that there were negative correlations between these soil parameters and main soil salinity (Fig. 5). In the biplot, the variance ratio of RDA1 (28.74%) reached a significant level ($P < 0.01$), and the salt indexes that had the greater negative impacts on the four soil parameters (SOM, AN, AP, and AK) were pH (0.961), the concentration of exchangeable Na_{ex}^+ (0.772), and CO_3^{2-} + HCO_3^- (0.761), in the sequence cited. Meanwhile, there were some positive correlations among the concentrations of SO_4^{2-} or $Ca^{2+} + Mg^{2+}$ and soil nutrients.

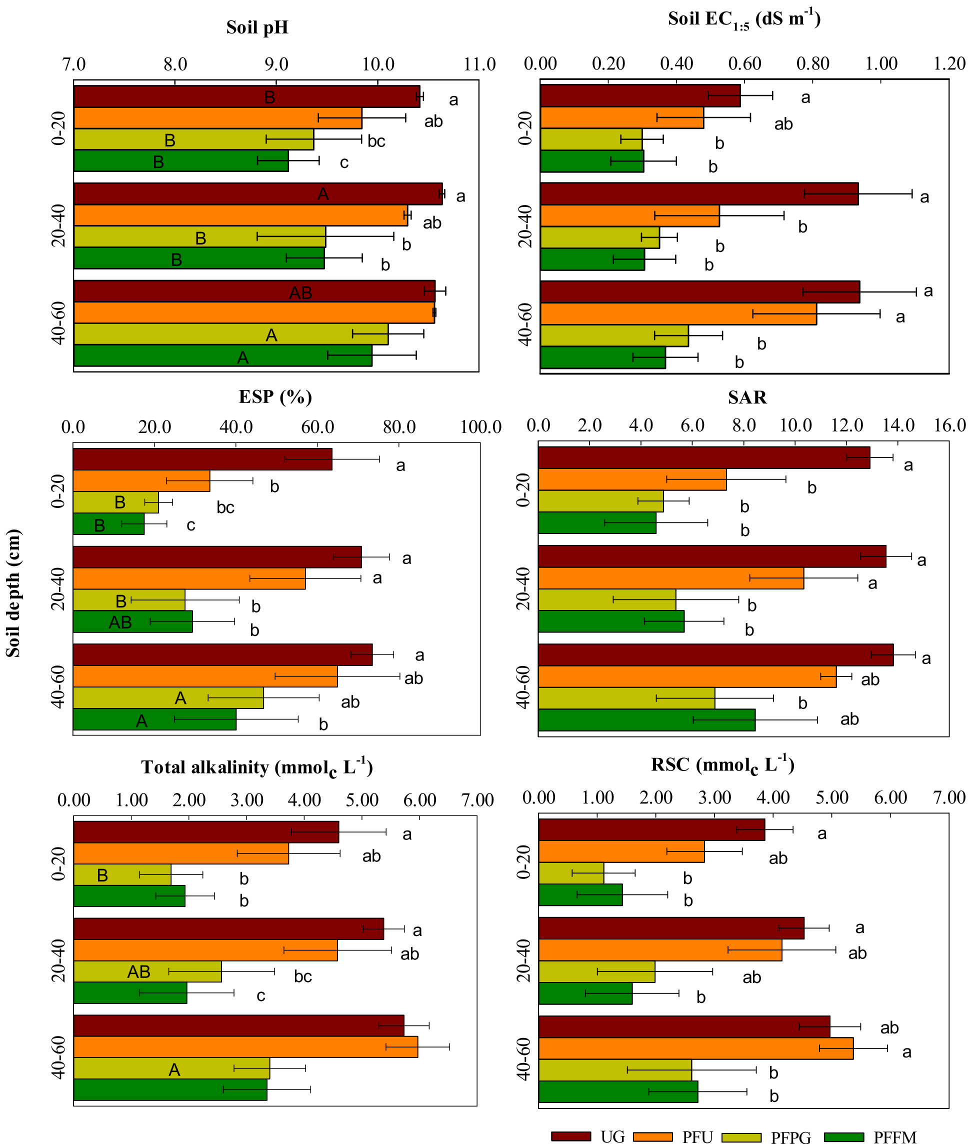

Fig. 3. Changes of soil salinization and alkalization parameters at different soil depths in the four experimental treatments. Bars represent means ± SEM, n = 4 (replicated plots). UG, uncultivated saline-sodic grassland; PFU, paddy fields untreated + tillage; PFPG, paddy fields with phosphogypsum + tillage; PFFM, paddy fields with farmyard manure + tillage; $EC_{1:5}$, electrical conductivity; ESP, exchangeable sodium percentage; SAR, sodium adsorption ratio; RSC, residual sodium carbonate; soil pH and $EC_{1:5}$ were measured at the ratio of soil to water of 1:5; total alkalinity is the sum of the concentrations of CO_3^{2-} and HCO_3^-. Capital letters on bars of the same color indicate a significant difference of the specific parameter among the different depths ($P < 0.05$); lowercase letters on the outside of bars indicate a significant difference of the specific parameter among different treatments, under the same depth ($P < 0.05$); no letters on bars of the same color or on the outside of the bar indicate that there was no significant difference in the corresponding specific parameter among the different soil depths or treatments. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 2
Pearson correlation analysis between soil salinization-alkalization parameters and various salt ion concentrations.

	EC	ESP	SAR	RSC	Na^+	K^+	Ca^{2+} + Mg^{2+}	Cl^-	SO_4^{2-}	CO_3^{2-} + HCO_3^-	Na_{ex}^+
pH	0.884**	0.983**	0.947**	0.962**	0.888**	0.389	-0.633*	0.745**	-0.782**	0.936**	0.924**
EC		0.912**	0.912**	0.933**	0.932**	0.377	-0.357	0.884**	-0.614*	0.955**	0.958**
ESP			0.972**	0.948**	0.928**	0.424	-0.517	0.810**	-0.769**	0.932**	0.948**
SAR				0.938**	0.961**	0.512	-0.477	0.869**	-0.710**	0.928**	0.920**
RSC					0.883**	0.417	-0.542	0.786**	-0.730**	0.986**	0.923**
Na^+						0.584*	-0.327	0.953**	-0.563	0.903**	0.933**
K^+							0.070	0.639*	0.112	0.446	0.366
Ca^{2+} + Mg^{2+}								-0.122	0.603*	-0.465	-0.372
Cl^-									-0.385	0.822**	0.865**
SO_4^{2-}										-0.669*	-0.684*
CO_3^{2-} + HCO_3^-											0.937**

Notes: EC, electrical conductivity; ESP, exchangeable sodium percentage; SAR, sodium adsorption ratio; RSC, residual sodium carbonate; Na_{ex}^+, exchange sodium; soil pH and EC were measured at the ratio of soil to water of 1:5. ** Correlation is significant at the 0.01 level (2-tailed), * Correlation is significant at the 0.05 level (1-tailed).

3.3. Long-term effects of tillage and amendments on heavy metal accumulation

Like salt ions, most of the five heavy metal concentrations had no significant changes at different depths of saline-sodic soil, and only the concentration of As in PFFM and of Pb in UG increased significantly ($P < 0.05$) with increasing soil depths (Table 3). After 10 years of tillage, amendment, and rice cultivation in saline-sodic soil, the concentration of As in soil, from 0 to 60 cm, decreased significantly ($P < 0.05$). Taking the 0–20 cm topsoil layer as an example, the concentrations of As in

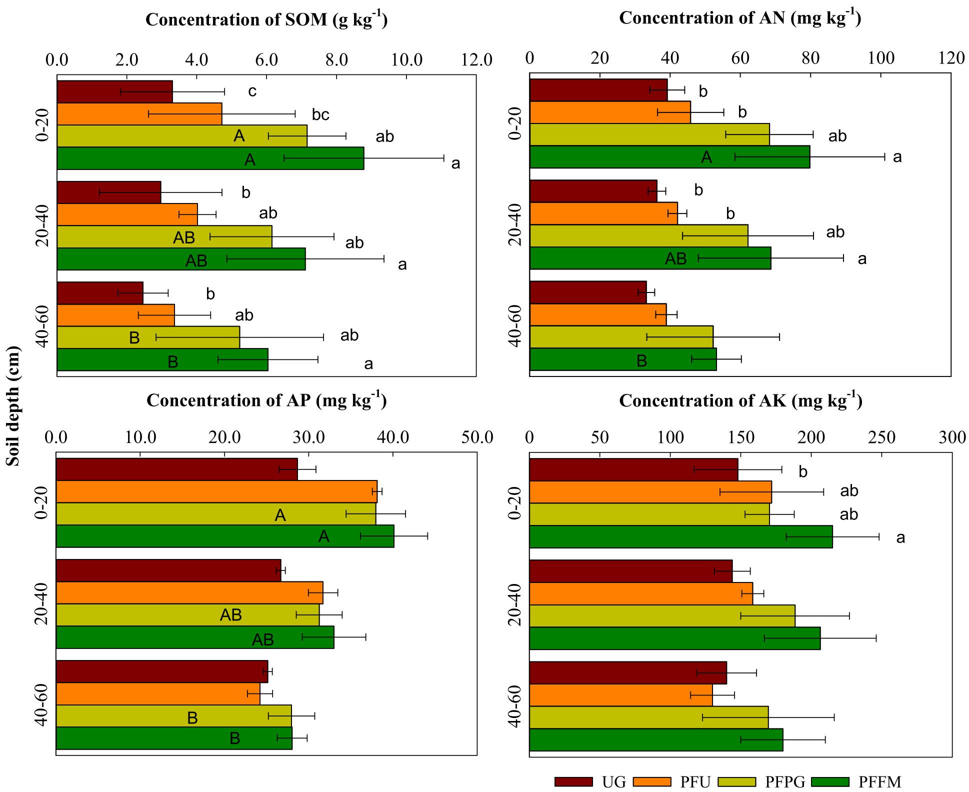

Fig. 4. Changes in the concentrations of soil organic matter (SOM), available nitrogen (AN), available phosphorus (AP), and available potassium (AK) at different soil depths in each of the four experimental treatments. Bars represent means ± SEM, n = 4 (replicated plots). UG, uncultivated saline-sodic grassland; PFU, paddy fields untreated + tillage; PFPG, paddy fields with phosphogypsum + tillage; PFFM, paddy fields with farmyard manure + tillage. Capital letters on bars of the same color indicate a significant difference in the nutrient concentration among the different depths ($P < 0.05$); lowercase letters on the outside of the bars indicate a significant difference in the nutrient concentration among the different treatments, under the same depth ($P < 0.05$); no letters on bars of the same color or on the outside of the bar indicate that there was no significant difference in the corresponding specific nutrient concentration among different soil depths or treatments. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

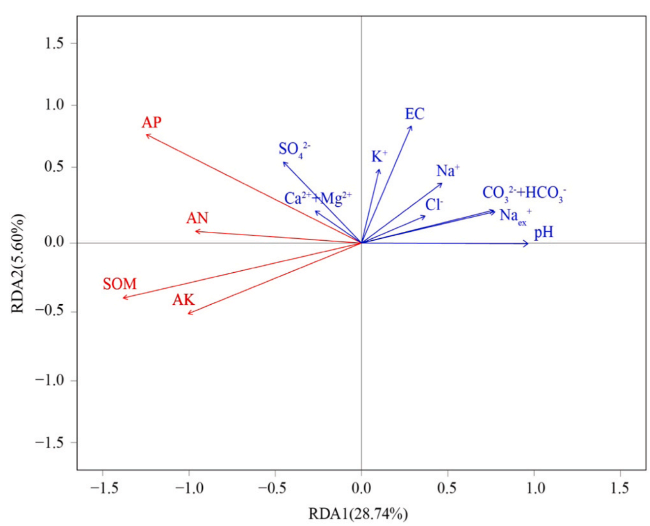

Fig. 5. Biplot for redundancy analysis (RDA1 & RDA2) for the effect of soil salt ions on soil nutrients concentrations of saline-sodic paddy soils. SOM, soil organic matter; AN, available nitrogen; AP, available phosphorus; AK, available potassium; EC, electrical conductivity. Soil pH and EC were measured at the ratio of soil to water of 1:5.

PFU, PFPG and PFFM decreased 2.6%, 36.6%, and 54.2%, respectively, compared with that in UG. In the 0–20 cm topsoil layer, the concentrations of Hg in PFPG and PFFM slightly increased compared with that in UG; slightly decreased between PFU and UG, and significantly decreased in PFFM compared with that in PFU ($P < 0.05$). However, the concentrations of Pb, Cd, and Cr in the same soil depth did not change among the four experimental treatments.

4. Discussion

4.1. Long-term effect of tillage combined with rice cultivation and amendments on soil salt ions, salinization, and alkalization parameters

It was expected that tillage and amendments would be beneficial to decrease soil salinization and sodicity, as demonstrated by previous studies (Tan and Kang, 2009; Chi et al., 2012; Huang et al., 2016; Sundha et al., 2020). However, with the growing use of tillage and amendments applied to saline-sodic soils before planting rice, we needed to verify which salt ions were being leached more efficiently with those practices. Our 10-year experiment established that the concentrations of Na^+, CO_3^{2-} + HCO_3^-, and Cl^- in the 0–20 cm topsoil layer in PFU decreased by 42.9%, 18.9%, and 64.6%, respectively, compared with those of UG (Fig. 2), which indicated that Cl^- and free Na^+ were easily leached from the soil. While soil salts were reduced by phosphogypsum or farmyard sheep manure, the concentrations of Na^+ and CO_3^{2-} + HCO_3^- were reduced at a higher degree than the concentration of Cl^-. In addition, the concentration of CO_3^{2-} + HCO_3^- decreased by 54.6% and 48.2% in the 0–20 cm topsoil layer in PFPG and PFFM compared with that in PFU (Fig. 2), indicating that soil amendments played an important role in reducing salt ion concentration of CO_3^{2-} + HCO_3^-.

In saline-sodic soils of Northeast China, Na^+ (mainly Na_{ex}^+) is often associated with CO_3^{2-} or HCO_3^-. Therefore, Na_2CO_3 and $NaHCO_3$ are also considered to be the main sodic salt components of this soil. Our results agree with this observation, more specifically in that the concentration of CO_3^{2-} + HCO_3^- was only positively correlated with the concentration of Na^+ or Na_{ex}^+ (r = 0.903 or 0.937, $P < 0.01$) among the four main soil cations (Table 2). The hydrolysis of inherent Na_2CO_3 and $NaHCO_3$ in the soil produced a large amount of CO_3^{2-} and HCO_3^-, increased sodium saturation in the system, and was accompanied by an increase in soil pH (Gupta et al., 1984; Sundha et al., 2018). High pH, the concentrations of soluble salts, and the accumulation of Na^+ generally increased osmotic stress and Na^+ toxicity, while decreasing mineral availability because of the competition between different ions (e.g., Na^+ vs. K^+) for plant

Table 3
Changes in the concentrations of arsenic (As), lead (Pb), cadmium (Cd), chromium (Cr) and mercury (Hg) at different soil depths under four treatments, after 10 years of tillage, amendments, and rice cultivation. Data are means ± SEM, n = 4 (replicated plots).

Items	Soil depth (cm)	Treatments				Compare with environmental quality standard for soils from China
		UG	PFU	PFPG	PFFM	
As (mg kg^{-1})	0–20	1.53 ± 0.10 A	1.49 ± 0.44 A	0.97 ± 0.27 B	0.70 ± 0.12 c B	≤ 15 mg kg^{-1}
	20–40	1.68 ± 0.06 A	1.45 ± 0.38 AB	0.96 ± 0.21 B	0.99 ± 0.29 b B	
	40–60	1.68 ± 0.31 AB	1.94 ± 0.05 A	0.99 ± 0.20 C	1.37 ± 0.26 a BC	
Pb (mg kg^{-1})	0–20	23.50 ± 0.32c	24.37 ± 2.38	24.40 ± 2.62	24.98 ± 1.81	≤ 35 mg kg^{-1}
	20–40	26.03 ± 0.38 b	24.52 ± 0.93	25.72 ± 1.39	25.58 ± 2.00	
	40–60	28.39 ± 0.91 a	28.28 ± 1.29	25.99 ± 2.56	27.42 ± 2.78	
Cd (μg kg^{-1})	0–20	16.07 ± 1.23	16.81 ± 0.30	15.68 ± 1.88	15.97 ± 0.55	≤ 200 μg kg^{-1}
	20–40	15.03 ± 0.65	14.09 ± 1.35	16.11 ± 1.67	15.01 ± 1.12	
	40–60	15.18 ± 1.59	16.20 ± 0.67	15.10 ± 0.67	15.42 ± 1.06	
Cr (mg kg^{-1})	0–20	15.55 ± 1.75	15.44 ± 0.12	16.20 ± 4.40	16.78 ± 3.65	≤ 90 mg kg^{-1}
	20–40	16.62 ± 1.35	19.99 ± 0.96	17.09 ± 2.42	19.04 ± 5.54	
	40–60	18.47 ± 1.17	22.61 ± 7.86	18.59 ± 2.71	18.43 ± 2.33	
Hg (μg kg^{-1})	0–20	16.11 ± 2.49 AB	12.15 ± 0.93 B	20.02 ± 6.42 AB	27.12 ± 9.44 A	≤ 150 μg kg^{-1}
	20–40	17.52 ± 1.82	12.91 ± 0.81	20.31 ± 5.27	20.27 ± 6.99	
	40–60	17.65 ± 1.46	14.54 ± 3.84	16.36 ± 4.55	13.77 ± 3.63	

Notes: lowercase letters indicate a significant difference of corresponding heavy metal concentration between different soil depths, within the same treatment columns ($P < 0.05$); capital letters indicate a significant difference of corresponding heavy metal concentration between different treatments, at the same soil depth ($P < 0.05$); no letters indicate that there was no significant difference in the concentration of the same heavy metal between different soil depths or different experimental treatments ($P \geq 0.05$). UG, uncultivated saline-sodic grassland; PFU, paddy fields untreated + tillage; PFPG, paddy fields with phosphogypsum + tillage; PFFM, paddy fields with farmyard manure + tillage.

absorption in these soils (Chhabra, 1996; Hillel, 1998; Rengasamy, 2006; Karlin et al., 2011; D'Odorico et al., 2013). Additionally, the excess of exchangeable Na^+ contributed to breaking up soil aggregates, diminishing aeration, and hydraulic conductivity (Bronick and Lal, 2005). All the above factors combined limit the growth of plants, making it necessary to improve saline-sodic soil chemical and physical characteristics to restore crop productivity potential.

As we have mentioned previously, there are many effective methods and alternative materials for the improvement of saline-sodic soils (Eynard et al., 2006; Sadiq et al., 2007; Chi et al., 2012; Shaaban et al., 2013a; Huang et al., 2016). Although it was difficult to determine which improvement methods or agents had the best effects, the key to mitigating soil salinity and sodicity was in knowing how much, and which ions can be removed from the soil. Our results confirmed that soils treated with amendments (PFPG and PFFM) had lower concentrations of Na^+, CO_3^{2-} + HCO_3^- and Cl^- (Fig. 2), indicating that the soil amendments phosphogypsum and farmyard manure can promote soil desalination during rice cultivation in saline-sodic soils. The specific principle of improving saline-sodic soil through the application of phosphogypsum or farmyard manure has been reported by many researchers (Bronick and Lal, 2005; Cruickshank, 2007; Chaganti et al., 2015; Sundha et al., 2020). Phosphogypsum is generally considered as a rich source of Ca^{2+}, and its use enhances Ca^{2+}, Mg^{2+}, and K^+ saturation at the exchange soil sites (Walker and Bernal, 2008), and it can replace exchangeable Na^+ with Ca^{2+} (El Hasini et al., 2019), which also benefits plant growth. Application of farmyard manure can increase the contents of soil organic matter and release free organic acids which neutralize alkalinity in soils (Chaganti et al., 2015). However, when applied in excess cow manure increased soil salinity and decreased crop yield (Gonçalo Filho et al., 2020). Both amendments can also accelerate the leaching of Na^+, reduce soil pH, ESP, EC, and improve the water-holding capacity and aggregate stability of soil (Bennett et al., 2015; Chaganti et al., 2015; Murphy, 2015; Singh et al., 2018).

Traditionally, soil salinity and alkalinity changes have been evaluated using soil samples through laboratory analyses (Rhoades et al., 1997). However, in practice, the analysis of many soil salinization and alkalization parameters is too complex and difficult; so, it is necessary to select a small number of representative, simple, and easy-to-analyze parameters. In our study, the trend changes of six soil salinization and alkalization parameters in different experimental treatments were consistent with the concentrations of main salt ions in the soil (Figs. 2 and 3). Correlation analysis also indicated that there were significant ($P < 0.01$) positive correlations among the six soil salinization parameters for pH, $EC_{1:5}$, ESP, SAR, RSC, CO_3^{2-} + HCO_3^- (Table 2). These significant correlations were observed between the six salinization parameters and the concentrations of Na^+ (especially Na_{ex}^+), CO_3^{2-} + HCO_3^- and Cl^-, which were the main salt ions in the saline-sodic soil under experimentation (Table 2). Among many salinization parameters and soil salt ions, pH and $EC_{1:5}$ are relatively easy to determine. Our previous studies also demonstrated that soil pH and $EC_{1:5}$ were the main sodicity and salinity indicators, respectively, that affected crop yield in saline-sodic soils (Huang et al., 2017, 2019). Therefore, soil pH and $EC_{1:5}$ can be used as two relatively simple and practical indicators to evaluate soil salinization and sodicity.

4.2. Long-term effects of tillage combined with rice cultivation and amendments on soil fertility

Numerous studies have demonstrated that tillage and the use of organic manure/chemical amendments not only decreased soil salinity but also improved soil fertility of salt-affected soils (Yaduvanshi and Sharma, 2008; Usman et al., 2016; Xie et al., 2020). Our previous investigations also confirmed that high pH and salt concentrations significantly inhibited nutrients absorption of rice and reduced soil fertility in saline-sodic soil of Northeast China (Huang et al., 2017; Chen et al., 2018). Our results showed that the concentrations of SOM, AN, AP, and AK were significantly increased ($P < 0.05$) after 10 years of planting rice in saline-sodic soil. In particular, the concentrations of SOM and AN in the 0–20 cm topsoil layer in the PFPG increased by 51.7% and 49.0% compared with those in the UG, respectively. Both SOM and AN increased by 86.0% and 74.0%, respectively, in PFFM compared with UG. These results illustrated that soil amendments can accelerate the improvement of soil fertility in the process of planting rice in saline-sodic soils. However, according to the national soil nutrient content classification formulated by the 2nd Soil Census in China (National Soil Survey Office, 1998), the saline-sodic paddy field belongs to the category of high-yield farmland in terms of the concentrations of AP and AK, while it is still classified as low-yield farmland in terms of its concentrations of SOM and AN. Consequently, research directed at increasing soil organic carbon and nitrogen should be the forefront priorities to improve saline-sodic paddy fields in the future.

The relationship between soil salinity and nutrients has attracted increasing attention in the past decades, but most of them focused on the effects of soil salinity/alkalinity on the absorption of mineral nutrients by plants (Grattan and Grieve, 1999; Hu and Schmidhalter, 2005). Information is scarce on salinity/alkalinity influence on the transformation and accumulation of soil nutrients in soils and absorption by plants. It is often reported that the concentrations of SOM and N were relatively low in saline-sodic soils in Northeast China (Huang et al., 2010, 2016; Chen et al., 2018). The experimental data from this study indicated that the concentrations of SOM and AN could not be increased to a satisfactory level even after 10 years of rice cultivation and the application of commercial fertilizers that provided 183 kg N ha^{-1}, 90 kg P_2O_5 ha^{-1}, and 100 kg K_2O ha^{-1} every year (Fig. 4). Some studies also indicated that the total N and inorganic N in soils significantly increased under the long-term repeated application of mineral and organic fertilizer (Ju et al., 2006; Zhou et al., 2013). However, our data indicated that the increased soil C and N in saline-sodic paddy fields was not ideal during rice cultivation, which may be related to an excessive loss of both C and N under high-pH in the soil environment (Li et al., 2017). The loss of N could also have been caused for its unavailability to plants under moderately high soil salinity, as reported previously for corn cultivated in a loamy sand soil (Lacerda et al., 2018). The RDA analysis demonstrated that high pH caused by high values of exchangeable Na_{ex}^{+} and CO_3^{2-} + HCO_3^{-} became the main factor leading to poor soil-fertility improvement (Fig. 5). The soil sodicity might be responsible for high mineralization and loss of SOM and N through the increased solubility of SOM and N in the presence of high pH and Na^{+} (Wong et al., 2010; Usman et al., 2016). Therefore, it is necessary to reduce soil salinity and alkalinity to improve the concentrations of SOM and N in saline-sodic paddy fields. Our results clearly indicated that soil tillage combined with both amendments was the most effective practice in reducing SOM and N losses caused by soil sodicity.

4.3. Long-term effects of tillage and amendments on heavy metal accumulation

Heavy metal contamination in farmland soils has attracted increasing attention of researchers worldwide, especially in areas where pesticides, herbicides, and chemical fertilizers are overused, and where there is the application of sewage irrigation, soil amendments, and industrial waste (Zhao et al., 2010; Lü et al., 2018; Zhou et al., 2019). In some acid soil paddy fields in southern China, the problem of Pb, Cr, As, Cd, and Hg exceeding the acceptable health standards established by local and/or federal government, was prominent and about 10% of agricultural soils were contaminated by these heavy metals (Kou et al., 2018; Ouyang et al., 2020). In salt affected soils, little information on soil heavy metal status has been reported. However, many industrial or agricultural wastes have been used for soil amelioration, such as the waste of desulfurization of gypsum, phosphogypsum, and from intensive livestock production systems, such as poultry manure.

It is well known that plants readily take up heavy metals from contaminated soils. Although the American Environmental Protection Agency (EPA) has finally moved towards banning the use of organic-arsenic-based coccidiostat drugs in poultry production systems in the US, this practice may continue in other countries and other compounds used in animal production systems may contaminate soils beyond what is acceptable by some health/environmental standards. Recently, several potentially toxic elements (PTEs) originated in poultry feed as additives or growth promoters, and transferred to crops and vegetables through manure amendments, have been reported to be responsible for levels of Cd, Cr, Cu, and Pb bioaccumulation in radish, garlic, barley, and wheat above the levels accepted by the Chinese EPA (SEPA) and WHO/FAO (Muhammad et al., 2020). Other studies have reported that the concentrations of heavy metals (Cd, As, Pb, Hg, and Cr) originated from desulfurized gypsum used in reclaimed soils and crops were far lower than the established standards and below detectable limits, indicating that certain industrial wastes could be applied safely in saline-alkaline soils (Wang et al., 2017; Zhao et al., 2018). Also, the concentrations of Zn and Cu increased or exceeded accepted standards when 20% of sludge compost was added to a saline-alkali soil (Zhu et al., 2012).

Our experimental results established that there were no significant differences in the concentrations of Pb, Cd, and Cr in soil from the two amendments applied to saline-sodic soil after 10 years. The concentrations of As decreased significantly when the soil was amended with phosphogypsum or sheep manure, and only the concentrations of Hg in the 0–20 cm topsoil layer in PFFM significantly increased compared with that of PFU. However, the concentrations of all these five heavy metals were far lower than that established by environmental quality standards for soils in China (Table 3).

Although the concentrations of some heavy metals in phosphogypsum and farmyard manure exceeded the soil quality standards, such as the concentration of Hg in phosphogypsum and the concentrations of Cd and Hg in sheep manure (Table 1), the levels of soil heavy metals (such as As, Cd, and Pb) generally decreased with increasing salinity. Heavy metals were negatively correlated with electrical conductivity (EC) but positively correlated with soil organic carbon (SOC) ($P < 0.05$) (Bai et al., 2019). Previous data indicated that the accumulation of heavy metals in soil also changes according to both pH and the nature of the heavy metal (Zhang et al., 2018). Soil pH and organic matter were also widely considered as factors with the strongest spatial correlation with heavy metal availability (Zhao et al., 2010). Therefore, to avoid the accumulation of heavy metals in saline-sodic paddy fields, it is necessary to gradually increase the content of soil organic matter when salinity and soil pH decrease in saline-sodic paddy fields. Our long-term data also helps to eliminate people's concerns that the application of phosphogypsum or sheep manure to reclaim saline-sodic soils into agricultural use could cause heavy metal pollution.

5. Conclusions

The present study described the changes in sodicity, salinity, mineral status, pH, and heavy metal concentrations in response to yearly tillage and single applications of two types of amendments to saline/sodic paddy soils cultivated with rice in Northeast China. Tillage and rice cultivation alone were not enough to reduce total soil salinity significantly at all soil depths examined. However, the combination of tillage and rice cultivation with either phosphogypsum or sheep manure, applied before each annual rice cultivation, significantly decreased salinization and alkalization parameters, increased soil fertility, and decreased soil pH without increasing the concentrations of five heavy metals beyond environmental quality standards in China. Soil pH and $EC_{1:5}$ proved to be two relatively simple and practical parameters to indicate changes in soil main salt ions and other salinization and alkalization parameters. High soil pH and sodicity significantly inhibited soil fertility improvement in sodic/saline paddy fields. Therefore, tillage and rice cultivation combined with phosphogypsum or farmyard sheep manure are deemed suitable and affordable measures to mitigate soil salinity and sodicity. These measures are available to farmers and can be used at a large scale to reclaim and improve saline-sodic soils destined for the cultivation of rice and other salt-sensitive crops.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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