



Relationship between spatio-temporal evolution of soil pH and geological environment/surface cover in the eastern Nenjiang River Basin of Northeast China during the past 30 years

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ABSTRACT

To illuminate the spatio-temporal variation characteristics and geochemical driving mechanism of soil pH in the Nenjiang River Basin, the National Multi-objective Regional Geochemical Survey data of topsoil, the Second National Soil Survey data and Normalized Difference Vegetation Index (NDVI) were analyzed. The areas of neutral and alkaline soil decreased by 21100 km² and 30500 km², respectively, while that of strongly alkaline, extremely alkaline, and strongly acidic soil increased by 19600 km², 18200 km², and 15500 km², respectively, during the past 30 years. NDVI decreased with the increase of soil pH when soil pH > 8.0, and it was reversed when soil pH < 5.0. There were significant differences in soil pH with various surface cover types, which showed an ascending order: Arbor < reed < maize < rice < high and medium-covered meadow < low-covered meadow < *Puccinellia*. The weathering products of minerals rich in K₂O, Na₂O, CaO, and MgO entered into the low plain and were enriched in different parts by water transportation and lake deposition, while Fe and Al remained in the low hilly areas, which was the geochemical driving mechanism. The results of this study will provide scientific basis for making scientific and rational decisions on soil acidification and salinization.

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1. Introduction

As an important basic property of soil, pH is the comprehensive result of various soil-forming factors in the process of soil formation and evolution (Xiong Y and Li QK, 1987). Soil acidification and alkalization restrict the bioavailability of chemical elements, such as selenium (Se), and the activity of soil microorganisms (Zhao BQ et al., 2010; Zhao J et al., 2011; Curtin D and Trolove S, 2013; Hou EQ et al., 2015; Yang Z et al., 2020; Liu XJ et al., 2022; Dong QY et al., 2022), affecting plant growth and seriously threatening productivity and ecological functions of normal soil (Hoegh-

Guldborg O et al., 2007; Pan YM et al., 2018; Zhang ZQ et al., 2018). Soil pH is mainly controlled by soil-forming factors in the natural state (Guo JH et al., 2010; Li PF et al., 2019), while human intervention will change the variation degree and rate of soil pH (Guo ZX et al., 2011; Minasny B et al., 2016; Li QQ et al., 2020; Wu YH et al., 2022). According to the statistics, there are distinguished differences in the variation degree of soil pH in different countries, eg. about 19% of farmland in Britain showed a decrease in soil pH from 1982 to 1988 (Goulding KWT and Blake L, 1998), and soil pH in South Korea increased by 0.3 units from 2000 to 2012 (Minasny B et al., 2016). The average topsoil pH of farmland in China decreased by 0.5 units, with a range of 0.13–0.80 units from 1980 to 2000 (Guo JH et al., 2010). The calcareous soil pH in northern China also showed a significant downward trend (Yang YH et al., 2012).

Songnen Plain, as an important commodity grain base in China, has displayed high soil alkalinity in Northeast China

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(Liu ZM et al., 2004; Wang Q et al., 2017). Meanwhile, the area of severely salinized land is still expanding at a rate of 99 km²/a in recent decades, and the degree of salinization is constantly increasing, due to the influence of natural and human factors (Li XJ, 2000; Sun GY and Wang HX, 2016). There is also a trend of soil acidification in the Liaohe Plain (Cheng HX et al., 2012; Bai SB et al. 2016; Wang Y et al., 2017). It is obvious that alkalization/acidification has become one of the important ecological problems of black soil in Northeast China. In this paper, the authors analyzed the spatio-temporal variation characteristics of soil pH in the Nenjiang River Basin since the 1980s and determined the geochemical controlling factors in soil pH variation based on the soil geochemical data from the Second National Soil Survey (1979–1986) (Second Soil Survey) and the National Multi-objective Regional Geochemical Survey (2006–2015) (Multi-objective Survey). The result of this study could provide an important theoretical basis for the conservation and sustainable utilization of black soil resources.

2. Methods

2.1. Geological setting of the study area

The study area is located in the east of Nenjiang River Basin, across Heilongjiang Province, Jilin Province, and Inner Mongolia Autonomous Region (121°39'–127°45' E and 44°01'–49°58' N), covering an area of 133000 km² (Fig. 1), and belongs to the Mesozoic depression area. The widely developed river alluvium and lake sediments of the Upper Pleistocene form a large valley alluvial plain. Topographically, the northern, western, and southern parts of the area are higher, and the eastern part is connected with the Songhua River Plain, forming the Songnen Plain. Geomorphologically, the area can be divided into low hills in the Daxing'an Mountains, platform, piedmont sloping plain, southern alluvial-lacustrine plain, interdune depression, floodplain, and low terrace from the north to the south, with an altitude of 140–520 m. The lower plain area in the south is low and flat, with poor drainage and a large number of closed



Fig. 1. Geographical location of the study area.

and semi-closed depressions. Composed of low mountains and hills, the northern part is higher and can be ascribed to the material denudation area. The geological and geomorphic conditions are favorable for the migration and enrichment of elements. The low plain is the enriched area of salt migration, while the low hilly turns to be the salt dilution area. Meadow soil and dark brown soil are the main soil types. Chernozem soil, black soil, aeolian sand soil, and marsh soil are also widely distributed. Most of the land use is cultivated land, accounting for 53.68%, followed by grassland, woodland, marsh, and saline-alkali land, accounting for 7.78%–12.06%. Other land use types are scattered in the study area, accounting for less than 5% (Fig. 2).

2.2. Sampling and analysis

The topsoil (0–20 cm) samples were collected at a density of one sample per km². Each soil sample was randomly composed of three sub-samples. The soil samples were collected into clean cloth bags. After being put into clean cloth bags, soil samples with high humidity were then isolated with plastic sample bags to prevent contamination between samples. The dry weight of the collected soil samples was more than 1 kg. Soil samples were dried without pollution and passed through a 20-mesh nylon sieve. After that, geochemical elements and pH were analyzed. A total of 32853 data were obtained. It includes 54 geochemical indexes such as pH, K₂O, Na₂O, CaO, MgO, Al₂O₃, Fe₂O₃, etc. Sampling and analysis are carried out according to the Specification for Multi-objective Regional Geochemical Investigation (1 : 250000) (Ministry of Land and Resources, PRC, 2015).

2.3. Data Sources

Other data sources used are listed as follows:

(i) Data of the Second Soil Survey: From the 2 km × 2 km

raster dataset of topsoil properties provided by Nanjing Institute of Soil Sciences, CAS. Using the co-kriging interpolation method, the dataset was constructed based on 2647 records of physical and chemical properties of soil profiles in Volume 1–6 of Soil Species Annals of China and the soil type map of China. The attribute field of the dataset contains pH, which could directly reflect the soil pH status in the 1980s and be used for comparative study.

(ii) Vector data of soil type map of Northeast China: From Nanjing Institute of Soil Science, CAS. The data is based on the 1 : 1000000 soil map of the People's Republic of China, organized by the National Soil Survey Office. The property field of the data contains the soil order, group, and subgroup.

(iii) Vector data of land use of Northeast China in 2010 (1 : 100000): From Institute of Geographic Sciences and Natural Resources Research, CAS. The data was jointly completed by the Institute of Geographic Sciences and Natural Resources Research, CAS, Institute of Remote Sensing Applications, CAS, Northeast Institute of Geography and Agroecology, CAS, and other institutes. The reconstruction of land use/cover data was based on Landsat TM/ETM remote sensing image.

(iv) Normalized Difference Vegetation Index (NDVI) data of Northeast China in 2010: From Resource and Environment Science and Data Center, CAS (<https://www.resdc.cn/>). It could accurately reflect the spatial and temporal distribution and variation of surface cover in different regions of China.

2.4. Data processing

The two-period data of soil pH at 32853 sites in the eastern Nenjiang River Basin were used as the original data. The raster data of the Second Soil Survey were extracted from the current sampling site. The inverse distance weighted interpolation method is used for spatial interpolation analysis in light of the geostatistical analysis module of ArcGIS10.2 (Peng M et al., 2019). Since the sampling sites in the study

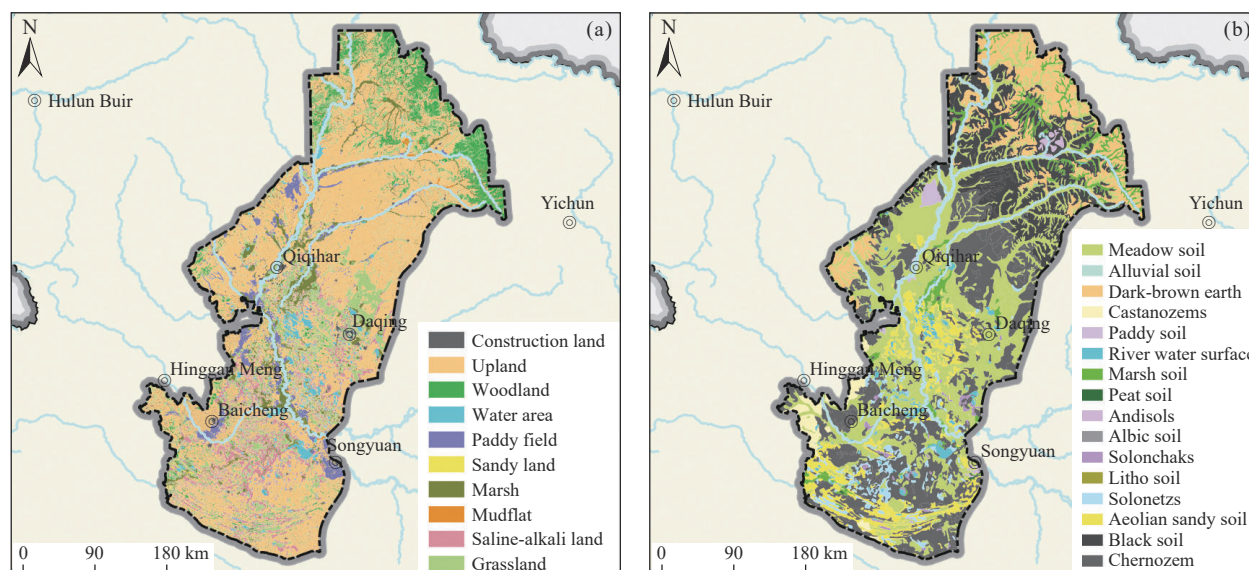


Fig. 2. Distribution map of land use types (a) and soil types (b) in the eastern Nenjiang River Basin.

area are evenly distributed, the authors use a fixed search radius of 5000 m, a search direction of 360°, the default power parameter of 2, and a neighbor smoothing coefficient of 0.2. The statistical analysis of soil pH data is completed using Excel 2019.

3. Results and discussion

3.1. Statistical parameters and variation of soil pH

3.1.1. Statistical distribution

The soil pH of the Second Soil Survey was obtained by extracting the raster data set using ArcGIS, and the extracting sites were in accordance with that of the Multi-objective Survey. The results of the normality test of all the soil pH in different soil types showed that the inspection P values were less than 0.05 (Table 1), and none of them fit the normal distribution. As a result, the authors chose the nonparametric test method to verify whether these two periods of soil pH datasets with different land uses and soil types showed a statistically significant difference.

The statistical results of soil pH in the two periods

revealed that the inspection P values of the non-parametric test were less than 0.05, indicating that there were statistically distinguished differences between them (Table 2). In the 1980s, the arithmetic means value of soil pH of the Second Soil Survey in the eastern Nenjiang River Basin was 7.48, with the minimum, maximum, and range values of 4.7, 10.4, and 5.7, respectively. While the arithmetic mean of soil pH of the Multi-objective Survey was 7.72, 0.24 units higher than that of the Second Survey. The minimum, maximum, and range values of soil pH were 10.97, 4.32, and 6.65, respectively. The significant increase in soil pH may imply the trend of acidic and alkaline polarization in the study area. As for the variation coefficient of soil pH in the two periods, it showed low variation intensity, which was higher than that of the Second Soil Survey, and indicated that the fluctuation of soil pH data showed an increasing trend in nearly 30 years. The authors carried out the structural analysis with the semi-variogram of soil pH in these two periods in the Nenjiang river valley, to determine the influence degree of structural factors (natural spatial variability) and random factors (sampling and measurement, etc.) that cause variations in these statistical parameters (Zhang C and Mcgrath D, 2004;

Table 1. Normality test results of soil pH in different land uses and soil types in the Nenjiang River Basin.

Classification	Sample classification	Sample number	Multi-objective Survey	Second Survey	Test method
			P	P	
Land use types	Total samples	32853	0	0	Kolmogorov-Smirnova
	Grassland	3915	<0.05	0	Kolmogorov-Smirnova
	Upland	16815	0	0	Kolmogorov-Smirnova
	Construction land	1135	<0.05	<0.05	Shapiro-Wilk
	Woodland	3371	0	0	Kolmogorov-Smirnova
	Sandy land	52	<0.05	<0.05	Shapiro-Wilk
	Paddy field	1146	<0.05	<0.05	Shapiro-Wilk
	Mudflat	319	<0.05	<0.05	Shapiro-Wilk
	Saline-alkali land	2529	<0.05	<0.05	Kolmogorov-Smirnova
	Marsh	2795	<0.05	0	Kolmogorov-Smirnova
Soil type	Dark-brown earth	3282	<0.05	0	Kolmogorov-Smirnova
	Meadow soil	11333	0	0	Kolmogorov-Smirnova
	Aeolian sandy soil	2418	<0.05	<0.05	Kolmogorov-Smirnova
	Chernozem	7273	<0.05	0	Kolmogorov-Smirnova
	Black soil	4638	<0.05	0	Kolmogorov-Smirnova
	Andisols	87	<0.05	<0.05	Shapiro-Wilk
	Solonchaks	771	<0.05	<0.05	Shapiro-Wilk
	Castanozems	535	<0.05	<0.05	Shapiro-Wilk
	Paddy soil	233	<0.05	<0.05	Shapiro-Wilk
	Alluvial soil	136	<0.05	<0.05	Shapiro-Wilk
Solonchaks	180	<0.05	<0.05	Shapiro-Wilk	
Marsh soil	1749	<0.05	<0.05	Shapiro-Wilk	

Table 2. Descriptive statistics of soil pH in the Nenjiang River Basin.

Data Source	Samplenumber	Minimum value	Maximum value	Mean ±Standard deviation	Variation Coefficient	Median	Test Statistics	
							Z	P
Second Soil Survey	32853	4.7	10.4	7.48±1.07	0.14	7.6	-33.096	<0.05
Multi-objective Survey	32853	4.32	10.97	7.72±1.67	0.22	8.25		

Notes: Mean value is the iteratively arithmetic mean after the outliers are eliminated. Second Soil Survey refers to Second National Soil Survey. Multi-objective Survey refers to the National Multi-objective Regional Geochemical Survey.

Xia XQ et al., 2017). The results showed that the nugget coefficient of soil pH in the Second Soil Survey was 36.74%, which proved the moderate intensity of spatial correlation (Table 3). It could be concluded that both structural and random factors influenced the soil pH in the Second Soil Survey, and structural factors were more effective. The nugget coefficient of soil pH in the Multi-objective Survey was less than 25%, which indicated that the spatial correlation was strong and structural factors were dominant. It could be demonstrated that the variations of soil pH statistical parameters during the two periods were chiefly influenced by structural factors.

3.1.2. pH variation in different soil types

Statistical results of soil pH in different soil types in the two periods (Table 4) showed that *P* values of non-parametric tests were all less than 0.05 (except paddy soil). It demonstrated that there were significant differences between the data in the two periods (except paddy soil), which was apparently verified by the box diagram of soil pH (Fig. 3). In the Second Soil Survey, the pH of Solonetz, Solonchaks, and Chernozem were relatively high, with a value of 8.77, 8.62 and 8.25, respectively, while that of dark brown earth, black soil, alluvial soil, and andisols were relatively low (all below 6.8). That of other soil types ranged from 7.0 to 7.9. After 30 years, the pH of andisols, dark brown earth, marsh soil and black soil decreased by 0.48–1.21 units in the Multi-objective Survey. Andisols decreased by 1.21 units (from 6.72 to 5.51), taking on a trend from a neutral level to an acidic level

significantly. Soil pH of meadow soil, chernozem, aeolian sandy soil, solonetz, castanozems, solonchaks, and alluvial soil increased by 0.42–1.67 units. The pH of meadow soil and chernozem widely distributed in the study area were 8.61 and 8.67, respectively. They increased by 0.55 and 0.42 units, 7.2% and 5.1%, respectively, compared with that in the 1980s. While pH of solonetz, solonchaks, and aeolian sandy soil increased significantly from 8.77, 8.62, and 7.51 to 9.62, 9.62, and 8.87, respectively, increasing by 0.85, 1, and 1.36 units, with the range of 9.7%–18.1%. It indicated that the pH of solonetz and solonchaks had changed from a strongly alkaline level to an extremely alkaline level, while aeolian sandy soil from an alkaline level to a strongly alkaline level. It can thus be recognized that the soil in the eastern Nenjiang River Basin was developing toward the polarization of acidification and alkalization.

3.1.3. Variations of soil pH in different land uses

Statistical results of soil pH in different land uses (Table 5) revealed that *P* values of nonparametric tests were less than 0.05, except for mudflat. Data in the two periods showed significant differences, which were validated by the soil pH box diagram in different land uses (Fig. 4). In the 1980s, the soil pH of sandy land, saline-alkali land, and grassland were 8.44, 8.38, and 7.76, respectively, while that of woodland was 6.80 and neutral level. That of the other types ranged from 7.2 to 7.76, mainly neutral and weakly alkaline levels. Soil pH of all land use types showed an increasing trend in varying

Table 3. Optimal semi-variation model and corresponding parameters of topsoil pH in the Nenjiang River Basin.

Data Source	Model	Nugget C_0	Sill C_0+C	Nugget coefficient $C_0/(C_0+C)$	Range/km	Residual SS	Goodness of fit test R^2
Second Soil Survey	Exponential	0.367	0.999	36.74%	300.9	0.0106	0.963
Multi-objective Survey	Gaussian	0.381	1.952	19.52%	264.5	0.0319	0.976

Notes: Second Soil Survey refers to Second National Soil Survey. Multi-objective Survey refers to the National Multi-objective Regional Geochemical Survey.

Table 4. Statistics analysis results of soil pH in different soil types in the eastern Nenjiang River Basin.

Soil type	Sample number	Second Soil Survey				Multi-objective Survey				Nonparametric Test Statistics	
		Minimum value	Maximum value	Mean± Standard deviation	Median	Minimum value	Maximum value	Mean± Standard deviation	Median	Z	P
Meadow soil	11333	5.3	9.6	7.66±1.16	8.2	4.37	10.97	8.21±1.55	8.57	-49.25	0
Chernozem	7273	5.3	10.4	8.25±0.48	8.3	5.38	10.7	8.67±0.80	8.61	-40.69	0
Black soil	4638	5.3	8.8	6.49±0.57	6.6	4.34	9.73	6.01±0.64	5.93	-38.38	0
Dark-brown earth	3282	4.7	8.8	6.45±0.41	6.5	4.32	8.85	5.57±0.57	5.48	-46.47	0
Aeolian sandy soil	2418	5.3	9.6	7.51±0.82	7.6	5.43	10.85	8.87±0.94	8.87	-37.97	0
Marsh soil	1749	5.3	9	7.02±0.70	6.8	4.34	10.74	6.43±1.66	5.62	-12.97	<0.05
Solonetz	771	5.5	10.3	8.77±0.68	8.7	6.73	10.61	9.62±0.55	9.75	-20.68	<0.05
Castanozems	535	5.3	10.3	7.83±0.47	7.8	6.37	10.42	8.56±0.44	8.45	-18.66	<0.05
Paddy soil	233	5.3	9	7.39±0.95	7.9	4.53	9.98	7.38±1.07	7.56	-1.50	0.13
Solonchaks	180	5.5	10.4	8.62±0.66	9	7.83	10.89	9.62±0.59	9.73	-10.36	<0.05
Alluvial soil	136	5.3	8.3	6.57±0.44	6.5	5.41	10.39	7.16±1.22	7.10	-5.52	<0.05
Andisol	87	6.5	7.4	6.72±0.23	6.7	4.9	6.49	5.51±0.30	5.46	-8.10	<0.05

Notes: Mean value is the iteratively arithmetic mean after the outliers are eliminated. Second Soil Survey refers to the Second National Soil Survey. Multi-objective Survey refers to the National Multi-objective Regional Geochemical Survey.

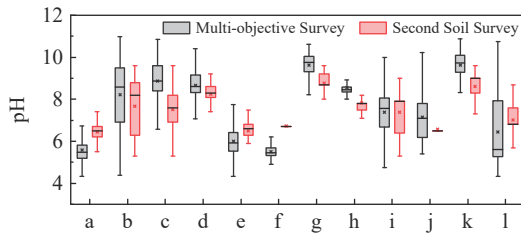


Fig. 3. Soil pH box diagram in different soil types. a–dark-brown soil; b–meadow soil; c–aeolian sandy soil; d–chernozem; e–black soil; f–andisols; g–solonetz; h–castanozem; i–paddy soil; j–alluvial soil; k–solonchaks; l–marsh soil.

degrees except that of woodland after 30 years. The pH of saline-alkali land increased the most, from 8.38 to 9.73, by 1.35 units and 16.1%. It was noted that saline-alkali land had changed from an alkaline level to an extremely alkaline level, which had toxic effects on most crops (Revision Committee of Dictionary of Water Conservancy, Hohai University, 2015). It also demonstrated that saline-alkali soil had experienced enrichment of other carbonate minerals (such as sodium carbonate) that are more soluble than calcium carbonate. The pH of sandy land, grassland, and paddy field increased by 8.53%, 7.47%, and 7.22%, respectively. That of upland, marsh, and construction land increased by 1.61%–3.26%. That of woodland decreased from 6.80 to 6.19, by 0.61 units and 8.97%. It indicated that the soil pH of the woodland had varied from a neutral level to an acidic level. This may be caused by a large number of vegetation litters covering the surface of woodland every year, providing the accumulation of soil organic carbon which will lead to soil acidification. The formation of a water-soluble complex between organic matter and non-acidic cations such as Ca^{2+} and Mg^{2+} accelerates the leaching of these cations in the soil. Meanwhile, H^+ could be dissociated from acidic groups with a large amount of organic matter. In addition, the metabolic capacity of the microbial community in the forest soil is greater with stronger respiration that could also promote soil acidification (Ji G et al., 2015).

3.2. Relationships between the spatial distribution of soil pH and surface cover

3.2.1. Relationships between soil pH and Normalized Difference Vegetation Index (NDVI)

The overall spatial distribution of soil pH in the two periods in the eastern Nenjiang River Basin was almost similar (Figs. 5a, b). The areas with high soil pH were basically distributed in the south alluvial-fluvial plain, which was low and had poor drainage and a large number of closed and semi-closed depressions. The NDVI in strongly alkaline and extremely alkaline areas was apparently low (Fig. 5c). It demonstrated that soil alkalization could significantly limit soil productivity. The areas with low soil pH were mainly distributed in the hilly areas with high altitudes, which was in the north of the study area. The NDVI in the Bei'an area, where strongly acidic soil was widely distributed, was also relatively low. It revealed that soil acidification could also have a definite impact on vegetation growth. The spatio-temporal distribution of soil pH reflected that alkaline salt from rocks or parent materials in the Nenjiang River Basin was transported, deposited, and enriched in the southern low-lying area with poor drainage. Moreover, the strong leaching of salt in the regions along with the water system resulted in the migration and depletion of salt ions from mineral weathering.

In the past 30 years, soil pH in the eastern Nenjiang River Basin showed a variation pattern, decreasing in the north and rising in the south (Fig. 6). Soil pH in the Nehe-Bei'an area, the northern Nenjiang River Basin, showed a decreasing trend, with an average of more than 1 unit, and was apparently characterized by soil acidification trend. In the southern Da'an-Tongyu region, the soil pH increased by more than 1 unit on average and was featured a strong alkalization trend. In the central Lindian-Yi'an region, the soil pH variation was not obvious, ranging from -0.25 to 0.25 .

There were considerable differences in the soil pH levels of the two periods (Fig. 7). According to the results of the

Table 5. Statistics analysis results of soil pH in different land use types in the eastern Nenjiang River Basin.

Land use type	Samples	Second Soil Survey				Multi-objective Survey				Nonparametric Test Statistics	
		Minimum value	Maximum value	Mean± Standard deviation	Median	Minimum value	Maximum value	Mean± Standard deviation	Median	Z	P
Grassland	3915	5.3	10.3	7.76±1.08	8.2	4.42	10.89	8.34±1.69	8.9	-27.60	<0.05
Upland	16815	4.7	10.4	7.46±1.03	7.6	4.34	10.87	7.58±1.46	8.05	-12.56	<0.05
Construction land	1135	5.3	9.6	7.66±1.00	8.1	4.72	10.31	7.91±1.29	8.35	-8.78	<0.05
Woodland	3371	4.7	9.6	6.80±0.82	6.5	4.32	10.6	6.19±1.51	5.52	-29.81	<0.05
Sandy land	52	7.2	9	8.44±0.34	8.5	7.16	10.23	9.16±0.65	9.335	-5.37	<0.05
Paddy field	1146	5.3	9.6	7.34±1.13	7.9	4.71	10.17	7.87±1.07	8.17	-13.50	<0.05
Mudflat	319	5.3	9.1	6.85±0.94	6.5	4.51	10.32	6.65±1.33	6.39	-1.71	0.086
Saline-alkali land	2529	5.3	10.3	8.38±0.69	8.6	6.76	10.97	9.73±0.52	9.85	-42.88	0
Marsh	2795	4.7	10.3	7.20±1.08	6.8	4.34	10.74	7.43±1.78	7.53	-7.91	<0.05

Note: Mean value is the iteratively arithmetic mean after the outliers eliminated.

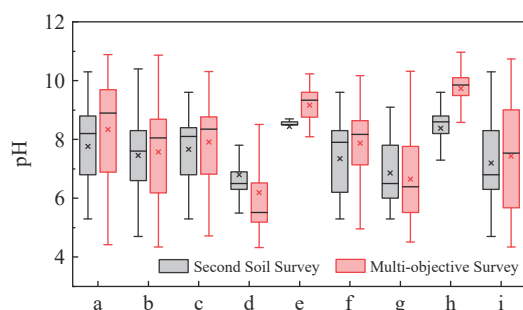


Fig. 4. Soil pH box diagram in different land use types. a–grassland; b–upland; c–construction land; d–woodland; e–sandy land; f–paddy field; g–mudflat; h–saline-alkali land; i–marsh.

Second Soil Survey, soil pH mainly ranged from 5.5–6.5 (acidic), 7.5–8.5 (alkaline), 6.5–7.5 (neutral) and 8.5–9.5 (strongly alkaline), accounting for 20.8%, 37.84%, 24.56% and 16.41% of the investigated areas, respectively. The strongly acidic and extremely alkaline soils (pH>9.5) accounted for 0.35% and 0.03% of the investigated areas, respectively.

After 30 years, the soil pH was strongly acidic and strongly alkaline. The distribution areas of the two periods were basically the same for the acidic soil, while the areas with the other pH had significant differences. The areas of neutral and alkaline soil were drastically reduced by 64.34% and 60.37%, from 32800 km² and 5000 km² in the Second Soil Survey to 11700 km² and 20000 km² in the Multi-objective Survey, respectively. The areas of strongly alkaline, extremely alkaline, and strongly acidic soil were greatly increased by 1.89 times, 455 times, 32 times, from 21900 km², 40 km², and 500 km² in the Second Soil Survey to 41500 km², 18200 km², and 16000 km² in the Multi-objective Survey, respectively. It showed that soil pH in the study area had changed from neutral level to acidic, alkaline, and strongly alkaline levels. The area of soil alkalization in the south and soil acidification in the north was likely to expand further.

Such pH variations directly affected soil chemical and biological processes and influenced its productivity function, which was further consolidated by the correlation analysis

between soil pH and NDVI (Fig. 8). Based on the analysis data of 32853 topsoil samples in the Nenjiang River Basin, the NDVI data was divided into 33 groups according to the pH interval of 0.2 units. When soil pH > 8.0, NDVI decreased significantly with increasing soil pH. When soil pH > 5.0, NDVI decreased with decreasing soil pH. It was due to the soil acidification and alkalization, which could reduce the bioavailability of nitrogen, phosphorus, potassium, iron, zinc, copper, boron, and other nutrient elements in soil, and affect crop growth and yield (Li LQ, 1986; Condon LM et al., 2005; Chen CC et al., 2009; Zheng SJ, 2010; Jia Ka LT et al., 2011; Yu TY et al., 2014; Baquy MAA et al., 2017; Weil RR and Brady NC, 2017). It can be recognized that soil alkalization and acidification in the Nenjiang River Basin have led to the reduction of soil productivity function during the past 30 years. In addition, soil acidification could also increase the availability of heavy metals in soil (Weil RR and Brady NC, 2017), and increase the probability of heavy metals entering crops and threatening human health. Therefore, scientists and government policymakers should attach great importance to these problems.

3.2.2. Relationships between soil pH and vegetation types in various ecosystems

(i) Soil pH characteristics in different vegetation types in various ecosystems

There are diverse vegetation types, various ecosystems, and abundant crops in the Nenjiang River Basin (Fig. 9). Arbors, steppe meadows, reeds, *Astragalus adsurgens*, and other vegetation types are widely distributed. It could be noted there are significant differences between soil pH and vegetation types (Table 6). The soil pH of the forest ecosystem with vegetation of arbor in the northern study area was the lowest and exhibited weak acid levels in the general. In the central part, the soil pH of the wetland ecosystem with vegetation of reeds and rushes was 7.43 and exhibited a weakly alkaline level. The soil pH of the agricultural ecosystem with vegetation of maize, soybean, and rice was 7.58, and 7.87, respectively, and exhibited alkaline levels. The soil pH of the grassland ecosystem with vegetation of high

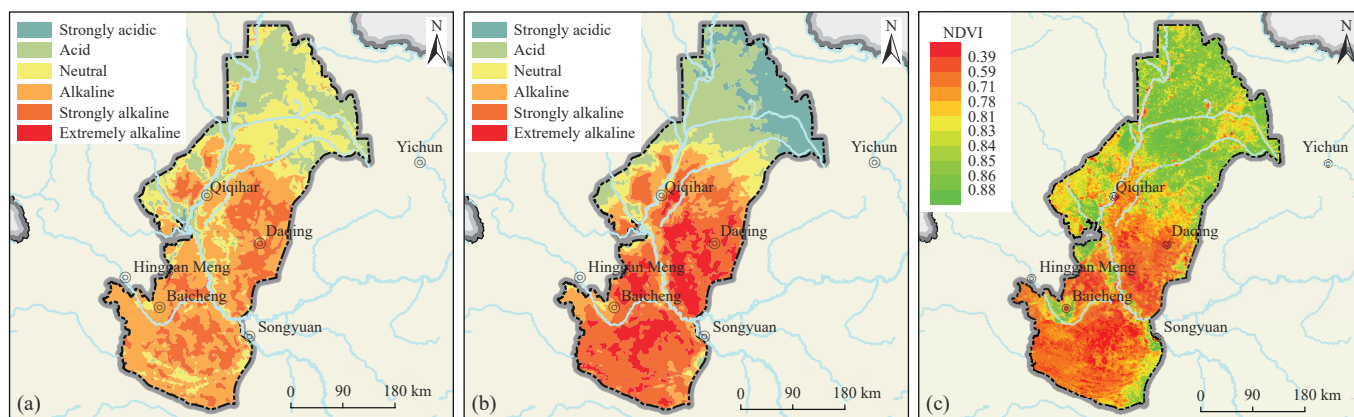


Fig. 5. Spatial distribution of soil pH and Normalized Difference Vegetation Index (NDVI). a–soil pH distribution in the 1980s; b–current soil pH distribution; c–current NDVI distribution.

and medium-covered meadow was 8.34 and exhibited an alkaline level. The soil pH of the sandy land with vegetation of low-covered meadows was 9.16 and of a strongly alkaline

level. The soil pH of the saline-alkali land with vegetation of *Puccinellia* and *Astragalus adsurgens* was 9.73 and exhibited the highest and most strongly alkaline level. To sum up, there was a significant correlation between soil pH and vegetation types in various ecosystems.

(ii) Vegetation growth in different land use types

Plants could normally grow and develop within a wide range of pH values. Different types of plants have favourable soil pH ranges. Extremely acidic and alkaline soil is enriched with H^+ and Na^+ , respectively, and lacks Ca^{2+} , which is not favourable for most plants (Weil RR and Brady NC, 2017). It is very difficult for most plants to grow in saline-alkali land with $pH > 9.5$ or extremely acidic land with $pH < 2.0$. Therefore, artificial intervention needs to be carried out to adjust the soil pH to restore land productivity and ecological function. According to the comprehensive analysis of soil pH distribution and surface cover structural characteristics (Table 7), the grassland area suitable for high and medium-covered meadows was 12012.4 km², accounting for 74.64% of the total grassland area. The area of the sandy land suitable for low-covered meadows was 162.6 km², accounting for 79.80% of the total area of sandy land. That of saline-alkali land

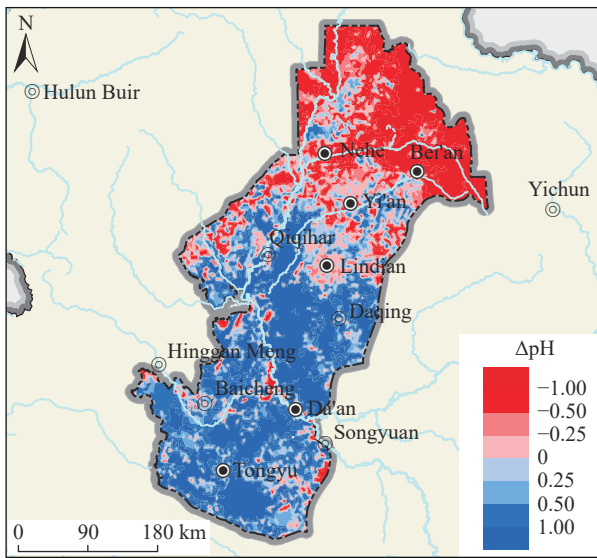


Fig. 6. Spatial variation of soil pH in the eastern Nenjiang River Basin in recent 30 years.

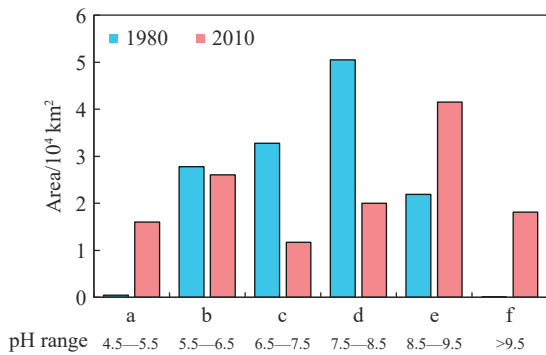


Fig. 7. Segmentation statistics of soil pH at different stages in the eastern Nenjiang River Basin. a—strongly acidic; b—acidic; c—neutral; d—alkaline; e—strongly alkaline; f—extremely alkaline.

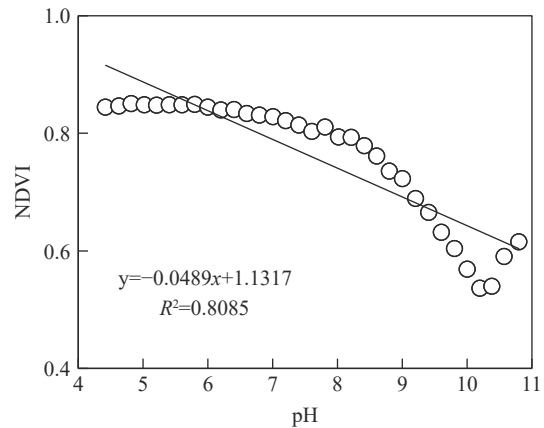


Fig. 8. Correlation analysis between Normalized Difference Vegetation Index (NDVI) and soil pH.

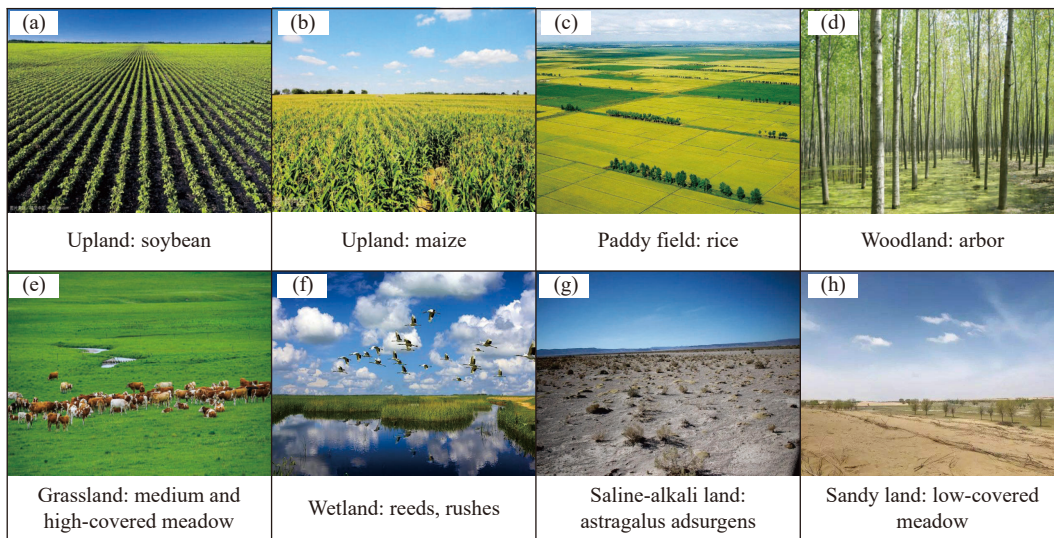


Fig. 9. Vegetation types in various ecosystems.

suitable for vegetation growth, such as *Puccinellia*, *Astragalus adsurgens*, was 4551.7 km², accounting for 43.83% of the total area of saline-alkali land. Marshes suitable for the growth of reeds and rushes covered an area of 10113 km², accounting for 87.09% of the total area of marshes. The cultivated land suitable for maize, soybean, and rice accounted for 93.69% of the total cultivated land. The distribution of cultivated land resources in the region was reasonable. All the land unsuitable for vegetation growth was caused by soil salinization. It indicates that soil salinization is a prominent ecological problem in the study area.

3.3. Influencing factors on soil pH distribution

3.3.1. Geochemical impacts

To establish a geochemical prediction model of soil elements related to soil acidification and alkalization, the data of 32853 topsoil samples in the Nenjiang River Basin were divided into 33 groups, and the average value of each element was calculated according to the pH interval of 0.2 units. The correlation analysis between soil pH and the contents of K,

Na, Ca, Mg, Fe, and Al oxides (measured by a mass fraction) was studied. The results showed that soil pH was significantly positively correlated with K₂O, Na₂O, CaO, and MgO, and negatively correlated with Al₂O₃ and Fe₂O₃ (Table 8; Fig. 10). The degree of topsoil alkalization in Nenjiang River Basin was obviously controlled by the concentration of salt ions, and different concentrations of salt ions had displayed different control degrees on soil alkalization. Meanwhile, it was recognized that there were nonlinear mutations between soil *w* (CaO) and pH. It was demonstrated that pH=8.48 and *w* (CaO)=5.05 % were the mutation points of the increasing of soil pH controlled by CaO content by the method of piecewise linear fitting. Considering the significant positive correlation between *w* (Na₂O, K₂O) and pH > 8.48, it was indicated that the increase of topsoil pH in the Nenjiang River Basin was controlled by Na₂CO₃ or K₂CO₃, which is more soluble than CaCO₃ (Weil RR and Brady NC, 2017). The acidification degree of topsoil in the Nenjiang River Basin was apparently controlled by the content of Al₂O₃ and Fe₂O₃. Al³⁺ has a strong hydrolyzing ability, which could promote the decomposition of water molecules into H⁺ and OH⁻. Moreover, Al³⁺ will combine with OH⁻, and then H⁺ will reduce soil pH. Fe³⁺ has the same properties as Al³⁺ and could also hydrolyze to produce H⁺ (Weil RR and Brady NC, 2017). The topsoil pH distribution pattern with an alkaline trend in the southern and an acidic trend in the northern Nenjiang River Basin was mainly controlled by the large occurrence of pyroclastic rocks, basic/medium-basic volcanic lavas, Hercynian and Yanshanian granites in the Daxing'an Mountains and Xiaoxing'an Mountains (the main provenance area). The geochemical compositions of these rocks were characterized by the enrichment of silica- and alkaline earth metals (Liu CM et al., 2007). When the Daxing'an Mountains and Xiaoxing'an Mountains were slowly uplifted, they were

Table 6. Statistics analysis results of soil pH in different vegetation types.

Vegetation type	Minimum value	Maximum value	Mean±Standard deviation
Arbor	4.32	10.60	6.19±1.51
Reed, rush	4.34	10.74	7.43±1.78
Maize, soybean	4.34	10.87	7.58±1.46
Rice	4.71	10.17	7.87±1.07
High and medium-covered meadow	4.42	10.89	8.34±1.69
Low-covered meadow	7.16	10.23	9.16±0.65
<i>Puccinellia</i> , <i>Astragalus adsurgens</i>	6.76	10.97	9.73±0.52

Table 7. Suitable areas of vegetation growth in different land use types.

Land use types	Vegetation types	Land areas/km ²	Suitable areas/km ²	Proportion of suitable areas/%
Grassland	High and medium-covered meadow	16093.2	12012.4	74.64
Upland	Maize, soybean, etc.	67027.1	62552.3	93.32
Woodland	Arbor	13304.5	12827.9	96.42
Sandy land	Low-covered meadow	203.7	162.6	79.80
Paddy field	Rice	4635.7	4588.2	98.98
Saline-alkali land	<i>Puccinellia</i> , <i>Astragalus adsurgens</i> , etc.	10383.7	4551.7	43.83
Marsh	Reed, rush, etc.	11612.3	10113.0	87.09

Table 8. Correlation analysis results between soil elements and soil pH in the eastern Nenjiang River Basin.

	pH	K ₂ O	Na ₂ O	CaO	MgO	Al ₂ O ₃	Fe ₂ O ₃
pH	1.000	0.980**	0.878**	0.901**	0.795**	-0.869**	-0.961**
K ₂ O	0.980**	1.000	0.896**	0.820**	0.809**	-0.799**	-0.917**
Na ₂ O	0.878**	0.896**	1.000	0.729**	0.959**	-0.762**	-0.857**
CaO	0.901**	0.820**	0.729**	1.000	0.680**	-0.965**	-0.957**
MgO	0.795**	0.809**	0.959**	0.680**	1.000	-0.699**	-0.776**
Al ₂ O ₃	-0.869**	-0.799**	-0.762**	-0.965**	-0.699**	1.000	0.968**
Fe ₂ O ₃	-0.961**	-0.917**	-0.857**	-0.957**	-0.776**	0.968**	1.000

Notes: 32853 of original topsoil data were calculated according to the arithmetic mean value of 33 data at pH intervals of 0.2 units for statistics. ** $\alpha = 0.01$ level extremely significant.

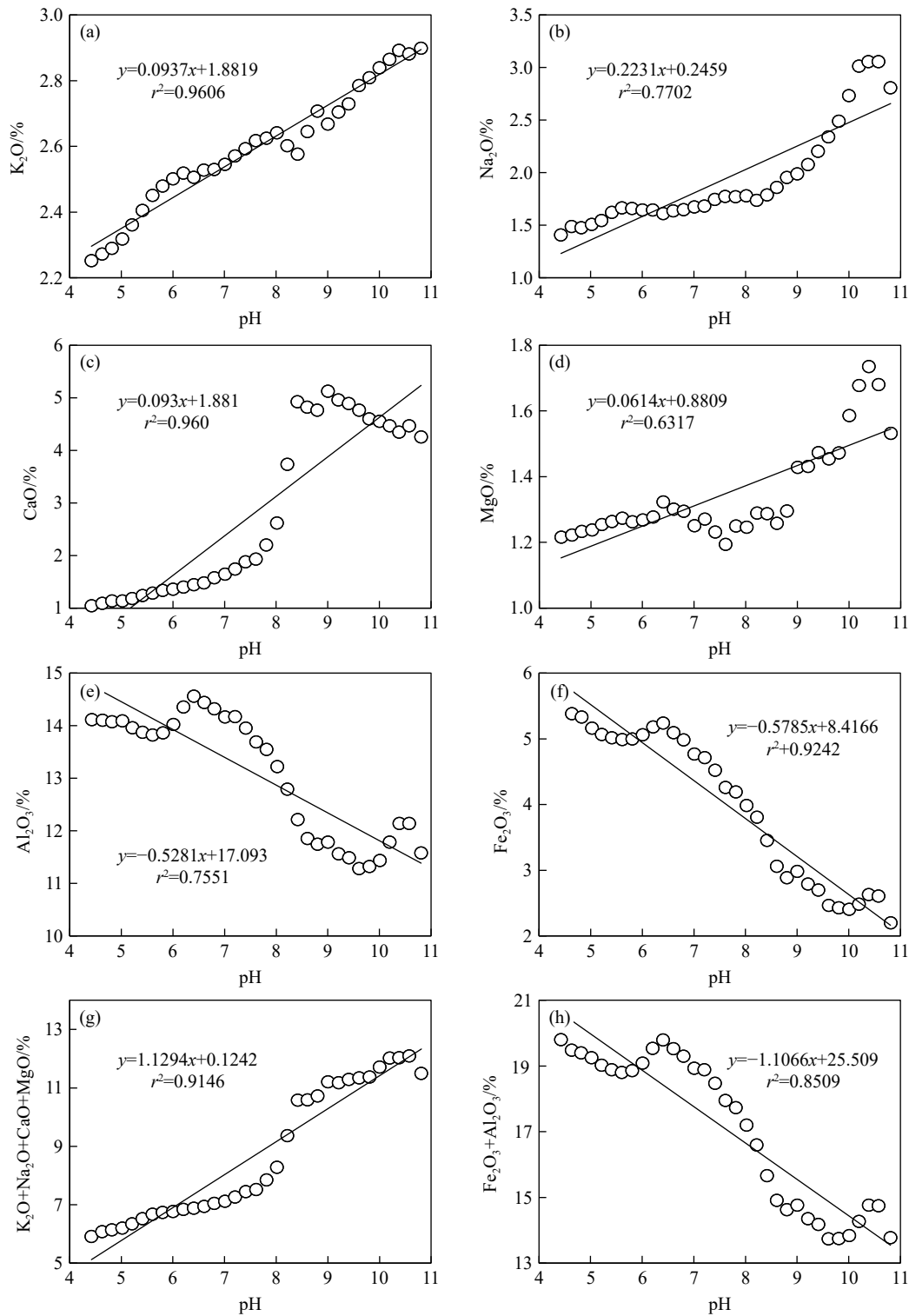


Fig. 10. Relationships models between soil elements and pH in the Nenjiang River Basin.

subjected to weathering and erosion for a long time. The weathering products of rocks and minerals which were rich in K_2O , Na_2O , CaO , and MgO became the main material flow and entered the southern low plain along the direction of surface runoff, which could be clearly demonstrated by the spatial distribution pattern of Na_2O (Fig. 10). Besides, since the low plains exhibited poor drainage, some materials were extracted from those rocks and converted into soluble ions (K^+ , Na^+ , Ca^{2+} , Mg^{2+} ions). They were carried out along the

surface water and groundwater runoff and then accumulated in the areas with poorer drainage. Continuous actions such as evaporation concentration would result in secondary salinization. Meanwhile, Fe and Al remained in the hilly regions for their relatively low activity (Fig. 11), and displayed secondary enrichment during the processes of weathering and soil formation, causing the current element distribution pattern and controlling the spatial distribution of soil pH.

3.3.2. Impact of geomorphology

Based on the classification of geomorphic unit, the average of the soil pH and the related elements were divided into 33 sets by every 20 m elevation intervals. The correlation analysis was carried out between the elevation and main controlling elements, K, Na, Ca, Mg, Fe and Al oxides content (mass fraction), in the topsoil in the eastern Nenjiang River Basin. The results showed that soil pH decreased gradually with the elevation (Fig. 12a). The soil pH in the plain with relatively low and flat terrain was the highest, with an average value of 8.53 (Fig. 12b), and exhibited strongly alkaline level. With the rising of the terrain elevation, the soil pH gradually varied from alkaline to acidic. In the low mountains and hilly areas with relatively high terrain, the soil pH was 6.49, 5.81, respectively. In the plains with relatively low elevation and higher platform, the soil pH was 6.71 and at a neutral level. It was caused by the different migration and enrichment locations of the main elements controlling soil pH. The most active K and Na migrated fastest and were enriched in the low-lying flat area with poor drainage, resulting in the highest soil pH in this area (Fig. 13). Ca was enriched in the transition zone between the high plain and platform, where the soil pH ranged from 7.0 to 8.4. Meanwhile, Fe and Al, which controlled soil acidification, were enriched in the low mountain-hilly area (above 240 m). The soil pH in this area

was 4.97–6.65. Consequently, it could be concluded that the geomorphology and the migration and enrichment of main elements were very important factors for controlling soil pH distribution. In addition, soil parent materials, hydrogeological conditions, climate conditions, soil types and land use types were also factors that could significantly affect the spatial distribution of soil pH (Zhang W et al., 2015).

3.3.3. Discussions on factors of human activities

Besides the natural factors, many researches have demonstrated that soil pH may change dramatically within one or two decades due to human activities (Goulding KWT and Blake L, 1998; Kirk GJD et al., 2009; Yang YH et al., 2012; Tian D and Niu S, 2015). On the one hand, excessive and irrational application of chemical nitrogen fertilizer has accelerated the decrease of soil pH in the pursuit of high yield (Li QQ et al., 2020). Meta-analysis showed that global soil pH decreased significantly by 0.26 units, due to the addition of N (Tian D and Niu S, 2015). Excessive application of nitrogen fertilizer could cause nitrate loss and alkali cation depletion, leading to soil acidification (Guo ZX et al., 2011; Tian D and Niu S, 2015). On the other hand, with the rapid development of industrialization and massive burning of fossil fuels, emissions of gases, such as CO₂, NO_x and SO₂, continue to increase, influencing soil pH in the form of acid rain (Aas W

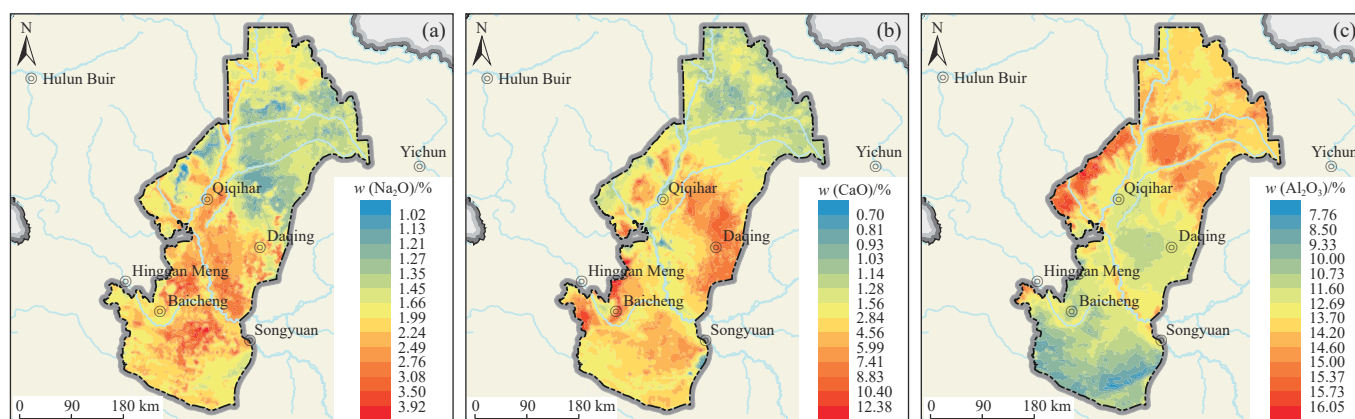


Fig. 11. Geochemical maps of Na₂O, CaO and Al₂O₃ for topsoil in Nenjiang River Basin.

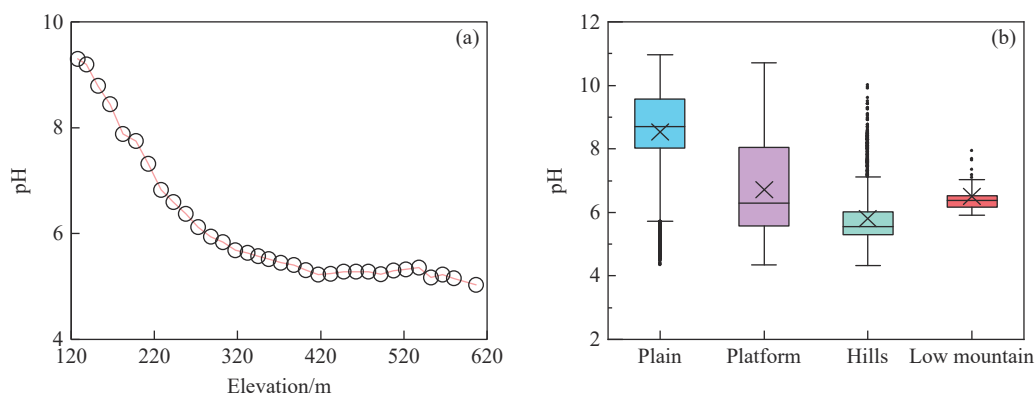


Fig. 12. Relationships between soil pH and elevation (a) and geomorphology (b). The lower and the upper edge lines in the rectangular box represent 5% and 95% of all data, respectively. The upper solid point is the outlier value. The upper and lower edges of the rectangular box represent the upper and lower quartiles, representing 75% and 25% of all data, respectively. The solid line represents the median, and the × represents the mean value.

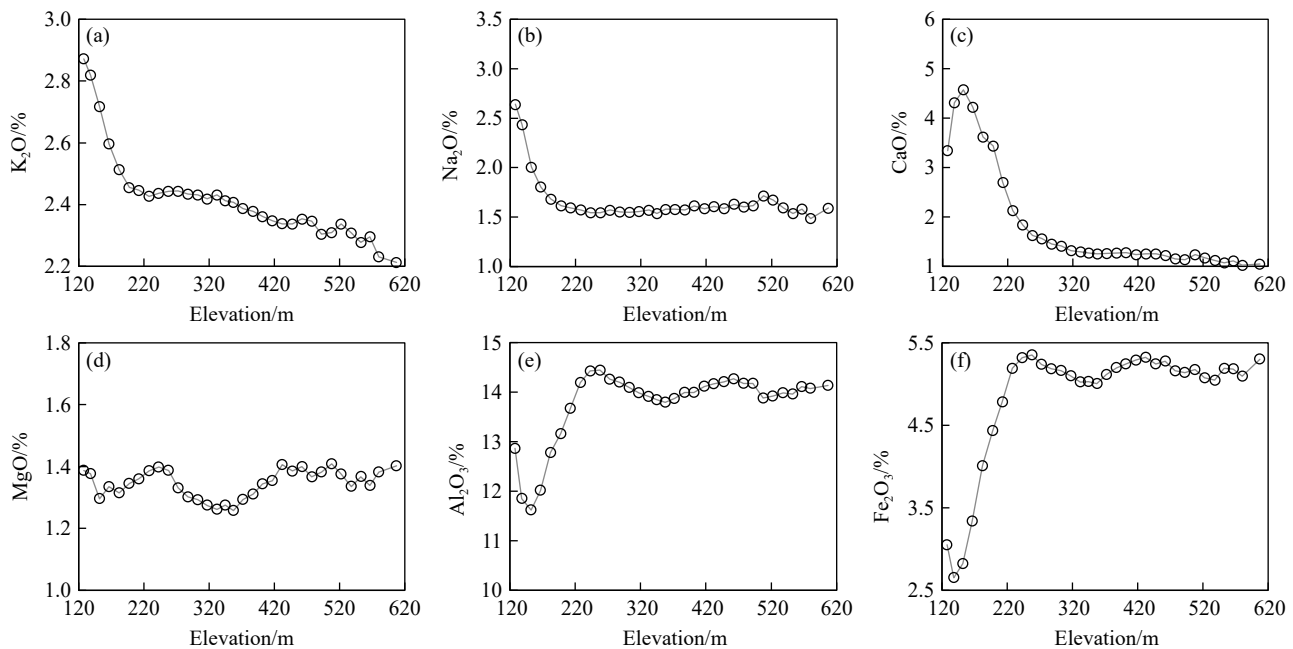


Fig. 13. Relationships between the main controlling elements oxides content (mass fraction) and the elevation in the topsoil.

et al., 2007). Additionally, a large number of exchangeable salt ions, such as K^+ , Ca^{2+} and Mg^{2+} , are carried away by crop harvest, which could result in the release of the same amount of H^+ into the soil (Tang CX et al., 2013; Li QQ et al., 2020). Subsequently, soil saline ion saturation degree and buffering performance decreases (Liao H, 2006), resulting in the acceleration of soil acidification.

4. Conclusions

Based on the soil geochemical data from the Multi-objective Survey, the current situation and spatial distribution of topsoil pH in the Nenjiang River Basin of Northeast China were studied. Combined with the Second National Soil Survey data, the evolution and driving factors of soil pH in recent decades were systematically analyzed. The main conclusions were listed as follows.

(i) The topsoil pH in Nenjiang River Basin varied from neutral, alkaline levels to acidic and strongly alkaline levels, respectively. Alkaline and saline soil had changed from strongly alkaline level to extremely alkaline level. In the northern part of the basin, the soil pH in the hilly region dominated by woodland showed a decreasing trend. The variation intensity and fluctuation of soil pH increased during the 30 years.

(ii) In the Nenjiang River Basin, the areas of neutral and alkaline soil decreased by 21100 km² and 30500 km², respectively, while that of strongly alkaline, extremely alkaline and strongly acidic soil increased by 19600 km², 18200 km² and 15500 km², respectively. There were significant differences in soil pH with various surface cover types, exhibiting an ascending sequence, arbor < reed < maize < rice < high and medium-covered meadow < low covered meadow < *Puccinellia*. The land unsuitable for vegetation growth was caused by salinization.

(iii) When soil pH > 8.0, the Normalized Difference Vegetation Index (NDVI) decreased with the increasing of soil pH, while it decreased with the decreasing of soil pH when soil pH < 5.0. Soil alkalization and acidification in the Nenjiang River Basin had resulted in the decrease of vegetation productivity function.

(iv) K^+ , Na^+ , Ca^{2+} and Mg^{2+} were the main saline ions controlling soil alkalization in the Nenjiang River Basin, while Al^{3+} and Fe^{3+} were the main elements controlling soil acidification. The degree of soil alkalization was distinctly controlled by the concentration of saline ions. When pH > 8.48, the increase of soil pH was controlled by Na_2CO_3 or K_2CO_3 which is more soluble than $CaCO_3$.

(v) The topsoil pH distribution pattern exhibited alkaline trend in the south and acidic tendency in the north and was mainly controlled by geochemical driving mechanism. As the main material flow, the weathering products of rocks and minerals rich in K_2O , Na_2O , CaO and MgO from Daxing'an Mountains and Xiaoxing'an Mountains entered the southern low plain along the direction of surface runoff and were enriched in the different parts. Fe and Al remained in the hilly regions for their relatively low activity.

CRedit authorship contribution statement

Guo-dong Liu, Ming-hui Wei, and Na-na Fang conceived and planned the experiments. Ze Yang contributed to sample preparation and the interpretation of the results. Guo-dong Liu wrote the manuscript with support from Na-na Fang and Hong-ye Xiao. Yi-he Zhang and Hong-ye Xiao supervised the project. All authors provided critical feedback and helped shape the research, analysis, and manuscript.

Declaration of competing interest

The authors declare no conflicts of interest.

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