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Contact UKCEH NORA team at noraceh@ceh.ac.uk

TITLE: Decadal shifts in soil pH and organic matter differ between land uses in contrasting regions in China RUNNING TITLE: pH and organic matter changes in Chinese soils Yiming Sun (1,2), Guanlin Guo (1)\*, Huading Shi (1), Mengjiao Liu (1), Aidan Keith (3), Hong Li (2,3) and Kevin C Jones (2)\* 1. State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy for Environmental Sciences, Beijing 100012.P.R.China 2. Lancaster Environment Centre (LEC), Lancaster University, Lancaster, LA1 4YO, UK 3. UK Centre for Ecology and Hydrology, Library Avenue, Bailrigg, Lancaster LA1 4AP, UK \*Author for correspondence: k.c.jones@lancaster.ac.uk and guogl@craes.org.cn **Highlights** Mean pH of paddy soils fell sharply over the two decades - from pH 5.81 to 5.19. Dry farmlands in the northern sampling area fell slightly - from pH 8.15 to 7.82. SOM content of dry farmland and woodland rose in north and south China. 

#### Abstract

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Soil organic matter (SOM) and pH are critical soil properties strongly linked to carbon storage, nutrient cycling and crop productivity. Land use is known to have a dominant impact on these key soil properties, but we often lack the ability to examine temporal trajectories across extensive spatial scales. Large-scale monitoring programmes provide the data to evaluate these longer-term changes, and under different climatic conditions. This study used data from Chinese soil surveys to examine changes in soil pH and SOM across different land uses (dry farmland, paddy fields, grassland, woodland, unused land), with surface soil (0-20 cm) collected in the periods 1985-90 (Survey 1; 890 samples) and 2006-10 (Survey 2; 5005 samples) from two contrasting areas. In the southern part of China the mean pH of paddy soils fell sharply over the two decades between surveys - from pH 5.81 to 5.19 (p<0.001), while dry farmlands in the northern sampling area fell slightly (from pH 8.15 to 7.82; p<0.001). The mean SOM content of dry farmland soil rose in both areas and the mean SOM of paddy fields in the southern area also rose (all p<0.001). Woodland soil pH in the south showed an increase from 4.71 to 5.29 (p<0.001) but no significant difference was measured in the woodlands of the northern area, although the trend increased. The SOM content of woodland top soils rose in the northern (p=0.003) and southern (p<0.001) study areas. The implications and potential causes of these changes over the two decade timespan between surveys are discussed and suggestions made as to how large scale soil sampling campaigns can be designed to monitor for changes and potential controlling factors.

Key words: Soil change; land use; soil surveys; woodland; paddy fields; agriculture

### 1. Introduction

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The scale of China's economic growth, the size of the country and its population, and the diversity of its climate and ecosystems mean there is great demand to understand the spatial and temporal variability in the Chinese environment. Following scientific and regulatory focus on China's air and water quality, the Government is now prioritising soil quality (State Council, 2016). Knowledge and effective management of China's basic soil resources is essential, requiring careful and systematic surveying of the terrestrial environment. Soil pH and soil organic matter (SOM) are critically important properties of soils. Understanding their variability, range and any underlying changes is fundamentally important for agriculture/food security, land use management and the environmental sciences. Soil pH is important for crop production, nutrient chemistry, soil organisms and in shaping plant community composition in natural ecosystems. SOM is critical for soil structure and workability, the ability of soils to store nutrients and water, and for the global C cycle. China's agricultural land is critical for food production and its diverse landscape is critical for the balance of natural ecosystems. China covers 7.7% of the world's total farmland (Cai and Barry, 1994) and therefore any systematic changes have global implications. Some recent and high profile studies have reported underlying rapid changes in Chinese soils. For example, Guo et al. (2011) reported significant acidification of major Chinese croplands between the 1980s and the early 2000s, while Fang et al. (2007) and Tang et al. (2018) presented evidence of the impacts of human activities on carbon sequestration in China's soils and ecosystems. In addition, soil acidification has been reported on agricultural and forest land in the UK (Blake, 1999; Blake, 2002; Goulding, 2016; Reynolds et al., 2013), North America and Europe (Reuss et al., 1987). However, there is still a shortage of systematic information from which to evaluate the spatio-temporal ranges and variations in the pH and SOM of Chinese soils across different land uses. Large-scale surveys have been undertaken in China at different times and co-ordinated by different Ministries but the datasets are not widely available or evaluated yet. Here we report on pH and SOM data obtained for two time periods (1985-90 and 2006-10) across two important and climatically different parts of China. These data sets provide the opportunity to evaluate temporal trajectories in key soil properties across land use types at an extensive spatial scale, thus critically advancing the knowledge base needed to manage China's vast soils and land resources. In this paper we therefore explore the distribution of pH and SOM values for the two surveys, and test whether changes over two decades are significant; importantly, we look at differences within the main broad land-use types to determine whether temporal changes are land-use specific and consistent across the two contrasting regions. The findings are discussed in relation to other studies for China and internationally, and consider the wider implications for China's land use management. Furthermore, we consider how future regional/national surveys of China's soil resources can be designed and coordinated in the light of international experiences, to ensure the most reliable information, capable of detecting underlying changes is obtained.

### 2. Material and methods

*2.1. Study areas* 

Two major surveys of Chinese soils have been conducted by Government Ministries. The first was 1985-90, the second was more comprehensive, with more samples taken over the period 2006-2010 (see **Table S1**). For this study, two regions were selected from these national surveys, one in the north and one in the south (see **Figure 1**). These two regions were selected because: (1) They are representative of economically developed regions in the north and south of China; (2) they each have

a comparable land mass to individual countries elsewhere in the world (e.g. Belgium = 30,700 km<sup>2</sup>, Netherlands = 40,600 km<sup>2</sup>) or even UK (= 242,500 km<sup>2</sup>) where national soil survey data are available, with each having comparable and sufficient regional coverage to show the range and characteristics of soil pH and organic matter; (3) the geographical, geological and vegetation types in the two areas provide sufficient variation in soil properties to avoid autocorrelation problems.

Area 1 (north) covers 218,000 km<sup>2</sup>. Land use types include dry farmland, paddy fields, woodland (including coniferous forest, broadleaf forest, coniferous-broadleaf forest, and shrub), grassland and unused land. Dry farmland dominates in Area 1, with wheat, maize, rice, beans and other crops being common. However, the land use in Area 1 has also undergone big changes (see **Table S2**); arable land, grassland and unused land have decreased, but woodland, garden and construction land have increased (Wu et al., 2015). Area 1 has a temperate semi-humid and semi-arid continental climate. Summers are hot and humid with high rainfall; winter is cold and dry. The most widely distributed soil types are brown earths. The main zonal soils also showed succession from the southeast (brown soils) to northwest (chestnut brown soil) (Hao et al., 2017).

Area 2 (south) covers 178,000 km<sup>2</sup> of varying terrain, with high land in the north and lower land in the south, near the coast. It has a tropical and subtropical monsoon maritime climate. Igneous rocks dominate around a third of the province. Elsewhere it has the full range from ultrabasic to acid rocks, with acidic granite a major component (Lin et al., 2006). Three main soil types occur - latosols (pH 4.5-5.5), lateritic red soils (pH 4.5-5.6) and red soils (pH 4.5-6) (Lian, 2002). Their formation is influenced by strong soil leaching, because of the sub-tropical high rainfall conditions (Lian, 2002). Major land use types include paddy fields, a range of fruit and vegetable crops (or collectively defined 'dry agricultural land'), woodlands (including coniferous forest, broadleaf forest, coniferous-broadleaf

forest, and shrub), grasslands and unused land. Paddy fields make up the largest type, accounting for 27% of the whole area (Guo et al., 2011). A huge urbanization programme and rapid development of the economy has had a significant effect in changing the composition of land use types. The composition of land use in Area 2 has changed significantly from the 1990s, with a decrease of arable land and the increase of urbanisation, industrial and mining land (Tang, 2008) (see **Table S2 and S3**).

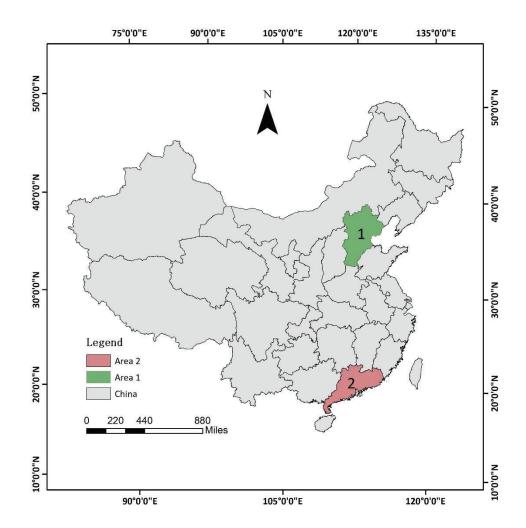


Figure 1: Soil sampling sites in north (Area 1) and south (Area 2) of China.

2.2. Soil surveys

The Chinese National Environmental Monitoring Centre (CNEMC), the Chinese Academy of Sciences (CAS), the MEP Chinese Research Academy of Environmental Sciences (CRAES) and a number of universities in China were also involved in these activities. Sampling sites were randomly selected using a grid method for the two surveys, with consideration of different environmental factors including soil types, vegetation types, land uses, soil texture etc (see Supplementary Information for further information). Topsoil (0-20 cm) was collected and stones, litter and large roots removed. Soil samples were dried at room temperature and then gently ground to pass through a 2 mm sieve. 100 g dry samples were used for chemical analysis. Soil pH was determined, depending on the salinity and OM status of the soils, as follows: a 2.5:1 ratio of water or saline solution for acid soils with 1 mol KCl/L, neutral and alkaline soils with 0.01 mol CaCl<sub>2</sub>/L); a ratio of 5:1 for saline soil; a ratio of 10:1 for litter-rich and peat soil. SOM (%) was determined by heated oxidation with K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>-H<sub>2</sub>SO<sub>4</sub> (185 °C), followed by back titration by FeSO<sub>4</sub> (see Table S1). The number of samples taken in the two surveys differed, with a more comprehensive survey conducted in 2006-2010. In summary, data was available as follows: Area 1: 1985-1990 - 500 samples, 2006-2010 - 3132 samples; Area 2: 1985-1990 – 390 samples, 2006-2010 – 1873 samples (**Table 1**).

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2.3. Data analysis

- Unpaired t-tests were used to examine differences in soil pH and SOM between surveys for whole areas and for separate land use types in these areas. The formula for the unpaired t-test is:
- 139  $t = \frac{\bar{x}_1 \bar{x}_2}{\sqrt{\frac{S_1^2}{N_1} + \frac{S_2^2}{N_2}}}$ , where  $\bar{x}_1$ ,  $s_1^2$  and N1 are the first sample mean, sample variance and sample size; x2,
- s<sub>2</sub><sup>2</sup> and N2 are the second sample mean, sample variance and sample size. R software was used for statistical analyses (R Core Team, 2016). The distribution of soil pH and SOM data for all samples

and samples from individual land use types were visualised in the ggplot2 package (Wickham, 2016) using geom\_density to produce smoothed sample densities for comparison of the surveys, and geom\_hex was used to plot relationships between soil pH and SOM within land use types.

# 3. Results

**Table 1** presents the summary of soil pH and SOM data from the surveys. **Table 2** and **3** give details of soil pH and SOM, respectively, according to land use type.

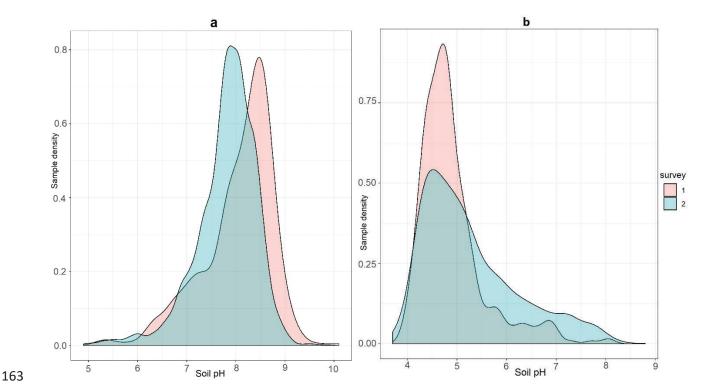
Table 1: Soil pH and organic matter in Area 1 (north) and Area 2 (south) from 1985-90 to 2006-10. Range value in brackets.

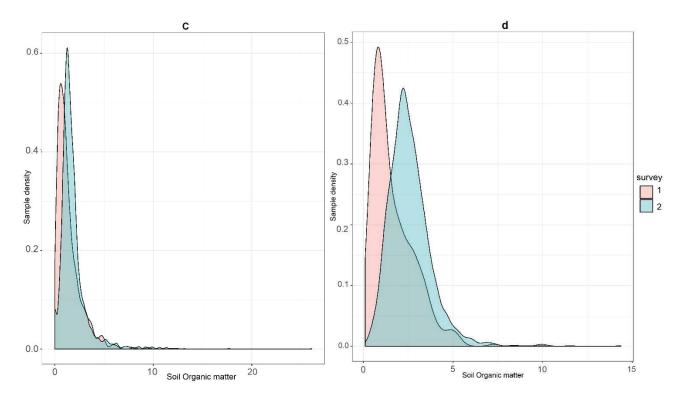
Site	Year	Sample	Soil	рН	Organic matter			
		number	Mean	Median	Mean	Median		
	1985-90	500	8.05	8.25	1.37	1.00		
Area 1			(6.7-8.9)		(0.23-3.7)			
	2006-10	3132	7.81	7.9	1.83	1.49		
			(6.7-8.6)		(0.48-4.31)			
	1985-90	390	4.90	4.8	1.65	1.23		
Area 2			(4.2-6.4)		(0.38-3.92)			
	2006-10	1873	5.26	5	2.58	2.41		
			(4.2-7.3)		(1.06-4.62)			

# 3.1. Characterization of pH and SOM distribution and variation

Mean (and median) pH values for all the soils sampled in Area 1 were 8.05 (8.25) in 1985-90 (n=500) and 7.81 (7.9) in 2006-10 (n = 3132) (i.e. an apparent decline). In Area 2 mean (and median) values for all the soil samples were 4.90 (4.8) in 1990 (n = 390) and 5.26 (5.0) in 2006 (n = 1873) (i.e. an apparent increase). However, it is important to note that the sites sampled and the distribution of samples across land uses differed between the surveys. The apparent overall differences in soil pH values between the two surveys are significant for soil pH (see **Table 2** for statistics; **Figure 2a, b**) and SOM (see **Table 2** for statistics; **Figure 2c, d**) but need to be seen as indicative only, with consideration given the shifts in land use composition.







**Figure 2:** Sample density of pH and SOM values from both surveys for the two study regions. a. soil pH in Area 1; b. soil pH in Area 2; c: SOM in Area 1; d: SOM in Area 2. Survey 1 (pink) carried out from 1985 to 1990; Survey 2 (blue) carried out from 2006 to 2010.

Emphasis can be placed on direct comparisons with those land use types that were most comprehensively sampled in both surveys. In this regard, the woodland (n = 101/515 in 1985-90/2006-10) and dry farmland soils (n = 334/2283) in Area 1 can be most confidently compared. At the level of land use type, the pH trends were different compared to each area overall, with dry farmland being significantly lower ( $t_{1,447}=9.05$ , p<0.0001) in 2006-10 (mean = 7.82) than 1985-90 (mean = 8.15). Woodland soils were not significantly different between surveys. Repeating the differences between the test of surveys, using only the subset of samples which were taken in the same locations (n = 73/27) also showed a significant reduction in soil pH from the first to the second survey for dry farmland ( $t_{1,47}=2.31$ , p = 0.025). There were not sufficient samples in the same locations to do this for the other land

use types. The grassland soils data summarised in **Table 2** also show an apparently significant decrease with time, but the number of samples available from 1990 was limited, so these grassland trends should be treated with some caution.

Table 2: Topsoil pH across different land use types in Area 1 and 2 in the 1985-90 and 2006-10 surveys. df = degrees of freedom.

	Land use	N		Estimate (mean)		T-value	95 ре	ercent	df	P-value
	type	1985-	2006-	1985-	2006-		confidence			
		90	10	90	10		interval			
	Dry	334	2283	8.15	7.82	9.05	0.26	0.40	447.37	<0.001
Area 1	farmland									
	Grassland	17	196	8.52	7.88	4.04	0.31	0.98	20.10	<0.001
	Paddy field	6	45	8.03	7.91	0.84	-0.19	0.44	10.63	0.42
	Unused	42	93	7.95	7.74	1.52	-0.07	0.47	49.03	0.14
	land									
	Woodland	101	515	7.70	7.82	-1.34	-0.29	0.06	115.34	0.18
Area 2	Dry	23	163	4.71	5.11	-2.89	-0.67	-0.12	53.81	0.005
	farmland									
	Grassland	0	3							
	Paddy field	66	1061	5.81	5.19	4.72	0.36	0.88	91.451	<0.001
	Unused	0	4							
	land									
	Woodland	301	642	4.71	5.29	-17.22	-0.65	-0.51	1251.2	<0.001

In Area 2, the woodland soils in 2006-10 (n = 642, mean = 5.29) were also higher ( $t_{1, 1251}$ = -17.22, p< 0.0001) than in 1985-90 (n = 301, mean = 4.71), while paddy field soils were markedly lower in

2006-10 (n = 1061, mean = 5.19) than in 1985-90 (5.81) ( $t_{1,\,91}$ =4.72, , p<0.0001). It is noted that these mean values are derived from a wide range of soil pH values in each survey/land use, as highlighted by **Figure 3**.

Other statistically significant differences over time are summarised in **Table 2**, but it should be noted that sample numbers were more limited in these cases.



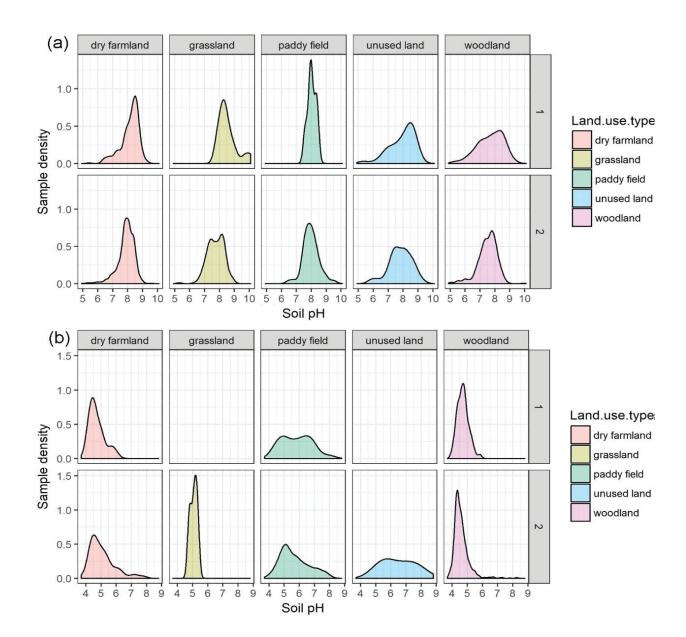


Figure 3: Sample density of soil pH values for each land use type in (a) Area 1 and (b) Area 2. 1: survey carried out from 1985 to 1990; 2: survey carried out from 2006 to 2010. There is no data

recorded in grassland and unused land during two soil surveys there is no data recorded in grassland and unused land during two soil surveys.

In general, soil pH in Area 1 is higher (range 6.7-8.9) than that in Area 2 (range 4-7). Area 1 has more saline soils with higher soil pH. The distribution of soil pH values in different land use types is shown in **Figure 3**. The most complete information (i.e. greatest number of samples) is available for paddy field soils, dry farmland and woodland soils. In Area 1 the soil pH range is similar across all land use types – for example the mean for both dry farmland and woodland was 7.82 in the 2006-2010 survey. In Area 2, although mean values in 2006-10 were similar (paddy field 5.19; woodland 5.29; dry farmland 5.11), the range of values were rather different (see **Figure 3**).

### 3.2. Land use and SOM

In Area 1 decreasing SOM followed the sequence woodland > dry farmland > paddy field (see **Table 3** and **Figure 4**). In Area 2, the sequence was less clear and showed some differences between the two surveys: in 1985-90, woodland > paddy field > dry farmland; in 2006-10, paddy field > dry farmland > woodland (see **Table 3** and **Figure 4**).

Table 3: Soil organic matter (0-20 cm) across different land use types in Areas 1 and 2 in the 1985-90 and 2006-10 surveys. df = degrees of freedom.

Site	Land use	N	Estimate	T-value	95 percent	df	P-value
	type		(mean)		confidence interval		

		1985-	2006-	1985-	2006-					
		90	10	90	10					
	Dry	334	2283	1.35	1.81	-6.69	-0.59	-0.32	561.43	<0.001
Area 1	farmland									
	Paddy field	6	45	1.22	1.74	-1.38	-1.41	0.37	7.00	0.21
	Woodland	101	515	1.39	1.89	-3.00	-0.81	-0.17	133.71	0.003
	Dry	23	163	1.23	2.59	-6.71	-1.77	-0.95	42.46	<0.001
Area 2	farmland									
	Paddy field	66	1061	1.63	2.67	-6.56	-1.35	-0.72	89.22	<0.001
	Woodland	301	642	1.68	2.55	-10.88	-1.03	-0.71	419.17	<0.001

The overall in mean SOM content increased from 1985-1990 to 2000-2006 in both Area 1 soils (mean of 1.37% (median = 1.00%) to 1.83% (1.49%), and Area 2 soils (1.65% (1.23%) to 2.58% (2.41%)). These represent large relative differences in the two decade time interval. However, as noted previously for overall differences in soil pH, the apparent overall change in SOM summarised in **Table** 1 and **Figure 2** need to be interpreted along with additional information, because the sites sampled and the distribution of samples across land uses differed between the surveys. It is therefore important to look at the land use types separately.

In Area 1, the statistically significant results were for dry farmland, woodland and grassland, with the caveat noted above about the limited number of grassland samples analysed from 1985-90. Dry farmland SOM increased from 1.35% to 1.81% (p<0.001), woodland from 1.39% to 1.89% (p=0.003) and grassland from 0.93 to 1.89% (p<0.001). In Area 2, dry farmland, paddy field and woodland SOM

all showed statistically significant (p<0.001) increases, from 1.23 to 2.59%, from 1.63 to 2.67% and from 1.68 to 2.55%, respectively (see **Table 3** and **Figure 4**). Repeating the test of differences between surveys using only the subset of samples which were taken in the same locations (n = 73/27) also showed a significant increase in SOM from 1985-90 to 2006-10 for dry farmland ( $t_{1,45} = 2.02$ , p = 0.049). As for soil pH, there were insufficient samples taken in the same locations to do this for the other land use types.

Previous studies have explored the relationship between SOM and pH for soils across China and different regions (e.g. see Dai et al. (2009)). The relation between these important two variables is complex and highly variable, because it depends on many factors — notably geology, climate, vegetation types, soil microbiology, and land use management. There were no clear relationships between SOM and pH within each land use types, neither by region or survey (see **Figure S1**).

In summary, the key results from this study are as follows:

Agricultural soils - the mean pH of paddy soils in Area 2 fell sharply (p<0.001) between 1985-90 and 2006-10 - from pH 5.81 to 5.19, while dry farmlands in the north fell slightly (8.15-7.82) but significantly (p<0.001) too. The mean SOM content of dry agricultural land rose sharply (p<0.001) in both Area 1 and Area 2. The mean SOM of the Area 2 paddy fields also rose significantly (p<0.001). Woodland soils — woodland soil pH in Area 2 showed a net increase (p<0.001) from 4.71 to 5.29; no statistically significant difference was measured in the woodlands of Area 1. The SOM content of woodland top soils, rose sharply, in the northern (p=0.003) and southern (p<0.001) study areas, respectively.

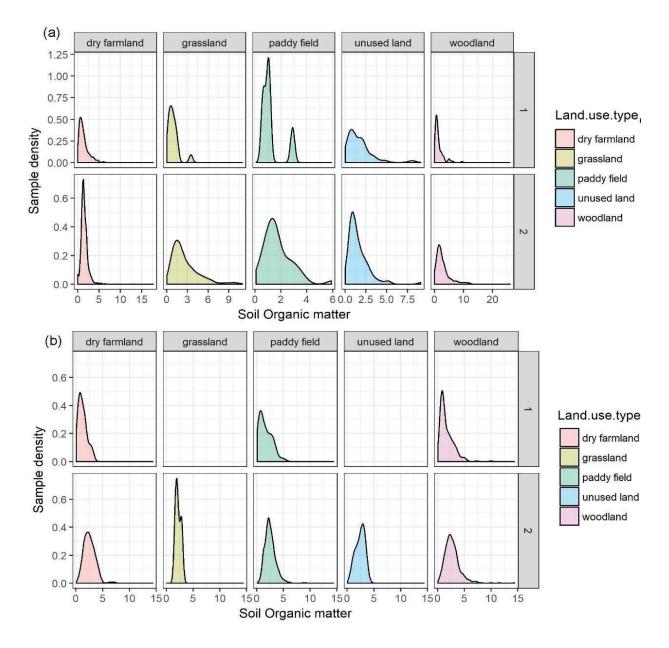


Figure 4: Sample density of soil organic matter values for each land use type in (a) Area 1 and (b) Area 2. 1: survey carried out from 1985 to 1990; 2: survey carried out from 2006 to 2010. There are no data recorded in grassland and unused land during the two soil surveys.

4. Discussions

The changes in soil pH and SOM across two contrasting regions of China represent major differences in the two decade time window of this study. They have significant implications for carbon storage, nutrient cycling and crop productivity, and need to be understood to optimise land management in different environmental contexts and avoid degradation of China's soil resources. Agricultural soils of the different regions demonstrated variable change depending on specific land use type; soil pH in dry farmlands decreased in the north and increased in the south, whereas paddy field soils decreased in both regions but to different extents. In woodland soils, there were increases in soil pH in both regions, though this was only significant in the south. Soil organic matter tended to increase in all land use types but to a greater extent in the south where soil types generally had lower pH and climate is sub-tropical. Interactions between the composition of land use and environmental conditions play a key role in determining the trajectory of soil quality at large spatial scales. Below we discuss these findings in more detail in terms of other large-scale studies of soil change, potential causes of change and the implications for future management and monitoring.

4.1. Have such rapid changes in soil pH and SOM been reported before?

Previous studies have reported underlying recent and rapid changes in soil pH in Chinese soils. For example, Guo et al. (2010) found soil pH in major Chinese crop-production areas significantly decreased from the 1980s to the 2000s. They compared cropland soil pH in the 1980s and 2000s using results from two nationwide surveys, 154 paired sites and long-term agricultural sites. They reported declines in pH under cash crop systems and under cereals, with the size of reduction influenced by soil type and soil pH range (i.e. some function of buffering capacity). For example, leached red soils (typically pH~5) in southern China declined by 0.23-0.30 pH units, while fluvo-aquic soils in the north

declined by 0.27-0.58 units. They were able to show the relative contributions of different processes to increased acidity followed the sequence: processes related to N-cycling > base cation uptake by crops > acid deposition. The widespread use of N fertilisers, they argued, accounted for most of the decline in soil pH. Guo et al. (2018) observed paddy soil pH decreased by an overall 0.6 pH units from 1980 to 2010 in Jiangxi Province. Guo et al. (2011) also reported soil pH in Guangdong Province decreased from 5.70 to 5.44 based on ca. 30-year data. The dataset reported here adds important information with a systematic assessment of soil pH and SOM in all the main land use types, highlighting temporal changes in agricultural and woodland soils. Yang et al. (2015) reported a significant decreasing trend in soil pH occurred in broadleaved forests and minor changes occurred in coniferous or mixed coniferous and broadleaved forests by using historical soil inventory data from the 1980s and a data set synthesized from literature published after 2000 in the forest ecosystem. Soil pH of tea plantation decreased from 1980s to 2010 based on 2058 soil samples from 19 provinces (Yan et al., 2020). With the change of agricultural land use, a significant pH decrease (1.2 to 0.68) was found in different soil depths based on a paired soil surveys from 1980s to 2010s in Chengdu Plain of China (Li et al., 2020). A recent survey from UK's Countryside Survey also found a recovery from acidification from 1978 to 2007 (acid to neutral) with declining SO<sub>4</sub> deposition (Thomas et al., 2020). Probably the world's most systematic assessments of long-term soil changes have been conducted in the UK, with a combination of long-term (>100 years) controlled arable and pasture grassland agricultural plot trials at Rothamsted Research station (Blake et al., 1999; Johnston et al., 1986) and the Great Britain Countryside Survey across a wide range of habitats, with several thousand samples taken in 1978-2007 (Keith et al., 2015; Reynolds et al., 2013). These provide support to our study with comparable changes across a similar time period, namely: the generally significant increase in pH

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across most UK habitats from 1978 to 2007, by up to 0.6-0.8 pH units for some; there are some differences comparing England and Scotland, highlighting broad regional differences. Soil C concentrations decreased in arable and horticulture habitats (considered most equivalent in terms of land use intensity to 'dry farmland' in this study), but increased under broadleaved/mixed woodlands (Reynolds et al., 2013). The controlled Rothamsted experiments provide the clearest controlled and quantifiable evidence of changes in pH linked to atmospheric deposition and N inputs (Hütsch et al., 1994), together with increasing soil C in response to organic matter amendments of farmland (e.g. addition of straw stubble and livestock manures) (Powlson et al., 2011a; Powlson et al., 2011b). Increases in soil pH in recent decades in some UK soils have been linked to reduced sulphur acid deposition inputs (Blake and Goulding, 2002; Emmett et al., 2010), as the UK's emissions from coal combustion, industry and domestic heating sources have declined (Emmett et al., 2010).

# 4.2. What factors could cause such changes?

Changes in topsoil pH and SOM over time are caused by a shift in the balance between inputs and losses. For pH, this is the balance between H ion inputs from soil weathering, acidifying atmospheric deposition and additions in fertilisers and plant residues. For SOM, it is the balance between the rate of accumulation of the C stock (from photosynthesis, C additions in leaf litter, stubble and residue incorporation) and the rate of decomposition/leaching/other losses. The systems studied here differ in their inputs/losses and their ability to buffer changes. Paddy field soils have very different inputs/losses to woodland systems, for example. To understand the changes seen in the systems studied here, it is therefore necessary to consider inputs/losses, and other large-scale environmental and management

factors, that have changed over recent decades to shift the balance of hydrogen ions and soil C stocks in the different Chinese ecosystems studied here.

The loss of soil C can be relatively rapid (e.g. after moving from grassland to arable, or following ploughing/disturbance), compared to the length of time and inputs required to build up soil C stocks. Active management of the C inputs added to agricultural soils can have major impacts on C stocks. A long-term study from Thomsen and Christensen (2004) reported SOM clearly and persistently increased with the annual application of straw and ryegrass. For example, when the amount of straw returned was 4 t/hm², 8 t/hm² and 12 t/hm², after 18 years, soil C increased by 12%, 21% and 30%, respectively. In addition, Thomas et al. (2020) suggested that change of topsoil organic carbons land uses was strongly affected by land use change, climatic variables (temperature and precipitation) and atmospheric deposition (e.g. SO4 deposition).

China's 'dry' agricultural lands have seen great changes in land management practices over recent decades, through the Land Reform, the drive towards agricultural self-sufficiency, greater use of fertilisers and pesticides, and often with changes in agricultural practices (Fei et al., 2010; Han et al., 2017; He et al., 2018; Zhao et al., 2018). Some of these changes have been imposed/adopted regionally. Such factors include: greater incorporation of crop residues; greater addition of livestock manures; high fertiliser loadings and use of pest control agents; mechanisation and changes in the crops grown and cropping patterns. Similarly, China's 'wet' agricultural lands (paddy fields) have also seen shifts in practice, which have resulted in dramatic gains in rice yields in China since the 1950s. These include: improved varieties of rice; changes to the incorporation of crop residues; much greater fertiliser use and changing inputs via atmospheric deposition; and changes in irrigation practice or cropping patterns.

These changes also differ between regions and land use types, which makes it difficult to predict how

the SOM inputs and C cycling have been impacted; China's agricultural extension service farm plots can potentially provide an important resource to conduct systematic studies of the factors influencing SOM (and pH) trends. Woodland systems and soils have also witnessed changes in several factors, which can influence the SOM dynamics of topsoils. These include: shifts in the proportions of primary and secondary woodland; the degree of active woodland management (e.g. clearance/felling/species mix/planting programmes); changing atmospheric loadings of CO<sub>2</sub> and nutrients, which can affect woodland productivity and C storage. Future work is needed, to systematically monitor soil changes and to assess the contribution of these drivers in controlling the pH and SOM content of China's soils resource, to help explain the trends seen here and in other studies.

Guo et al. (2010) published a comprehensive survey of soil pH in Area 2, where they were able to compare soil types from the 1980s with data from 2002-07. They focussed on trend differences between soil types. Alluvial soils from river valleys and the Pearl River Delta increased in soil pH, while red soils and paddy soils decreased. They also noted how major land use changes and agricultural practices, including urbanisation, acid mine drainage and excessive fertiliser use, had influenced the province. These important factors cannot easily be studied with our survey results, because precise information on soil types, locations and agricultural inputs are not known. However, the survey data presented here adds to the body of evidence showing rapid changes in critical soil properties in Chinese soil systems.

4.3. What are the main implications of the changes reported here?

This study shows that the basic properties of Chinese soils are changing quickly - they are dynamic, not static, systems. Rates of change in soil pH are fast and in line with some other recent published

work from China and the UK that demonstrate significant change on decadal timescales. Perhaps the greatest concern is that agricultural soil pH is declining, notably that of paddy field soils, which supply rice – the key staple foodstuff – to much of China's population. Greater acidity, particularly in the pH 4–6 range, can induce Al and Fe toxicity in crop plants, affect nutrient availability, soil fertility and crop yields. Reversing agricultural soil acidification is costly and labour/resource intensive.

4.4. How can future surveys be conducted to verify underlying trends and shed light on causes?

China is committed to soil surveys — with large resources and man-power at its disposal. This is clear from the scale and intensity of the national surveys already conducted. For example, the most recent national survey of soil pollutant quality (for selected heavy metals and organic contaminants) in the 2000s took many thousands of samples across China. Indeed, another national survey is being conducted now. However, what this study shows is that it is critical to be able to improve the quality of information obtained from such surveys, to give definitive information on the extent and scale of underlying changes in soil pH and SOM, and to yield information to explain the causes, in a way that is not possible from this study. This needs very careful design, handling and analysis, to ensure thorough statistical interpretation can be assured, capable of detecting underlying changes and their causes. This is not simply a matter of analysing large numbers of samples. Knowledge of other national soil monitoring programmes and experience operating the long-running GB Countryside Survey in the UK are valuable in guiding future soil monitoring programmes in China, and the following aspects of monitoring are considered important:

Sampling strategy: Survey designs for national sampling strategies across Europe include, amongst others, systematic or gridded sampling and stratified random sampling (Van Leeuwen, 2017). These

designs allow selection of sampling locations to be representative of the prevailing composition of land uses and soil types, and provide unbiased estimates to enable upscaling. Since land use can change over time, a survey sampling design which is not based on land use types is more flexible and temporal estimates can be reported with and without land use change. The Countryside Survey uses the ITE Land Classification (Bunce et al., 1996) which stratifies Great Britain according to major environmental gradients (e.g. climate, geology, topography). In a stratified random survey, it is important to consider sample replication within strata and power analyses may be needed for different reporting classifications and metrics, particularly if devolved or regional reporting is required.

Co-location of data: Measurements taken from the same sampling locations provide the basis for robust integrated modelling of different data. The most effective soil monitoring programmes would combine collections of biological, chemical and physical properties, along with functional measures of the soil, and the assessment of the plant community. The unit of replication for strata is a 1 km square in the survey design of the Countryside Survey but, for soil monitoring, there are five sampling plots within each 1 km square; soil, vegetation and habitat data are linked in these plots and this colocation has been exploited in a variety of integrated modelling activities (Caruso et al., 2019; Maskell et al., 2013; Norton et al., 2018; Reynolds et al., 2013). It is important to capture detailed data on the plant community in conservation areas or national parks, where indirect drivers may be causing changes in vegetation composition that are not picked up in intensively managed habitat or with a coarse land use type. Other data such as climate and landscape-level metrics are linked at the 1 km resolution.

Sample archives: The Countryside Survey has air-dried and frozen soil samples, which are catalogued and stored in dedicated archives. This means that new analyses can be undertaken on stored samples and, importantly, comparisons of methods can be made when they are updated or change. Repeated sampling: Large-scale monitoring often evaluates data as a population of samples, for example those from different land uses as done in this study. Sampling the same set of locations over time (e.g. every 5–10 years) provides the strongest statistical basis to analyse changes over time. In order to do this, it is important that precise sampling locations can be re-located in subsequent surveys; this is done using GPS coordinates, detailed written descriptions and plot and landscape photographs for CS. Statistical analyses, however, should be flexible enough to accommodate a mixture of old, repeat and new sampling locations (Scott, 2008); it is therefore very important to have a systematic schema for uniquely identifying sampling locations, so that data can be efficiently handled and combined for analyses. Recent Chinese papers discuss some of these issues in detail (Peng et al., 2016; Song et al., 2017).

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### References

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- Blake, L., Goulding, K., 2002. Effects of atmospheric deposition, soil pH and acidification on heavy metal contents in soils and vegetation of semi-natural ecosystems at Rothamsted Experimental Station, UK. Plant and soil 240(2), 235-251.
- Blake, L., Goulding, K., Mott, C., Johnston, A., 1999. Changes in soil chemistry accompanying acidification over more than 100 years under woodland and grass at Rothamsted Experimental Station, UK. European Journal of Soil Science 50(3), 401-412.

- Bunce, R.G.H., J. Barr, C., Clarke, R., Howard, D., M. J. Lane, A., 1996. ITE Merlewood Land Classification of Great Britain, 23.
- Cai, Y., Barry, S., 1994. Sustainability in Chinese agriculture: challenge and hope. Agriculture, ecosystems & environment 49(3), 279-288.
- Caruso, T., Schaefer, I., Monson, F., Keith, A.M., 2019. Oribatid mites show how climate and latitudinal gradients in organic matter can drive large-scale biodiversity patterns of soil communities. Journal of Biogeography 46(3), 611-620.
- Dai, W., Huang, Y., Wu, L., Yu, J., 2009. Relationships between soil organic matter content (SOM) and pH in topsoil of zonal soils in China. Acta Pedologica Sinica 46(5), 851-860.
- Emmett, B., Reynolds, B., Chamberlain, P., Rowe, E., Spurgeon, D., Brittain, S., Frogbrook, Z., Hughes, S., Lawlor, A., Poskitt, J., 2010. Countryside survey: soils report from 2007.
- Fang, J.Y., Guo, Z.D., Piao, S.L., Chen, A.P., 2007. Terrestrial vegetation carbon sinks in China, 1981–2000. Science in China 50(9), 1341-1350.
- Fei, L., Wang, X.K., Bing, H., Ouyang, Z.Y., Duan, X.N., Hua, Z., Hong, M., 2010. Soil carbon sequestrations by nitrogen fertilizer application, straw return and no-tillage in China's cropland. Global Change Biology 15(2), 281-305.
- Goulding K.W.T., 2016. Soil acidification and the importance of liming agricultural soils with particular reference to the United Kingdom. Soil use and management 32, 390-399
- Guo, J., Liu, X., Zhang, Y., Shen, J., Han, W., Zhang, W., Christie, P., Goulding, K., Vitousek, P., Zhang, F., 2010.
   Significant acidification in major Chinese croplands. Science, 1182570.
- Guo, X., Li, H., Yu, H., Li, W., Ye, Y., Biswas, A., 2018. Drivers of spatio-temporal changes in paddy soil pH in Jiangxi Province, China from 1980 to 2010. Scientific reports 8(1), 2702.
- Guo, Z., Wang, J., Chai, M., Chen, Z., Zhan, Z., Zheng, W., Wei, X., 2011. Spatiotemporal variation of soil pH in Guangdong Province of China in past 30 years. Chinese Journal of Applied Ecology 22(2), 425-430.
- Hütsch, B.W., Webster, C.P., Powlson, D.S., 1994. Methane oxidation in soil as affected by land use, soil pH and N
   fertilization. Soil Biology and Biochemistry 26(12), 1613-1622.
- Han, D., Wiesmeier, M., Conant, R.T., KüHnel, A., Sun, Z., Kögel-Knabner, I., Hou, R., Cong, P., Liang, R., Ouyang,
   Z., 2017. Large soil organic carbon increase due to improved agronomic management in the North China
   Plain from 1980s to 2010s. Global Change Biology 24(3).
- Hao, R., Wang, W., Hao, J., 2017. Classification of climate types and dynamic change in Beijing-Tianjin-Hebei region.
   Journal of Anhui Agricultural University 44(4), 670-676.
- He, Y.T., He, X.H., Xu, M.G., Zhang, W.J., Yang, X.Y., Huang, S.M., 2018. Long-term fertilization increases soil
   organic carbon and alters its chemical composition in three wheat-maize cropping sites across central and
   south China. Soil & Tillage Research 177, 79-87.
- Johnston, A., Goulding, K., Poulton, P., 1986. Soil acidification during more than 100 years under permanent grassland and woodland at Rothamsted. Soil Use and Management 2(1), 3-10.
- Keith, A.M., Griffiths, R.I., Henrys, P.A., Hughes, S., Lebron, I., Maskell, L.C., Ogle, S.M., Robinson, D.A., Rowe,
   E.C., Smart, S.M., 2015. Monitoring soil natural capital and ecosystem services by using large-scale survey
   data. Soil Ecosystems Services (soilecosystemsservices).
- Li, Q., Li, A., Yu, X., Dai, T., Peng, Y., Yuan, D., Zhao, B., Tao, Q., Wang, C., Li, B., 2020. Soil acidification of the soil profile across Chengdu Plain of China from the 1980s to 2010s. Science of The Total Environment 698, 134320.
- Lian, J., 2002. Classification of the natural vegetation and its distribution in Guangdong Province, Sun Yat-Sen University.

- Lin, B., Yang, S., Zhu, B., Wu, H., 2006. Geological structure and basic geotechnical characteristics in Guangdong Province. Chinese Journal of Rock Mechanics and Engineering 25(z2), 3337-3346.
- Maskell, L.C., Crowe, A., Dunbar, M.J., Emmett, B., Henrys, P., Keith, A.M., Norton, L.R., Scholefield, P., Clark,
   D.B., Simpson, I.C., 2013. Exploring the ecological constraints to multiple ecosystem service delivery and
   biodiversity. Journal of Applied Ecology 50(3), 561-571.
- Murtaza G., Usman Y., Niazi N.K., Usman M., Hussain T., 2017. Bioaccumulation of potentially toxic elements in cereal and legume crops: a review. CLEAN–Soil, Air, Water 45: 1700548.
- Norton, L.R., Smart, S.M., Maskell, L.C., Henrys, P.A., Wood, C.M., Keith, A.M., Emmett, B.A., Cosby, B.J., Thomas, A., Scholefield, P.A., 2018. Identifying effective approaches for monitoring national natural capital for policy use. Ecosystem Services 30, 98-106.
- Peng, S., Wen, D., He, N., Yu, G., Ma, A., Wang, Q., 2016. Carbon storage in China's forest ecosystems: estimation by different integrative methods. Ecology & Evolution 6(10), 3129-3145.
- Powlson, D.S., Glendining, M.J., Coleman, K., Whitmore, A.P., 2011a. Implications for Soil Properties of Removing Cereal Straw: Results from Long-Term Studies 1. Agronomy Journal 103(1), 279-287.
- Powlson, D.S., Whitmore, A.P., Goulding, K.W., 2011b. Soil carbon sequestration to mitigate climate change: a critical re-examination to identify the true and the false. European Journal of Soil Science 62(1), 42-55.
- 492 R Core Team, 2016. R: A language and environment for statistical computing. R Foundation for Statistical Computing, 493 Vienna, Austria. URL https://www.R-project.org/.
- Reynolds, B., Chamberlain, P., Poskitt, J., Woods, C., Scott, W., Rowe, E., Robinson, D., Frogbrook, Z., Keith, A., Henrys, P., 2013. Countryside Survey: National "Soil Change" 1978–2007 for Topsoils in Great Britain acidity, carbon, and total nitrogen status. Vadose Zone Journal 12(2), 1-15.
- Reuss J., Cosby B., Wright R., 1987. Chemical processes governing soil and water acidification. Nature 329: 27-32.
- 498 Scott, W.A., 2008. Countryside Survey. Statistical Report.
- Song, Y.Q., Yang, L.A., Li, B., Hu, Y.M., Liu, Y.L., 2017. Spatial prediction of soil organic matter using a hybrid geostatistical model of an extreme learning machine and ordinary kriging. Sustainability 9(5), 754.
- 501 State Council, 2016. Action plan for soil pollution prevention and control.
- Tang, Q., 2008. The study on the relationship between the urbanlization and the land use change of Guangdong province, Institute of Geographic Science and Natural Resource Research, Chinese Academy of Sciences, 90 pp.
- Tang, X., Zhao, X., Bai, Y., Tang, Z., Wang, W., Zhao, Y., Wan, H., Xie, Z., Shi, X., Wu, B., 2018. Carbon pools in
   China's terrestrial ecosystems: New estimates based on an intensive field survey. Proceedings of the National
   Academy of Sciences 115(16), 4015-4020.
- Thomas, A., Cosby, B. J., Henrys. P., Emmett, B., 2020. Patterns and trends of topsoil carbon in the UK: Complex interactions of land use change, climate and pollution. Science of The Total Environment, 729, 138330.
- Thomsen, I.K., Christensen, B.T., 2004. Yields of wheat and soil carbon and nitrogen contents following long-term incorporation of barley straw and ryegrass catch crops. Soil Use and Management 20(4), 432-438.
- Van Leeuwen, J.P., Saby, N.P.A., Jones, A., Louwagie, G., Micheli, E., Rutgers, M., Schulte, R.P.O., Spiegel, H., Toth,
   G., Creamer, R.E., 2017. Gap assessment in current soil monitoring networks across Europe for measuring
   soil functions. Environmental Research Letters 12, 124007.
- Wickham, H., 2016. ggplot2: Elegant Graphics for Data Analysis. Springer-Verlag, New York.
- Wu, J., Cao, Q., Shi, S., Huang, X., Lu, Z., 2015. Spatio-temporal variability of habitat quality in Beijing-Tianjin-Hebei Area based on land use change. Chinese Journal of Applied Ecology 26(11), 3457-3466.
- Yan, P., Wu, L., Wang, D., Fu, J., Shen, C., Li, X., Zhang, L., Fan, L., Wenyan, H., 2020. Soil acidification
  in Chinese tea plantations. Science of The Total Environment 715, 136963.

Yang, Y., Li, P., He, H., Xia, Z., Datta, A., Ma, W., Ying, Z., Liu, X., Han, W., Wilson, M.C., 2015. Long-term changes in soil pH across major forest ecosystems in China: Soil pH dynamics in forests. Geophysical Research Letters 42(3), 933-940.
 Zhao, Y., Wang, M., Hu, S., Zhang, X., Ouyang, Z., Zhang, G., Huang, B., Zhao, S., Wu, J., Xie, D., 2018. Economics-and policy-driven organic carbon input enhancement dominates soil organic carbon accumulation in Chinese croplands. Proceedings of the National Academy of Sciences115(16), 4045-4050.