(1) Soil carbon concentrations were measured using two different methods (pp. 16501-16502) that have shown to differ from each other. Most of the samples were analyzed by the potassium dichromate method and these samples were concentrated near the beginning and middle of the 20 year study period. However, soil samples collected towards the end of the 30 year study period were measured by dry combustion. Dry combustion usually yields higher carbon concentrations than wet chemical digestions, and this is documented in perhaps 20-30 different reports in the literature. For example, see Tivet et al. (2012) Soil Sci. Soc. Am. J. 76: 1048-1059; and, Islam (2006) Encyclopedia of Soil Science (R. Lal, ed.), pp. 1164-1167. Does this important methodological difference have any bearing on the rise in SOC concentration and density towards the end of the 30 year study period as illustrated in Figure 1 on page 16523 of the manuscript?

[AUTHORs]: We did a pre-experiment to compare the two different methods on the soil used in our study. Our result indicated that SOC contents determined by dry combustion and potassium dichromate titrimetric method are comparable and not significant different within the SOC level 7-12 g kg⁻¹. This meant that the SOC differences due to analytical methods are negligible for the soil in our study (Wang Pan-lei, Qin Feng-qin, Cai Pei, Meng Fan-qiao, Zhang Min, Comparison of Acidification and Soil Organic Carbon Determination for Semihumid Soils in North China, Chinese Journal of Soil Science, 2014, 45(4): 880-887) and will not lead to significant increase of SOC due to different method. However, we agreed with the reviewer that for SOC at high level (<12 g kg⁻¹), this difference should be taken into account.

(2) The authors indicate that bulk density was "interpolated" for apparently all of the soil samples taken during the 1982-2011 study period (page 16503, lines 8-9). I assume this means that there were either none or very few direct measurement of soil bulk density to accompany the soils that were collected for measurement of carbon concentration. Since bulk density has a very large and important impact on the calculation of soil carbon stocks, this strikes me as an important limitation to the value of this data set. Furthermore, bulk density can vary substantially across a landscape in response to soil physical characteristics, organic matter production and decay, land management practices, and variation in these factors through time. So, trying to simply interpolate this very important number could give rise to large and unknowable errors in the estimate of soil carbon stocks (mass per unit area).

[AUTHORs]: Our study aimed to analyze the impacts of agricultural intensification on soil carbon, and the relationship between SOC content and driving factors (climate and farming managements). The SOC content and its relationship with driving factors are not influenced by soil bulk density. The soil bulk density was included in calculation of the change of SOC storage within the past three decades and the minor error of soil bulk density will not change the general conclusion of this study. However, we revised the text of Discussion section at line 348-350 to highlight this issue.

Minor comments

Page 16499, Line 7: I'm not sure why there should be a tilde () in between the two numbers 1.3 and 21.2. This is also done on Page 16501 Line 14. Should this actually be a dash, or some other

symbol?

[AUTHORs]: Tilde has been replaced by dash at Line 51 and 113, and modification has also been done at other relevant lines of the MS.

Page 16499, Line 7: The units "million T C annually" are used in the middle of this line. It might be better to transform this into Teragrams (Tg) of C since you use Tg throughout the remainder of the paper.

[AUTHORs]: This has been revised at Line 51.

Page 16502, Line 4: The term "SOC content" is used here and in many subsequent locations throughout the manuscript. This is a vague term and should be replaced with "SOC concentration".

[AUTHORs]: We have consulted several scientists on this issue and the responses are different. Some scientist thought that concentration is more appropriate for liquid. The term of "SOC content" or "carbon content" has also been in many papers, for example: Guohan Song, Lianqing Li, Genxing Pan, et al. Topsoil organic carbon storage of China and its loss by cultivation. Biogeochemistry, 2005, 74: 47-62; Pat H. Bellamy, Peter J. Loveland, R. Ian Bradley, et al. Carbon losses from all soils across England and Wales 1978–2003. Nature, 2005, 437(8): 245-248; Catherine E. Stewart, Keith Paustian, Richard T. Conant, et al. Soil carbon saturation: concept, evidence and evaluation. Biogeochemistry, 2007, 86:19-31. So in our MS we kept the expression of "SOC content".

Referee #4

The paper analysed the change in soil carbon stocks and content over a period of three decades for a county in northern China. Although the paper is well written, and the authors collected a relevant data set, the paper lacks details on the methodology and data sources, which should be added. The subject of the paper is interesting, but does not really provide a substantial contribution to scientific progress, as there have been more studies that showed that agricultural intensification in China can lead to increased SOC levels.

[AUTHORs]: We hope our study can contribute to deeper and wider understanding of the impact of agricultural intensification on SOC levels in the flowing perspectives: 1) Long-term monitoring of SOC change at regional level. The study period in the study was 30 years (from 1982 to 2011), covering the most dynamic stage of Chinese agricultural development. Most studies on SOC change in China are around 20 years (Table 3); 2) High densities of soil sampling. The least number of samples was 199 in 2003 and the largest number was 3637 in 2006. Average number of soil samplings was about 800 sampling annually. 3) Evolvement of straw incorporation. Wheat straw incorporation started in 1988 and the farmland area under incorporation gradually increased gradually. Maize straw incorporation reached 70% of total farmland in 2008. These three features are quite unique among studies on relationship between SOC level and farming managements and will provide an interesting and important scientific evidences for better management of SOC.

For the methodology and data sources, we gave more details at line 113-115.

Minor comments:

* Page 16499, line 7: Unit not clear

[AUTHORs]: This has been revised at Line 51.

* Page 16499, line 25: here is mentioned that few if any study exist on the SOC content and stock change, however, later in Table 3 several studies are mentioned for Northern China, thus change this text.

[AUTHORs]: This has been revised accordingly at Line 70-72.

* Page 16500: Add also some information on the total size of the county

[AUTHORs]: We have added the data of 509 km² at Line 82.

* Page 16501: How many soil samples were derived from the Annual Soil Fertility Survey for each year?

[AUTHORS]: Information of soil sampling was included in a table in the early edition of this paper and the table was deleted according to the editor's suggestion. More information of annual soil fertility survey was added at line 114-115.

* Page 16501: It is not clear whether land use is also reported in the soil survey

[AUTHORs]: Land use was reported in the annual soil survey and was categorized to types of cropland, vegetable land and construction land.

* Page 16502: According to Pribyl (2010) a conversion factor of 0.5 would in most cases be more appropriate

[AUTHORs]: In China, 0.58 is a most commonly accepted conversion coefficient in soil organic matter/carbon research, and still adopted now in official soil organic matter monitoring program (Bao, S. D. Soil Agro-chemistry Analysis. Beijing: China Agriculture Press. 2005; Wu, H. B., Guo, Z. T., Peng, C. H. Land use induced changes of organic carbon storage in soils of China. Global Change Biology, 2003, 9: 305-315). We also used this coefficient in this paper to keep the consistence with the data of SOC in other studies like in Table 3. Thank the reviewer for this comment and in later research, we will consider the coefficient proposed by Pribyl (2010).

* Page 16502: At which level was the climate data obtained, average for the county or higher resolution? And temporal resolution?

[AUTHORs]: Climate data obtained is average for the county level. The data of temperature and precipitation is the daily values. We revised the text at Line 149-150.

* Page 16503: Data analysis section should be extended, explaining better how the data were calculated, how many samples, and average for each land use?

[AUTHORs]: Equation (3) and (4) are used for SOC density and SOC storage, respectively, applicable for cropland, vegetable land and construction land. During the years when the annual soil survey was implemented and detailed soil data was obtained, the number of soil

samplings ranged between 199 and 3637 with an average of 800 samples per year. Number of vegetable soils averaged 79. We added the information at Line 164-166.

*Page 16505 and Figure 2: How was the SOC stock under construction land determined? I would expect that this should be lower, as the top layer is often removed.

[AUTHORs]: Farmland protection is a basic policy in Huantai and China. When farmland is transferred to construction land, the top soil (0-30 cm) will be removed to other farmland or greenhouse. This meant that the process of construction/industrialization will not result in the significant loss of soil carbon. So the SOC content in the previous year was viewed as the SOC content of construction land after transformed from farmland.

* Page 16506, line 20: Not by precipitation, as that was not a significant correlation according to Table 1

[AUTHORs]: We revised the text at Line 256-258 as this is a general statement.

* Page 16508, line 1: Why was the C input from organic fertilizers not included in the analysis, although not significant, it could be added to Table 1.

[AUTHORs]: In Huantai, organic fertilizer was not applied in cropland since 1980s except in vegetable land and orchard. Our paper mainly analysis the relationship between SOC level and farming managements on cropland, so the C input from organic fertilizer was not included in the analysis.

* Page 16508: The N2O emissions will be even higher, here only the direct emissions are accounted for, but including the indirect emissions (ammonia volatilization and leaching and runoff) and the emissions from crop residues will double this value

[AUTHORs]: We agree with the reviewer's point. The text was revised at Line 307-309.

References:

Pribyl, D.W. 2010. A critical review of the conventional SOC to SOM conversion factor. Geoderma 156, 75-83.

Increase in soil organic carbon by agricultural intensification in Northern China

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Abstract. Agricultural intensification has contributed greatly to the sustained food supply of China's 1.3 billion population over the 30 year period during 1982-2011. Intensification has several and widely recognized negative environmental impacts including depletion of water resources, pollution of water bodies, greenhouse gas emissions and soil acidification. However, there have been few studies over this period on the impacts of intensification on soil organic carbon (SOC) at the regional level. The present study was conducted in Huantai county, a typical intensive farming region in Northern China, to analyze the temporal dynamics of SOC influenced by climate and farming practices. The results indicate that from 1982 to 2011, SOC content and stock in the 0-20 cm layer of the cropland increased from 7.8 ± 1.6 to $11.0 \pm 2.3 \text{ g} \cdot \text{kg}^{-1}$ (41%) and $21 \pm 4.3 \text{ to } 33.0 \pm 7.0 \text{ Mg} \cdot \text{ha}^{-1}$ (54%), respectively. The SOC stock (0-20 cm) of the farmland for the entire county increased from 0.75 to 1.2 Tg (59%). Correlation analysis revealed that incorporation of crop residues significantly increased SOC, while increase in the mean annual temperature decreased the SOC level. Therefore, agricultural intensification has increased crop productivity and contributed to SOC sequestration in Northern China. In the near future, more appropriate technologies and practices must be developed and implemented for a 正版用户 1/25/2015 2:49 PM

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36 maintenance or enhancement of SOC in this region and elsewhere in Northern China,

that also reduce non-CO₂ greenhouse gas emissions, since the climate benefit from the

additional SOC storage is estimated to be smaller than the negative climate impacts of

39 N₂O from N fertilizer additions.

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Keywords: Soil organic carbon, agricultural production intensification, crop residue

42 incorporation, nitrogen fertilizer, temperature, Northern China

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1 Introduction

Increasing soil organic matter (SOM) storage in arable lands can ensure the sustained supply of nitrogen (N) and other nutrients to crop growth and maintain appropriate soil quality such as aeration, permeability, water-holding capacity and nutrient preserving capacity (Smith et al., 2012). Globally, accumulation of SOM or soil organic carbon (SOC) stock in arable lands, which contributes to the mitigation of greenhouse effect and a concomitant improvement in soil fertility (Matson et al., 1997; Sainju et al., 2009), may be achieved by a range of improved farming practices. These practices include adoption of high-yielding crop varieties, balanced fertilization, crop residue incorporation, no-till (NT) or reduced tillage, optimal irrigation, high cropping intensity (Matson et al., 1997; Kucharik, et al., 2001). For instance, agricultural soils in US had a carbon sink capacity of 1.3-21.2 Tg C annually from 1982 to 1997, due to land use, NT, higher cropping intensity etc. (Eve et al., 2002; Ogle et al., 2003). The rate of increase in SOC stock in Canada was 5.7 Tg C yr⁻¹ during 1991-2001 (Vanden Bygaart et al., 2004). In The Netherlands, the SOC content of arable land increased by about 0.08 g kg⁻¹ yr⁻¹ between 1984 and 2004 (Reijneveld et al., 2009). Benbi and Brar (2009) reported that SOC in the Punjab state of India increased from 2.9 to 4.0 g kg⁻¹ (38%) between 1981 and 2006, largely resulting from irrigation, optimal fertilization, and an increase in crop productivity.

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and 35-40% of nation's wheat (Triticum aestivum L.) and maize (Zea mays L.),

respectively (NBSC, 2014). Similar to other regions in Northern China, Huantai

Northern China is one of the most important agricultural regions, producing 60-80%

county has experienced the agricultural intensification process including a high cropping intensity (200%, winter wheat—summer maize), high fertilizer rate (500,600 kg N ha⁻¹ yr⁻¹), frequent irrigation and tillage, and an increasing ratio of crop residues incorporation since the 1980s (Shi et al., 2013; Kong et al., 2014). Agricultural intensification increased crop yield within a short period. For example, Huantai county achieved production of >15 Mg grains (wheat + maize) ha⁻¹ yr⁻¹ for all of its farmland since 1990. The effect of agricultural intensification on increasing crop yields has been well documented, however, most of the research done on SOC sequestration in agricultural soils is confined to long-term plot-scale experiments. Studies available at national or region levels were within short period, especially in Northern China (<20 years) where is characterized by low levels of SOM.

We collected three-decades of data of climate, farm management and crop yield from Huantai county, to: 1) analyze the evolution of SOC at the regional level from 1982 to 2011, and 2) establish the cause-effect relationship between the driving forces and SOC change. The results derived from this study may contribute to improved farm management for the long-term sustainable agricultural development in the intensive farming of Northern China and elsewhere.

2 Materials and methods

2.1 Study area

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Huantai county, with an area of 509 km², is located in Northern China (36°51′50″-37°06′00″ N latitude and 117°50′00″-118°10′40″ E longitude). It is characterized by a warm temperate continental monsoon climate, with annual average temperature of 13.4 °C and annual precipitation of 604 mm. The rainfall occurs mainly in June, July and August; with the annual frost-free season of about 198 days. Slope gradient of the landscape is low in northern and high in southern regions, with an average altitude of 6.5-29.5 m, falling gently from southwest to northeast. The main soil types, according to the US soil classification system, include Haplustalfs, Aquents and Vertisols. The household contract responsibility system was implemented in Huantai county in 1980, and land productivity has increased

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significantly with an increase in fertilizer input, frequent irrigation and adoption of high-yielding varieties. In 1990, Huantai county became the first *Dun Liang County* which achieved high productivity of one Mg of grain per mu for the whole county (1 mu=1/15 ha) in China. Vegetable production has also been intensified in the county since 1990s.

2.2 Data Collection

The soil, climate and farming data were collected between 2011 to 2013 from the sources as described below:

- *The 2nd National Soil Survey*: The 2nd National Soil Survey in China was conducted during 1981-1983, and was undertaken in 1982 for Huantai county. The survey collected and analyzed soil samples for genesis, physical, chemical and biological properties. The SOM content was tested using the potassium dichromate titrimetric method (Jankauskas et al., 2006). The SOM data in this study was obtained for 258 soil samples (0-20 cm), together with the corresponding GPS location coordinates.
- Annual Soil Fertility Survey: The annual SOM data of farmland soil (0-20 cm) were collected from the Soil Fertility Survey, a program undertaken by the county agricultural extension since 1987. Every year, samples from the topsoil layer were collected after the autumn harvest and analyzed for pH, SOM and nutrient contents. Soil sampling in the survey was stratified according to the division of administrative villages. Each village had at least one composite soil sample, representing 6.7-33.3 ha of farmland. The number of soil samples was from 199 (in 2003) to 3637 (in 2007) and the average was 786 each year. The SOM content was determined by the potassium dichromate titrimetric method (Jankauskas et al., 2006). The data for 1988, 1991, 2000, 2001, 2004, 2005 and 2010 were for each town as the survey was only done at the township level. As there were no data for the period 1983 to 1986, it was assumed that a steady change in SOC occurred during this period. Thus, the average SOC content for the whole county was obtained by interpolation based on the data from 1982 and 1987.

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- *Soil Sampling and Measurement Program*: In September 2011, a soil survey was implemented for the whole county. Soil samples were obtained from 0-20 cm depth of the farmland (including cropland and vegetable land), in an evenly distributed 2 × 2 km grid. Each sample was composited from three collection points. The GPS location was recorded for each sampling point. Soil was ground and passed through a 0.15 mm sieve. For the SOC analysis, 2, 3 g of the soil sample was weighed into the beaker, and then 20 mL HCl of 0.5 mol·L⁻¹ was added for acidification and removal of carbonates. Samples were then transferred into a rotary oscillator (Ronghua, HY-B) to shake for 30 min. After standing for 12 h, deionized water was used to remove the acid from soil samples until neutral pH was obtained. Soil samples were dried in the oven at 60 °C and weighed. The SOC content was determined by a C and N Elemental Analyzer (Thermo EA flash 1112). As acidification led to soil mass loss, the SOC data were corrected using Eq. (1):

$$W_{soil2} = \frac{m_{soil1} \times W_{soil1}}{m_{soil2}} \tag{1}$$

Where, W_{soil2} is the corrected SOC content, m_{soil1} is the soil mass after acidification, W_{soil1} is the SOC content after acidification and m_{soil2} is the soil mass before acidification. The comparative study of SOC content determined by Elemental Analyzer and potassium dichromate titrimetric method indicated that these two methods are comparable and the differences in between are not significant (Wang et al., 2014).

- Climate and farm management: Data were obtained for land use (e.g. area under crops, vegetables and urban use), grain and straw yields of wheat and maize, nitrogen fertilizer rate, and the rate of straw incorporation from the Huantai agricultural yearbook of 1982 - 2011. Climate data for temperature and precipitation were obtained from the China Climatic Data Center, National Meteorological Information Center, CMA (http://cdc.cma.gov.cn). The temperature and precipitation data was

daily value for the county level.

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- Calculation of SOC density and stock: The Van Bemmelen conversion factor, namely that SOM contains 58% carbon (Page et al., 1982), was used to compute SOC content in (Eq. (2)). The SOC density and stock were computed by Eq. (3) and Eq. (4), respectively:

$$SOC = SOM \times 0.58 \times 10 \tag{2}$$

$$SOCD = SOC \times \gamma \times H \times 10^4$$
 (3)

$$SOCS = SOCD \times S \times 10^{-9}$$
 (4)

where, SOC is the soil organic carbon content (g kg⁻¹), SOM is the soil organic matter content (%), *SOCD* is the SOC density (kg ha⁻¹), γ is the soil bulk density (BD, g cm⁻³), H is the thickness (m) of soil layer (0-20 cm), *SOCS* is the SOC stock (Tg) for the whole county, and S is the farmland area (ha). Soil BD values were interpolated over years from measured values taken in 1982 and 2011. Average number of cropland and vegetable land soil samples was 786 and 79, respectively. For the calculation of SOCS, we assumed that the SOC content of land for industrial or

residential use was maintained at the same level after the farmland was converted.

2.3 Data analysis

The Kolmogorov-Smirnov test was used in the SPSS Statistics 17.0 package to determine if SOC content followed a normal distribution. Central tendency, dispersion degree and distribution characteristics of SOC data were calculated. Pearson correlation analysis and/or partial correlation analysis were conducted between SOC content and driving factors including mean annual temperature, mean annual precipitation, grain yield, nitrogen fertilizer rate and straw C incorporation. A multivariate regression model was developed to account for the impact of these factors on temporal SOC change in Huantai county.

3 Results

205 3.1 Evolution of SOC content and density between 1982 and 2011

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Figure 1 presents the dynamic changes of topsoil SOC content and density for the cropland (winter wheat-summer maize) from 1982 to 2011. The mean increase in cropland from 1982 to 2011 was 7.8 ± 1.6 g kg⁻¹ to 11.0 ± 2.3 g kg⁻¹ for SOC content and, 21.4 ± 4.3 Mg ha⁻¹ to 33.0 ± 7.0 Mg ha⁻¹ for SOC density, with rates of increase of 41% and 54%, respectively. The mean SOC content of vegetable land increased similarly as cropland, i.e., from 7.8 ± 1.6 g kg⁻¹ to 11.0 ± 2.8 g kg⁻¹ (data not shown). This trend indicates that the rate of increase in SOC content and density of cropland (0-20 cm) in Huantai county since the early 1980s was 0.11 g kg⁻¹ yr⁻¹ and 0.40 Mg ha⁻¹ yr⁻¹, respectively. The growth of SOC density was significantly related to increasing SOC content, but also supported by the increase in soil BD in the 0-20 cm layer of the farmland (1.4 g cm⁻³ in 1982 to 1.5 g cm⁻³ in 2011).

3.2 Change of SOC stock in Huantai county from 1982 to 2011

An adjustment in the local agricultural sector altered the land use between 1982 and 2011 in Huantai county. The farmland area in 1982 was 35204 ha, of which more than 99% was under winter wheat - summer maize cropping (cropland). Due to the expansion of vegetable production since 1990s, land used for vegetable production increased to about 20% by the early 2000s, and has remained constant until 2011 at about 6000 ha. The average SOC content of vegetable land is not significant different from that of cropland (11.0 vs. 11.0 g kg⁻¹ in 2011 for whole county). Some farmland was converted to construction use with the expansion of industry and urban land uses. Of the 31% reduction of farmland area between 1982 and 2011, 16% was used for vegetable farming and 15% converted to construction land.

Although area under cropland decreased from 35,204 ha in 1982 to 24,343 ha in 2011, the SOC stock of cropland (0-20 cm) increased from 0.75 ± 0.15 to 0.80 ± 0.17 Tg C (an increase by 6.7%. Fig. 2). When the SOC stock in vegetable land and construction land (converted from cropland and for industrial and residential use. The area increased every year as urbanization took place) was also included, total SOC stock of the farmland in the Huantai county was estimated to be 1.2 Tg C in 2011,

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3.3 The cause-effect relationship governing change in SOC level

Among the natural/climate forces which can influence SOC level, mean air temperature in Huantai county increased within the period of 1982 to 2011, with a relationship represented by the regression equation y = 0.073x + 12.2 ($R^2 = 0.67$, P < 0.0001, Fig. 3a). However, there was no significant change in precipitation over this period. Nitrogen fertilizer input for farmland was 400 kg N ha⁻¹ yr⁻¹ in the 1980s and peaked at about 600 kg N ha⁻¹ yr⁻¹ in 1994, followed by a decline to 500 kg N ha⁻¹ yr⁻¹ in 2011. There were also significant increases in grain yield over this period ($R^2 = 0.63$, P < 0.0001, Fig. 3b). The carbon input between 1982 and 1987 was estimated at about 800 to 1000 kg C ha⁻¹ yr⁻¹, mainly through roots. However, crop production experienced a rapid growth after 1988, and crop residues (mainly wheat straw) were returned to the soil, leading to a significant increase in the input of biomass-C, especially the aboveground C. As much as 70% of maize straw was also incorporated to the farmland from 2007 onward, leading to a total C input of > 8000 kg ha⁻¹ yr⁻¹ ($R^2 = 0.90$, P < 0.0001, Fig. 3c).

Correlation and regression analyses were performed between SOC of cropland and driving factors, i.e., temperature, precipitation, crop (wheat and maize) yield, N fertilizer rate and C input from crop residues (Table 1). There was a highly significant correlation (P<0.01) between SOC content and temperature, crop yield, C input from crop residues, with correlation coefficients (r) of 0.55, 0.79 and 0.91, respectively. The correlation between SOC and N fertilizer rate was also significant (r=0.38, P<0.05). However, there was no significant correlation between SOC content and the mean annual precipitation.

A partial correlation analysis was conducted to determine the relationship between SOC content of cropland with any one major driving factor, as the effect of a set of controlling random variables removed (Table 2). The data indicated a highly significant and positive correlation between SOC content and the C input from crop residues (r=0.80, *P*<0.0001), but a negative correlation between SOC content and

annual mean temperature (r=-0.42, *P*=0.027). The weak positive correlation between SOC content and N fertilizer rate (r=0.03, *P*=0.86), and weak negative correlation between SOC and crop yield (r=-0.08, *P*=0.70), indicated that N fertilizer and increasing crop yield did not contributed to the augment of SOC in Huantai from 1982 to 2011. As the effect of C input from crop residues was removed during partial correlation, the rising temperature during the past 3 decades significantly decreased the cropland SOC content in Huantai, or promoted the SOC decomposition. In addition to the partial correlation analysis, a multivariate regression model was also developed as follows:

Y=12.0 - 0.31 Temperature - 0.0003 Precipitation - 0.0006 Nitrogen + 0.0005 C input - 0.0001 Yield (r^2 =0.88)

where, Y is the SOC content in g kg⁻¹. The multi-variate regression analysis also confirmed the positive impact of the input of crop residue carbon and negative impact of the increase in temperature.

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4 Discussion

4.1 Driving factors for SOC accumulation

The SOC level of farmland was influenced by climate (mainly temperature and precipitation) and farming practices, including crop residue incorporation, N fertilizer use, crop yield etc. (Khan et al. 2007; Ladha et al. 2011).

Climate factors: Climate warming may increase the rate of SOM decomposition, while the effect of precipitation on SOC is mostly known to be positive because in general SOC increases with increase in precipitation (Post et al., 1982). Whereas air temperature in the Huantai region has significantly increased since the 1980s, the precipitation has not. There is a significant positive correlation between the temperature and SOC content (Table 1), however the partial correlation analysis showed that the correlation was negative (r=-0.42, p=0.03, Table 2), indicating that SOC in Huantai county was enhanced by factors other than temperature and precipitation. Indeed, it may take much longer (~50 yrs) to observe the effect of climate change on SOC level (De Bruijn et al., 2012), indicating a strong need for



long-term research.

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Nitrogen fertilizer: In general, N is the most limiting nutrient in crop production systems (Robertson and Vitousek, 2009). It promotes the production of crop dry matter (and therefore C input to the soil) while chemically stabilizing C in the soil, thereby potentially increasing soil C storage (Paustian et al., 1997). A high input of N fertilizer was a prominent feature of farming in Huantai county, where average N fertilizer rate increased from 400 kg N ha⁻¹ yr⁻¹ in the 1980s to 600 kg N ha⁻¹ yr⁻¹ in the 1990s. However, the rate of N fertilizer gradually declined and stabilized at 500 kg N ha⁻¹ yr⁻¹ in the 2010s. Such a trend is attributed to the increased use of machinery in agriculture and the extension of formula fertilization techniques adopted in the region. The strategy was to balance the N fertilization rate in consideration of the high grain output (>15 Mg grain ha⁻¹ yr⁻¹). However, the rate of SOC increase was still high during this period, as indicated by a weak positive correlation between N fertilizer rate and SOC level over the 30-year period (Table 1). The vegetable production in Huantai county is open-field and there is less organic fertilizer input (3-5 t ha⁻¹ yr⁻¹) compared with other regions like Shouguang (>15 t ha⁻¹ yr⁻¹) in China, SOC increase due to organic fertilizers is not significant and at the same level as cropland (11.0 vs. 10.9 g kg⁻¹ in 2011).

A few studies (Khan et al., 2007; Mulvaney et al., 2009) have reported that even the long-term input of a massive amount of residue-C and synthetic N fertilizer do not sequester SOC; this was not the case with intensification of cropland in Northern China. A major factor lies in the concentration of the principal parameter (i.e., SOM) which was significantly lower in cropland soils of Northern China than those of the U.S. Corn Belt (9 vs. 25 g kg⁻¹, Ludwig et al., 2011) or countries of the western Europe, because large areas of farmlands in Northern China were affected by saline-alkaline processes prior to 1970s. The initial low crop productivity in the temperate region where N fertilization rate was low resulted in higher SOC level in Northern China with increasing biomass input achieved by higher input of N. Similar trends have been reported by other studies from around the world (Song et al., 2005;

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of mineral N not only increases the production cost, but also exacerbates the negative environment effects, including nitrate contamination of ground and surface waters and N₂O emission to the atmosphere (Triberti et al., 2008). Carbon sequestration effects should be valued in a systematic approach (Wang and Cao, 2011). Indeed, the climate benefit from the additional SOC storage is smaller than the climate damage caused by N₂O from N fertilizer additions (N₂O emissions are estimated (conservatively) to be ~57 kt CO₂-eq. yr⁻¹, assuming the smallest farmland area of 24343 ha and current stable N application rate of 500 kg N ha⁻¹ yr⁻¹ using the IPCC Tier 1 default Emission Factor of 0.01; soil C storage over the period is equivalent to 54 kt CO₂-eq. yr⁻¹). If the indirect emissions (ammonia volatilization, leaching and runoff) are considered, the environmental effect of nitrogen fertilizer application will be more serious.

Balanced fertilization should be widely promoted, for optimization of the integrated economic benefits and ecosystem services. It is important to understand that increasing input of N fertilizer increased SOC only when crop residues were returned to the soil. There may have been either no, or only a slight increase in SOC level, if the aboveground crop residues were removed or burnt (Alvarez, 2005) so N fertilization in itself is not a suitable strategy to increase SOC, particularly considering the over-riding effects of N₂O emissions from the N fertilizer.

Grain yield: Grain yield in Huantai county increased from 7200 kg ha⁻¹ yr⁻¹ in 1982 to 16117 kg ha⁻¹ yr⁻¹ in 2011 (an increase of 124%). The highly significant correlation (r=0.79, *P*<0.01) between SOC content and grain yield indicates the importance of SOM to achieving high crop productivity, and *vice versa* (Pan et al., 2009). Indeed, the interdependence between crop yield and SOM content is widely recognized (Lal, 2002; 2013). Increase in the SOC pool of 1 Mg C ha⁻¹ in the root zone can increase annual food production by 30-50 million Mg in developing countries (Lal, 2013). Meta-analysis indicated that crop yield greatly increased by crop residue retention (*P*<0.001), particularly in upland China (Liu et al., 2014). Qiu et al. (2009) estimated that for every increase of 1 g C kg⁻¹ of SOC in Huantai county, grain yield could increase by 454 kg ha⁻¹. In Northern China, beneficial effects of crop

yield increase will be enhanced as the strategy of returning crop residues is extended to other regions of Northern China.

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Carbon input from crop residues: Carbon input is one of the most efficient factors for the accretion of SOC, which is also confirmed by the highly significant correlation between SOC content and C input from crop residue incorporation (r=0.80, P<0.0001, Table 2). Similar results have also been reported by other scientists (Freibauer et al., 2004). Smith et al. (2005; 2012) reported that input of crop residues could attain the highest rate of C sequestration (0.7 Mg C ha yr⁻¹) in comparison with that of merely 0.2 Mg C ha⁻¹ yr⁻¹ with the input of mineral N fertilizer. In our study, there are two SOC increase stages, one early (1987-1992) and one late (2007-2012), which the early one coincides nicely with increased wheat residue incorporation and the late one with the increased maize residue incorporation. Again, it highlights the significance of crop straw input for the building of soil organic matter. In comparison with data from other countries like India (~1 Mg C ha⁻¹ yr⁻¹, Srinivasarao et al., 2014) or US (~4.2 Mg C ha-1 yr-1, Johnson et al., 2006), the input of residue-C in Northern China is much higher (~8 Mg C ha⁻¹ yr⁻¹, Fig. 3c), for maintaining a rapid rate of increase of SOM. Thus, rate of input of residue-C is the principal determinant of the rate of increase of SOM: the annual rate of SOM increase being lower in India, intermediate in the U.S. and high in the North China Plains. Since retention of the entire amount of residues of maize in North China started from 2007/2008, the accretion of SOC is projected to continue for another two to three decades until the mid-2040s. It should also be noted that in our study, soil bulk density from 1983 to 2011 was interpolated and this could give rise to errors in the SOC storage estimation.

Miao et al. (2011) reported that the significance of N fertilizer and crop residues incorporation to the maintenance and increase of the SOM. Retention of residues (wheat and maize) in conjunction with appropriate rate of N fertilization have been properly implemented in Northern China where the antecedent levels SOM are much lower than those in northern America and western Europe. Therefore, a judicious continuation of these practices will continue to accumulate SOM for a long time to come. However, similar trends may not occur under all situations. For example, Khan

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et al. (2007) explained that after attaining a steady state, it is unlikely that SOC will continue to increase, and may even decline with continuous use of synthetic N, because of the enhanced activities of heterotrophic soil microorganisms in using crop-derived residues or SOM. In the context of Northern China, therefore, it is likely that the increase in SOC level will continue because of the improved crop productivity and retention of crop residues, but it will eventually attain a new equilibrium. Some other SOC-enhancing practices including application of residue-based animal manure derived from the same land unit, contribute further to SOC sequestration. In addition, higher level of mechanization during the agricultural intensification process may increase the soil BD (about 7% in our study), and this also contributed to the increase of SOC stock and should not be ignored when quantifying of farmland carbon sequestration.

4.2 Comparison of SOC accretion through agricultural intensification in different regions of China

Agriculture in China has grown rapidly over the three decades since 1982, primarily because of the Household Contract Responsibility System and adoption of the open policy. Further, the intensification process achieved some economic and environmental benefits (Firbank et al., 2013). In the case of Northern China, however, there were also problems with increases in soil compaction and water pollution over the period of three decades.

Among all agronomic regions, Northern China registered the highest rate of SOC sequestration. In contrast, however, the SOC level has declined in Northeast China since the 1980s (Table 3). Northeast China, one of the few world regions characterized by the black soil (Phaeozems) and cold climate, has high antecedent SOC content (~ 20 g kg⁻¹ in uncultivated soils). Thereafter, cultivation and intensive farming after the 1980s have increased the rate of decomposition of SOM along with a low input of organic materials and biomass-C. The SOC level in Northern China was extremely low (5 to 10 g kg⁻¹) in comparison with the soils of Northeast China and other regions. This trend can be explained by the fact that Northern China has a

long history of low crop productivity, long dry season (Stockmann et al., 2013), sandy soil texture and low input of organic amendments. Application of N fertilizer and retention of crop residues has increased the SOC level more in dry lands of Northern China than in those of paddy soils of the lowlands (Liu et al., 2014). Huantai county and other agronomic regions (except Northeast China) in China also witnessed increase in SOC level, mainly attributed to the practices of fertilization (synthetic and organic), increase in crop yield, retention of crop residues, adoption of conservation tillage and use of organic amendments (Table 3). The 0-30 cm layer of soils of cropland in China have gained SOC at the rate of 17-28 Tg C yr⁻¹ between 1980 and 2000 (Huang et al., 2010), which is similar to the estimates of 25-37 Tg C yr⁻¹ reported by Lal (2002).

Agricultural intensification in China is an ongoing process, and is progressively evolving over time. For instance, since 2012, maize residues from some cropland have been harvested by Huantai farmers for use as cattle feed and the eventual return of the animal manure to cropland. It is possible that the efficiency of SOM accretion through animal manure is higher than that of returning maize straw (Wilhelm et al., 2007), which may result in yet another period of SOM accretion at the regional level with proper dissemination of this technology (Ladha et al., 2011). Similar to northern China, other important grain production region like Midwest US also experienced the stage of SOC accumulation, although the practices (residue management, non- or reduced tillage and crop rotation) are different (Ogle et al., 2003; Jelinski and Kucharik, 2009). Adoption of conservation agriculture (NT or minimum tillage) may be another option for SOC sequestration. However, its applicability and efficiency need to be validated through long-term research.

453 5 Conclusions

The study of the impact of agricultural intensification on SOC content and stock was conducted in the Huantai county, which is a representative region in Northern China. The farmland SOC stock of the whole county increased by more than 50% over three decades from 1982-2011. Among several improved farming practices,

M 1/23/2015 3:57 PM 已删除: M 1/23/2015 3:58 PM 已删除: M 1/23/2015 3:58 PM 已删除: M 1/23/2015 3:58 PM 已删除: retention of crop residues strongly contributed to the restoration of SOC, but there was no synergistic effect between N fertilization rate and crop yield on increase in SOC. The SOC content decreased with increase in mean annual temperature. The temporal change in SOC was significantly influenced by the evolution of the practice of retention of crop residues through implementation of some local farming policies. The data support the conclusion that agricultural intensification may both increase crop productivity and enhance some ecosystem services, such as SOC sequestration in croplands of Northern China. However, current farming practice (e.g., retention of crop residues) may not always linearly increase SOC over time, indicating a strong need for a long-term research. Furthermore, there is also a need to explore other options such as the application of manure through integration of crop and animal production. Research on the use of animal manure within the region is a priority, because of its multiple benefits for grain production, the economy and ecosystem services such as SOC sequestration.

Author contribution

Fanqiao Meng, Wenliang Wu and Yan Liao designed the experiments and Yan Liao carried them out. Yan Liao and Pete Smith performed the calculations and data analysis. Yan Liao, Fanqiao Meng, Pete Smith and Rattan Lal prepared the manuscript with contributions from all co-authors.

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	SOC	Temperature	Precipitation	Crop yield	N fertilizer rate	C input from crop residues
SOC	1					
Temperature	0.55**	1				
Precipitation	0.30	0.09	1			
Crop yield	0.79^{**}	0.62**	0.30	1		
N fertilizer rate	0.38^{*}	0.01	0.48**	0.55**	1	
C input from crop residues	0.91**	0.73**	0.35	0.89**	0.43*	1

^{**} means a highly significant correlation at the level of p<0.01; * means a significant correlation at the level of p<0.05

Table 2 Partial correlation analysis between SOC content of cropland and driving factors in Huantai county

	Partial correlation coefficient between SOC and the					
Control variables	driving factor*					
Crop yield, C input from	SOC vs. Temperature	-0.42 (p=0.03, df=26)				
crop residues						
Crop yield, C input from	SOC vs. N fertilizer	0.03 (p=0.86, df=26)				
crop residues	rate					
C input from crop residues,	SOC vs. Crop yield	-0.08 (p=0.70, df=25)				
N fertilizer rate,						
Temperature						
N fertilizer rate,	SOC vs. C input from	0.80 (p<0.0001, df=25)				
Temperature, crop yield	crop residues					

^{*}All of the partial correlation coefficients are 2-tailed.

Table 3 Topsoil (0-20 cm) SOC change from 1980s to 2000s in different agronomic regions of China*

	regions of	Cillia								
						SOC	SOCD	SOCS		
			Soil type				Mg ha ⁻¹			
	Region	Site	(FAO)	Climate	Period	%	yr ⁻¹	Tg yr ⁻¹	Literature	
	Our study	_	_			41	0.40	0.15	_	
1	North	Quzhou	Fluvisols	Warm,	1980-2000	31			Liu, et al., 20	
	China			semi-arid,						M 1/23/
				temperate						已删除
				monsoon						
1		Daxing	Fluvisols	Warm,	1981-2000	33			Hu, et al., 20	06
ı		Č		semi-humid,	₹					M 1/23/
				temperate						已删除
				monsoon						
1		Hebei	Calcaric	Warm,	1984-2004		0.34	2.2	Xi, et al., 201	13
ı		110001	Cambisol	semi-arid,	1,01,2001		0.5 .		111, 00 u.i., 20	M 1/23/
			Camoisoi	temperate						已删除
				monsoon						
ı		Henan	Fluvisols	Warm,	1984-2004		0.32	2.8	Xi, et al., 201	12
ı		Helian	1 14 13013	semi-humid,	1704-2004		0.52	2.0	A1, ct a1., 201	M 1/23/
				temperate						已删除
				monsoon						
1		Luancheng	Calcaria	Warm,	1979-2000	38			Zhang et al.,	2002
		Luaneneng	Cambisol	semi-arid,	1979-2000	30	▼		Zhang et al.,	M 1/23/
I			Califolisot	temperate	▼				//_ [已删除
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1	N. d.		CI.	0 11 11	1004 2004		0.55	2.0	W: 1 1 20	M 1/23/
I	Northeast	Liaoning	Chernozems	Semi-humid,	1984-2004		-0.57	-2.9	Xi, et al., 20	已删除
	China			temperate						M 1/23/
				continental					\\\	已删除
1				monsoon						M 1/23/
		Jilin	Albic luvisols	Semi-humid	1985-2005		-0.81	-7.2	Xi, et al., 20	己删除
				temperate					\ \ \	
1				monsoon					\ \	M 1/23/
		Heilongjiang	Phaeozems	Cold	1986-2006		-0.70	-5.5	Xi, et al., 20	
				temperate					\ \	M 1/23/
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		Heilongjiang	Phaeozems	Cold	1982-2002	-14			Yu et al., 20	
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	Northwest	Yining	Calciustoll	Arid	1981-2001	-9.3			Hou et al., 2	已删除
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		Akesu	Calcaric	Arid, warm	1982-2001	10			Li et al., 2002	2

		fluvisols	temperate continental						
	Huangshui	Eutric cambisols	monsoon Arid, warm temperate continental	1981-2001	28			Chen et al, 200)3
	Zhangye	Eutric cambisols	monsoon Arid, warm temperate continental monsoon	1982-2003		0.2		Zhang et al., 20	009
East Chin	a Jiangsu	Fluvisols/can bisols		1980-2000	34			Yu et al., 2003	
	Jinhua	Haplic alisols		1982-2002	2.6			Xie et al., 2003	3
	Xuzhou	Fluvisols	Warm, semi-humid, temperate monsoon	1981-2001		0.1~0.5		Zhang et al., 20	009
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	Zhengzhou	Fluvisols	Warm, semi-humid, temperate monsoon	1982-2003	35			Fu et al., 20	L删除: Hunan E版用户 1/25/2015 2:44 PM L删除: Haplic alisols
	Taoyuan	Haplic alisols	s Humid, mid-subtropic al monsoon	1979-2003	32			Liu et al., 20	E版用户 1/25/2015 2:45 PM 品删除: Humid, E版用户 1/25/2015 2:45 PM
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	Pixian	Haplic	Sub-tropical	1981-2002	9.0		Wei et al.,	2004		
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692	Figure captions
693	
694	Figure 1 Evolution of SOC content and density for cropland from 1982 to 2011 in
695	Huantai County. Error bars are highlighted only for the years which soil survey is
696	undertaken.
697	
698	Figure 2 Change of SOC stock for farmland (cropland, vegetable land, farmland
699	converted to construction land) from 1982 to 2011 in Huantai County.
700	
701	Figure 3 Regression analysis of driving factors with the years in Huantai. a)
702	temperature vs. year, b) yield vs. year, and c) carbon input vs. year.







