### Response to comment of BGD-12-C182-2015

### Comment of Referee #1:

The research implemented a very unique N2O sampling network consisting of 147 sampling locations on a single field of 4 ha of tea. This level of sampling requires an extraordinary effort, and the data so obtained are of unique value for evaluating spatial variability of N2O emissions from agricultural soils. The manuscript is worth publishing.

### Author's Response:

We highly appreciated the encouragement made by the referee #1.

## Author's Changes to the Manuscript:

No change.

### Response to comment of BGD-12-C194-2015

### Comment of Referee #2:

This paper by Fu et al. reports a study on the spatial variability of N2O emissions from a tea field during the wet season in subtropical China. The authors measured N2O emissions using 147 chambers and applied three spatial interpolation methods to estimate the spatial distribution of the emissions. The findings highlight the importance of selecting an appropriate spatial interpolation method to provide a reliable estimation of regional N2O emissions.

The paper makes a contribution to knowledge. The methodology is sound and the conclusions are supported by the results. The paper is well written and deserves publication.

Just one minor edit: P.1488, L14: change "The" to "the".

#### Author's Response:

We highly appreciated the encouragement made by the referee #2, and many thanks for the minor comment. We made the correction in the revised manuscript.

## Author's Changes to the Manuscript:

See change in P.18, L377 in the revised manuscript.

### Responses to comments of BGD-12-C456-2015

### Comment I of the Editor:

- I.1) Please elaborate in detail the major novelty of this paper compared to the study from Li et al., 2013. Simply referring to dry season in one paper and to wet season in the other is not sufficient.
- I.2) In addition, comparing dry and wet season should be done carefully as half an hour sampling cannot represent an entire season.
- I.3) It is suggested in the abstract that soil properties should be included in interpolation methods. To my opinion, such an achievement would be required before final publication.

### Author's Response:

I.1) In subtropical central China, the temperature and precipitation are very different between wet season and dry season as well as subsequent field management, e.g., fertilization and weeding, which may result in strong seasonal variations of  $N_2O$  emissions from tea-planted soils with totally different spatial structures and controlling factors. Therefore, this wet season study is a companion investigation of the dry season study (Li et al., 2013) for looking at the spatial variability of  $N_2O$  emissions from tea field soils.

Compared to Li et al. (2013), the major novelties in this study may include the following three aspects. Firstly, the sampling chambers were redesigned (please see P8-9 L162-168 of Section 2.3 in the revised manuscript) to be more practical. Basically, the bases and the sampling chambers are separated. When in field operation, the chambers are clipped on the bases with sponge seals in between to stop the gas leaking. Thus, the volume of each sampling chamber is kept almost the same. Secondly, the sampling grids of this study (15 m S-N x 15 m E-W) were slightly denser than the dry-season study (15 m S-N x 20 m E-W). Finally, although we found that the spatial structures of N<sub>2</sub>O emissions were comparable during both the dry and wet seasons (with an effective range of approximately 28.0 m and 25.3 m, respectively), the influencing factors of N<sub>2</sub>O emissions were quite different, of which soil properties (soil ammonium, soil nitrate and soil organic carbon) and elevation had significant impacts on N<sub>2</sub>O emissions during the wet and dry seasons, respectively. In addition, the total amount of N<sub>2</sub>O emissions in the wet season was almost seven times

more than that during the dry season for a 30-min snapshot (10:00-10:30 a.m.) over the studied 4.0 ha tea-planted catchment.

I.2) Yes, we agreed that a 30-min measurement cannot fully represent the  $N_2O$  emissions for an entire season. However, the main objective of this study was to investigate the spatial structure and controlling factors of  $N_2O$  emissions from tea-planted soil, while the seasonal variation of the amounts of  $N_2O$  emissions was regarded important, but less concerned.

I.3) Yes, we totally agreed. In the cokriging interpolation approach, soil properties such as soil organic carbon, soil ammonium-N and soil nitrate-N (which had significant correlations with  $N_2O$  fluxes) were chosen as the co-variables for estimating the spatial distribution of  $N_2O$  emissions (see Sections 3.3 and 4.3 in the revised manuscript).

### Author's Changes to the Manuscript:

No change.

### Comment II of the Editor:

II) The paper should refer to and discuss process-based biogeochemical modeling approaches (e.g., DNDC). Which implication do the results of the paper have for existing models and process understanding (see e.g., Butterbach-Bahl et al., 2013)?

## Author's Response:

The main objective of this paper was to explore the spatial structure and controlling factors of  $N_2O$  emissions from tea-planted soils over a catchment. The outcomes of

such a research may help us to design the static chamber positioning scheme when a continuous measurement of  $N_2O$  emissions from tea fields is carried out, and provide a more accurate estimation of  $N_2O$  emissions at regional scales. Although the main controlling factors of  $N_2O$  emissions from tea fields found in this study were helpful for the relationship development, there were few direct contributions to understand the biogeochemical processes simulated in the ecosystem models such as DNDC and DAYCENT.

## Author's Changes to the Manuscript:

No change.

### Comment III of the Editor:

III) Provide more details on the size of the static chamber (e.g., volume). Please also explain why the chamber volumes were not ventilated (air mixing to avoid concentration gradients)? Could this potentially cause random sampling errors that have influenced the spatial distribution of the emissions?

### Author's Response:

The static chambers were divided into two parts: base and chamber. The base was 0.15 m in diameter and 0.05 m high. The chamber was 0.15 m in diameter and 0.15 m high. In the field operation, the base was gently inserted vertically into the soil, and the chamber was clipped on the base with the sponge seals in between to stop gas leaking. Therefore, the effective static chambers volume was equal to the chamber volume of 0.002651 m<sup>3</sup>.

Theoretically, these static chambers should be ventilated for balancing air pressure at operation. However, our pre-experiment of gas sampling had proven that the ventilation of the chamber had little impact on the N<sub>2</sub>O concentration gradients during a short incubation time period of 30-min. Therefore, to avoid the unnecessary complexity of the gas sampling chamber, the ventilation device was not applied in our study.

### Author's Changes to the Manuscript:

please see changes in P.8-9, L162-171 of Section 2.3 in the revised manuscript.

### Comment IV of the Editor:

IV) It would be important to see the spatial distribution of the error of the interpolation (kriging variance) on a map.

# Author's Response:

Yes, we agreed. A new kriging variance map (Fig. 10) for Fig. 9 was produced and its relevant description was also made in the revised manuscript.

### Author's Changes to the Manuscript:

The new kriging variance map (Fig. 10) was added in P.49 in the revised manuscript, and the relevant description was added in P.14, L293-294 in Section 3.3 in the revised manuscript. The map caption was also added in P.39 L674-680.

1 Wet-season spatial variability of N<sub>2</sub>O emissions from a tea field in subtropical central China 2 Xiaoqing Fu, Xinliang Liu, Yong Li \*, Jianlin Shen, Yi Wang, Ganghua Zou, Hang Li, Lifang 3 Song, Jinshui Wu 4 5 Changsha Research Station for Agricultural & Environmental Monitoring and 6 Key Laboratory of Agro-ecological Processes in Subtropical Regions, 7 Institute of Subtropical Agriculture, Chinese Academy of Sciences, 8 9 Hunan 410125, China These two authors contributed equally to this work. 10 \*Correspondence to: Professor Yong Li 11 Institute of Subtropical Agriculture, 12 Chinese Academy of Sciences, Hunan 410125, China 13 Tel: +86-731-8461-5291 14 Fax: +86-731-8461-2685 15 16 E-mail: yli@isa.ac.cn

#### Abstract

18

Tea fields emit large amounts of nitrous oxide (N<sub>2</sub>O) to the atmosphere. Obtaining accurate 19 estimations of N<sub>2</sub>O emissions from tea-planted soils is challenging due to strong spatial 20 variability. We examined the spatial variability of N<sub>2</sub>O emissions from a red-soil tea field in 21 Hunan province, China, on 22 April 2012 (in a wet season) using 147 static mini chambers 22 approximately regular gridded in a 4.0 ha tea field. The N<sub>2</sub>O fluxes for a 30-min snapshot 23 (10:00-10:30 am) ranged from -1.73 to 1,659.11 g N ha<sup>-1</sup> d<sup>-1</sup> and were positively skewed with 24 an average flux of 102.24 g N ha<sup>-1</sup>d<sup>-1</sup>. The N<sub>2</sub>O flux data were transformed to a normal 25 distribution by using a logit function. The geostatistical analyses of our data indicated that the 26 logit-transformed N<sub>2</sub>O fluxes (FLUX30t) exhibited strong spatial autocorrelation, which was 27 characterized by an exponential semivariogram model with an effective range of 25.2 m. As 28 observed in the wet season, the logit-transformed soil ammonium-N (NH4Nt), soil nitrate-N 29 30 (NO3Nt), soil organic carbon (SOCt), total soil nitrogen (TSNt) were all found to be significantly correlated with FLUX30t (r=0.57-0.71, p<0.001). Three spatial interpolation 31 methods (ordinary kriging, regression kriging and cokriging) were applied to estimate the 32 33 spatial distribution of N<sub>2</sub>O emissions over the study area. Cokriging with NH4Nt and NO3Nt as covariables (r=0.74 and RMSE=1.18) outperformed ordinary kriging (r=0.18 and 34 RMSE=1.74), regression kriging with the sample position as a predictor (r=0.49 and 35 RMSE=1.55) and cokriging with SOCt as a covariable (r=0.58 and RMSE=1.44). The 36 predictions of the three kriging interpolation methods for the total N<sub>2</sub>O emissions of 4.0 ha 37 tea field ranged from 148.2 to 208.1 g N d<sup>-1</sup>, based on the 30 min snapshots obtained during 38

the wet season. Our findings suggested that to accurately estimate the total  $N_2O$  emissions

over a region, the environmental variables (e.g., soil properties) and the current land use

pattern (e.g., tea row transects in the present study) must be included in spatial interpolation.

Additionally, compared with other kriging approaches, the cokriging prediction approach

showed great advantages in being easily deployed, and more importantly providing accurate

regional estimation of N<sub>2</sub>O emissions from tea-planted soils.

### Introduction

According to the latest data, which show rapid increases in their atmospheric concentrations (IPCC, 2013), nitrous oxide (N<sub>2</sub>O), carbon dioxide (CO<sub>2</sub>) and methane (CH<sub>4</sub>) are three major greenhouse gases in the atmosphere that significantly contribute to global warming. Among these major greenhouse gases, N<sub>2</sub>O has a very high radiative forcing per unit mass (265-fold stronger than CO<sub>2</sub> on a 100 year horizon) and plays an important role in ozone depletion in the stratosphere (Ravishankara et al., 2009). The primary sources of N<sub>2</sub>O are from agriculture development and the subsequent increased use of chemical N fertilizers (Ambus and Christensen, 1994; Mosier et al., 1996, 1998; Yanai et al., 2003; Tokuda and Hayatsu, 2004; Akiyama et al., 2006; Ravishankara et al., 2009). Agricultural soils produce 2.8 (1.7–4.8) Tg of N<sub>2</sub>O-N yr<sup>-1</sup> (IPCC, 2013). The N<sub>2</sub>O is emitted from soils via the microbial processes of nitrification under aerobic conditions and denitrification under anaerobic conditions (Firestone and Davidson, 1989; Wrage et al., 2004). The magnitude of soil N<sub>2</sub>O emissions is highly variable and strongly influenced by changes in environmental conditions.

Among the different agricultural soils, tea-planted soils are important sources of N<sub>2</sub>O that are rapidly attracting attention due to recent large increases in the number of tea plantations and large N fertilizer inputs (Akiyama et al., 2006; Lin and Han, 2009; Fu et al., 2010, 2012; Hirono and Nonaka, 2012; Han et al., 2013; Li et al., 2013). In China, the total tea-planted area was approximately 2.10 million ha (mostly distributed in Fujian, Anhui, Zhejiang and Hunan) in 2013 (NBSC, 2014). Compared with other agricultural soils, tea-planted soils provide optimal conditions (e.g., low soil pH, high temperature and ample moisture) for microbes to emit significant amounts of N<sub>2</sub>O (Hayatsu, 1993; Venterea and Rolston, 2000; Li et al., 2013). However, because few measurements of N<sub>2</sub>O emissions from tea-planted soils have been reported in China (Fu et al., 2012; Li et al., 2013; Han et al., 2013), it is difficult to conduct precise spatial and temporal evaluations of N<sub>2</sub>O emissions from tea-planted soils. To estimate the N<sub>2</sub>O emissions from tea-planted soils accurately and to understand the roles that tea plantations play in global warming, it is necessary to investigate the spatial and temporal patterns and related mechanisms of N<sub>2</sub>O emissions from tea fields. This information will lead to the development of effective land management options for mitigating N<sub>2</sub>O emissions from a significant source, tea plantation. The N<sub>2</sub>O fluxes have large spatial variability in agricultural soils (Konda et al., 2008, 2010; Meda et al., 2012; Li et al., 2013). Many previous studies in tea fields have found pronounced seasonal fluctuations in N<sub>2</sub>O fluxes, with higher N<sub>2</sub>O emissions during the wet season than during the dry season (Fu et al., 2012; Han et al., 2013). The seasonal and spatial variability of N<sub>2</sub>O emissions significantly contributes to the uncertainty when estimating the

60

61

62

63

64

65

66

67

68

69

70

71

72

73

74

75

76

77

78

79

contributions of subtropical tea-planted ecosystems to N<sub>2</sub>O flux. Moreover, most of our knowledge regarding seasonal changes and the spatial variability of N<sub>2</sub>O fluxes is based on a small number of measurements taken from tea-planted soils. Li et al. (2013) investigated the spatial structure of N<sub>2</sub>O fluxes for tea-planted soils during the dry season in October 2010 and found that the spatial distribution of the N<sub>2</sub>O fluxes was primarily associated with field elevation (r=-0.42, p<0.001). The other soil properties (e.g., soil organic carbon, soil water and soil mineral nitrogen) were not significantly related to N<sub>2</sub>O flux. To obtain a more accurate evaluation of annual N<sub>2</sub>O emissions from tea-planted soils, a study on the spatial structure and distribution of N<sub>2</sub>O emissions during a wet season (in contrast to the dry season) is necessary. To understand the structure of the spatially distributed data and to predict the N<sub>2</sub>O fluxes at the unsampled locations, geostatistical analyses can be useful (Goovaerts, 1997; Webster and Oliver, 2001). Geostatistics provide statistical tools for describing the quantitative spatial variability of field observations for the accurate mapping and planning of rational sampling schemes that efficiently utilize the available labor (Webster, 1985). Several geostatistical methods are used to examine the spatial variability of N<sub>2</sub>O fluxes, including simple kriging (SK), ordinary kriging (OK), regression kriging (RK) and cokriging (CK). The most commonly used method is OK (Clemens et al., 1999; Röver et al., 1999; Mathieu et al., 2006; Konda et al., 2008, 2010), which uses the derived theoretical semivariogram models to interpolate the spatial distribution of N<sub>2</sub>O fluxes. However, research has demonstrated that RK and CK approaches, which use related auxiliary variables, improve the prediction

81

82

83

84

85

86

87

88

89

90

91

92

93

94

95

96

97

98

99

100

accuracy (Goovaerts, 1997; Webster and Oliver, 2001; Hengl et al., 2004). The RK method combines multiple regressions, including linear regressions, generalized linear models, generalized added models and regression tree models, with the auxiliary variables used for kriging (Odeh et al., 1994). In the RK method, linear regressions are commonly used. The CK approach uses correlations that may exist between the predicted variables and other more easily measured variables. These variables can be measured at the same points as the predicted variable, at other points, or at both. Compared with the RK approach, the CK approach is commonly applied when the measurement of a covariable is less expensive than the cost of a predicted variable (Stein et al., 1988; Odeh et al., 1995). In addition to the feature correlation as a criterion for selecting covariables, the CK approach also requires that both of the predicted variable and covariables have similar spatial structures (Odeh et al., 1994). In this study, we used three interpolation methods (OK, RK and CK) to estimate the spatial distribution of N<sub>2</sub>O fluxes in a tea field. In contrast with the dry season, the spatial variability of the N<sub>2</sub>O emissions was investigated during the wet season in April 2012 from the same tea-planted catchment that was studied by Li et al. (2013). The catchment consisted of a completely independent hydrological system. Thus, the spatial distribution of the N<sub>2</sub>O emissions within the catchment was expected to have intrinsic characteristics. The objectives of this study were to (i) evaluate the spatial variability of N<sub>2</sub>O emissions from soils planted with tea in subtropical central China during the wet season, (ii) determine the key environmental factors controlling N<sub>2</sub>O

emissions, and (iii) assess the prediction efficiency of three kriging interpolation methods.

102

103

104

105

106

107

108

109

110

111

112

113

114

115

116

117

118

119

120

121

#### 2. Materials and Methods

2.1 Site description

The field experiment was conducted in a small catchment (4.0 ha) in Jinjing, Changsha, in Hunan province, China (28°32′50″N and 113°19′58″ E and elevation 90 to 111 m) (Fig. 1). The region has a subtropical monsoon climate with a mean annual air temperature of 17.5°C and a mean annual precipitation of 1400 mm (average from 1979 to 2012). The site had four distinct seasons: spring (February to April), summer (May to July), autumn (August to November), and winter (December to January). On average, 70% of the annual precipitation occurred in April, May and June. The daily air temperature and precipitation for 2012 were recorded by an automatic weather station (Intelimet A, IMET-ADV2, Dynamax, USA) located next to the studied catchment (Fig. 2). The soil of the catchment was a Haplic Alisol (FAO/UNESCO soil taxonomy) that was derived from a granitic parental material. Tea (Camellia sinensis L., cv. Baihaozao) was contour-planted 10 years ago using an inter-row spacing of 0.5 m in the catchment.

<Insert Figs. 1 & 2 near here>

### 2.2 Sampling positions

In the 4.0 ha tea-planted catchment, 1964 evenly-distributed points with plane coordinates and elevation values and 456 centerlines of tea tree row were recorded by locally calibrated differential Geographic Positioning System (DGPS) receiver (Sanding Southern Survey Co.,

China), and then were used to develop the local DEM and land use data (at a spatial resolution of 0.1 m, respectively, as shown in Fig. 1c and d). The land use data showed the four positions where the chambers were placed, including the inter-row, fertilization point, under tea tree and in tea tree row, as described in Li. et al. (2013). The spatial positions of the gas sampling points in a 15 m × 15 m regular grid over the catchment were originally determined using a DGPS receiver on 20 April 2012. Some of the chamber positions were slightly adjusted (because of a lack of space in the tea tree rows or to avoid roads and trenches). Thus, the chambers were placed in one of four locations mentioned above (Fig. 1d). Overall, 147 sampling points were determined, and the Euclidean distances between each point and its nearest neighbors ranged from 14.6 m to 16.7 m. The x-y coordinates, the gas sampling position information (the inter-row, fertilization point, under tea tree and in tea tree row along tea row transects), and the elevations at the sampling points were recorded.

# 2.3 Gas and soil properties measurements

Gas and soil samples were collected at each grid point on 22 April 2012 using a closed mini chamber technique. A mini chamber set was composed of PVC and had two parts (base and chamber). The base was 0.15 m in diameter and 0.05 m high. The chamber was 0.15 m in diameter and 0.15 m high, and was equipped with rubber septa on the top for gas sampling. In the field operation, the base was gently inserted vertically into the soil on 20 April 2012, and the chamber was clipped on the base with the sponge seals in between to stop gas leaking before gas sampling on 22 April 2012. Therefore, the effective static chambers volume was

equal to the chamber volume of 0.002651 m<sup>3</sup>. The mini chambers were composed of PVC and had two parts (base and lid). The bases were 0.15 m tall and were gently inserted into the soilvertically to a depth of 0.05 m on 20 April 2012. The lids with rubber septa for gas sampling were screwed onto the bases when the gas was sampled on 22 April 2012. Gas samples were collected from the headspace between 10:00 and 10:30 am. For simultaneous sampling, 25 skilled gas sampling persons helped to accomplish the field sampling. Each person only took care of one column containing 4 to 8 sampling positions (see Fig. 1), and started sampling at the same time of 10 am. At each point, three gas sample replicates were collected from the headspace into pre-evacuated 12 mL vials (Exetainers, Labco, UK) at 0 and 30 min after the chamber body was clipped. After collecting the gas samples, the air temperature in each chamber was measured for subsequent correction of the flux calculation, and then three replicate soil cores, 0.05 m in diameter and 0.20 m in depth, were collected from the soils inside the mini chambers. Soil samples were put straight into clean zip-lock bags for avoiding soil moisture loss, and quickly transported back to the laboratory in thermal insulation boxes and stored in a refrigeration room at 4 °C for preventing any microbial activity (such as mineralization, nitrification and denitrification). The N<sub>2</sub>O concentrations of the gas samples were analyzed using a gas chromatograph (Agilent 7890A, Agilent, USA) that was fit with a <sup>63</sup>Ni-electron capture detector and an automatic sample injector system. The N<sub>2</sub>O fluxes (FLUX30, g N ha<sup>-1</sup> d<sup>-1</sup>) were calculated as described by Li et al. (2013). The soil physical/chemical properties determined by using fresh soil, e.g., the soil ammonium content (NH4N), soil nitrate content (NO3N), soil dissolved organic carbon content (DOC), soil

166

167

168

169

170

171

172

173

174

175

176

177

178

179

180

181

182

183

184

185

volumetric water content (SWC), and soil bulk density (BD), were measured within three days after sampling, while those using air-dried soil, e.g., total soil nitrogen content (TSN), soil organic carbon content (SOC) and soil clay/silt/sand content (CLAY, SILT and SAND), were determined within two weeks after the field work.

### 2.4 Data analyses

The descriptive statistical and geostatistical analyses were performed using R (R Development Core Team, 2014) with the gstat package (DGUU, 2010).

Descriptive statistical analyses were used to determine the mean, median, minimum and maximum values, SD, coefficient of variation (CV), and skewness of the original and logit-transformed data. These analyses were based on the four chamber placement positions. Because the FLUX30, NH4N, NO3N, SOC, TSN and SWC data were highly skewed, these values were transformed by using a logit function (Hengl et al., 2004). The transformed variables were named FLUX30t, NH4Nt, NO3Nt, SOCt, TSNt and SWCt. Using a Pearson's correlation, the relationships between FLUX30t, NH4Nt, NO3Nt, SOCt, TSNt, SWCt, DOC, BD, SAND, SILT, and CLAY were tested. The significance of the differences in the FLUX30t and environmental factors (NH4Nt, NO3Nt, SOCt, TSNt and DOC) between any two of the different chamber positions along the entire tea-tree row transect were evaluated using the Tukey's Honest Significant Difference method.

In the geostatistical analyses, an experimental semivariogram of FLUX30t was calculated, and the theoretical semivariogram models were fit. The ratio of the partial sill to

the total sill was used as an index of spatial dependence. Armstrong (1998) stated that a variable with a higher ratio of partial sill to sill and a longer semivariogram range were more structured. The spatial distribution of FLUX30t across the catchment was predicted using three kriging interpolation methods (OK, RK and CK). These data were transformed back to the original scale of FLUX30 for mapping. The Leave-One-Out cross-validation method was used to evaluate the accuracy of interpolating FLUX30t using the three different kriging methods.

## 3. Results

3.1 Exploratory data analyses

In the 4.0 ha tea-planted catchment, the  $N_2O$  fluxes during the 30-min one-time measurements performed on 22 April 2012 ranged from -1.73 to 1,659.11 g N ha<sup>-1</sup> d<sup>-1</sup>, with a median value of 27.56 g N ha<sup>-1</sup> d<sup>-1</sup> and a CV of 234.7 % (Table 1). The  $N_2O$  flux data were positively skewed (Table 1 and Fig. 3a), and their logit-transformations were approximately normally distributed (Table 1 and Fig. 3b). From Table 3, the logit-transformed  $N_2O$  fluxes (FLUX30t) were the highest in the fertilization points, and the differences in the FLUX30t values among the chamber placement positions were statistically significant (p < 0.001).

< Insert Table 1 & Fig. 3 near here>

The ELEVATION, BD, DOC, SWC, SAND, SILT, and CLAY were approximately normally distributed, with skewness values of less than 1 (Table 1). Additionally, DOC displayed a moderate CV of 34.6 %, and the other variables had lower CVs (4.1–23.8 %). The NH4N, NO3N, SOC and TSN were positively skewed, and the logit-transformations (NH4Nt, NO3Nt, SOCt and TSNt) had approximately normal distributions (Table 1). The NH4N and NO3N had very high CVs (190.8 % and 141.6 %, respectively), and the SOC and TSN had moderate CVs (50.1 % and 38.3 %, respectively).

The NH4Nt, NO3Nt, SOCt, TSNt and SWC were significantly correlated with the  $N_2O$  fluxes (Fig. 5), and the NH4Nt, NO3Nt and TSNt had strong positive relationships with  $N_2O$  (r = 0.71, 0.70 and 0.57, respectively, p < 0.001). The  $N_2O$  emissions and some soil properties (NH4N, NO3N, SOC, TSN and SWC) in the fertilization points were significantly different (p < 0.001) from the other three chamber placement positions (Fig. 6). These variables were used as auxiliary covariables for the CK approach.

### <Insert Figs. 4 & 5 near here>

3.2 Spatial variability of  $N_2O$  emissions and related environmental factors

Because most of the soil properties were significantly correlated with the chamber placement positions, two types of semivariogram models were calculated for the N<sub>2</sub>O and soil parameters (correlated with N<sub>2</sub>O fluxes) in the wet season (Table 2). The FLUX30t exhibited strong spatial autocorrelation and was characterized by an exponential semivariogram model,

a theoretical distance parameter of 8.40 m (equivalent to an effective range of 25.2 m) and a zero nugget. The NH4Nt, SWCt, SAND and SILT showed almost no spatial dependency, while NO3Nt and TSNt demonstrated weak spatial dependency with a range parameter of 91.9 and 58.0 m, respectively (equivalent to an effective range of 163.7 and 102.6 m, respectively). The SOCt exhibited a moderate spatial dependency within 93.0 m. By detrending the influence of the chamber placement position, large changes in the semivariogram models occurred regarding the above variables. Although the semivariograms of the regression residuals of FLUX30t, NH4Nt, NO3Nt and SOCt were best-fit with the same semivariogram model (exponential) with a similar range of 17.4 m (equivalent to an effective range of 52.1 m), the spatial dependencies of those variables were different (Table 2). Of the soil properties, only SOCt had a similar spatial structure to FLUX30t when the influence of the chamber placement position was detrended (Table 2). Based on these correlation analyses and spatial variability analyses, the covariables for the CK method were determined.

### <Insert Table 2 near here>

3.3 Spatial interpolation of  $N_2O$  emissions by three methods

Three spatial interpolation methods were used in this study to predict the spatial distribution of  $N_2O$  emissions from tea soils in the catchment. In the first method, the derived theoretical semivariogram model for FLUX30t that is presented in Table 2 was used for the OK

prediction. In the second method, RK was used and the chamber placement position was identified as the auxiliary regression predictor. Thus, the semivariogram of the regression residuals of FLUX30t were calculated and best-fit with the theoretical semivariogram model shown in Fig. 6. In the third method, CK involved two groups of covariables. As described previously, because SOCt (detrending the influence of chamber placement position) showed a similar spatial structure to FLUX30t (detrending the influence of chamber placing position), a CK process was performed using SOCt as the covariable. Firstly, the direct and cross-semivariograms of FLUX30t and SOCt (detrending the influence of the chamber placement position) were calculated and best-fit with a linear model for co-regionalization (LMC). Next, the fitted LMC was used to predict the spatial surface of N<sub>2</sub>O emissions. Because NH4Nt and NO3Nt were significantly correlated with FLUX30t (Fig. 5), a second CK with NH4Nt and NO3Nt as the covariables was processed, similarly to that of the CK with SOCt. However, these covariables had different spatial structures (Table 2). As reflected by the lower root mean squared error (RMSE) and higher r values (Table 4), the CK method performed better than the other spatial interpolation methods. Furthermore, the CK with NH4Nt and NO3Nt as two covariables outperformed the CK with SOCt as the covariable.

286

270

271

272

273

274

275

276

277

278

279

280

281

282

283

284

285

# <Insert Figs.6-9 near here>

288

289

290

287

As shown in Fig. 9, the surface map for the spatial distribution of N<sub>2</sub>O emissions interpolated by OK was rougher than the maps obtained from the other interpolation

approaches. The kriging error maps were showed in Fig. 10, also indicating the CK method outperforming the other spatial interpolation methods. The four kriging interpolations of OK, RK, CK with SOCt as the covariable and CK with NH4Nt and NO3Nt as the covariables were able to predict that the total amount of N<sub>2</sub>O emissions in the tea fields during the wet season were 208.1 g N d<sup>-1</sup>, 148.2 g N d<sup>-1</sup>, 149.7 g N d<sup>-1</sup> and 150.5 g N d<sup>-1</sup>, respectively. From the performance evaluations of the four spatial interpolations, the total N<sub>2</sub>O emissions from the tea field on 22 April 2012 during the wet season were approximately 150 g N d<sup>-1</sup>.

## <Insert Fig. 10 near here>

### Discussion

4.1 Seasonal differences of  $N_2O$  fluxes in the red soil planted with tea

The N<sub>2</sub>O emissions from soils have obvious seasonal fluctuations, with emissions that are significantly higher during the wet season than during the dry season (Konda et al., 2010). To understand the seasonal changes in the spatial structures of N<sub>2</sub>O fluxes, we compared the N<sub>2</sub>O emissions between the wet (this study) and dry (Li et al., 2013) seasons. In general, the mean, SD and coefficient of variation (102.24, 239.96 g N ha<sup>-1</sup> d<sup>-1</sup> and 234.7 %, respectively) of the N<sub>2</sub>O fluxes in the wet season were all higher than those (2.88, 8.94 g N ha<sup>-1</sup> d<sup>-1</sup> and 152.0 %, respectively) during the dry season (Table 3). Furthermore, in contrast with the dry season, the N<sub>2</sub>O fluxes during the wet season were significantly different among the four chamber placement positions, with the highest fluxes occurring at the fertilization points and the inter-row positions (Table 3). During the wet season, the high N<sub>2</sub>O fluxes at the

fertilization points and the inter-row positions resulted from the high soil moisture, due to more rainfall, and from the fertilization that occurred on 19 February 2012 (Fig. 2). The soil N and the soil organic C availability are directly increased by the application of chemical and organic N fertilizers. The additional in the available C and N supplied by fertilization resulted in increased soil microbial activity, which stimulated the nitrification and denitrification processes that contribute to soil N<sub>2</sub>O emissions (Davidson et al., 1993; Kiese et al., 2003; Werner et al., 2007).

### <Insert Table 3 near here>

4.2 Spatial structure of N<sub>2</sub>O emissions from red soils planted with tea

Soil type, topography and land management (fertilization, tillage and irrigation) are the primary factors that affect the spatial structures of N<sub>2</sub>O emissions (Folorunso and Rolston, 1984; Clemens et al., 1990; Velthof et al., 1996; Konda et al., 2008). During the wet season, the N<sub>2</sub>O fluxes showed a strong spatial dependence (with a range of approximately 25.3 m) that was similar to the dry season range of approximately 28.0 m in the tea-planted fields (Li et al., 2013). These results indicated that the spatial dependence of N<sub>2</sub>O fluxes at the current spatial sampling scale was comparable between seasons. Our findings for a fertilized tea field were similar to those of Konda et al. (2010) for a tropical forest. However, these results contrasted those of many previous investigations for agricultural fields, including winter wheat (Ball et al., 1997; Clemens et al., 1999; Röver et al., 1999; Mathieu et al., 2006),

summer maize (Clements et al., 1999), onion (Yanai et al., 2003), and grassland (Ambus and Christensen, 1994; Velthof et al., 1996; van den Pol-van Dasselaar et al., 1998; Turner et al., 2008) fields, in which the N<sub>2</sub>O flux presented no, weak or moderate spatial dependence. This discrepancy primarily occurred because of the unique geographical characterization and land management of the tea plantation. Compared with other agricultural fields in flat areas, tea fields are always distributed in hills or mountains. Therefore, the contributions of the topography to the spatial dependence of the N<sub>2</sub>O flux were strong (Li et al., 2013). Additionally, tea is a perennial plant. Thus, apart from fertilization and weeding, the soil disturbance in tea fields is always very low. During the dry season, the topography (elevation) had a significant effect on the spatial pattern of N<sub>2</sub>O fluxes in the tea-planted fields (Li et al., 2013). Similar spatial patterns of N<sub>2</sub>O fluxes with topography were also observed in forest soils (Van Kessel et al., 1993; Konda et al., 2010). Theoretically, the SWC varies with the topography and affects the spatial pattern of N<sub>2</sub>O fluxes by controlling the conditions for soil nitrification and denitrification (Firestone and Davidson, 1989; Wrage et al., 2004). Although the SWC had no relationships with N<sub>2</sub>O and elevation during the dry season (Li et al., 2013), a correlation existed in the present study (Fig. 5). The microstructures of the tea tree-row transect and the land management practices of tea production were the primary influences on the spatial pattern of soil water in the tea-planted fields (Li et al., 2013). During the wet season, fertilization contributed to the spatial pattern of N<sub>2</sub>O fluxes in the tea-planted fields, with the highest averaged fluxes at the fertilization sites (198.81 g N ha<sup>-1</sup> d<sup>-1</sup>) (Table 3). Fertilization resulted in similar spatial

334

335

336

337

338

339

340

341

342

343

344

345

346

347

348

349

350

351

352

353

patterns of N<sub>2</sub>O fluxes in other agricultural soils (Ball et al., 1997; Clements et al., 1999;

Röver et al., 1999; Mathieu et al., 2006; Yanai et al., 2003).

In view of the analysis of the primary factors that affected the spatial pattern of  $N_2O$  fluxes, we detrended the influences of the environmental factors when the  $N_2O$  flux semivariograms were calculated to more deeply explore the spatial structures of the  $N_2O$  emissions in the tea-planted fields. For example, during the dry and wet seasons, the spatial influences of elevation (Li et al., 2013) and chamber placement position, respectively, were detrended when computing the  $N_2O$  flux semivariograms. Because the relationship between chamber placement position and  $N_2O$  flux was more relevant than the relationship between elevation and  $N_2O$  flux, the effect of detrending the influence of chamber placement position during the wet season was more obvious than that of detrending the influence of elevation during the dry season (Li et al., 2013). This effect was also reflected in the evaluation of the performance of the RK method for the wet and dry seasons (Table 4).

4.3 Spatial interpolations of  $N_2O$  emissions by three methods

The three interpolation methods (OK, RK and CK) were used to predict the spatial distributions of N<sub>2</sub>O emissions from the red soils planted with tea during dry (Li et al., 2013) and wet seasons (this study). However, these three methods resulted in significantly different performances between the dry and wet seasons (Table 4). We conducted comparative analyses for the performance of the three interpolation methods using two aspects: different seasons and different methods. Firstly, **T**the OK method performed better when predicting the

spatial distribution of N<sub>2</sub>O fluxes for the dry season relative to the wet season. Because the OK method directly used the fitted theoretical semivariogram model of the target variable to predict the spatial distribution, its performance reflected the predictive ability of the original data (Goovaerts, 1997). During the wet season, more factors (e.g., NH4N, NO3N, SOC, TSN and SWC) influenced the spatial distributions of the N<sub>2</sub>O fluxes than the dry season (Table 2 and Fig. 5). The values of the original data were concealed. Thus, other sophisticated kriging methods, such as RK and CK, which reconcile the relationships between N<sub>2</sub>O fluxes and environmental factors, could be useful. The RK method performed better when elevation was used as an auxiliary regression predictor during the dry season than when the chamber placement position was used during the wet season (Table 4). This finding primarily occurred because the chamber placement position was a categorical variable with a lower regression fitting ability than elevation, which was a continuous variable (Goovaerts, 1997). The performances of the CK with two groups of covariables during the wet season were better than those of the CK with three groups of covariables during the dry season (Table 4). Particularly, the CK with strongly correlated covariables of NO3N and NH4N (r = 0.70-0.71and p < 0.001) (Fig. 5) performed the best (r = 0.74 and RMSE = 1.04) (Table 4). Secondly, by comparing the performances of the three interpolation methods, the RK and CK methods, which are more sophisticated kriging technologies, performed better than the OK method for the dry and wet seasons. Similar results were obtained by previous researchers (Stein et al., 1988; Odeh et al., 1995; Goovaerts, 1997; Hengl et al., 2004). When comparing the performances of RK and CK, no differences were observed for the dry season.

376

377

378

379

380

381

382

383

384

385

386

387

388

389

390

391

392

393

394

395

However, during the wet season, the CK significantly outperformed the RK (Table 4). Overall, few attempts have been made to provide a good method for selecting interpolation methods between RK and CK (Kontters et al., 1995; Odeh et al. 1995). Li et al. (2013) suggested that RK was a good choice because of the performance of the two interpolation methods and the difficulties encountered when applying CK. However, in this study, the CK method was better than the RK method because of its high predictive performance (Table 4), its readily available required covariables (e.g., NH4N, NO3N and SOC) at co-locations, and because expensive surface data were not needed (e.g., DEM and land use data, which are required by RK) (Goovaerts, 1997; Webster and Oliver, 2001). Our conclusions were similar to those of many previous studies that found that CK was the most versatile and rigorous statistical technique for estimating spatial points (Stein et al., 1988; Odeh et al., 1995; Webster and Oliver, 2001). For the application of CK, the covariables must show a correlation with the target variable and present a similar spatial structure as the target variable (Odeh et al., 1995; Goovaerts, 1997; Webster and Oliver, 2001). Therefore, we further compared the effects of the two groups of covariables for CK in this study. We found that CK method with NH4Nt and NO3Nt (showed significant correlations with FLUX30t) as covariables outperformed the CK method with SOCt (presented a similar spatial structure to FLUX30t) as a covariable, indicating that the feature correlation was more important than the similarity of the spatial structure when selecting CK covariables. This finding can be regarded as a prerequisite for selecting covariables for CK application.

417

397

398

399

400

401

402

403

404

405

406

407

408

409

410

411

412

413

414

415

The three spatial interpolation methods predicted similar total N<sub>2</sub>O emissions from the tea-planted red soils in the 4.0 ha catchment on 30 October 2010 (in the dry season) and on 22 April 2012 (in the wet season), ranging from 21.2 to 22.1 g N d<sup>-1</sup> and from 148.2 to 208.1 g N d<sup>-1</sup> (Table 4), respectively. The predicted errors during the wet season were higher than those of the dry season (Table 4). This result mainly occurred because fertilization was a major factor that affected the N<sub>2</sub>O emissions from the tea fields during the wet season. Following fertilization, the horizontal and vertical movement of NH4N and NO3N in the topsoil of the tea fields potentially produced the strong spatial heterogeneity of N<sub>2</sub>O emissions. In addition, it is possible that the variations in the availability of oxygen in the soils was regulated by soil moisture, which determined the spatio-temporal heterogeneity of N<sub>2</sub>O emissions by inducing different degrees of soil nitrification and denitrification (Davidson et al., 2000; Konda et al., 2010). Thus, spatial interpolation methods must be chosen carefully to accurately estimate the spatial distribution of N<sub>2</sub>O emissions when the emissions are high and have strong spatial variability in the fields.

### **5 Conclusions**

During the wet season of 2012, a 30-min one-time measurement of  $N_2O$  emissions from a 4.0 ha red-soil tea field in the subtropical region of central China were determined at 147 points. The  $N_2O$  fluxes significantly varied with space. In addition, the  $N_2O$  fluxes were significantly

correlated with the NH4N, NO3N, SOC and TSN contents (r > 0.27 and p < 0.001). The logit-transformed N<sub>2</sub>O fluxes demonstrated a strong spatial dependency and were characterized by an exponential semivariogram model with an effective range of 25.2 m. Three spatial interpolation methods (OK, RK and CK) were used to predict the spatial distribution of N<sub>2</sub>O emissions. The RK and CK methods were relatively accurate for predicting results. Although the N<sub>2</sub>O emissions were much higher during the wet season than in the dry season, the N<sub>2</sub>O emissions exhibited similar spatial structure during both seasons. Such a phenomenon was mainly attributed to the low soil disturbance (e.g., only fertilizing in a very small proportion of area and weeding) in the tea field.

To effectively mitigate high  $N_2O$  emissions from the tea field soils, the biological and chemical mechanisms of  $N_2O$  emissions must be deeply explored. In addition, the responsive land management practices, such as biochar application, deep fertilization (under 20 cm), the use of controlled-release fertilizers and ecological engineering, must be recommended and deployed, especially during the wet season.

## Acknowledgements

The National Basic Research Program of China (2012CB417105) and the National Natural Science Foundation of China (41171200) financially supported this research.

References

- Akiyama, H., Yan, X. Y., and Yagi, K.: Estimations of emission factors for fertilizer-induced
- direct N<sub>2</sub>O emissions from agricultural soils in Japan: Summary of available data, Soil Sci.
- 460 Plant Nutr., 52, 774-787, 2006.
- Ambus, P. and Christensen, S.: Measurement of N<sub>2</sub>O emission from a fertilized grassland: an
- analysis of spatial variability, J. Geophys. Res., 99, 16557-16567, 1994.
- Armstrong, M.: Basic linear Geostatistics, Springer Verlag, Berlin, 153 pp., 1998.
- Ball, B. C., Horgan, G. W., Clayton, H., and Parker, J. P.: Spatial variability of nitrous oxide
- fluxes and controlling soil and topographic properties, J. Environ. Qual., 26, 1399-1409,
- 466 1997.
- Clemens, J. Schillinger, M. P., Goldbach, H., and Huwe, B.: Spatial variability of N<sub>2</sub>O
- emissions and soil parameters of an arable silt loam a field study, Bio. Fert. Soils, 28,
- 469 403-406, 1999.
- Davidson, E. A., Matson, P. A., Vitousek, P. M., Riley, R., Dunkin, K., García-Méndez, G.,
- and Maass, J. M.: Processes regulating soil emissions of NO and N<sub>2</sub>O in a seasonally dry
- tropical forest, Ecology, 74, 130-139, 1993.
- Davidson, E. A., Keller, M., Erickson, H. E., Verchot, L. V., and Veldkamp, E.: Testing a
- conceptual model of soil emissions of nitrous and nitric oxides, Bioscience, 50, 667-680,
- 475 2000.
- DGUU (Department of Geography, Utrecht University): Introduction for Gstat, available at:
- http://www.gstat.org/index.html (last access: 15 December 2010), 2010.

- 478 Firestone, M., and Davidson, E.: Microbial basis of NO and N<sub>2</sub>O production and
- consumption, in: Exchange of Trace Gases Between Ecosystems and the Atmosphere,
- edited by: Andreae, M.O. and Schimel, D.S., John Wiley, Chichester, 7-21, 1989.
- Folorunso, O. A., and Rolston, D. E.: Spatial variability of field measured denitrification gas
- 482 fluxes, Soil Sci. Soc. Am. J., 48, 1214-1219, 1984.
- Fu, X., Li, Y., Xiao, R., Tong, C., and Wu, J.: N<sub>2</sub>O emissions from a tea field in subtropical
- China. In: Proceedings of the 19th World Congress of Soil Science, Soil Solutions for a
- Changing World, 1–6 August 2010, Brisbane (published on CDROM), 161-163, 2010.
- Fu, X., Li, Y., Su, W., Shen, J., Xiao, R., Tong, C., and Wu, J.: Annual dynamics of N2O
- emissions from a tea field in southern subtropical China, Plant Soil Environ., 58,
- 488 373-378, 2012.
- Goovaerts, P.: Geostatistics for Natural Resources Evaluation, Oxford University Press, New
- 490 York, 483 pp., 1997.
- 491 Gorres, J. H., Dichiaro, M. J., and Lyons, J. A.: Spatial and temporal patterns of soil
- biological activity in a forest and an old field, Soil Biol. Biochem., 30, 219-230, 1998.
- 493 Han, W. Y., Xu, J. M., Wei, K., Shi, W. Z., and Ma, L. F.: Estimation of N<sub>2</sub>O emission from
- tea garden soils, their adjacent vegetable garden and forest soils in eastern China, Environ.
- 495 Earth Sci., 70, 2495-2500, 2013.
- Hayatsu, M.: The lowest limit of pH for nitrification in tea soil and isolation of an acidophilic
- ammonia oxidizing bacterium, Soil. Sci. Plant Nutr., 39, 219-226, 1993.

- Hengl, T., Heuvelink, G. B. M., and Stein, A.: A generic framework for spatial prediction of
- soil variables based on regression-kriging, Geoderma, 120, 75-93, 2004.
- Hirono, Y., and Nonaka, K.: Nitrous oxide emissions from green tea fields in Japan:
- contribution of emissions from soil between rows and soil under the canopy of tea plants,
- 502 Soil. Sci. Plant Nutr., 58, 384-392, 2012.
- 503 IPCC: Climate change 2013: the physical science basis. Contribution of working group I, in:
- Fourth assessment report of the intergovernmental panel on climate change, edited by:
- Solomon S., Qin D., Manning, M., Chen Z., Marquis, M., Averyt, K.B., Tignor, M.,
- Miller, H.L., Cambridge University Press, Cambridge, 996 pp., 2013.
- 507 ISM (Institute for Statistics and Mathematics): The R Project for Statistical Computing,
- available at: http://www.r-project.org/ (last access: 15 December 2010), 2010.
- Kiese, R., Hewett, B., Graham, A., and Butterbach-Bahl, K.: Seasonal variability of N<sub>2</sub>O
- emissions and CH<sub>4</sub> uptake by tropical rainforest soils of Queensland, Australia. Global
- Biogeochem. Cy., 17, 1043, doi:10. 1029/2002GB002014, 2003.
- Konda, R., Ohta, S., Ishizuka, S., Arai, S., Ansori, S., Tanaka, N., and Hardjono, A.: Spatial
- structures of N<sub>2</sub>O, CO<sub>2</sub>, and CH<sub>4</sub> fluxes from Acacia mangium plantation soils during a
- relatively dry season in Indonesia, Soil Biol. Biochem., 40, 3021-3030, 2008.
- Konda, R., Ohta, S., Ishizuka, S., Heriyanto, J., and Wicaksono, A.: Seasonal changes in the
- spatial structures of N<sub>2</sub>O, CO<sub>2</sub> and CH<sub>4</sub> fluxes from Acacia mangium plantation soils in
- 517 Indonesia, Soil Biol. Biochem., 42, 1512-1522, 2010.

- Li, Y., Fu, X., Liu, X., Shen, J., Luo, Q., Xiao, R., Li, Y., Tong, C., and Wu, J.: Spatial
- variability and distribution of N<sub>2</sub>O emissions from a tea field during the dry season in
- subtropical central China, Geoderma, 193, 1-12, 2013.
- Lin, Y., and Han, W.: N<sub>2</sub>O emissions from different soils, Chinese Journal of Tea Science, 29,
- 522 456-464, 2009.
- Mathieu, O., Lévêque, J., Hénault, C., Milloux, M. J., Bizouard, F., and Andreux, F.:
- Emissions and spatial variability of N<sub>2</sub>O, N<sub>2</sub> and nitrous oxide mole fraction at the field
- scale, revealed with <sup>15</sup>N isotopic techniques, Soil Biol. Biochem., 38, 941-951, 2006.
- Meda, B., Flechard, C. R., Germain, K., Robin, P., Walter, C., and Hassouna, M.:
- Greenhouse gas emissions from the grassy outdoor run of organic broilers,
- Biogeosciences, 9, 1493-1508, doi:10.5194/bg-9-1493-2012, 2012.
- Mosier, A. R., Duxbury, J. M., Freney, J. R., Heinemeyer, O., and Minami, K.: Nitrous oxide
- emissions from agricultural fields: assessment, measurement and mitigation. Plant Soil,
- 531 181, 95-181, 1996.
- Mosier, A. R., Kroeze, C., Nevison, C., Oenema, O., Seitzinger, S., and van Cleamput, O.:
- Closing the global N<sub>2</sub>O budget: nitrous oxide emissions through the agricultural nitrogen
- 534 cycle, Nutr. Cycl. Agroecosys., 52, 225-248, 1998.
- NBSC (a): China Statistical Yearbook, annual publication, National Bureau of Statistics of
- China, Beijing, 2014.

- Odeh, I. O. A., McBratney, A. B., and Chittleborough, D. J.: Spatial prediction of soil
- properties from landform attributes derived from a digital elevation model, Geoderma, 63,
- 539 197-214, 1994.
- Odeh, I. O. A., McBratney, A. B., and Chittleborough, D. J.: Further results on prediction of
- soil properties from terrain attributes: heterotopic cokriging and regression kriging,
- 542 Geoderma, 67, 215-226, 1995.
- R Development Core Team: R: a language and environment for statistical computing. R
- Foundation for Statistical Computing, 2014.
- Ravishankara, A. R., Daniel, J. S., and Portmann, R. W.: Nitrous oxide (N<sub>2</sub>O): the dominant
- ozone-depleting substance emitted in the 21st century, Science, 326, 123-125, 2009.
- Röver, M., Heinemeyer, O., Munch, J. C., and Kaiser, E. A.: Spatial heterogeneity within the
- plough layer: high variability of N<sub>2</sub>O emission rates, Soil Biol. Biochem., 31, 167-173,
- 549 1999.
- 550 Stein, A., van Dooremolen, W., Bouma, J., and Bregt, A. K.: Cokriging point data on
- moisture deficit. Soil Sci. Soc. Am. J., 52, 1418-1423, 1988.
- Tokuda, S. I., and Hayatsu, M.: Nitrous oxide flux from a large amount of nitrogen fertilizer
- and soil environmental factors controlling the flux, Soil. Sci. Plant Nutr., 50, 365-374,
- 554 2004.
- Turner, D. A., Chen, D., Gellbally, I. E., Li, Y., Edis, R. B., Leuning, R., Kelly, K., and
- Phillips, F.: Spatial variability of nitrous oxide emissions from an Australian irrigated
- dairy pasture, Plant soil, 309, 77-88, 2008.

- Van den Pol-van Dasselaar, A., Corré, W. J., Klemedtsson, A., Weslien, P., Stein, A.,
- Klemedtsson, L., and Oenema, O.: Spatial variability of methane, nitrous oxide, and
- carbon dioxide emissions from drained grasslands, Soil Sci. Soc. Am. J., 62, 810-817,
- 561 1998.
- Van Kessel, C., Pennock, D.J., and Farrell, R.E.: Seasonal-variations in denitrification and
- nitrous oxide evolution at the landscape scale, Soil Sci. Soc. Am. J., 57, 988-995, 1993.
- Velthof, G. L., Jarvis, S. C., Stein, A., Allen, A. G., and Oenema, O.: Spatial variability of
- nitrous oxide fluxes in mown and grazed grasslands on a poorly drained clay soil, Soil
- Biol. Biochem., 28, 1215-1225, 1996.
- Venterea, R. T., and Rolston, D. E.: Mechanisms and kinetics of nitric and nitrous oxide
- production during nitrification in agricultural soil, Glob. Change Biol., 6, 303-316, 2000.
- Webster, R.: Quantitative spatial analysis of soil in the field, in: Advances in Soil Science,
- edited by: Stewart, B.A., Springer, New York, 1-70, 1985.
- Webster, R., and Oliver, M. A.: Geostatistics for Environmental Scientists, John Wiley &
- Sons, Chichester, 2001.
- Werner, C., Kiese, R., and Butterbach-Bahl, K.: Soil-atmosphere exchange of N<sub>2</sub>O, CH<sub>4</sub>, and
- 574 CO<sub>2</sub> and controlling environmental factors for tropical rain forest sites in western Kenya,
- J. Geophys Res., 112, D03308, doi:10.1029/2006JD007388, 2007.
- Wrage, N., Velthof, G. L., Laanbroek, H. J., and Oenema, O.: Nitrous oxide production in
- grassland soils: assessing the contribution of nitrifier denitrification, Soil Biol. Biochem.,
- 578 36, 229-236, 2004.

Yanai, J., Lee, C. K., Umeda, M., and Kosaki, T.: Spatial variability of soil chemical properties in a paddy field. Soil Sci. Plant Nutr., 46, 473-482, 2000.

Yanai, J., Sawamoto, T., Oe, T., Kusa, K., Yamakawa, K., Sakamoto, K., Naganawa, T.,

Inubushi, K., Hatano, R., and Kosaki, T.: Atmospheric pollutants and trace gases: spatial variability of nitrous oxide emissions and their soil-related determining factors in an agricultural field, J. Environ. Qual., 32, 1965-1977, 2003.

Table 1 Descriptive statistics of the N<sub>2</sub>O fluxes and environmental factors.

Variable <sup>a</sup>	Mean	Minimum	Maximum	CV	Skewness of the	Skewness of the
				(%)	original	logit-transformed data
					data	
FLUX30	102.24 <sup>b</sup>	-1.73	1,659.11	234.7	4.37	0.6
ELEVATION	80.64	74.25	87.96	4.1	0.04	-
BD	1.26	0.90	1.56	10.1	-0.28	-
DOC	185.56	43.70	424.14	34.6	0.75	-
NH4N	62.33	1.89	842.55	190.8	3.28	0.17
NO3N	21.54	0.48	135.29	141.6	1.85	0.28
SOC	13.33	5.11	52.52	50.1	2.27	-0.44
TSN	1.52	0.81	4.12	38.3	1.73	-0.01
SWC	0.33	0.19	0.47	16.6	0.07	-
SAND	39.73	16.98	63.79	23.8	0.02	-
SILT	47.15	26.78	64.17	16.1	-0.29	-
CLAY	13.12	8.68	21.68	21.5	1.00	-

<sup>&</sup>lt;sup>a</sup> FLUX30 is the N<sub>2</sub>O flux (g N ha<sup>-1</sup> d<sup>-1</sup>); ELEVATION is the elevation (m); and BD, DOC, NH4N, NO3N, SOC, TSN, SWC, SAND, SILT and CLAY are the soil bulk density (Mg m<sup>-3</sup>), soil dissolved organic carbon (mg C kg<sup>-1</sup> soil), soil ammonium (mg N kg<sup>-1</sup> soil), soil nitrate (mg N kg<sup>-1</sup> soil), soil organic carbon (g C kg<sup>-1</sup> soil), soil total nitrogen (g N kg<sup>-1</sup> soil),

- 591 gravimetric soil water (g H<sub>2</sub>O g<sup>-1</sup> soil), soil sand particle (%), soil silt particle (%) and soil
- clay particle (%) content, respectively, of the 0-20 cm of topsoil.
- <sup>b</sup> The median and standard deviation of the FLUX30 were 27.56 and 239.96 g N ha<sup>-1</sup> d<sup>-1</sup>,
- respectively.

Table 2 Semivariogram models for  $N_2O$  fluxes and the environmental factors.

Variable	Model	Nugget	Partial	Sill(nugget+	Distance	Effective	Partial
			sill	partial sill)	Parameter (m)	range (m)	sill/sill
FLUX30t <sup>a</sup>	Exp	0	3.7186	3.7186	8.40	25.2	1.00
NH4Nt <sup>a</sup>	$ND^{c}$	$ND^{c}$	ND <sup>c</sup>	$ND^{c}$	$ND^{c}$	$ND^{c}$	$ND^{c}$
NO3Nt <sup>a</sup>	Ste	4.0794	0.6113	4.6907	91.92	163.7	0.13
SOCt <sup>a</sup>	Sph	1.1198	0.7744	1.8942	92.96	93.0	0.41
TSNt <sup>a</sup>	Ste	1.0422	0.2816	1.3238	57.97	102.6	0.21
SWCt <sup>a</sup>	$ND^{c}$	$ND^{c}$	$ND^c$	$ND^{c}$	$ND^{c}$	$ND^{c}$	$ND^{c}$
SAND <sup>a</sup>	$ND^{c}$	$ND^{c}$	ND <sup>c</sup>	$ND^{c}$	$ND^{c}$	$ND^{c}$	$ND^{c}$
SILT <sup>a</sup>	ND <sup>c</sup>	$ND^{c}$	ND <sup>c</sup>	ND <sup>c</sup>	ND <sup>c</sup>	ND <sup>c</sup>	$ND^{c}$
FLUX30t <sup>b</sup>	Exp	1.1911	2.0560	3.2471	17.36	52.1	0.63
NH4Nt <sup>b</sup>	Exp	2.0473	0.7185	2.7658	17.36	52.1	0.26
NO3Nt <sup>b</sup>	Exp	1.6241	1.1188	2.7429	17.36	52.1	0.41
SOCt <sup>b</sup>	Exp	0.6043	1.0777	1.6820	17.36	52.1	0.64
$TSNt^b$	Ste	0.9347	0.3114	1.2461	59.53	105.4	0.25
$SWCt^b$	$ND^{c}$	$ND^c$	$ND^c$	$ND^{c}$	$ND^{c}$	$ND^{c}$	$ND^{c}$
$SAND^b$	ND	ND	ND	ND	ND	ND	ND
SILT <sup>b</sup>	ND	ND	ND	ND	ND	ND	ND

ND, not determined.

<sup>&</sup>lt;sup>a</sup> Semivariogram models for the OK method.

- <sup>b</sup> Semivariogram models for the RK method using the chamber placement position as the
- 599 auxiliary regression predictor.
- 600 <sup>c</sup> Spatial structures were not apparent.

Table 3 Statistics for  $N_2O$  fluxes during the dry and wet seasons.

Sample position	Mean	SD	Median	Max.	Min.	CV (%)
Dry season						
Inter-row (58)	5.15	4.95	4.09	22.43	-2.83	96.1
Fertilization point (50)	7.19	12.04	4.34	79.56	-6.42	167.4
Under tree (28)	3.58	2.91	2.36	10.28	0.68	81.3
In tree row (11)	5.95	10.38	3.98	52.17	-5.69	174.5
Wet season						
Inter-row (45)	101.69	287.23	27.56	1,659.11	-0.81	282.5
Fertilization point (45)	198.81	295.70	73.42	1,404.32	0.85	148.7
Under tree (22)	16.74	17.00	10.64	61.24	-1.73	101.6
In tree row (33)	28.30	38.34	14.72	177.08	0.19	135.5

The numbers in the parentheses represent the sample numbers for each chamber placement position.

Table 4 Cross-validations of the three different kriging interpolations for  $N_2O$  fluxes during the dry and wet seasons.

Method of spatial	Auxiliary	ME	RMSE	r	Predicted total
interpolation	variable	(no dimension)	(no dimension)		N <sub>2</sub> O emissions
					$(g N d^{-1})$
Dry season					
OK	-	0.0002	0.102	0.52	22.1 <sup>a</sup>
RK	ELEVATION	0.0008	0.098	0.57	21.1 <sup>a</sup>
CK	SOCt	0.0006	0.103	0.51	22.0 <sup>a</sup>
CK	ELEV	0.0008	0.099	0.57	21.5 <sup>a</sup>
CK	SOCt and	0.0009	0.098	0.57	21.2ª
	ELEV				
Wet season					
OK	-	-0.0005	1.739	0.18	208.1
RK	POSITION	-0.0006	1.549	0.49	148.2
CK	SOCt	0.0020	1.439	0.58	149.5
	(POSITION)				
CK	NH4Nt	0.0001	1.185	0.74	150.5
	(POSITION)				
	and NO3Nt				
	(POSITION)				

OK, RK and CK correspond to ordinary kriging, regression kriging and cokriging, respectively; For the dry season campaign, ELEVATION, SOCt and ELEV are the normalized elevation, the normalized soil organic carbon content and the inverse of the normalized elevation, respectively. For the wet season campaign, SOCt, NH4Nt and NO3Nt are the logit-transformations of soil organic carbon, soil ammonium and soil nitrate concentrations, respectively. "POSITION" (in the parentheses) indicates the process of detrending the influence of chamber placement position. The ME, RMSE, and r are the mean prediction error, the root mean squared error (the mean squared deviation ratio of the prediction residuals to the kriging standard errors), and the Pearson's correlation coefficient between the observations and the predictions, respectively.

The predicted total N<sub>2</sub>O emissions during the dry season were recalculated because the study area changed from 4.8 ha to 4.0 ha for the wet season.

## Figure captions

Figure 1. (**a**, **b**) Location and (**c**, **d**) digital elevation model and land use map of the tea planted catchment. The red circles in (**c**, **d**) represent the sample points. The catchment is located in Jinjing town, which is 70 km northeast of Changsha, the capital city of Hunan Province, China.

Figure 2. Daily (a) air temperatures and (b) precipitation during 2012.

Figure 3. Histograms of (**a**) the original N<sub>2</sub>O fluxes (FLUX30) and (**b**) the logit-transformed N<sub>2</sub>O fluxes (FLUX30t).

Figure 4. The Tukey's Honest Significant Difference analysis for FLUX30t, NH4Nt, NO3Nt, SOCt, TSNt and SWCt based on the four-chamber placement positions (R, inter-row; F, fertilization point; U, under tea tree; and I, in the tea tree row).

Figure 5. Correlation matrix with the Pearson's correlation coefficients (r) of the N<sub>2</sub>O fluxes and the environmental factors. All of the variables in the correlation matrix are normally distributed. FLUX30 represents the N<sub>2</sub>O flux (g N ha<sup>-1</sup> d<sup>-1</sup>); ELEVATION is the elevation (m); and BD, DOC, NH4N, NO3N, SOC, TSN, SWC, SAND, SILT and CLAY are the soil bulk density (Mg m<sup>-3</sup>), soil dissolved organic carbon (mg C kg<sup>-1</sup> soil), soil ammonium (mg N kg<sup>-1</sup>

soil), soil nitrate (mg N kg<sup>-1</sup> soil), soil organic carbon (g C kg<sup>-1</sup> soil), total soil nitrogen (g N kg<sup>-1</sup> soil), gravimetric soil water (g H<sub>2</sub>O g<sup>-1</sup> soil), soil sand particle (%), soil silt particle (%) and soil clay particle (%) contents of the top 0-20 cm of the soil, respectively. Furthermore, \*, \*\* and \*\*\* represent the statistical significance at probability levels of 0.05, 0.01 and 0.001, respectively. The lowercase letter t represents the logit transformation.

Figure 6. Semivariograms (open circles) and best-fitted models (solid lines) of the normal logit-transformed N<sub>2</sub>O fluxes (FLUX30t) (no dimension) for ordinary kriging (**a**) and the regression residuals of FLUX30t (no dimension) with chamber placement position as the predictor for regression kriging (**b**).

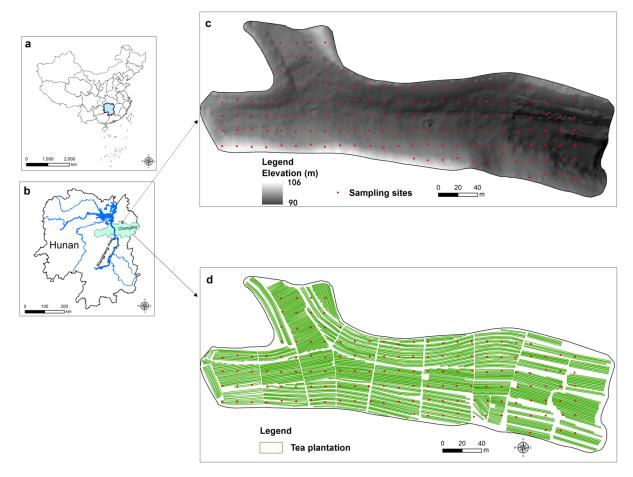
Figure 7. Direct and cross-semivariograms (open circles, detrending the influence of chamber placement position for cokriging) and the best-fitted linear model of the co-regionalization (solid lines) of the normal logit-transformed N<sub>2</sub>O fluxes (FLUX30t) (no dimension) and the normal SOC (SOCt, no dimension). The linear model of co-regionalization was characterized by using the same range and different sills for its component models.

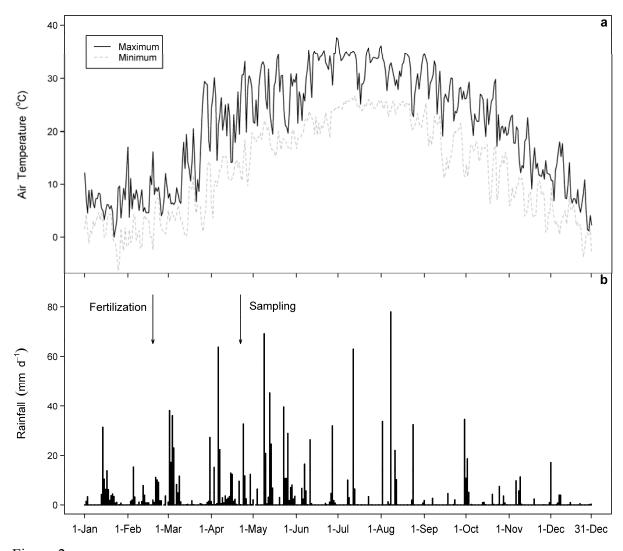
Figure 8. Direct and cross-semivariograms (open circles, detrending the influence of chamber placement position for cokriging) and the best-fit linear model of co-regionalization (solid lines) for the normal logit-transformed N<sub>2</sub>O fluxes (FLUX30t) (no dimension), NH4N (NH4Nt, no dimension) and NO3N (NO3Nt, no dimension). The linear model of

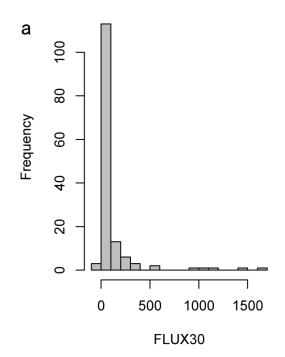
co-regionalization was characterized by the same range and different sills for its component models.

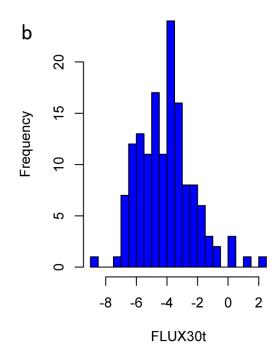
Figure 9. Spatial distributions of the N<sub>2</sub>O fluxes as predicted by (a) OK, (b) RK with chamber placement position as the regression predictor, (c) CK with SOCt (with the influence of chamber placement position detrended) as the covariable, and (d) CK with NH4Nt (with the influence of chamber placement position detrended) and NO3Nt (with the influence of chamber placement position detrended) as two covariables. Here, SOCt, NH4Nt and NO3Nt represent the logit-transformed soil organic carbon, soil ammonium and soil nitrate content, respectively.

Figure 10. Spatial distributions of kriging variance of the N<sub>2</sub>O fluxes as predicted by (a) OK, (b) RK with chamber placement position as the regression predictor, (c) CK with SOCt (with the influence of chamber placement position detrended) as the covariable, and (d) CK with NH4Nt (with the influence of chamber placement position detrended) and NO3Nt (with the influence of chamber placement position detrended) as two covariables. Here, SOCt, NH4Nt and NO3Nt represent the logit-transformed soil organic carbon, soil ammonium and soil nitrate content, respectively.









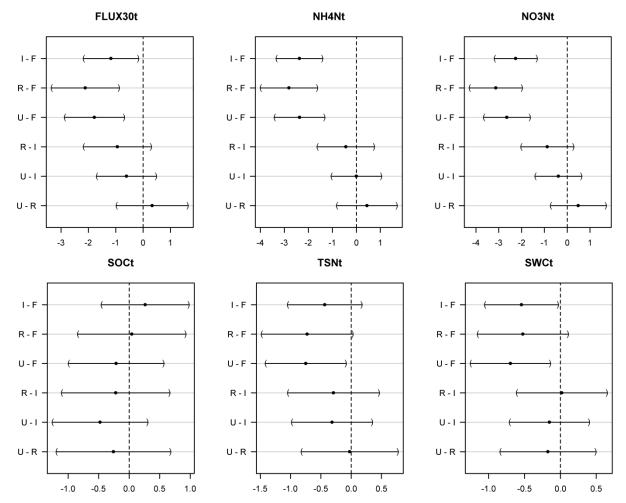
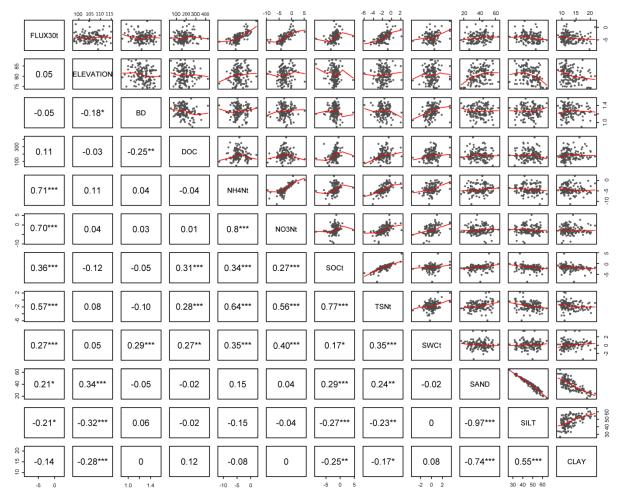
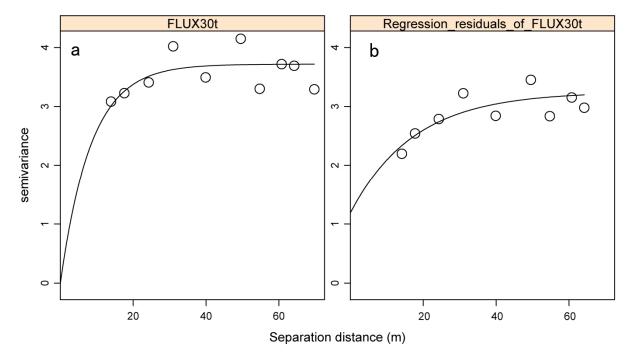
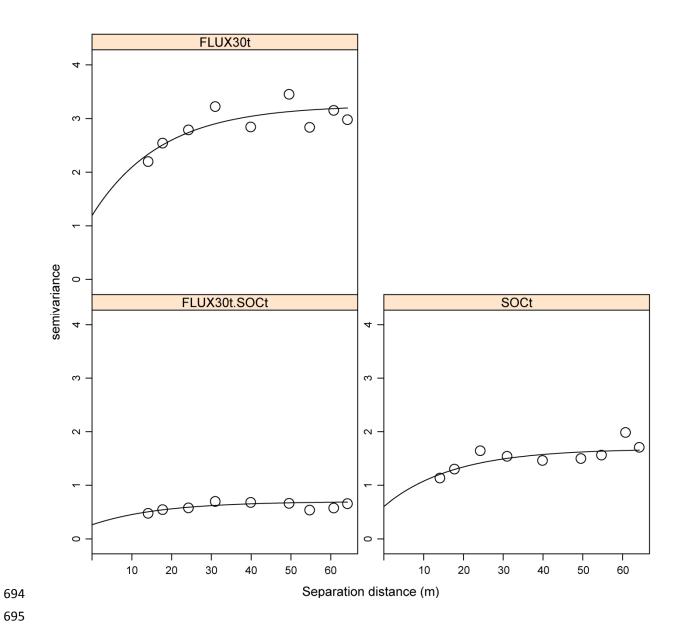


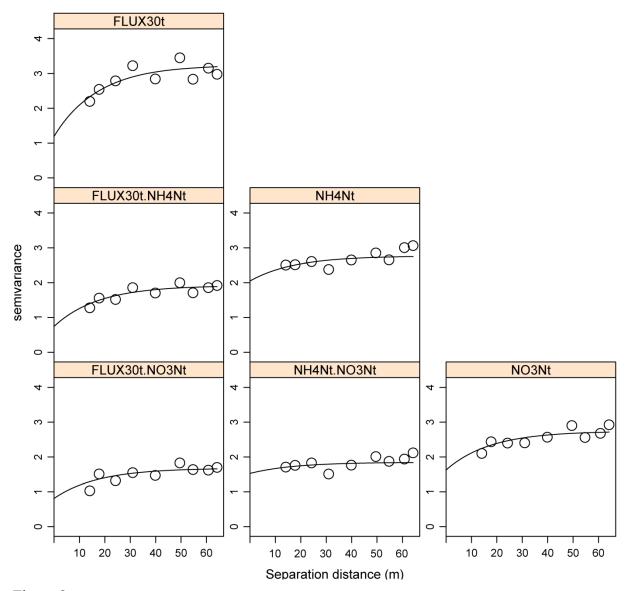
Figure 4











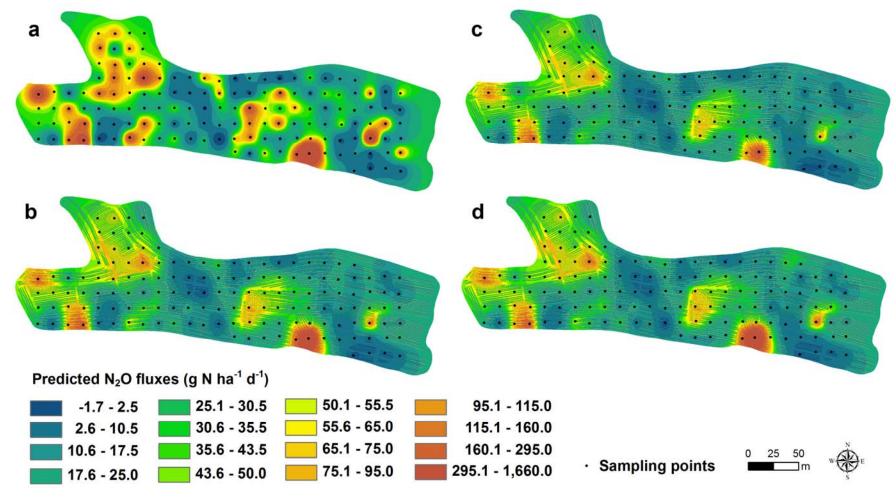


Figure 9

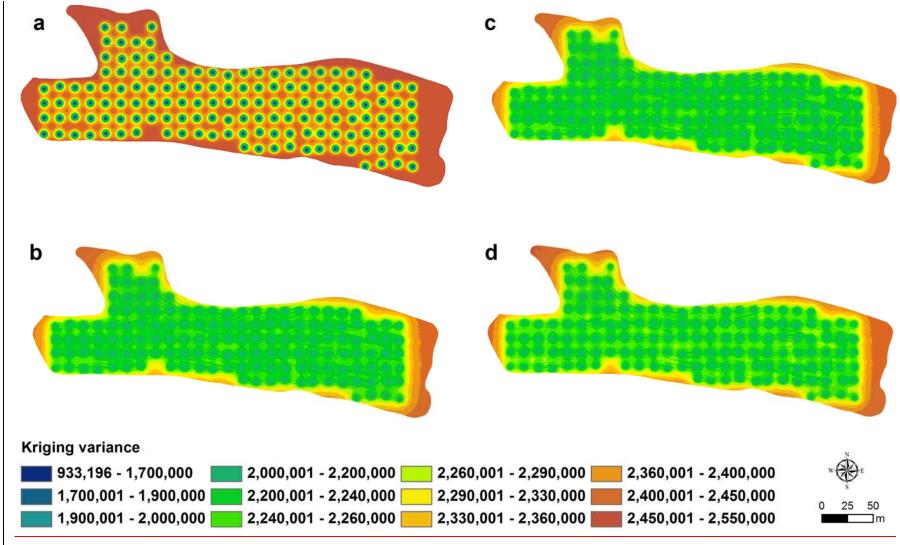


Figure 10