Wet-season spatial variability of N_2O emissions from a tea field in subtropical central China

Xiaoqing Fu, Xinliang Liu, Yong Li *, Jianlin Shen, Yi Wang, Ganghua Zou, Hang Li, Lifang Song, Jinshui Wu

Changsha Research Station for Agricultural & Environmental Monitoring and
Key Laboratory of Agro-ecological Processes in Subtropical Regions,
Institute of Subtropical Agriculture, Chinese Academy of Sciences,
Hunan 410125, China

These two authors contributed equally to this work.

*Correspondence to: Professor Yong Li
Institute of Subtropical Agriculture,
Chinese Academy of Sciences, Hunan 410125, China
Tel: +86-731-8461-5291
Fax: +86-731-8461-2685
E-mail: yli@isa.ac.cn

Abstract

Tea fields emit large amounts of nitrous oxide (N_2O) to the atmosphere. Obtaining accurate estimations of N_2O emissions from tea-planted soils is challenging due to strong spatial variability. We examined the spatial variability of N_2O emissions from a red-soil tea field in Hunan province, China, on 22 April 2012 (in a wet season) using 147 static mini chambers approximately regular gridded in a 4.0 ha tea field. The N_2O fluxes for a 30-min snapshot (10:00-10:30 am) ranged from -1.73 to 1,659.11 g N ha^{-1} d^{-1} and were positively skewed with an average flux of 102.24 g N $ha^{-1}d^{-1}$. The N_2O flux data were transformed to a normal distribution by using a logit function. The geostatistical analyses of our data indicated that the logit-transformed N_2O fluxes (FLUX30t) exhibited strong spatial autocorrelation, which was characterized by an exponential semivariogram model with an effective range of 25.2 m. As observed in the wet season, the logit-transformed soil ammonium-N (NH4Nt), soil nitrate-N (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 methods (ordinary kriging, regression kriging and cokriging) were applied to estimate the spatial distribution of N_2O 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 RMSE=1.74), regression kriging with the sample position as a predictor (r=0.49 and RMSE=1.55) and cokriging with SOCt as a covariable (r=0.58 and RMSE=1.44). The predictions of the three kriging interpolation methods for the total N_2O emissions of 4.0 ha tea field ranged from 148.2 to 208.1 g N d^{-1}, based on the 30 min snapshots obtained during

41 the wet season. Our findings suggested that to accurately estimate the total N_2O emissions
42 over a region, the environmental variables (e.g., soil properties) and the current land use
43 pattern (e.g., tea row transects in the present study) must be included in spatial interpolation.
44 Additionally, compared with other kriging approaches, the cokriging prediction approach
45 showed great advantages in being easily deployed, and more importantly providing accurate
46 regional estimation of N_2O emissions from tea-planted soils.
47
48 **Introduction**
49 According to the latest data, which show rapid increases in their atmospheric concentrations
50 (IPCC, 2013), nitrous oxide (N_2O), carbon dioxide (CO_2) and methane (CH_4) are three major
51 greenhouse gases in the atmosphere that significantly contribute to global warming. Among
52 these major greenhouse gases, N_2O has a very high radiative forcing per unit mass (265-fold
53 stronger than CO_2 on a 100 year horizon) and plays an important role in ozone depletion in
54 the stratosphere (Ravishankara et al., 2009). The primary sources of N_2O are from agriculture
55 development and the subsequent increased use of chemical N fertilizers (Ambus and
56 Christensen, 1994; Mosier et al., 1996, 1998; Yanai et al., 2003; Tokuda and Hayatsu, 2004;
57 Akiyama et al., 2006; Ravishankara et al., 2009). Agricultural soils produce 2.8 (1.7–4.8) Tg
58 of N_2O-N yr^{-1} (IPCC, 2013). The N_2O is emitted from soils via the microbial processes of
59 nitrification under aerobic conditions and denitrification under anaerobic conditions
60 (Firestone and Davidson, 1989; Wrage et al., 2004). The magnitude of soil N_2O emissions is
61 highly variable and strongly influenced by changes in environmental conditions.

Among the different agricultural soils, tea-planted soils are important sources of N_2O 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_2O (Hayatsu, 1993; Venterea and Rolston, 2000; Li et al., 2013). However, because few measurements of N_2O 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_2O emissions from tea-planted soils. To estimate the N_2O 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_2O emissions from tea fields. This information will lead to the development of effective land management options for mitigating N_2O emissions from a significant source, tea plantation.

The N_2O 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_2O fluxes, with higher N_2O 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_2O emissions significantly contributes to the uncertainty when estimating the

83 contributions of subtropical tea-planted ecosystems to N_2O flux. Moreover, most of our
84 knowledge regarding seasonal changes and the spatial variability of N_2O fluxes is based on a
85 small number of measurements taken from tea-planted soils. Li et al. (2013) investigated the
86 spatial structure of N_2O fluxes for tea-planted soils during the dry season in October 2010
87 and found that the spatial distribution of the N_2O fluxes was primarily associated with field
88 elevation (r=-0.42, p<0.001). The other soil properties (e.g., soil organic carbon, soil water
89 and soil mineral nitrogen) were not significantly related to N_2O flux. To obtain a more
90 accurate evaluation of annual N_2O emissions from tea-planted soils, a study on the spatial
91 structure and distribution of N_2O emissions during a wet season (in contrast to the dry season)
92 is necessary.

93 To understand the structure of the spatially distributed data and to predict the N_2O fluxes
94 at the unsampled locations, geostatistical analyses can be useful (Goovaerts, 1997; Webster
95 and Oliver, 2001). Geostatistics provide statistical tools for describing the quantitative spatial
96 variability of field observations for the accurate mapping and planning of rational sampling
97 schemes that efficiently utilize the available labor (Webster, 1985). Several geostatistical
98 methods are used to examine the spatial variability of N_2O fluxes, including simple kriging
99 (SK), ordinary kriging (OK), regression kriging (RK) and cokriging (CK). The most
100 commonly used method is OK (Clemens et al., 1999; Röver et al., 1999; Mathieu et al., 2006;
101 Konda et al., 2008, 2010), which uses the derived theoretical semivariogram models to
102 interpolate the spatial distribution of N_2O fluxes. However, research has demonstrated that
103 RK and CK approaches, which use related auxiliary variables, improve the prediction

104 accuracy (Goovaerts, 1997; Webster and Oliver, 2001; Hengl et al., 2004). The RK method
105 combines multiple regressions, including linear regressions, generalized linear models,
106 generalized added models and regression tree models, with the auxiliary variables used for
107 kriging (Odeh et al., 1994). In the RK method, linear regressions are commonly used. The
108 CK approach uses correlations that may exist between the predicted variables and other more
109 easily measured variables. These variables can be measured at the same points as the
110 predicted variable, at other points, or at both. Compared with the RK approach, the CK
111 approach is commonly applied when the measurement of a covariable is less expensive than
112 the cost of a predicted variable (Stein et al., 1988; Odeh et al., 1995). In addition to the
113 feature correlation as a criterion for selecting covariables, the CK approach also requires that
114 both of the predicted variable and covariables have similar spatial structures (Odeh et al.,
115 1994). In this study, we used three interpolation methods (OK, RK and CK) to estimate the
116 spatial distribution of N_2O fluxes in a tea field.
117 In contrast with the dry season, the spatial variability of the N_2O emissions was
118 investigated during the wet season in April 2012 from the same tea-planted catchment that
119 was studied by Li et al. (2013). The catchment consisted of a completely independent
120 hydrological system. Thus, the spatial distribution of the N_2O emissions within the catchment
121 was expected to have intrinsic characteristics. The objectives of this study were to (i) evaluate
122 the spatial variability of N_2O emissions from soils planted with tea in subtropical central
123 China during the wet season, (ii) determine the key environmental factors controlling N_2O
124 emissions, and (iii) assess the prediction efficiency of three kriging interpolation methods.

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 Fig. 1 & Fig. 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.,

147 China), and then were used to develop the local DEM and land use data (at a spatial
148 resolution of 0.1 m, respectively, as shown in Fig. 1c and d). The land use data showed the
149 four positions where the chambers were placed, including the inter-row, fertilization point,
150 under tea tree and in tea tree row, as described in Li. et al. (2013). The spatial positions of the
151 gas sampling points in a 15 m × 15 m regular grid over the catchment were originally
152 determined using a DGPS receiver on 20 April 2012. Some of the chamber positions were
153 slightly adjusted (because of a lack of space in the tea tree rows or to avoid roads and
154 trenches). Thus, the chambers were placed in one of four locations mentioned above (Fig. 1d).
155 Overall, 147 sampling points were determined, and the Euclidean distances between each
156 point and its nearest neighbors ranged from 14.6 m to 16.7 m. The x-y coordinates, the gas
157 sampling position information (the inter-row, fertilization point, under tea tree and in tea tree
158 row along tea row transects), and the elevations at the sampling points were recorded.
159
160 2.3 *Gas and soil properties measurements*
161 Gas and soil samples were collected at each grid point on 22 April 2012 using a closed mini
162 chamber technique. A mini chamber set was composed of PVC and had two parts (base and
163 chamber). The base was 0.15 m in diameter and 0.05 m high. The chamber was 0.15 m in
164 diameter and 0.15m high. In the field operation, the base was gently inserted vertically into
165 the soil on 20 April 2012, and the chamber was clipped on the base with the sponge seals in
166 between to stop gas leaking before gas sampling on 22 April 2012. Therefore, the effective
167 static chambers volume was equal to the chamber volume of 0.002651 m^3. 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 ^{o}C for preventing any microbial activity (such as mineralization, nitrification and denitrification). The N_2O concentrations of the gas samples were analyzed using a gas chromatograph (Agilent 7890A, Agilent, USA) that was fit with a ^{63}Ni-electron capture detector and an automatic sample injector system. The N_2O fluxes (FLUX30, g N ha^{-1} d^{-1}) 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 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.

189

190 2.4 *Data analyses*

191 The descriptive statistical and geostatistical analyses were performed using R (R

192 Development Core Team, 2014) with the gstat package (DGUU, 2010).

193 Descriptive statistical analyses were used to determine the mean, median, minimum and

194 maximum values, SD, coefficient of variation (CV), and skewness of the original and

195 logit-transformed data. These analyses were based on the four chamber placement positions.

196 Because the FLUX30, NH4N, NO3N, SOC, TSN and SWC data were highly skewed, these

197 values were transformed by using a logit function (Hengl et al., 2004). The transformed

198 variables were named FLUX30t, NH4Nt, NO3Nt, SOCt, TSNt and SWCt. Using a Pearson's

199 correlation, the relationships between FLUX30t, NH4Nt, NO3Nt, SOCt, TSNt, SWCt, DOC,

200 BD, SAND, SILT, and CLAY were tested. The significance of the differences in the FLUX30t

201 and environmental factors (NH4Nt, NO3Nt, SOCt, TSNt and DOC) between any two of the

202 different chamber positions along the entire tea-tree row transect were evaluated using the

203 Tukey's Honest Significant Difference method.

204 In the geostatistical analyses, an experimental semivariogram of FLUX30t was

205 calculated, and the theoretical semivariogram models were fit. The ratio of the partial sill to

206 the total sill was used as an index of spatial dependence. Armstrong (1998) stated that a

207 variable with a higher ratio of partial sill to sill and a longer semivariogram range were more

208 structured. The spatial distribution of FLUX30t across the catchment was predicted using

209 three kriging interpolation methods (OK, RK and CK).These data were transformed back to

210 the original scale of FLUX30 for mapping. The Leave-One-Out cross-validation method was
211 used to evaluate the accuracy of interpolating FLUX30t using the three different kriging
212 methods.
213
214 **3. Results**
215 3.1 *Exploratory data analyses*
216 In the 4.0 ha tea-planted catchment, the N_2O fluxes during the 30-min one-time
217 measurements performed on 22 April 2012 ranged from -1.73 to 1,659.11 g N ha^{-1} d^{-1}, with a
218 median value of 27.56 g N ha^{-1} d^{-1} and a CV of 234.7 % (Table 1). The N_2O flux data were
219 positively skewed (Table 1 and Fig. 3a), and their logit-transformations were approximately
220 normally distributed (Table 1 and Fig. 3b). From Table 3, the logit-transformed N_2O fluxes
221 (FLUX30t) were the highest in the fertilization points, and the differences in the FLUX30t
222 values among the chamber placement positions were statistically significant ($p < 0.001$).
223
224 *<Insert Table 1 & Fig. 3 near here>*
225
226 The ELEVATION, BD, DOC, SWC, SAND, SILT, and CLAY were approximately
227 normally distributed, with skewness values of less than 1 (Table 1). Additionally, DOC
228 displayed a moderate CV of 34.6 %, and the other variables had lower CVs (4.1–23.8 %). The
229 NH4N, NO3N, SOC and TSN were positively skewed, and the logit-transformations (NH4Nt,
230 NO3Nt, SOCt and TSNt) had approximately normal distributions (Table1). 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 Fig. 4 & Fig. 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_2O and soil parameters (correlated with N_2O 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

252 detrending the influence of the chamber placement position, large changes in the
253 semivariogram models occurred regarding the above variables. Although the semivariograms
254 of the regression residuals of FLUX30t, NH4Nt, NO3Nt and SOCt were best-fit with the
255 same semivariogram model (exponential) with a similar range of 17.4 m (equivalent to an
256 effective range of 52.1 m), the spatial dependencies of those variables were different (Table
257 2). Of the soil properties, only SOCt had a similar spatial structure to FLUX30t when the
258 influence of the chamber placement position was detrended (Table 2). Based on these
259 correlation analyses and spatial variability analyses, the covariables for the CK method were
260 determined.
261
262 *<Insert Table 2 near here>*
263
264 3.3 *Spatial interpolation of N_2O emissions by three methods*
265 Three spatial interpolation methods were used in this study to predict the spatial distribution
266 of N_2O emissions from tea soils in the catchment. In the first method, the derived theoretical
267 semivariogram model for FLUX30t that is presented in Table 2 was used for the OK
268 prediction. In the second method, RK was used and the chamber placement position was
269 identified as the auxiliary regression predictor. Thus, the semivariogram of the regression
270 residuals of FLUX30t were calculated and best-fit with the theoretical semivariogram model
271 shown in Fig. 6. In the third method, CK involved two groups of covariables. As described
272 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_2O 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.

<Insert Figs.6-8 near here>

As shown in Fig. 9, the surface map for the spatial distribution of N_2O 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_2O emissions in the tea fields during the wet season were 208.1 g N d^{-1}, 148.2 g N d^{-1}, 149.7 g N d^{-1} and 150.5 g N d^{-1}, respectively. From

294 the performance evaluations of the four spatial interpolations, the total N_2O emissions from
295 the tea field on 22 April 2012 during the wet season were approximately 150 g N d^{-1}.
296
297 *<Insert Figs. 9-10 near here>*
298
299 **Discussion**
300 4.1 *Seasonal differences of N_2O fluxes in the red soil planted with tea*
301 The N_2O emissions from soils have obvious seasonal fluctuations, with emissions that are
302 significantly higher during the wet season than during the dry season (Konda et al., 2010). To
303 understand the seasonal changes in the spatial structures of N_2O fluxes, we compared the
304 N_2O emissions between the wet (this study) and dry (Li et al., 2013) seasons. In general, the
305 mean, SD and coefficient of variation (102.24, 239.96 g N ha^{-1} d^{-1} and 234.7 %, respectively)
306 of the N_2O fluxes in the wet season were all higher than those (2.88, 8.94 g N ha^{-1} d^{-1} and
307 152.0 %, respectively) during the dry season (Table 3). Furthermore, in contrast with the dry
308 season, the N_2O fluxes during the wet season were significantly different among the four
309 chamber placement positions, with the highest fluxes occurring at the fertilization points and
310 the inter-row positions (Table 3). During the wet season, the high N_2O fluxes at the
311 fertilization points and the inter-row positions resulted from the high soil moisture, due to
312 more rainfall, and from the fertilization that occurred on 19 February 2012 (Fig. 2). The soil
313 N and the soil organic C availability are directly increased by the application of chemical and
314 organic N fertilizers. The additional in the available C and N supplied by fertilization resulted
315 in increased soil microbial activity, which stimulated the nitrification and denitrification

316 processes that contribute to soil N_2O emissions (Davidson et al., 1993; Kiese et al., 2003;
317 Werner et al., 2007).
318
319 *<Insert Table 3 near here>*
320
321 4.2 *Spatial structure of N_2O emissions from red soils planted with tea*
322 Soil type, topography and land management (fertilization, tillage and irrigation) are the
323 primary factors that affect the spatial structures of N_2O emissions (Folorunso and Rolston,
324 1984; Clemens et al., 1990; Velthof et al., 1996; Konda et al., 2008). During the wet season,
325 the N_2O fluxes showed a strong spatial dependence (with a range of approximately 25.3 m)
326 that was similar to the dry season range of approximately 28.0 m in the tea-planted fields (Li
327 et al., 2013). These results indicated that the spatial dependence of N_2O fluxes at the current
328 spatial sampling scale was comparable between seasons. Our findings for a fertilized tea field
329 were similar to those of Konda et al. (2010) for a tropical forest. However, these results
330 contrasted those of many previous investigations for agricultural fields, including winter
331 wheat (Ball et al., 1997; Clemens et al., 1999; Röver et al., 1999; Mathieu et al., 2006),
332 summer maize (Clements et al., 1999), onion (Yanai et al., 2003), and grassland (Ambus and
333 Christensen, 1994; Velthof et al., 1996; van den Pol-van Dasselaar et al., 1998; Turner et al.,
334 2008) fields, in which the N_2O flux presented no, weak or moderate spatial dependence. This
335 discrepancy primarily occurred because of the unique geographical characterization and land
336 management of the tea plantation. Compared with other agricultural fields in flat areas, tea

337 fields are always distributed in hills or mountains. Therefore, the contributions of the
338 topography to the spatial dependence of the N_2O flux were strong (Li et al., 2013).
339 Additionally, tea is a perennial plant. Thus, apart from fertilization and weeding, the soil
340 disturbance in tea fields is always very low.

341 During the dry season, the topography (elevation) had a significant effect on the spatial
342 pattern of N_2O fluxes in the tea-planted fields (Li et al., 2013). Similar spatial patterns of N_2O
343 fluxes with topography were also observed in forest soils (Van Kessel et al., 1993; Konda et
344 al., 2010). Theoretically, the SWC varies with the topography and affects the spatial pattern
345 of N_2O fluxes by controlling the conditions for soil nitrification and denitrification (Firestone
346 and Davidson, 1989; Wrage et al., 2004). Although the SWC had no relationships with N_2O
347 and elevation during the dry season (Li et al., 2013), a correlation existed in the present study
348 (Fig. 5). The microstructures of the tea tree-row transect and the land management practices
349 of tea production were the primary influences on the spatial pattern of soil water in the
350 tea-planted fields (Li et al., 2013). During the wet season, fertilization contributed to the
351 spatial pattern of N_2O fluxes in the tea-planted fields, with the highest averaged fluxes at the
352 fertilization sites (198.81 g N ha^{-1} d^{-1}) (Table 3). Fertilization resulted in similar spatial
353 patterns of N_2O fluxes in other agricultural soils (Ball et al., 1997; Clements et al., 1999;
354 Röver et al., 1999; Mathieu et al., 2006; Yanai et al., 2003).

355 In view of the analysis of the primary factors that affected the spatial pattern of N_2O
356 fluxes, we detrended the influences of the environmental factors when the N_2O flux
357 semivariograms were calculated to more deeply explore the spatial structures of the N_2O

358 emissions in the tea-planted fields. For example, during the dry and wet seasons, the spatial
359 influences of elevation (Li et al., 2013) and chamber placement position, respectively, were
360 detrended when computing the N_2O flux semivariograms. Because the relationship between
361 chamber placement position and N_2O flux was more relevant than the relationship between
362 elevation and N_2O flux, the effect of detrending the influence of chamber placement position
363 during the wet season was more obvious than that of detrending the influence of elevation
364 during the dry season (Li et al., 2013). This effect was also reflected in the evaluation of the
365 performance of the RK method for the wet and dry seasons (Table 4).
366
367 *4.3 Spatial interpolations of N_2O emissions by three methods*
368 The three interpolation methods (OK, RK and CK) were used to predict the spatial
369 distributions of N_2O emissions from the red soils planted with tea during dry (Li et al., 2013)
370 and wet seasons (this study). However, these three methods resulted in significantly different
371 performances between the dry and wet seasons (Table 4). We conducted comparative
372 analyses for the performance of the three interpolation methods using two aspects: different
373 seasons and different methods. Firstly, the OK method performed better when predicting the
374 spatial distribution of N_2O fluxes for the dry season relative to the wet season. Because the
375 OK method directly used the fitted theoretical semivariogram model of the target variable to
376 predict the spatial distribution, its performance reflected the predictive ability of the original
377 data (Goovaerts, 1997). During the wet season, more factors (e.g., NH4N, NO3N, SOC, TSN
378 and SWC) influenced the spatial distributions of the N_2O fluxes than the dry season (Table 2

379 and Fig. 5). The values of the original data were concealed. Thus, other sophisticated kriging
380 methods, such as RK and CK, which reconcile the relationships between N_2O fluxes and
381 environmental factors, could be useful. The RK method performed better when elevation was
382 used as an auxiliary regression predictor during the dry season than when the chamber
383 placement position was used during the wet season (Table 4). This finding primarily occurred
384 because the chamber placement position was a categorical variable with a lower regression
385 fitting ability than elevation, which was a continuous variable (Goovaerts, 1997). The
386 performances of the CK with two groups of covariables during the wet season were better
387 than those of the CK with three groups of covariables during the dry season (Table 4).
388 Particularly, the CK with strongly correlated covariables of NO3N and NH4N (r = 0.70-0.71
389 and $p < 0.001$) (Fig. 5) performed the best (r = 0.74 and RMSE = 1.04) (Table 4).
390 Secondly, by comparing the performances of the three interpolation methods, the RK
391 and CK methods, which are more sophisticated kriging technologies, performed better than
392 the OK method for the dry and wet seasons. Similar results were obtained by previous
393 researchers (Stein et al., 1988; Odeh et al., 1995; Goovaerts, 1997; Hengl et al., 2004).When
394 comparing the performances of RK and CK, no differences were observed for the dry season.
395 However, during the wet season, the CK significantly outperformed the RK (Table 4). Overall,
396 few attempts have been made to provide a good method for selecting interpolation methods
397 between RK and CK (Kontters et al., 1995; Odeh et al. 1995). Li et al. (2013) suggested that
398 RK was a good choice because of the performance of the two interpolation methods and the
399 difficulties encountered when applying CK. However, in this study, the CK method was

400 better than the RK method because of its high predictive performance (Table 4), its readily
401 available required covariables (e.g., NH4N, NO3N and SOC) at co-locations, and because
402 expensive surface data were not needed (e.g., DEM and land use data, which are required by
403 RK) (Goovaerts, 1997; Webster and Oliver, 2001). Our conclusions were similar to those of
404 many previous studies that found that CK was the most versatile and rigorous statistical
405 technique for estimating spatial points (Stein et al., 1988; Odeh et al., 1995; Webster and
406 Oliver, 2001). For the application of CK, the covariables must show a correlation with the
407 target variable and present a similar spatial structure as the target variable (Odeh et al., 1995;
408 Goovaerts, 1997; Webster and Oliver, 2001). Therefore, we further compared the effects of
409 the two groups of covariables for CK in this study. We found that CK method with NH4Nt
410 and NO3Nt (showed significant correlations with FLUX30t) as covariables outperformed the
411 CK method with SOCt (presented a similar spatial structure to FLUX30t) as a covariable,
412 indicating that the feature correlation was more important than the similarity of the spatial
413 structure when selecting CK covariables. This finding can be regarded as a prerequisite for
414 selecting covariables for CK application.
415
416 *<Insert Table 4 near here>*
417
418 The three spatial interpolation methods predicted similar total N_2O emissions from the
419 tea-planted red soils in the 4.0 ha catchment on 30 October 2010 (in the dry season) and on
420 22 April 2012 (in the wet season), ranging from 21.2 to 22.1 g N d^{-1} and from 148.2 to 208.1

421 g N d^{-1} (Table 4), respectively. The predicted errors during the wet season were higher than
422 those of the dry season (Table 4). This result mainly occurred because fertilization was a
423 major factor that affected the N_2O emissions from the tea fields during the wet season.
424 Following fertilization, the horizontal and vertical movement of NH4N and NO3N in the
425 topsoil of the tea fields potentially produced the strong spatial heterogeneity of N_2O
426 emissions. In addition, it is possible that the variations in the availability of oxygen in the
427 soils was regulated by soil moisture, which determined the spatio-temporal heterogeneity of
428 N_2O emissions by inducing different degrees of soil nitrification and denitrification
429 (Davidson et al., 2000; Konda et al., 2010). Thus, spatial interpolation methods must be
430 chosen carefully to accurately estimate the spatial distribution of N_2O emissions when the
431 emissions are high and have strong spatial variability in the fields.
432
433 **5 Conclusions**
434 During the wet season of 2012, a 30-min one-time measurement of N_2O emissions from a 4.0
435 ha red-soil tea field in the subtropical region of central China were determined at 147 points.
436 The N_2O fluxes significantly varied with space. In addition, the N_2O fluxes were significantly
437 correlated with the NH4N, NO3N, SOC and TSN contents ($r > 0.27$ and $p < 0.001$). The
438 logit-transformed N_2O fluxes demonstrated a strong spatial dependency and were
439 characterized by an exponential semivariogram model with an effective range of 25.2 m.
440 Three spatial interpolation methods (OK, RK and CK) were used to predict the spatial
441 distribution of N_2O emissions. The RK and CK methods were relatively accurate for

predicting results. Although the N_2O emissions were much higher during the wet season than in the dry season, the N_2O 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.

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581 variability of nitrous oxide emissions and their soil-related determining factors in an
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583

584 Table 1. Descriptive statistics of the N_2O fluxes and environmental factors.

Variable[a]	Mean	Minimum	Maximum	CV (%)	Skewness of the original data	Skewness of the logit-transformed data
FLUX30	102.24[b]	-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	-

585 [a]FLUX30 is the N_2O flux (g N ha^{-1} d^{-1}); ELEVATION is the elevation (m); and BD, DOC,
586 NH4N, NO3N, SOC, TSN, SWC, SAND, SILT and CLAY are the soil bulk density (Mg m^{-3}),
587 soil dissolved organic carbon (mg C kg^{-1} soil), soil ammonium (mg N kg^{-1} soil), soil nitrate
588 (mg N kg^{-1} soil), soil organic carbon (g C kg^{-1} soil), soil total nitrogen (g N kg^{-1} soil),

589 gravimetric soil water (g H_2O g^{-1} soil), soil sand particle (%), soil silt particle (%) and soil

590 clay particle (%) content, respectively, of the 0-20 cm of topsoil.

591 [b] The median and standard deviation of the FLUX30 were 27.56 and 239.96 g N ha^{-1} d^{-1},

592 respectively.

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

Variable	Model	Nugget	Partial sill	Sill(nugget+ partial sill)	Distance Parameter (m)	Effective range (m)	Partial sill/sill
FLUX30t[a]	Exp	0	3.7186	3.7186	8.40	25.2	1.00
NH4Nt[a]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]
NO3Nt[a]	Ste	4.0794	0.6113	4.6907	91.92	163.7	0.13
SOCt[a]	Sph	1.1198	0.7744	1.8942	92.96	93.0	0.41
TSNt[a]	Ste	1.0422	0.2816	1.3238	57.97	102.6	0.21
SWCt[a]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]
SAND[a]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]
SILT[a]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]	ND[c]
FLUX30t[b]	Exp	1.1911	2.0560	3.2471	17.36	52.1	0.63
NH4Nt[b]	Exp	2.0473	0.7185	2.7658	17.36	52.1	0.26
NO3Nt[b]	Exp	1.6241	1.1188	2.7429	17.36	52.1	0.41
SOCt[b]	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[b]	ND	ND	ND	ND	ND	ND	ND

594 ND, not determined.

595 [a]Semivariogram models for the OK method.

[b]Semivariogram models for the RK method using the chamber placement position as the auxiliary regression predictor.

[c]Spatial structures were not apparent.

599 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

600

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

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

Method of spatial interpolation	Auxiliary variable	ME (no dimension)	RMSE (no dimension)	*r*	Predicted total N_2O emissions (g N d^{-1})
Dry season					
OK	-	0.0002	0.102	0.52	22.1[a]
RK	ELEVATION	0.0008	0.098	0.57	21.1[a]
CK	SOCt	0.0006	0.103	0.51	22.0[a]
CK	ELEV	0.0008	0.099	0.57	21.5[a]
CK	SOCt and ELEV	0.0009	0.098	0.57	21.2[a]
Wet season					
OK	-	-0.0005	1.739	0.18	208.1
RK	POSITION	-0.0006	1.549	0.49	148.2
CK	SOCt (POSITION)	0.0020	1.439	0.58	149.5
CK	NH4Nt (POSITION) and NO3Nt (POSITION)	0.0001	1.185	0.74	150.5

605 OK, RK and CK correspond to ordinary kriging, regression kriging and cokriging,
606 respectively; For the dry season campaign, ELEVATION, SOCt and ELEV are the normalized
607 elevation, the normalized soil organic carbon content and the inverse of the normalized

608 elevation, respectively. For the wet season campaign, SOCt, NH4Nt and NO3Nt are the
609 logit-transformations of soil organic carbon, soil ammonium and soil nitrate concentrations,
610 respectively. "POSITION" (in the parentheses) indicates the process of detrending the
611 influence of chamber placement position. The ME, RMSE, and r are the mean prediction
612 error, the root mean squared error (the mean squared deviation ratio of the prediction
613 residuals to the kriging standard errors), and the Pearson's correlation coefficient between the
614 observations and the predictions, respectively.
615 [a]The predicted total N_2O emissions during the dry season were recalculated because the study
616 area changed from 4.8 ha to 4.0 ha for the wet season.

617

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

622

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

624

625 Figure 3. Histograms of (**a**) the original N_2O fluxes (FLUX30) and (**b**) the logit-transformed
626 N_2O fluxes (FLUX30t).

627

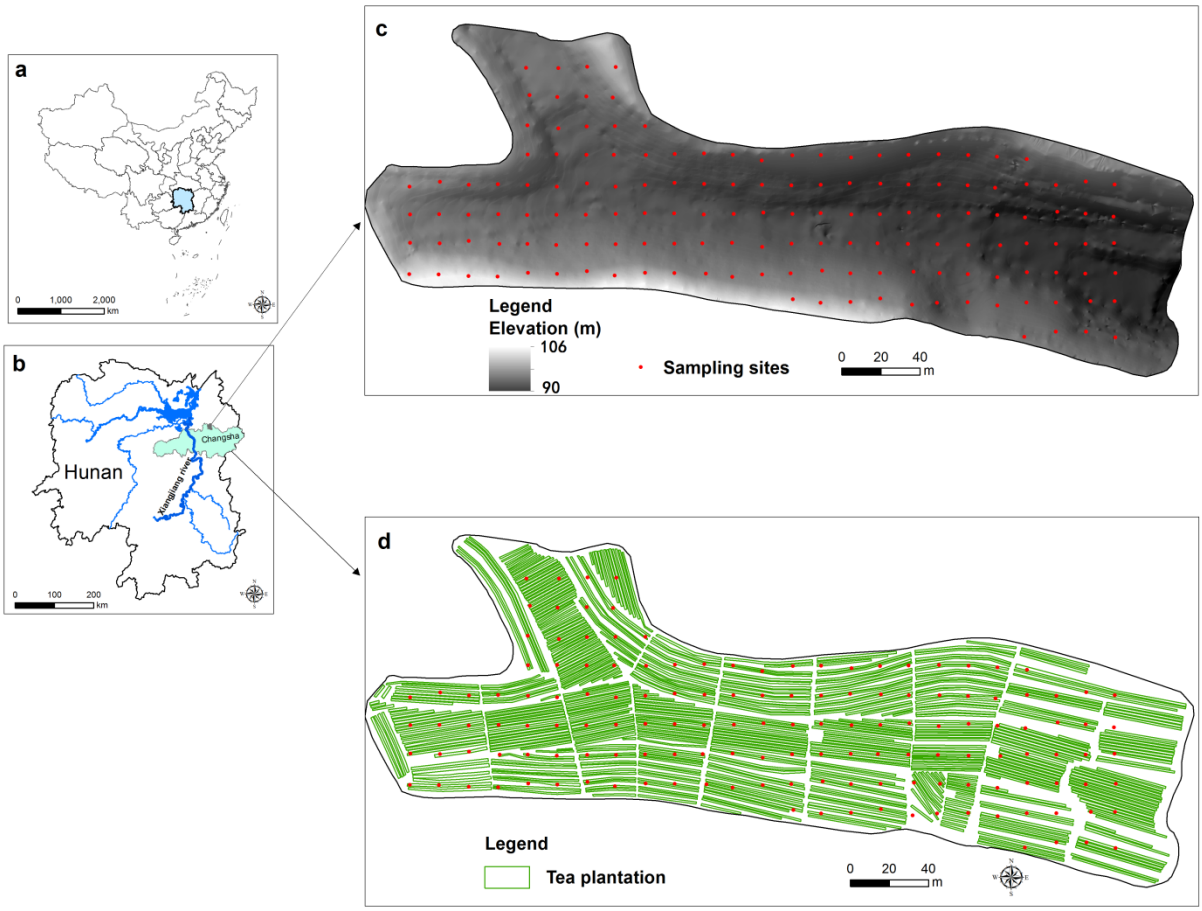
628 Figure 4. The Tukey's Honest Significant Difference analysis for FLUX30t, NH4Nt, NO3Nt,
629 SOCt, TSNt and SWCt based on the four-chamber placement positions (R, inter-row; F,
630 fertilization point; U, under tea tree; and I, in the tea tree row).

631

632 Figure 5. Correlation matrix with the Pearson's correlation coefficients (r) of the N_2O fluxes
633 and the environmental factors. All of the variables in the correlation matrix are normally
634 distributed. FLUX30 represents the N_2O flux (g N ha^{-1} d^{-1}); ELEVATION is the elevation (m);
635 and BD, DOC, NH4N, NO3N, SOC, TSN, SWC, SAND, SILT and CLAY are the soil bulk
636 density (Mg m^{-3}), soil dissolved organic carbon (mg C kg^{-1} soil), soil ammonium (mg N kg^{-1}
637 soil), soil nitrate (mg N kg^{-1} soil), soil organic carbon (g C kg^{-1} soil), total soil nitrogen (g N

638 kg^{-1} soil), gravimetric soil water (g H_2O g^{-1} soil), soil sand particle (%), soil silt particle (%)
639 and soil clay particle (%) contents of the top 0-20 cm of the soil, respectively. Furthermore, *,
640 ** and *** represent the statistical significance at probability levels of 0.05, 0.01 and 0.001,
641 respectively. The lowercase letter t represents the logit transformation.
642
643 Figure 6. Semivariograms (open circles) and best-fitted models (solid lines) of the normal
644 logit-transformed N_2O fluxes (FLUX30t) (no dimension) for ordinary kriging (**a**) and the
645 regression residuals of FLUX30t (no dimension) with chamber placement position as the
646 predictor for regression kriging (**b**).
647
648 Figure 7. Direct and cross-semivariograms (open circles, detrending the influence of chamber
649 placement position for cokriging) and the best-fitted linear model of the co-regionalization
650 (solid lines) of the normal logit-transformed N_2O fluxes (FLUX30t) (no dimension) and the
651 normal SOC (SOCt, no dimension). The linear model of co-regionalization was characterized
652 by using the same range and different sills for its component models.
653
654 Figure 8. Direct and cross-semivariograms (open circles, detrending the influence of chamber
655 placement position for cokriging) and the best-fit linear model of co-regionalization (solid
656 lines) for the normal logit-transformed N_2O fluxes (FLUX30t) (no dimension), NH4N
657 (NH4Nt, no dimension) and NO3N (NO3Nt, no dimension). The linear model of

658 co-regionalization was characterized by the same range and different sills for its component
659 models.
660
661 Figure 9. Spatial distributions of the N_2O fluxes as predicted by (**a**) OK, (**b**) RK with
662 chamber placement position as the regression predictor, (**c**) CK with SOCt (with the influence
663 of chamber placement position detrended) as the covariable, and (**d**) CK with NH4Nt (with
664 the influence of chamber placement position detrended) and NO3Nt (with the influence of
665 chamber placement position detrended) as two covariables. Here, SOCt, NH4Nt and NO3Nt
666 represent the logit-transformed soil organic carbon, soil ammonium and soil nitrate content,
667 respectively.
668
669 Figure 10. Spatial distributions of kriging variance of the N_2O fluxes as predicted by (**a**) OK,
670 (**b**) RK with chamber placement position as the regression predictor, (**c**) CK with SOCt (with
671 the influence of chamber placement position detrended) as the covariable, and (**d**) CK with
672 NH4Nt (with the influence of chamber placement position detrended) and NO3Nt (with the
673 influence of chamber placement position detrended) as two covariables. Here, SOCt, NH4Nt
674 and NO3Nt represent the logit-transformed soil organic carbon, soil ammonium and soil
675 nitrate content, respectively.
676
677



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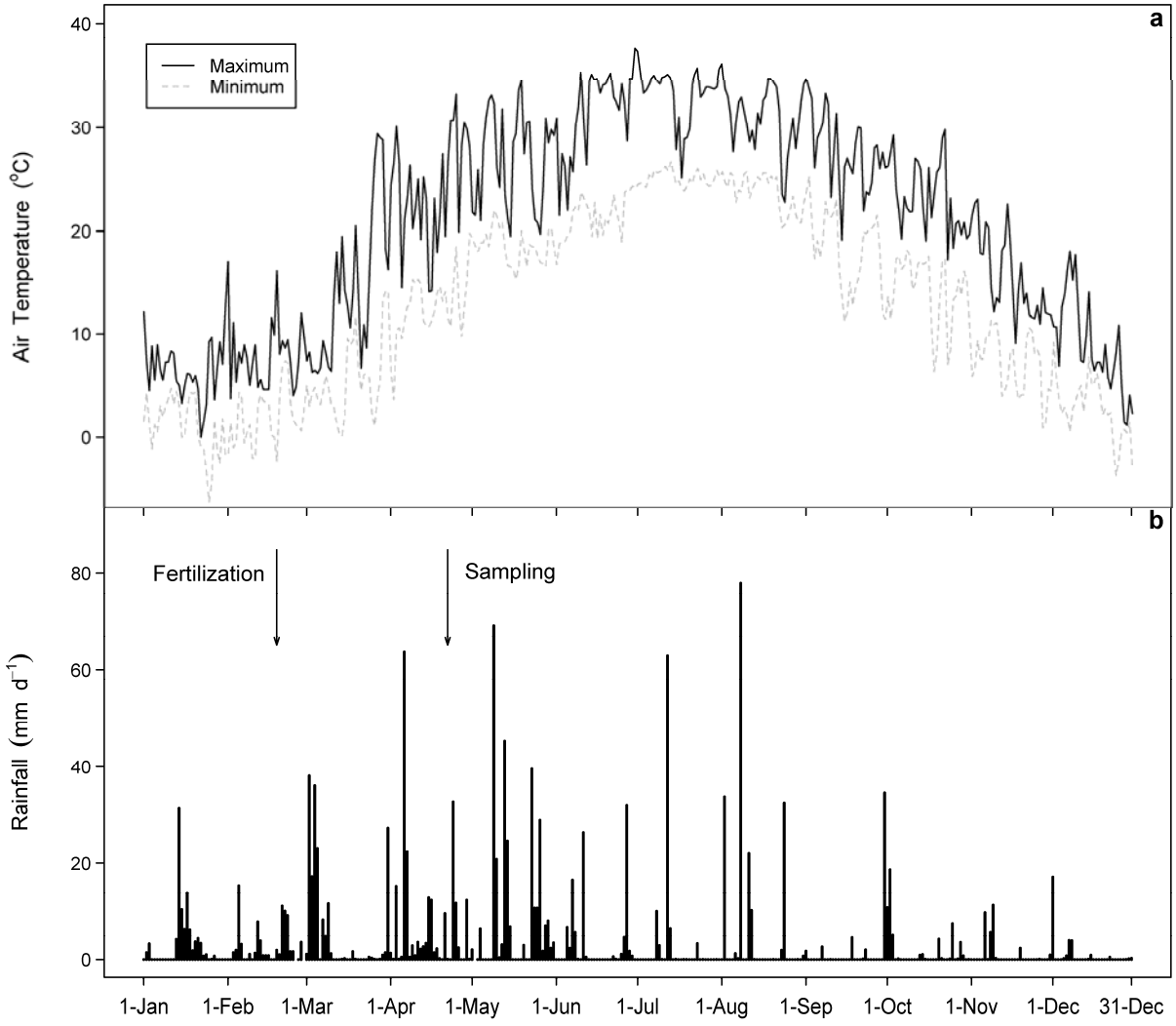
679 Figure 1.



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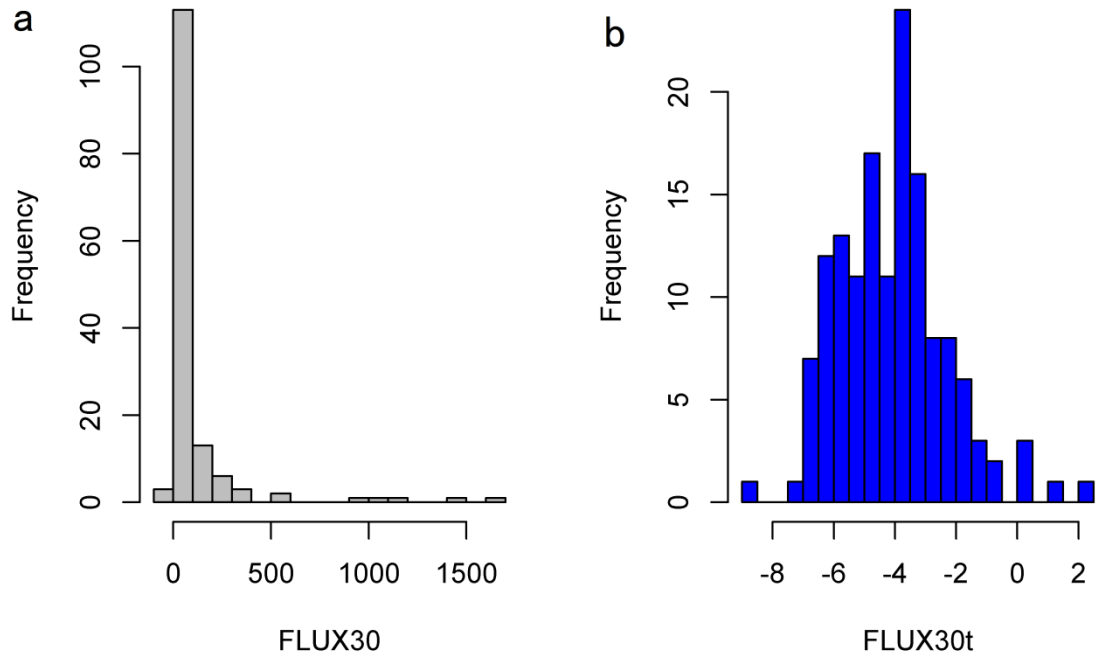
681 Figure 2.



682

683 Figure 3.

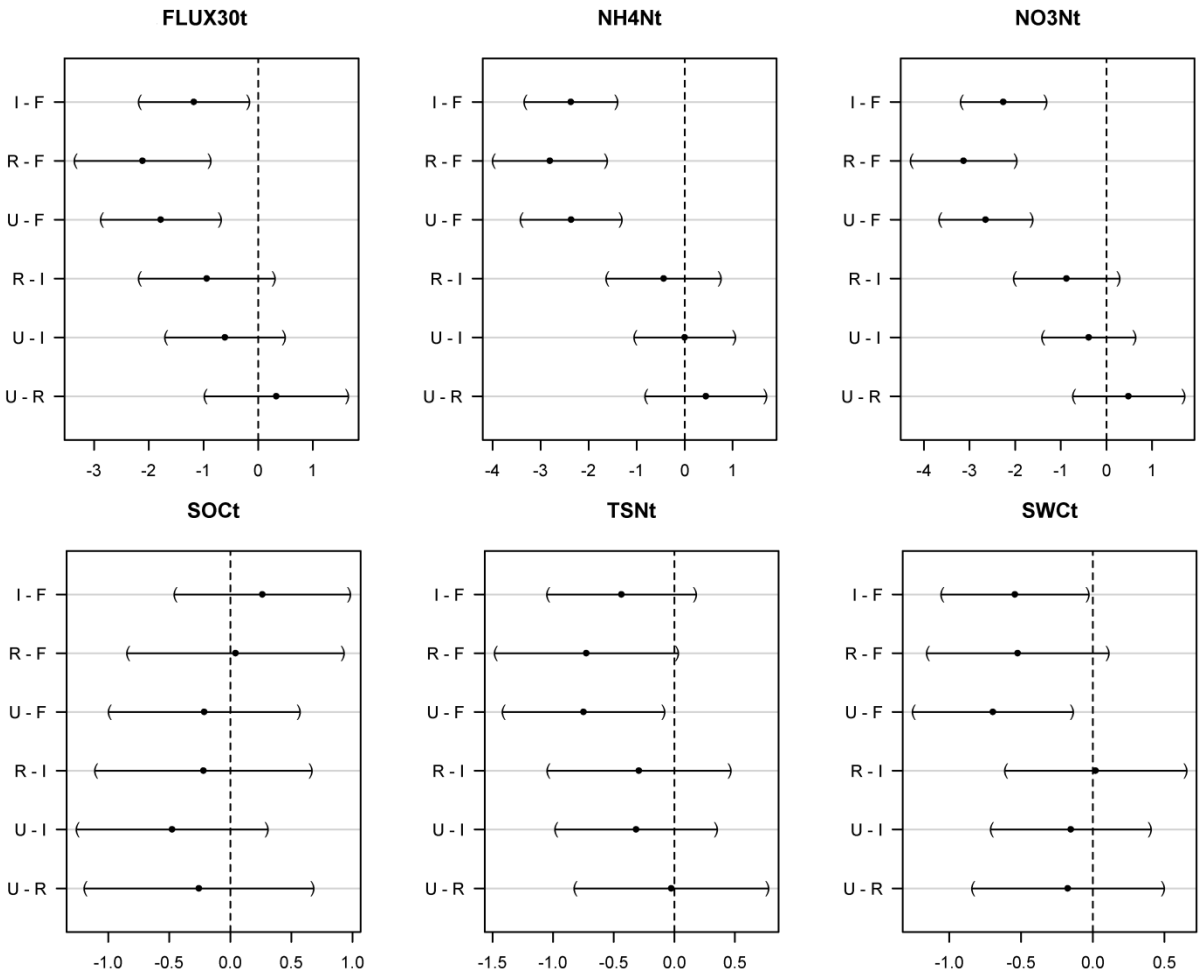


Figure 4.

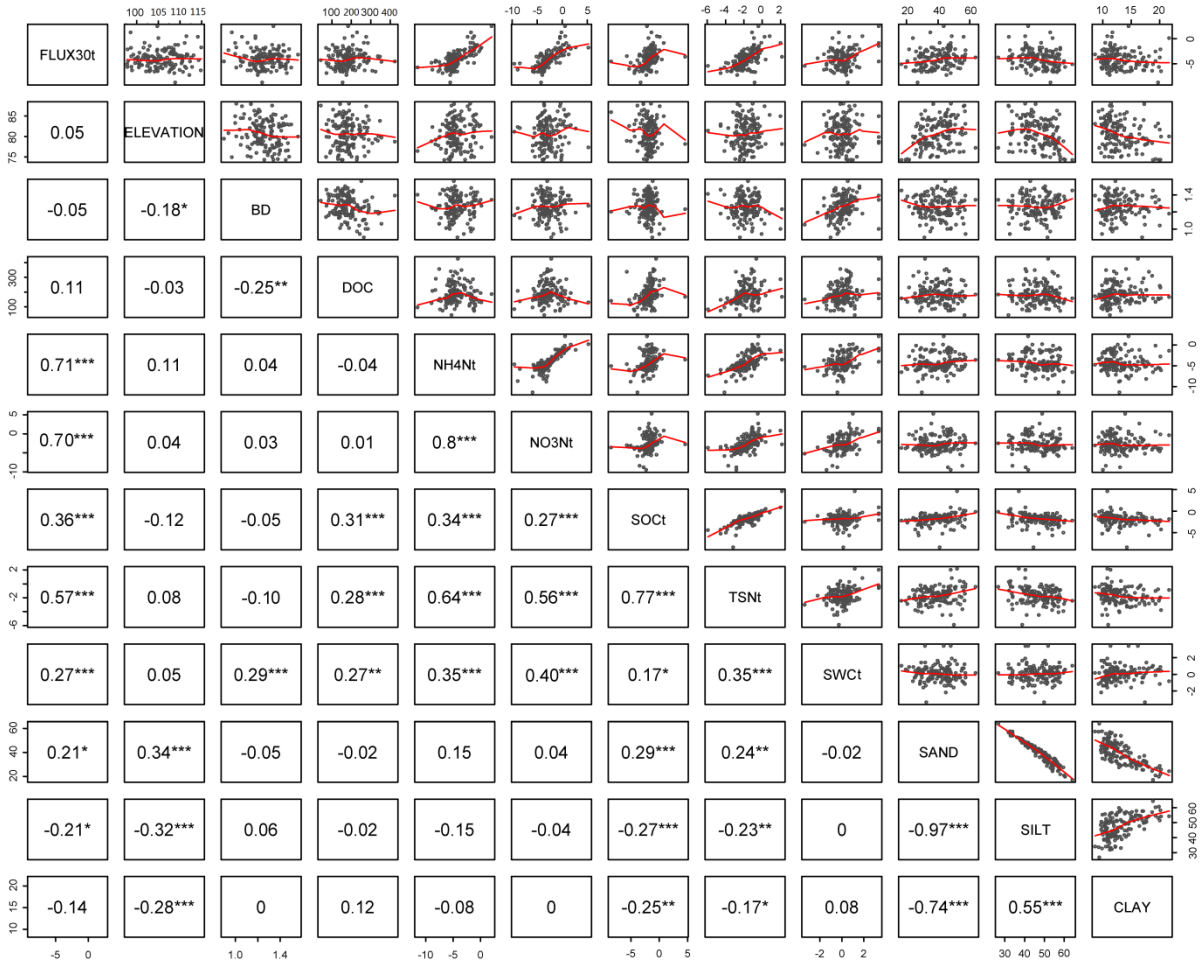
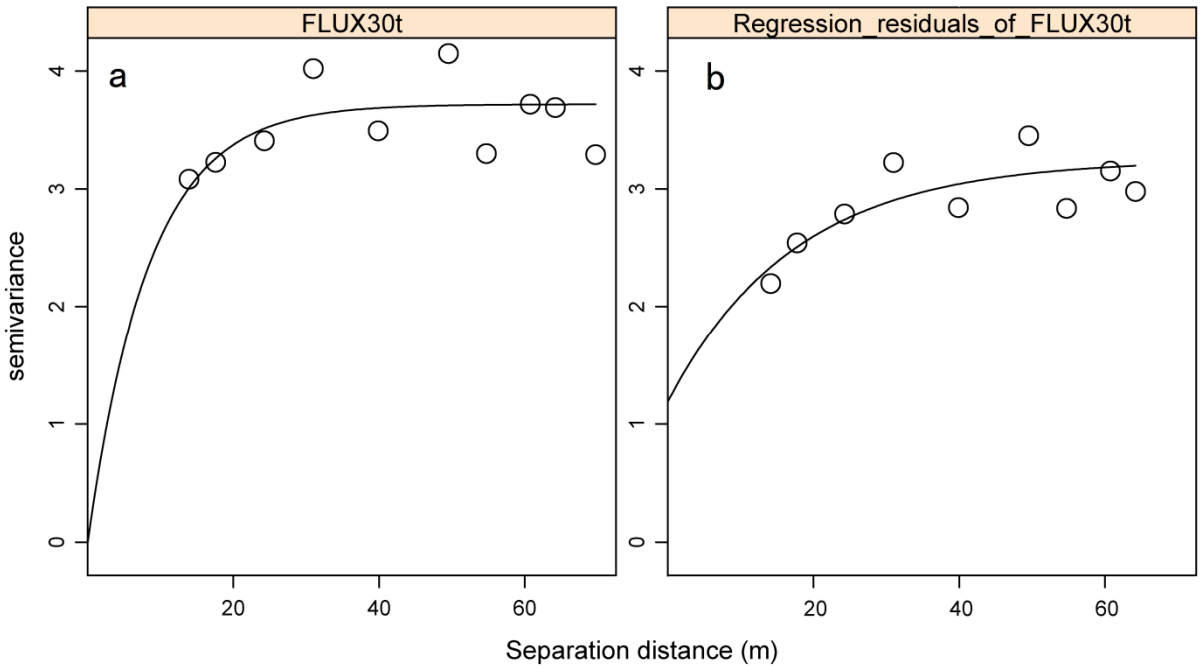

FLUX30t
ELEVATION
BD
DOC
NH4Nt
NO3Nt
SOCt
TSNt
SWCt
SAND
SILT
CLAY
0.05
-0.05
-0.18*
0.11
-0.03
-0.25**
0.71***
0.11
0.04
-0.04
0.70***
0.04
0.03
0.01
0.8***
0.36***
-0.12
-0.05
0.31***
0.34***
0.27***
0.57***
0.08
-0.10
0.28***
0.64***
0.56***
0.77***
0.27***
0.05
0.29***
0.27**
0.35***
0.40***
0.17*
0.35***
0.21*
0.34***
-0.05
-0.02
0.15
0.04
0.29***
0.24**
-0.02
-0.21*
-0.32***
0.06
-0.02
-0.15
-0.04
-0.27***
-0.23**
0
-0.97***
-0.14
-0.28***
0
0.12
-0.08
0
-0.25**
-0.17*
0.08
-0.74***
0.55***

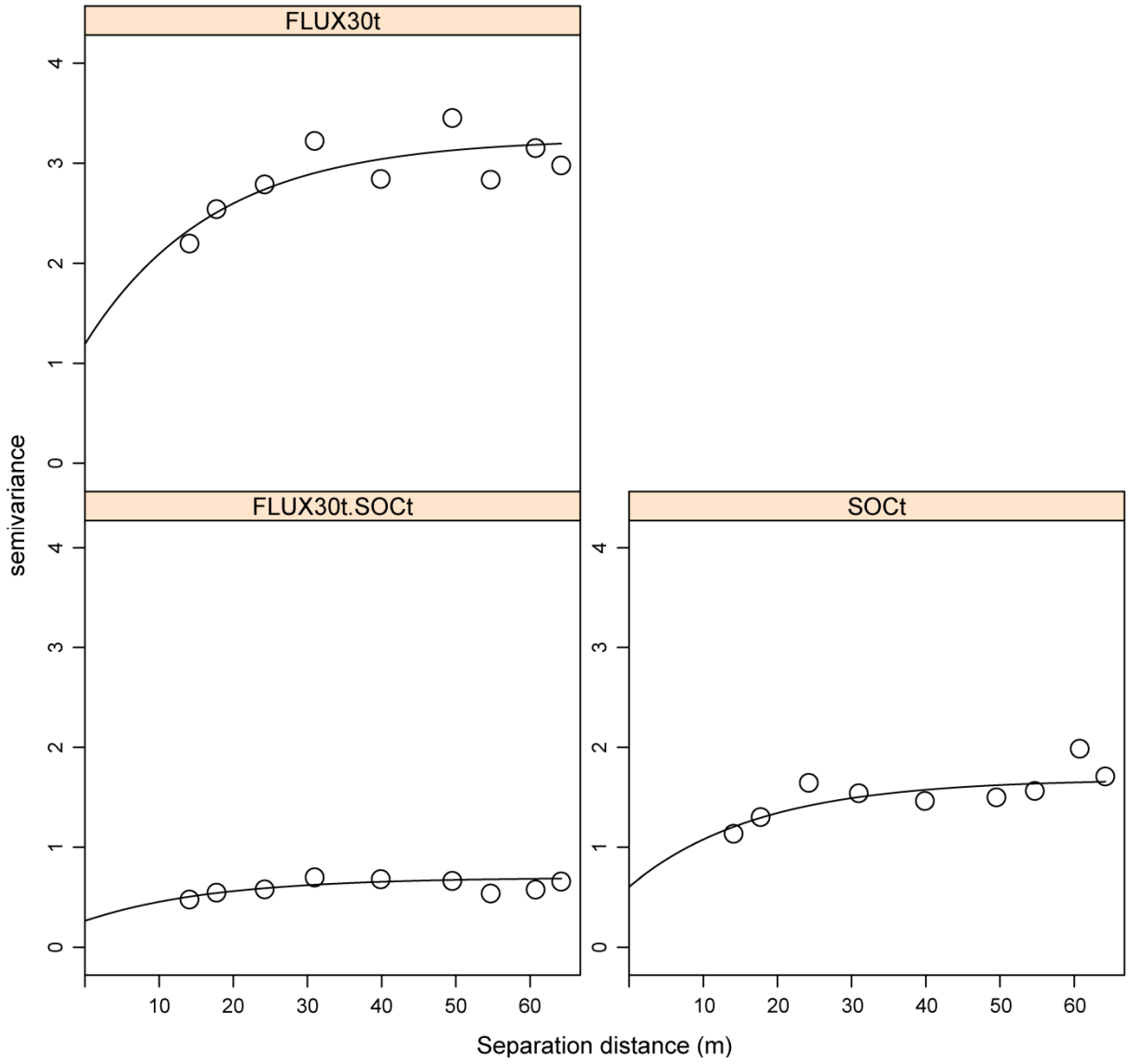
Figure 5.

689



690

691 Figure 6.

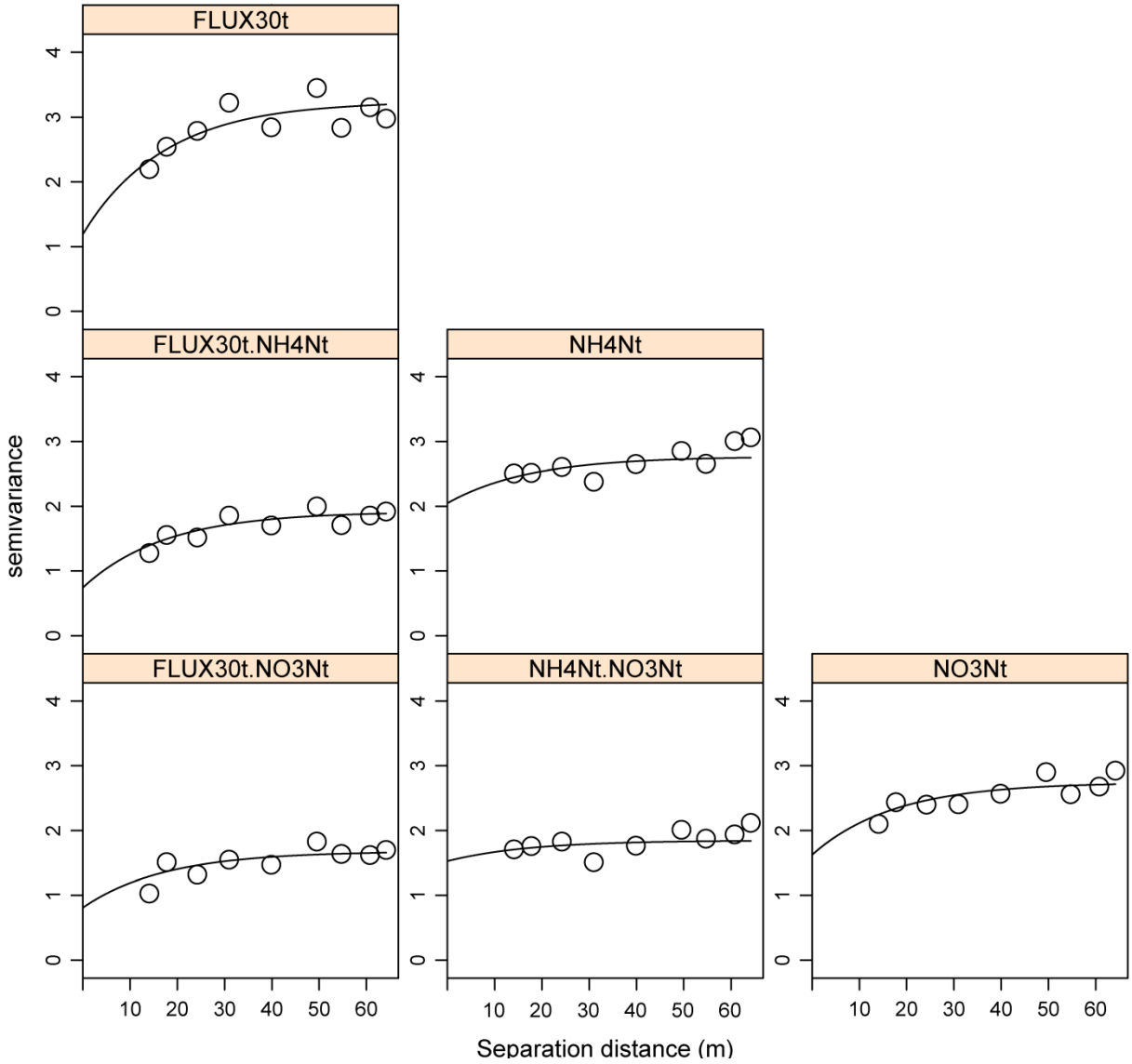


692

693

694 Figure 7.

695



696

697 Figure 8.

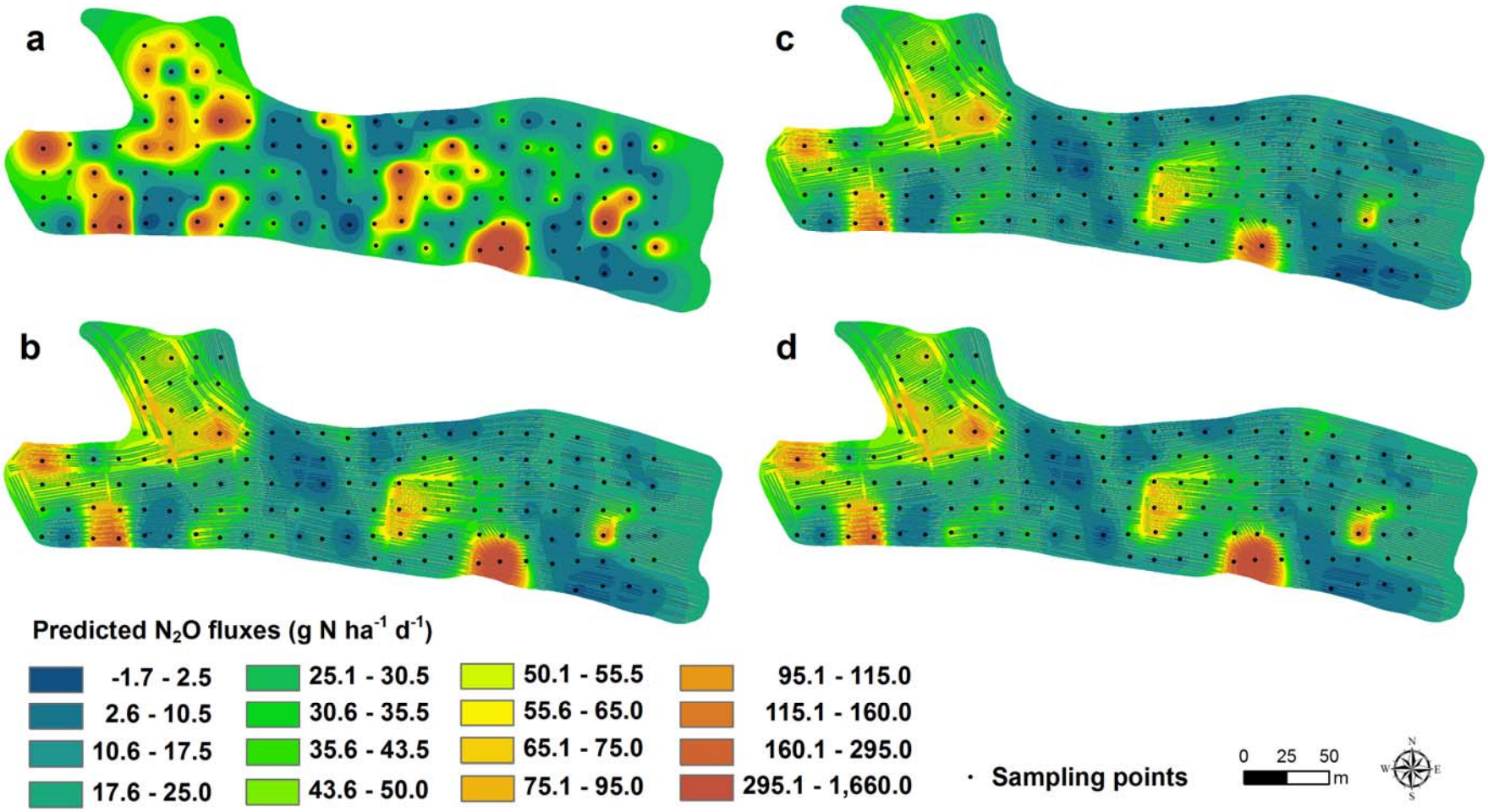

a
c
b
d
Predicted N_2O fluxes (g N ha^{-1} d^{-1})
-1.7 - 2.5
2.6 - 10.5
10.6 - 17.5
17.6 - 25.0
25.1 - 30.5
30.6 - 35.5
35.6 - 43.5
43.6 - 50.0
50.1 - 55.5
55.6 - 65.0
65.1 - 75.0
75.1 - 95.0
95.1 - 115.0
115.1 - 160.0
160.1 - 295.0
295.1 - 1,660.0
Sampling points
0 25 50
m
N
W
E
S

Figure 9.

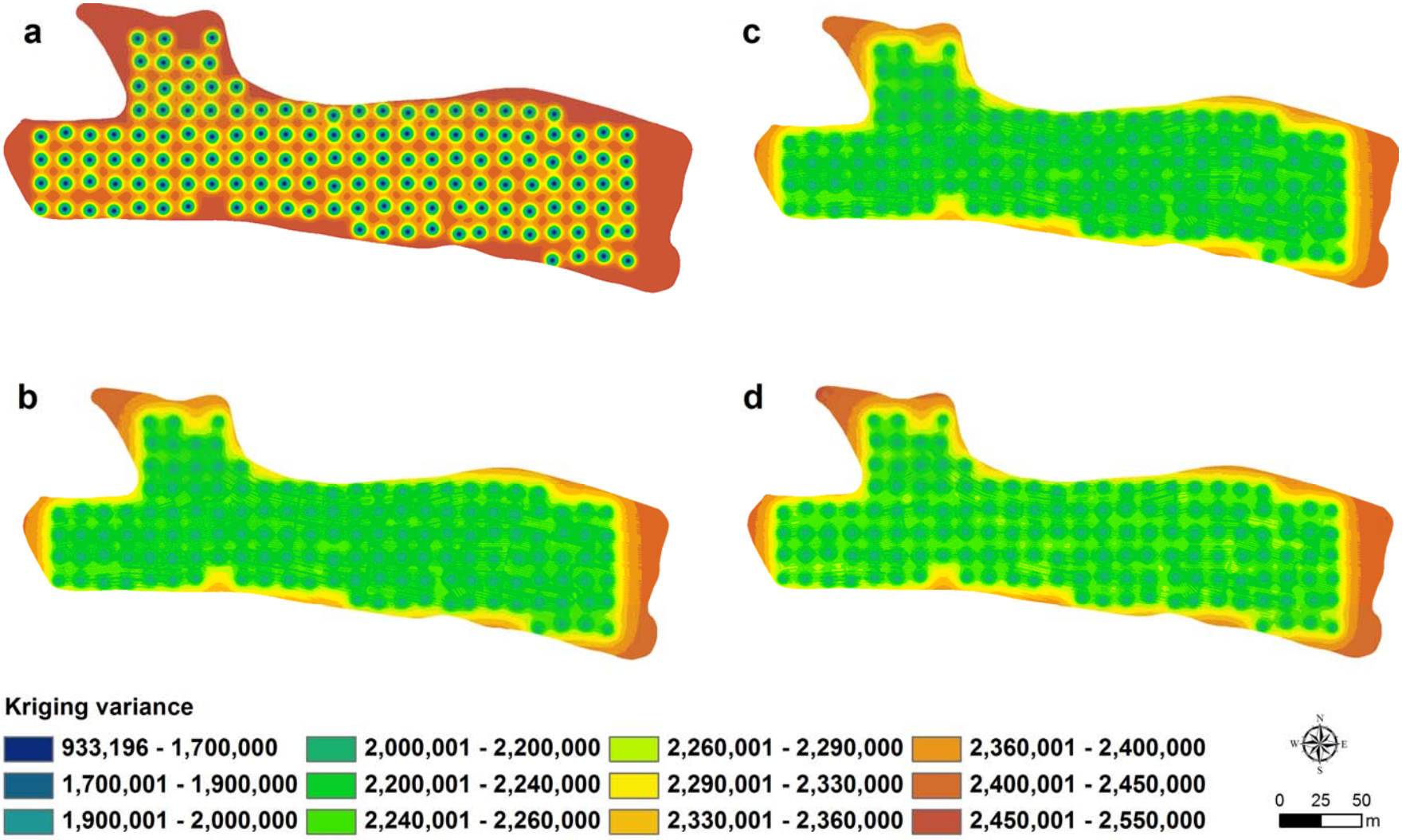

a
c
b
d
Kriging variance
933,196 - 1,700,000
1,700,001 - 1,900,000
1,900,001 - 2,000,000
2,000,001 - 2,200,000
2,200,001 - 2,240,000
2,240,001 - 2,260,000
2,260,001 - 2,290,000
2,290,001 - 2,330,000
2,330,001 - 2,360,000
2,360,001 - 2,400,000
2,400,001 - 2,450,000
2,450,001 - 2,550,000
N
W
E
S
0 25 50
m

Figure 10.