

1 Wet-season spatial variability of N₂O emissions from a tea field in subtropical central China

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18 **Abstract**

19 Tea fields emit large amounts of nitrous oxide (N₂O) to the atmosphere. Obtaining accurate
20 estimations of N₂O emissions from tea-planted soils is challenging due to strong spatial
21 variability. We examined the spatial variability of N₂O emissions from a red-soil tea field in
22 Hunan province, China, on 22 April 2012 (in a wet season) using 147 static mini chambers
23 approximately regular gridded in a 4.0 ha tea field. The N₂O fluxes for a 30-min snapshot
24 (10:00-10:30 am) ranged from -1.73 to 1,659.11 g N ha⁻¹ d⁻¹ and were positively skewed with
25 an average flux of 102.24 g N ha⁻¹ d⁻¹. The N₂O flux data were transformed to a normal
26 distribution by using a logit function. The geostatistical analyses of our data indicated that the
27 logit-transformed N₂O fluxes (FLUX30t) exhibited strong spatial autocorrelation, which was
28 characterized by an exponential semivariogram model with an effective range of 25.2 m. As
29 observed in the wet season, the logit-transformed soil ammonium-N (NH₄Nt), soil nitrate-N
30 (NO₃Nt), soil organic carbon (SOct), total soil nitrogen (TSNt) were all found to be
31 significantly correlated with FLUX30t ($r=0.57-0.71$, $p < 0.001$). Three spatial interpolation
32 methods (ordinary kriging, regression kriging and cokriging) were applied to estimate the
33 spatial distribution of N₂O emissions over the study area. Cokriging with NH₄Nt and NO₃Nt
34 as covariables ($r=0.74$ and RMSE=1.18) outperformed ordinary kriging ($r=0.18$ and
35 RMSE=1.74), regression kriging with the sample position as a predictor ($r=0.49$ and
36 RMSE=1.55) and cokriging with SOct as a covariable ($r=0.58$ and RMSE=1.44). The
37 predictions of the three kriging interpolation methods for the total N₂O emissions of 4.0 ha
38 tea field ranged from 148.2 to 208.1 g N d⁻¹, based on the 30 min snapshots obtained during

39 the wet season. Our findings suggested that to accurately estimate the total N₂O emissions
40 over a region, the environmental variables (e.g., soil properties) and the current land use
41 pattern (e.g., tea row transects in the present study) must be included in spatial interpolation.
42 Additionally, compared with other kriging approaches, the cokriging prediction approach
43 showed great advantages in being easily deployed, and more importantly providing accurate
44 regional estimation of N₂O emissions from tea-planted soils.

45

46 **Introduction**

47 According to the latest data, which show rapid increases in their atmospheric concentrations
48 (IPCC, 2013), nitrous oxide (N₂O), carbon dioxide (CO₂) and methane (CH₄) are three major
49 greenhouse gases in the atmosphere that significantly contribute to global warming. Among
50 these major greenhouse gases, N₂O has a very high radiative forcing per unit mass (265-fold
51 stronger than CO₂ on a 100 year horizon) and plays an important role in ozone depletion in
52 the stratosphere (Ravishankara et al., 2009). The primary sources of N₂O are from agriculture
53 development and the subsequent increased use of chemical N fertilizers (Ambus and
54 Christensen, 1994; Mosier et al., 1996, 1998; Yanai et al., 2003; Tokuda and Hayatsu, 2004;
55 Akiyama et al., 2006; Ravishankara et al., 2009). Agricultural soils produce 2.8 (1.7–4.8) Tg
56 of N₂O-N yr⁻¹ (IPCC, 2013). The N₂O is emitted from soils via the microbial processes of
57 nitrification under aerobic conditions and denitrification under anaerobic conditions
58 (Firestone and Davidson, 1989; Wrage et al., 2004). The magnitude of soil N₂O emissions is
59 highly variable and strongly influenced by changes in environmental conditions.

60 Among the different agricultural soils, tea-planted soils are important sources of N₂O that
61 are rapidly attracting attention due to recent large increases in the number of tea plantations
62 and large N fertilizer inputs ([Akiyama et al., 2006](#); [Lin and Han, 2009](#); [Fu et al., 2010, 2012](#);
63 [Hirono and Nonaka, 2012](#); [Han et al., 2013](#); [Li et al., 2013](#)). In China, the total tea-planted
64 area was approximately 2.10 million ha (mostly distributed in Fujian, Anhui, Zhejiang and
65 Hunan) in 2013 ([NBSC, 2014](#)). Compared with other agricultural soils, tea-planted soils
66 provide optimal conditions (e.g., low soil pH, high temperature and ample moisture) for
67 microbes to emit significant amounts of N₂O ([Hayatsu, 1993](#); [Venterea and Rolston, 2000](#); [Li](#)
68 [et al., 2013](#)). However, because few measurements of N₂O emissions from tea-planted soils
69 have been reported in China ([Fu et al., 2012](#); [Li et al., 2013](#); [Han et al., 2013](#)), it is difficult to
70 conduct precise spatial and temporal evaluations of N₂O emissions from tea-planted soils. To
71 estimate the N₂O emissions from tea-planted soils accurately and to understand the roles that
72 tea plantations play in global warming, it is necessary to investigate the spatial and temporal
73 patterns and related mechanisms of N₂O emissions from tea fields. This information will lead
74 to the development of effective land management options for mitigating N₂O emissions from
75 a significant source, tea plantation.

76 The N₂O fluxes have large spatial variability in agricultural soils ([Konda et al., 2008](#),
77 [2010](#); [Meda et al., 2012](#); [Li et al., 2013](#)). Many previous studies in tea fields have found
78 pronounced seasonal fluctuations in N₂O fluxes, with higher N₂O emissions during the wet
79 season than during the dry season ([Fu et al., 2012](#); [Han et al., 2013](#)). The seasonal and spatial
80 variability of N₂O emissions significantly contributes to the uncertainty when estimating the

81 contributions of subtropical tea-planted ecosystems to N₂O flux. Moreover, most of our
82 knowledge regarding seasonal changes and the spatial variability of N₂O fluxes is based on a
83 small number of measurements taken from tea-planted soils. [Li et al. \(2013\)](#) investigated the
84 spatial structure of N₂O fluxes for tea-planted soils during the dry season in October 2010
85 and found that the spatial distribution of the N₂O fluxes was primarily associated with field
86 elevation ($r=-0.42$, $p<0.001$). The other soil properties (e.g., soil organic carbon, soil water
87 and soil mineral nitrogen) were not significantly related to N₂O flux. To obtain a more
88 accurate evaluation of the interannual variability of N₂O emissions from tea-planted soils, a
89 study on the spatial structure and distribution of N₂O emissions during a wet season (in
90 contrast to the dry season) is necessary.

91 To understand the structure of the spatially distributed data and to predict the N₂O fluxes
92 at the unsampled locations, geostatistical analyses can be useful ([Goovaerts, 1997](#); [Webster
93 and Oliver, 2001](#)). Geostatistics provide statistical tools for describing the quantitative spatial
94 variability of field observations for the accurate mapping and planning of rational sampling
95 schemes that efficiently utilize the available labor ([Webster, 1985](#)). Several geostatistical
96 methods are used to examine the spatial variability of N₂O fluxes, including simple kriging
97 (SK), ordinary kriging (OK), regression kriging (RK) and cokriging (CK). The most
98 commonly used method is OK ([Clemens et al., 1999](#); [Röver et al., 1999](#); [Mathieu et al., 2006](#);
99 [Konda et al., 2008, 2010](#)), which uses the derived theoretical semivariogram models to
100 interpolate the spatial distribution of N₂O fluxes. However, research has demonstrated that
101 RK and CK approaches, which use related auxiliary variables, improve the prediction

102 accuracy (Goovaerts, 1997; Webster and Oliver, 2001; Hengl et al., 2004). The RK method
103 combines multiple regressions, including linear regressions, generalized linear models,
104 generalized added models and regression tree models, with the auxiliary variables used for
105 kriging (Odeh et al., 1994). In the RK method, linear regressions are commonly used. The
106 CK approach uses correlations that may exist between the predicted variables and other more
107 easily measured variables. These variables can be measured at the same points as the
108 predicted variable, at other points, or at both. Compared with the RK approach, the CK
109 approach is commonly applied when the measurement of a covariable is less expensive than
110 the cost of a predicted variable (Stein et al., 1988; Odeh et al., 1995). In addition to the
111 feature correlation as a criterion for selecting covariables, the CK approach also requires that
112 both of the predicted variable and covariables have similar spatial structures (Odeh et al.,
113 1994). In this study, we used three interpolation methods (OK, RK and CK) to estimate the
114 spatial distribution of N₂O fluxes in a tea field.

115 In contrast with the dry season, the spatial variability of the N₂O emissions was
116 investigated during the wet season in April 2012 from the same tea-planted catchment that
117 was studied by Li et al. (2013). The catchment consisted of a completely independent
118 hydrological system. Thus, the spatial distribution of the N₂O emissions within the catchment
119 was expected to have intrinsic characteristics. The objectives of this study were to (i) evaluate
120 the spatial variability of N₂O emissions from soils planted with tea in subtropical central
121 China during the wet season, (ii) determine the key environmental factors controlling N₂O
122 emissions, and (iii) assess the prediction efficiency of three kriging interpolation methods.

123

124 **2. Materials and Methods**

125 *2.1 Site description*

126 The field experiment was conducted in a small catchment (4.0 ha) in Jinjing, Changsha, in
127 Hunan province, China (28°32'50"N and 113°19'58" E and elevation 90 to 111 m) (Fig. 1).

128 The region has a subtropical monsoon climate with a mean annual air temperature of 17.5°C
129 and a mean annual precipitation of 1400 mm (average from 1979 to 2012). The site had four
130 distinct seasons: spring (February to April), summer (May to July), autumn (August to
131 November), and winter (December to January). On average, 70% of the annual precipitation
132 occurred in April, May and June. The daily air temperature and precipitation for 2012 were
133 recorded by an automatic weather station (Intelimet A, IMET-ADV2, Dynamax, USA)
134 located next to the studied catchment (Fig. 2). The soil of the catchment was a Haplic Alisol
135 (FAO/UNESCO soil taxonomy) that was derived from a granitic parental material. Tea
136 (*Camellia sinensis L., cv. Baihaozao*) was contour-planted 10 years ago using an inter-row
137 spacing of 0.5 m in the catchment.

138

139 *<Insert Fig. 1 & Fig. 2 near here>*

140

141 *2.2 Sampling positions*

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

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

157

158 *2.3 Gas and soil properties measurements*

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

166 equal to the chamber volume of 0.002651 m³. Gas samples were collected from the
167 headspace between 10:00 and 10:30 am. For simultaneous sampling, 25 skilled gas sampling
168 persons helped to accomplish the field sampling. Each person only took care of one column
169 containing 4 to 8 sampling positions (see Fig. 1), and started sampling at the same time of 10
170 am. At each point, three gas sample replicates were collected from the headspace into
171 pre-evacuated 12 mL vials (Exetainers, Labco, UK) at 0 and 30 min after the chamber body
172 was clipped. After collecting the gas samples, the air temperature in each chamber was
173 measured for subsequent correction of the flux calculation, and then three replicate soil cores,
174 0.05 m in diameter and 0.20 m in depth, were collected from the soils inside the mini
175 chambers. Soil samples were put straight into clean zip-lock bags for avoiding soil moisture
176 loss, and quickly transported back to the laboratory in thermal insulation boxes and stored in
177 a refrigeration room at 4 °C for preventing any microbial activity (such as mineralization,
178 nitrification and denitrification). The N₂O concentrations of the gas samples were analyzed
179 using a gas chromatograph (Agilent 7890A, Agilent, USA) that was fit with a ⁶³Ni-electron
180 capture detector and an automatic sample injector system. The N₂O fluxes (FLUX30, g N ha⁻¹
181 d⁻¹) were calculated as described by Li et al. (2013). The soil physical/chemical properties
182 determined by using fresh soil, e.g., the soil ammonium content (NH₄N), soil nitrate content
183 (NO₃N), soil dissolved organic carbon content (DOC), soil volumetric water content (SWC),
184 and soil bulk density (BD), were measured within three days after sampling, while those
185 using air-dried soil, e.g., total soil nitrogen content (TSN), soil organic carbon content (SOC)
186 and soil clay/silt/sand content (CLAY, SILT and SAND), were determined within two weeks

187 after the field work.

188

189 2.4 Data analyses

190 The descriptive statistical and geostatistical analyses were performed using R ([R](#)

191 [Development Core Team, 2014](#)) with the gstat package ([DGUU, 2010](#)).

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

203 In the geostatistical analyses, an experimental semivariogram of FLUX30t was
204 calculated, and the theoretical semivariogram models were fit. The ratio of the partial sill to
205 the total sill was used as an index of spatial dependence. [Armstrong \(1998\)](#) stated that a
206 variable with a higher ratio of partial sill to sill and a longer semivariogram range were more
207 structured. The spatial distribution of FLUX30t across the catchment was predicted using

208 three kriging interpolation methods (OK, RK and CK). These data were transformed back to
209 the original scale of FLUX30 for mapping. The Leave-One-Out cross-validation method was
210 used to evaluate the accuracy of interpolating FLUX30t using the three different kriging
211 methods.

212

213 **3. Results**

214 *3.1 Exploratory data analyses*

215 In the 4.0 ha tea-planted catchment, the N₂O fluxes during the 30-min one-time
216 measurements performed on 22 April 2012 ranged from -1.73 to 1,659.11 g N ha⁻¹ d⁻¹, with a
217 median value of 27.56 g N ha⁻¹ d⁻¹ and a CV of 234.7 % (Table 1). The N₂O flux data were
218 positively skewed (Table 1 and Fig. 3a), and their logit-transformations were approximately
219 normally distributed (Table 1 and Fig. 3b). From Table 3, the logit-transformed N₂O fluxes
220 (FLUX30t) were the highest in the fertilization points, and the differences in the FLUX30t
221 values among the chamber placement positions were statistically significant ($p < 0.001$).

222

223 *<Insert Table 1 & Fig. 3 near here>*

224

225 The ELEVATION, BD, DOC, SWC, SAND, SILT, and CLAY were approximately
226 normally distributed, with skewness values of less than 1 (Table 1). Additionally, DOC
227 displayed a moderate CV of 34.6 %, and the other variables had lower CVs (4.1–23.8 %). The
228 NH₄N, NO₃N, SOC and TSN were positively skewed, and the logit-transformations (NH₄Nt,

229 NO₃Nt, SO₄Ct and TSNt) had approximately normal distributions (Table 1). The NH₄N and
230 NO₃N had very high CVs (190.8 % and 141.6 %, respectively), and the SOC and TSN had
231 moderate CVs (50.1 % and 38.3 %, respectively).
232 The NH₄Nt, NO₃Nt, SO₄Ct, TSNt and SWC were significantly correlated with the N₂O fluxes
233 (Fig. 5), and the NH₄Nt, NO₃Nt and TSNt had strong positive relationships with N₂O ($r =$
234 0.71, 0.70 and 0.57, respectively, $p < 0.001$). The N₂O emissions and some soil properties
235 (NH₄N, NO₃N, SOC, TSN and SWC) in the fertilization points were significantly different
236 ($p < 0.001$) from the other three chamber placement positions (Fig. 6). These variables were
237 used as auxiliary covariables for the CK approach.

238

239 *<Insert Fig. 4 & Fig. 5 near here>*

240

241 *3.2 Spatial variability of N₂O emissions and related environmental factors*

242 Because most of the soil properties were significantly correlated with the chamber placement
243 positions, two types of semivariogram models were calculated for the N₂O and soil
244 parameters (correlated with N₂O fluxes) in the wet season (Table 2). The FLUX30t exhibited
245 strong spatial autocorrelation and was characterized by an exponential semivariogram model,
246 a theoretical distance parameter of 8.40 m (equivalent to an effective range of 25.2 m) and a
247 zero nugget. The NH₄Nt, SWCt, SAND and SILT showed almost no spatial dependency,
248 while NO₃Nt and TSNt demonstrated weak spatial dependency with a range parameter of
249 91.9 and 58.0 m, respectively (equivalent to an effective range of 163.7 and 102.6 m,

250 respectively).The SOCt exhibited a moderate spatial dependency within 93.0 m. By
251 detrending the influence of the chamber placement position, large changes in the
252 semivariogram models occurred regarding the above variables. Although the semivariograms
253 of the regression residuals of FLUX30t, NH4Nt, NO3Nt and SOCt were best-fit with the
254 same semivariogram model (exponential) with a similar range of 17.4 m (equivalent to an
255 effective range of 52.1 m), the spatial dependencies of those variables were different (Table
256 2). Of the soil properties, only SOCt had a similar spatial structure to FLUX30t when the
257 influence of the chamber placement position was detrended (Table 2). Based on these
258 correlation analyses and spatial variability analyses, the covariables for the CK method were
259 determined.

260

261 *<Insert Table 2 near here>*

262

263 *3.3 Spatial interpolation of N₂O emissions by three methods*

264 Three spatial interpolation methods were used in this study to predict the spatial distribution
265 of N₂O emissions from tea soils in the catchment. In the first method, the derived theoretical
266 semivariogram model for FLUX30t that is presented in Table 2 was used for the OK
267 prediction. In the second method, RK was used and the chamber placement position was
268 identified as the auxiliary regression predictor. Thus, the semivariogram of the regression
269 residuals of FLUX30t were calculated and best-fit with the theoretical semivariogram model
270 shown in Fig. 6. In the third method, CK involved two groups of covariables. As described

271 previously, because SOCt (detrending the influence of chamber placement position) showed a
272 similar spatial structure to FLUX30t (detrending the influence of chamber placing position), a
273 CK process was performed using SOCt as the covariable. Firstly, the direct and
274 cross-semivariograms of FLUX30t and SOCt (detrending the influence of the chamber
275 placement position) were calculated and best-fit with a linear model for co-regionalization
276 (LMC). Next, the fitted LMC was used to predict the spatial surface of N₂O emissions.
277 Because NH₄Nt and NO₃Nt were significantly correlated with FLUX30t (Fig. 5), a second
278 CK with NH₄Nt and NO₃Nt as the covariables was processed, similarly to that of the CK
279 with SOCt. However, these covariables had different spatial structures (Table 2). As reflected
280 by the lower root mean squared error (RMSE) and higher *r* values (Table 4), the CK method
281 performed better than the other spatial interpolation methods. Furthermore, the CK with
282 NH₄Nt and NO₃Nt as two covariables outperformed the CK with SOCt as the covariable.

283

284 *<Insert Figs.6-9 near here>*

285

286 As shown in Fig. 9, the surface map for the spatial distribution of N₂O emissions
287 interpolated by OK was rougher than the maps obtained from the other interpolation
288 approaches. The kriging standard deviation maps were showed in Fig. 10, and clearly
289 indicated that the RK and CK methods with lower kriging standard deviations outperformed
290 the OK method with higher kriging standard deviations. The four kriging interpolations of
291 OK, RK, CK with SOCt as the covariable and CK with NH₄Nt and NO₃Nt as the covariables

292 were able to predict that the total amount of N₂O emissions in the tea fields during the wet
293 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 N₂O emissions from
295 the tea field on 22 April 2012 during the wet season were approximately 150 g N d⁻¹.

296

297 *<Insert Fig. 10 near here>*

298

299 **Discussion**

300 *4.1 Seasonal differences of N₂O fluxes in the red soil planted with tea*

301 The N₂O 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 N₂O fluxes, we compared the
304 N₂O 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 N₂O 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 N₂O 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 N₂O 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 N₂O 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 N₂O 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 N₂O emissions (Folorunso and Rolston,
324 1984; Clemens et al., 1990; Velthof et al., 1996; Konda et al., 2008). During the wet season,
325 the N₂O 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 N₂O 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 N₂O 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 N₂O 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 N₂O fluxes in the tea-planted fields (Li et al., 2013). Similar spatial patterns of N₂O
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 N₂O 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 N₂O
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 N₂O 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 N₂O 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 N₂O
356 fluxes, we detrended the influences of the environmental factors when the N₂O flux
357 semivariograms were calculated to more deeply explore the spatial structures of the N₂O
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 N₂O flux semivariograms. Because the relationship between
361 chamber placement position and N₂O flux was more relevant than the relationship between
362 elevation and N₂O 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 N₂O emissions by three methods*

368 The three interpolation methods (OK, RK and CK) were used to predict the spatial
369 distributions of N₂O 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 N₂O 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 N₂O 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 N₂O 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 and
391 CK methods, which are more sophisticated kriging technologies, performed better than the
392 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 N₂O 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 N₂O 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 N₂O
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 N₂O 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 N₂O 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 N₂O 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 N₂O fluxes significantly varied with space. In addition, the N₂O fluxes were significantly
437 correlated with the NH₄N, NO₃N, SOC and TSN contents ($r > 0.27$ and $p < 0.001$). The
438 logit-transformed N₂O 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 N₂O emissions. The RK and CK methods were relatively accurate for
442 predicting results. Although the N₂O emissions were much higher during the wet season than
443 in the dry season, the N₂O emissions exhibited similar spatial structure during both seasons.
444 Such a phenomenon was mainly attributed to the low soil disturbance (e.g., only fertilizing
445 in a very small proportion of area and weeding) in the tea field.

446 To effectively mitigate high N₂O emissions from the tea field soils, the biological and
447 chemical mechanisms of N₂O emissions must be deeply explored. In addition, the responsive
448 land management practices, such as biochar application, deep fertilization (under 20 cm), the
449 use of controlled-release fertilizers and ecological engineering, must be recommended and
450 deployed, especially during the wet season.

451

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583

584 Table 1 Descriptive statistics of the N₂O 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 ^aFLUX30 is the N₂O 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 ($\text{g H}_2\text{O g}^{-1}$ soil), soil sand particle (%), soil silt particle (%) and soil
590 clay particle (%) content, respectively, of the 0-20 cm of topsoil.

591 ^bThe median and standard deviation of the FLUX30 were 27.56 and 239.96 $\text{g N ha}^{-1} \text{d}^{-1}$,
592 respectively.

593 Table 2 Semivariogram models for N₂O 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 ^aSemivariogram models for the OK method.

596 ^bSemivariogram models for the RK method using the chamber placement position as the
597 auxiliary regression predictor.

598 ^cSpatial structures were not apparent.

599

600 Table 3 Statistics for N₂O 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

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

603

604 Table 4 Cross-validations of the three different kriging interpolations for N₂O fluxes during
 605 the dry and wet seasons.

Method of spatial interpolation	Auxiliary variable	ME (no dimension)	RMSE (no dimension)	<i>r</i>	Predicted total N ₂ O 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

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

616 ^aThe predicted total N₂O emissions during the dry season were recalculated because the study
617 area changed from 4.8 ha to 4.0 ha for the wet season.

618

619 **Figure captions**

620

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

625

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

627

628 Figure 3. Histograms of **(a)** the original N₂O fluxes (FLUX30) and **(b)** the logit-transformed
629 N₂O fluxes (FLUX30t).

630

631 Figure 4. The Tukey's Honest Significant Difference analysis for FLUX30t, NH₄Nt, NO₃Nt,
632 SO₄t, TSNt and SWCt based on the four-chamber placement positions (R, inter-row; F,
633 fertilization point; U, under tea tree; and I, in the tea tree row).

634

635 Figure 5. Correlation matrix with the Pearson's correlation coefficients (r) of the N₂O fluxes
636 and the environmental factors. All of the variables in the correlation matrix are normally
637 distributed. FLUX30 represents the N₂O flux (g N ha⁻¹ d⁻¹); ELEVATION is the elevation (m);
638 and BD, DOC, NH₄N, NO₃N, SOC, TSN, SWC, SAND, SILT and CLAY are the soil bulk
639 density (Mg m⁻³), soil dissolved organic carbon (mg C kg⁻¹ soil), soil ammonium (mg N kg⁻¹

640 soil), soil nitrate (mg N kg^{-1} soil), soil organic carbon (g C kg^{-1} soil), total soil nitrogen (g N
641 kg^{-1} soil), gravimetric soil water ($\text{g H}_2\text{O g}^{-1}$ soil), soil sand particle (%), soil silt particle (%)
642 and soil clay particle (%) contents of the top 0-20 cm of the soil, respectively. Furthermore, *,
643 ** and *** represent the statistical significance at probability levels of 0.05, 0.01 and 0.001,
644 respectively. The lowercase letter t represents the logit transformation.

645

646 Figure 6. Semivariograms (open circles) and best-fitted models (solid lines) of the normal
647 logit-transformed N_2O fluxes (FLUX30t) (no dimension) for ordinary kriging (**a**) and the
648 regression residuals of FLUX30t (no dimension) with chamber placement position as the
649 predictor for regression kriging (**b**).

650

651 Figure 7. Direct and cross-semivariograms (open circles, detrending the influence of chamber
652 placement position for cokriging) and the best-fitted linear model of the co-regionalization
653 (solid lines) of the normal logit-transformed N_2O fluxes (FLUX30t) (no dimension) and the
654 normal SOC (SOCt, no dimension). The linear model of co-regionalization was characterized
655 by using the same range and different sills for its component models.

656

657 Figure 8. Direct and cross-semivariograms (open circles, detrending the influence of chamber
658 placement position for cokriging) and the best-fit linear model of co-regionalization (solid
659 lines) for the normal logit-transformed N_2O fluxes (FLUX30t) (no dimension), NH_4N
660 (NH_4Nt , no dimension) and NO_3N (NO_3Nt , no dimension). The linear model of

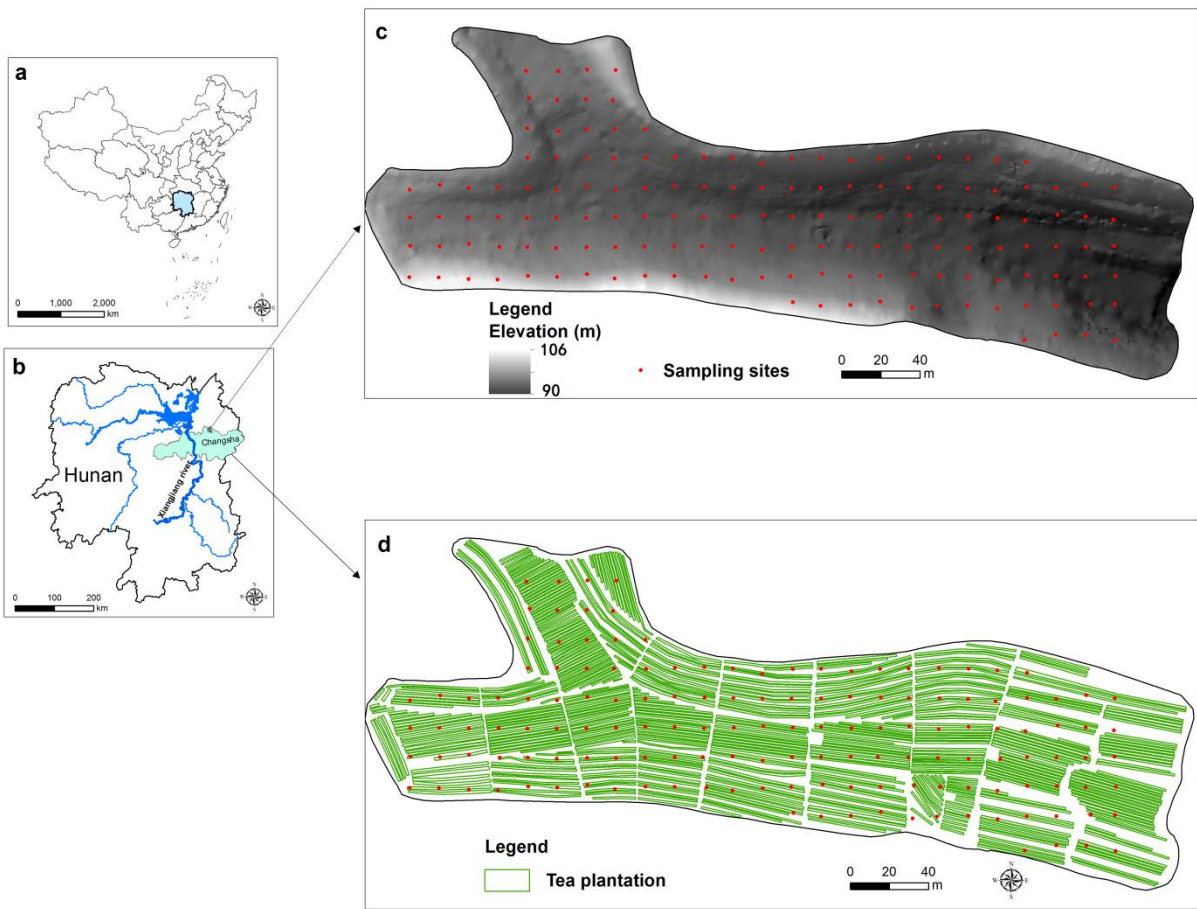
661 co-regionalization was characterized by the same range and different sills for its component
662 models.

663

664 Figure 9. Spatial distributions of the N₂O fluxes as predicted by **(a)** OK, **(b)** RK with
665 chamber placement position as the regression predictor, **(c)** CK with SOCt (with the influence
666 of chamber placement position detrended) as the covariable, and **(d)** CK with NH₄Nt (with
667 the influence of chamber placement position detrended) and NO₃Nt (with the influence of
668 chamber placement position detrended) as two covariables. Here, SOCt, NH₄Nt and NO₃Nt
669 represent the logit-transformed soil organic carbon, soil ammonium and soil nitrate content,
670 respectively.

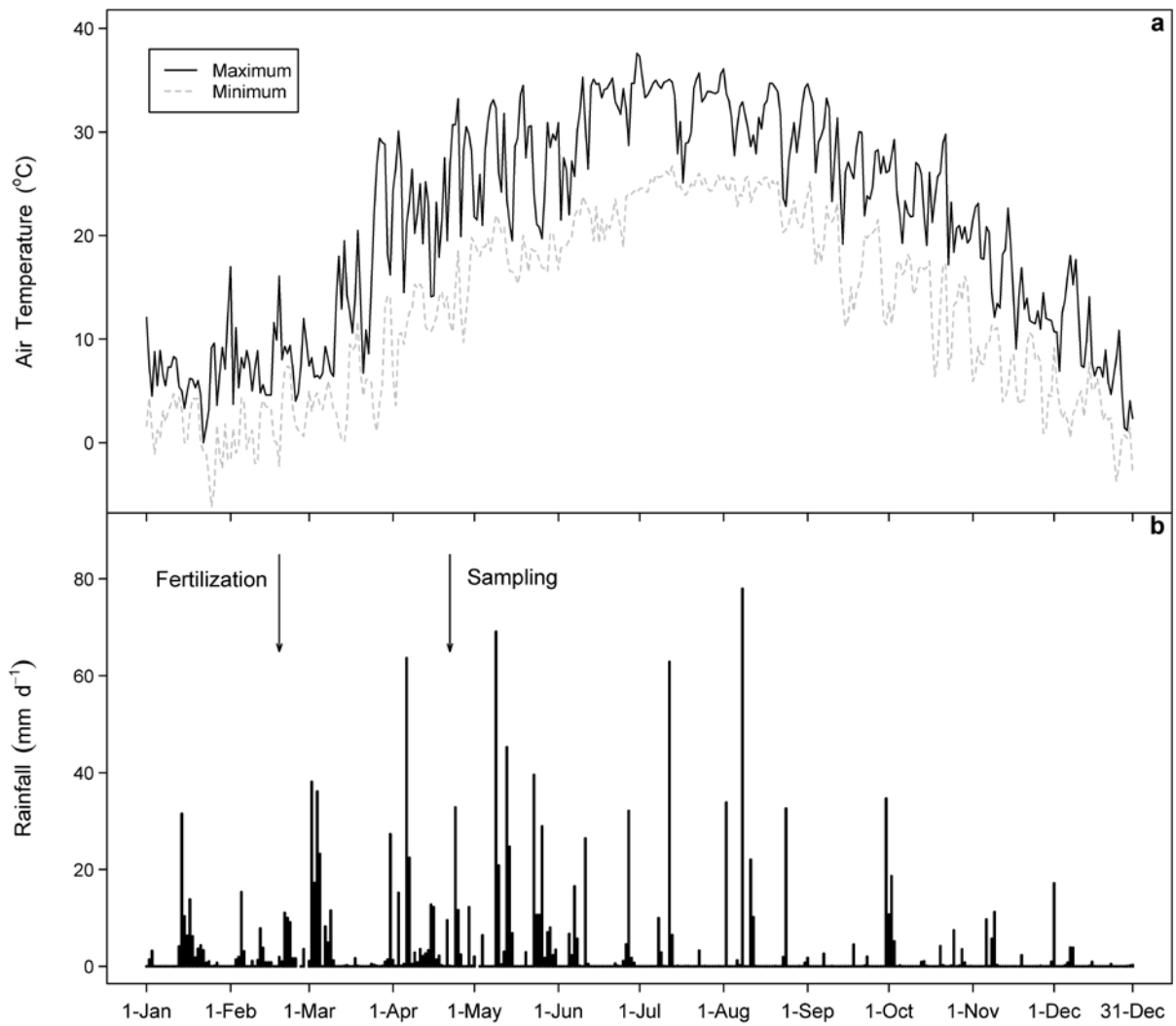
671

672 Figure 10. Spatial distributions of kriging standard deviations of the predicted N₂O fluxes by
673 **(a)** OK, **(b)** RK, **(c)** CK with SOCt as the covariable, and **(d)** CK with NH₄Nt and NO₃Nt as
674 two covariables.



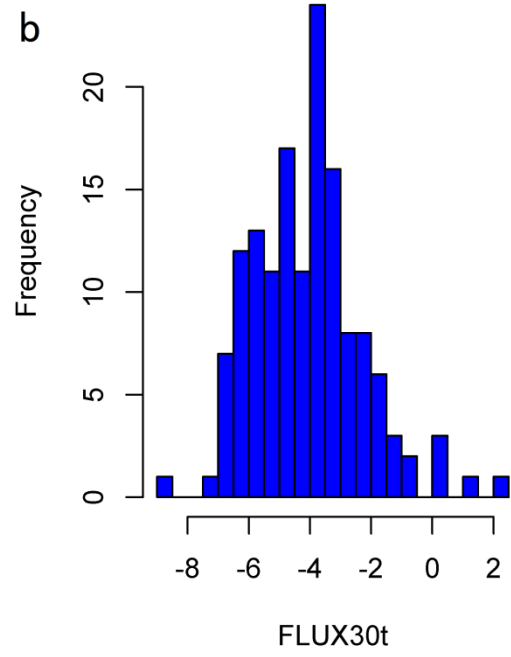
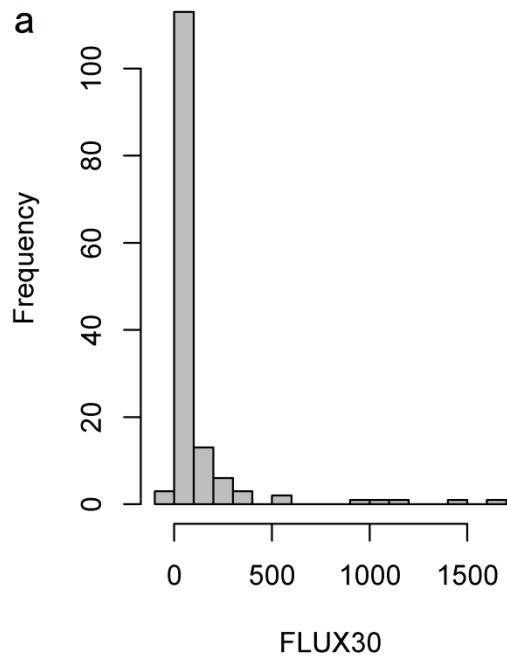
675

676 Figure 1



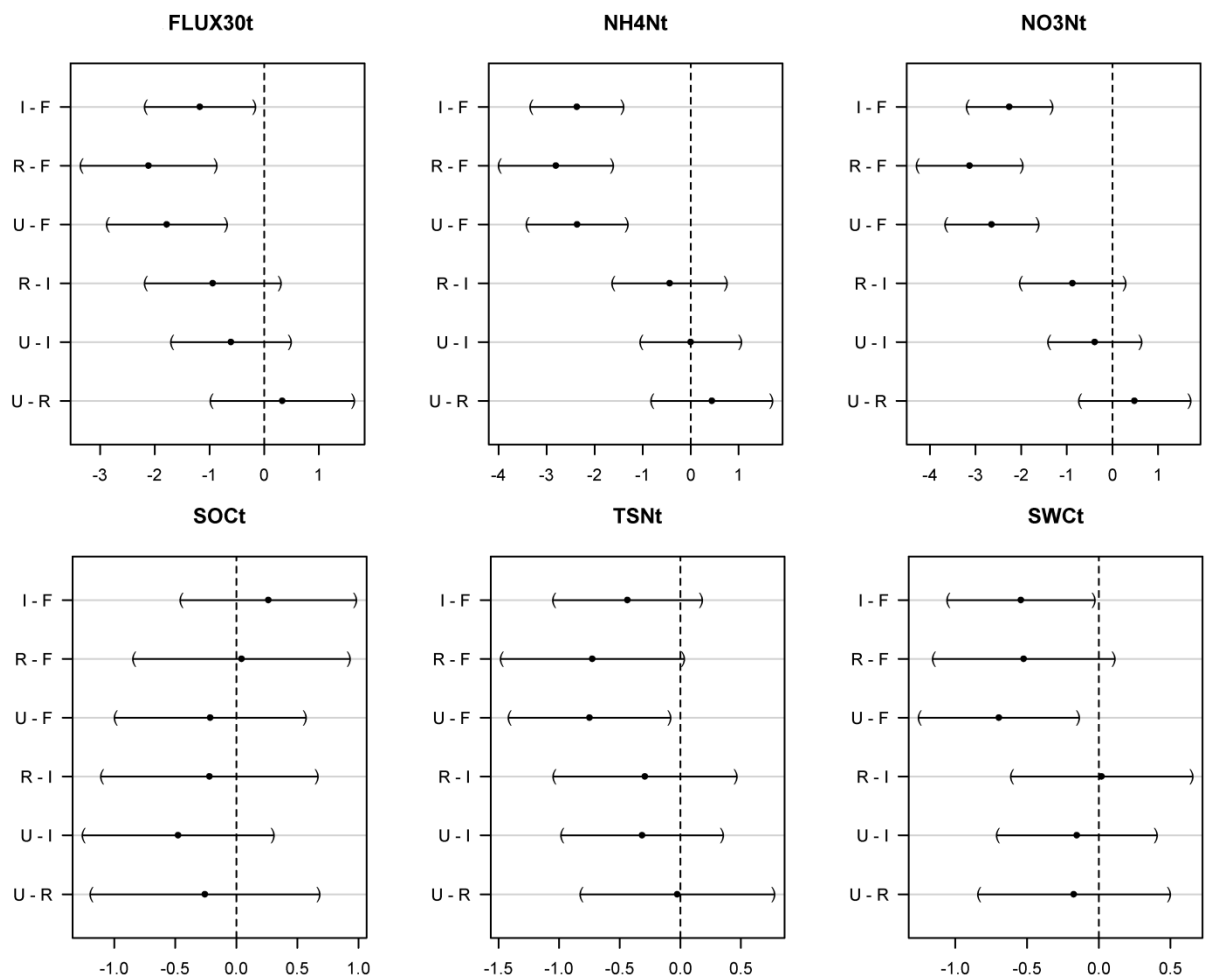
677

678 Figure 2



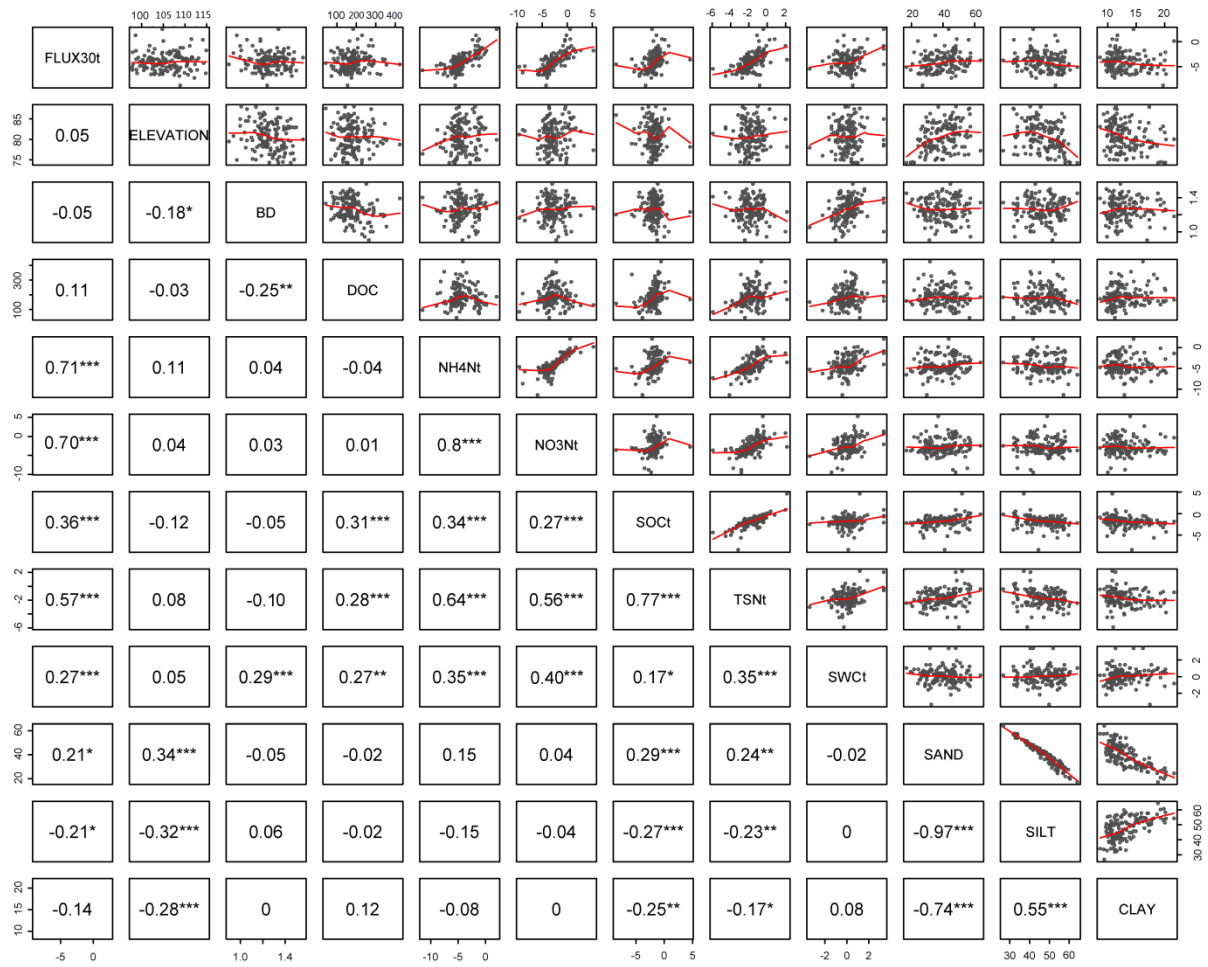
679

680 Figure 3



681

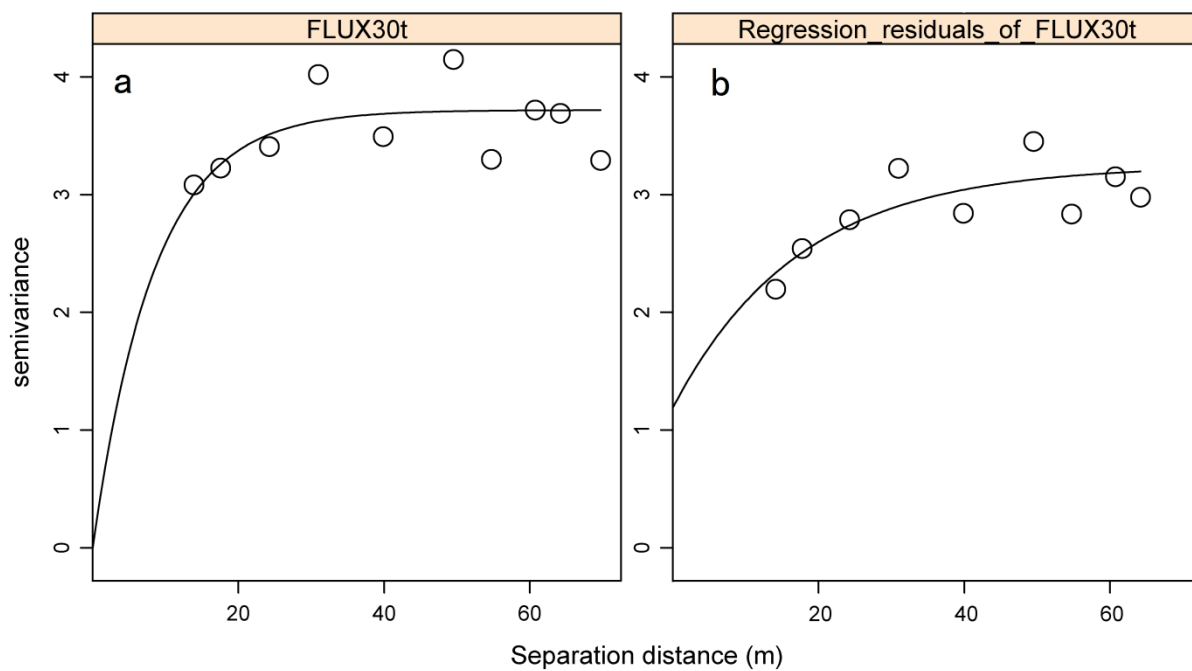
682 Figure 4



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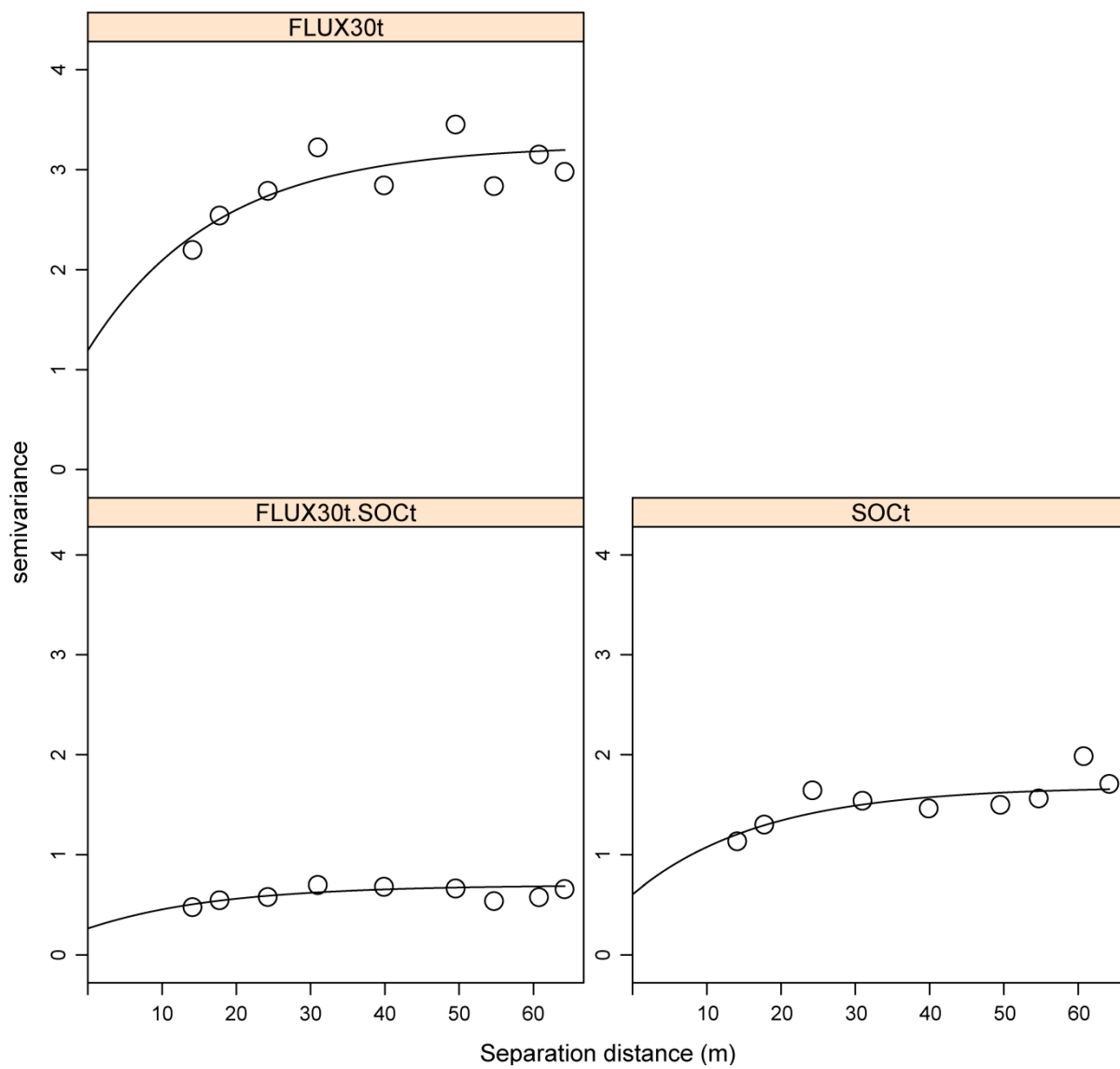
684 Figure 5

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686

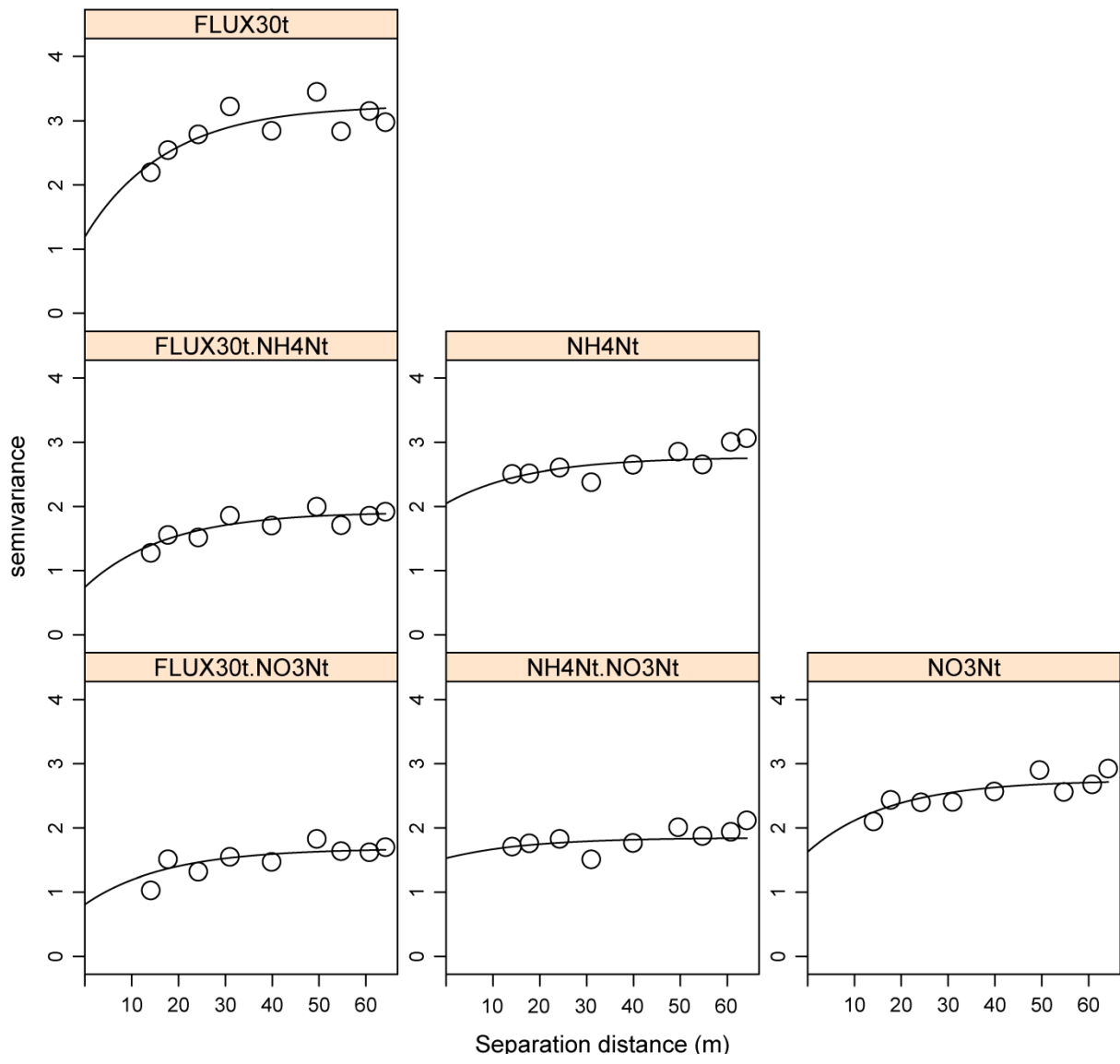
687 Figure 6



688

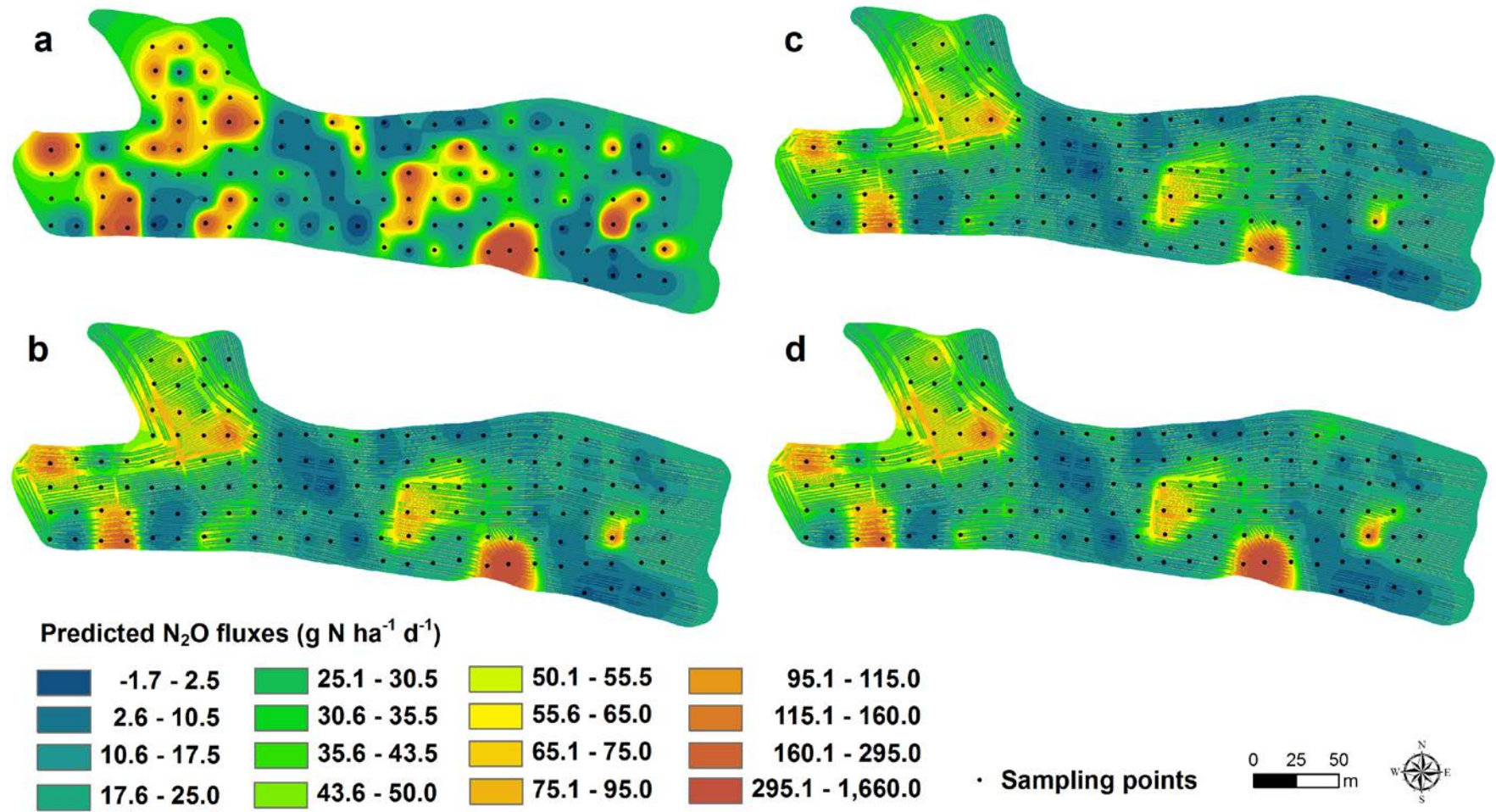
689 Figure 7

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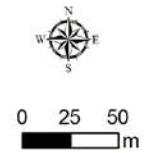
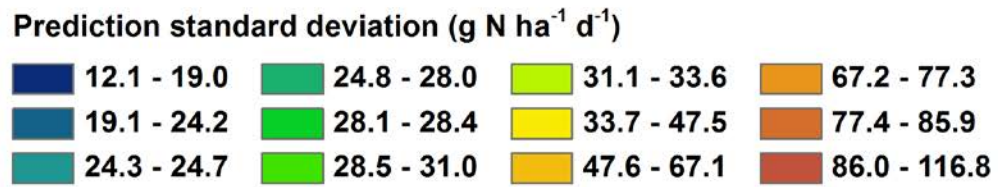
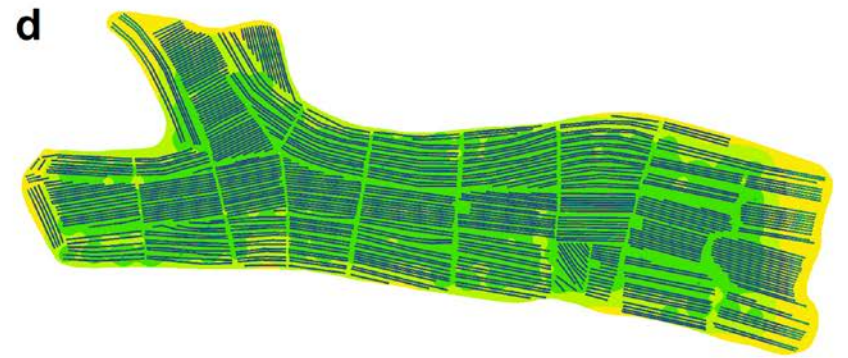
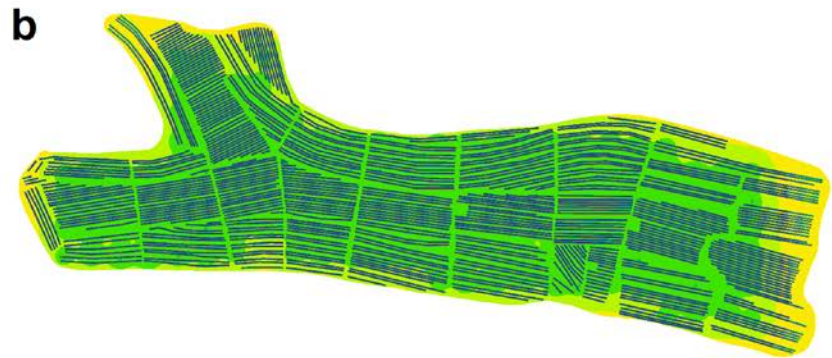
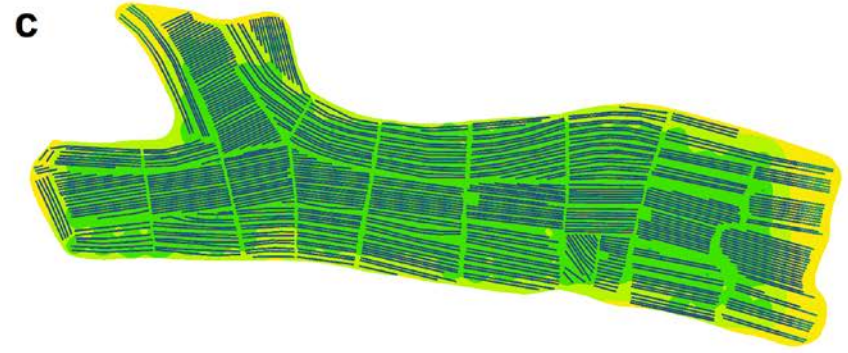
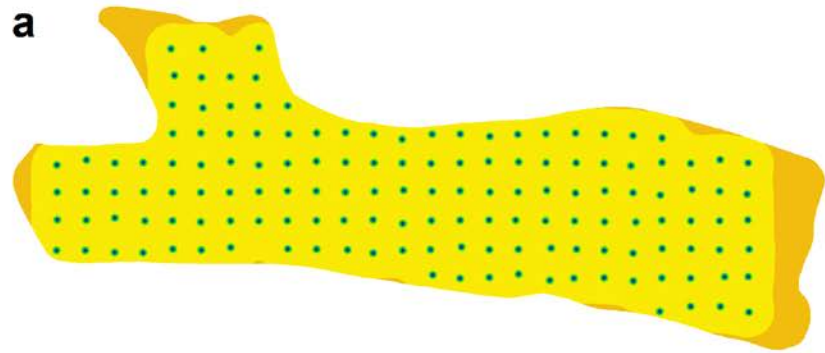
692 Figure 8



693

694 Figure 9

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696
697 Figure 10