

Scotland's Rural College

Gross N transformation rates and related N₂O emissions in Chinese and UK agricultural soils

Zhu, Gaodi; Song, Xiaotong; Ju, Xiaotang; Zhang, Jinbo; Muller, Christoph; Sylvester-Bradley, Roger; Thorman, Rachel; Bingham, IJ; Rees, RM

Published in:

Science of the Total Environment

DOI:

[10.1016/j.scitotenv.2019.02.241](https://doi.org/10.1016/j.scitotenv.2019.02.241)

Print publication: 20/05/2019

Document Version

Peer reviewed version

[Link to publication](#)

Citation for published version (APA):

Zhu, G., Song, X., Ju, X., Zhang, J., Muller, C., Sylvester-Bradley, R., Thorman, R., Bingham, IJ., & Rees, RM. (2019). Gross N transformation rates and related N₂O emissions in Chinese and UK agricultural soils. *Science of the Total Environment*, 666, 176-186. <https://doi.org/10.1016/j.scitotenv.2019.02.241>

General rights

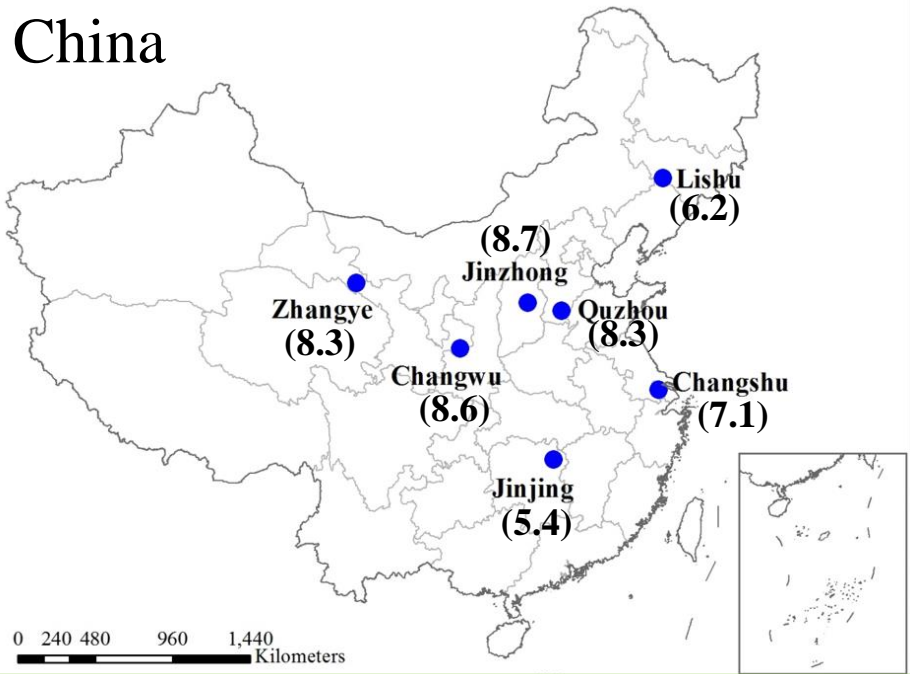
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

- Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
- You may not further distribute the material or use it for any profit-making activity or commercial gain
- You may freely distribute the URL identifying the publication in the public portal ?

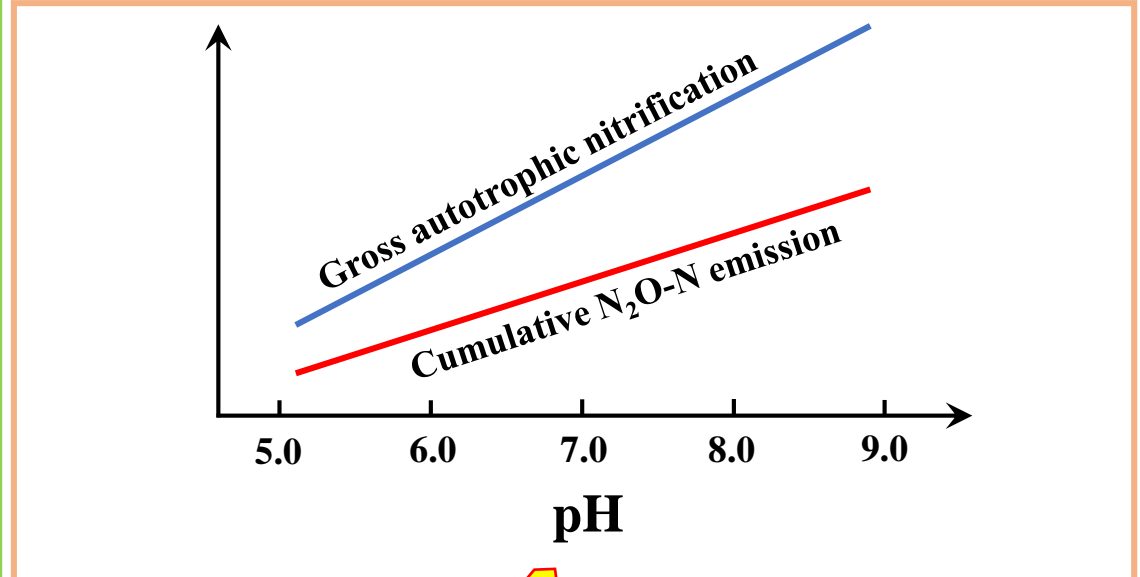
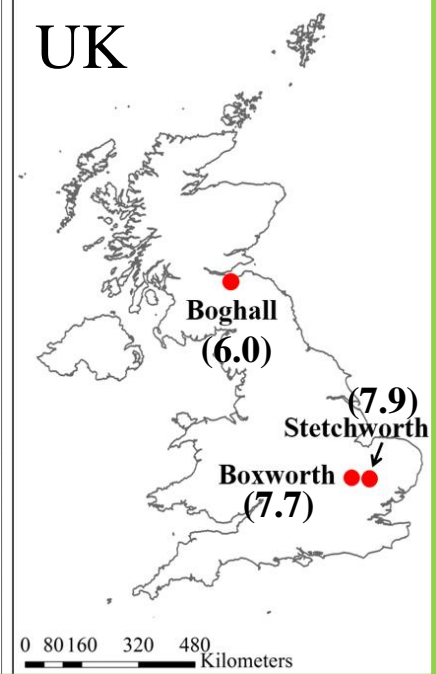
Take down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

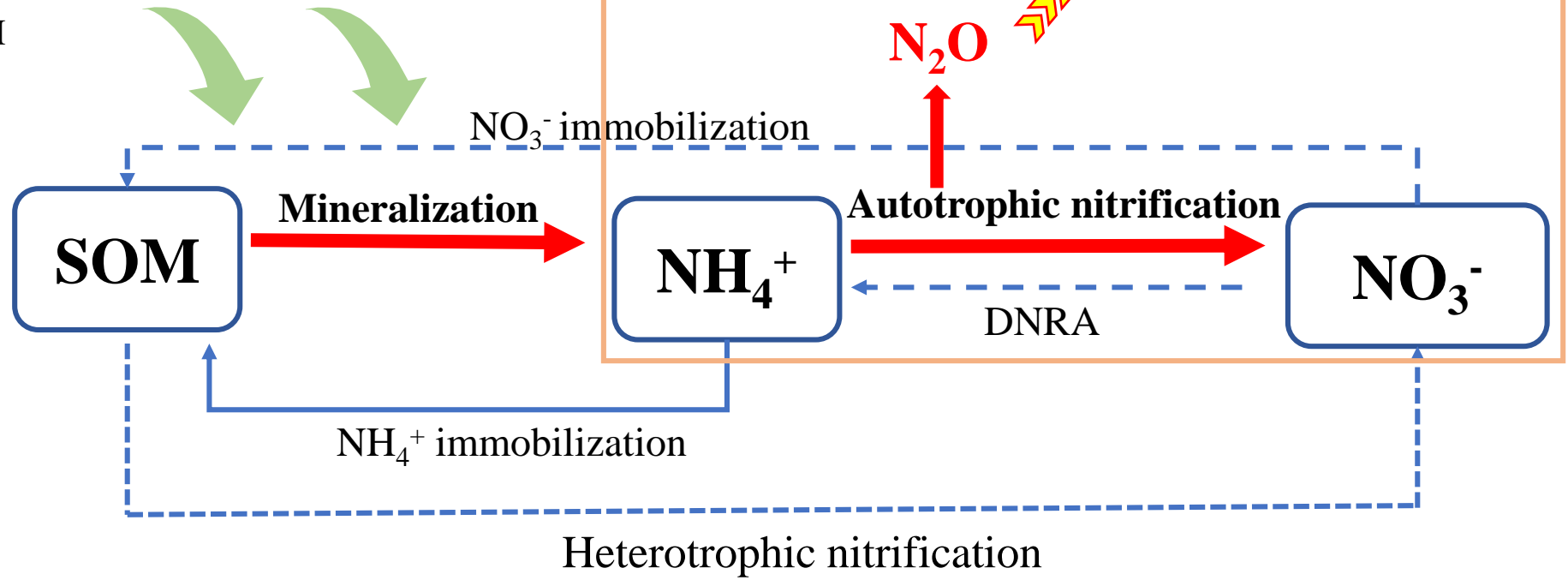
China



UK



Note: values in the brackets are soil pH



Highlights

We examined gross N transformations in Chinese and UK agricultural soils.

Autotrophic nitrification and mineralization were the key N transformation processes.

Autotrophic nitrification was important for N₂O emission under aerobic conditions.

pH was the primary factor controlling autotrophic nitrification and N₂O emissions.

Gross N transformation rates were not related to the origin of soil sampling sites.

Gross N transformation rate and related N₂O emission in seven Chinese agricultural soils as comparison with three UK agricultural soils

Authors: Gaodi Zhu^a, Xiaotong Song^a, Xiaotang Ju^{a*}, Jinbo Zhang^b, Christoph Müller^{c,d}, Roger Sylvester-Bradley^e, Rachel E. Thorman^e, Ian Bingham^f, Robert M. Rees^f

Affiliations:

^a *College of Resources and Environmental Sciences, China Agricultural University, 2 Yuanmingyuan West Road, Beijing 100193, China*

^b *School of Geography Sciences, Nanjing Normal University, Nanjing 210023, China*

^c *Department of Plant Ecology (IFZ), Justus-Liebig University Giessen, Heinrich-Buff-Ring 26, 35392 Giessen, Germany*

^d *School of Biology and Environmental Science, University College Dublin, Belfield, Dublin 4, Ireland*

^e *ADAS Boxworth, Battlegate Road, Boxworth, Cambridge CB23 4NN, UK*

^f *SRUC, West Mains Rd. Edinburgh, EH9 3JG, Scotland, UK*

***: Corresponding author**

XTJ: juxt@cau.edu.cn **Tel.:** +86 10 6273 2006; **Fax:** +86 10 6273 1016

Submitted to: Science of the Total Environment

Type of contribution: Research paper

Date of preparation: December 21, 2018

Number of text pages: 39

Number of figures: 6

Number of tables: 2

1 **Gross N transformation rates and related N₂O emissions in Chinese and UK**
2 **agricultural soils**

3 **Abstract**

4 The properties of agricultural soils in various regions of the world are variable
5 and can have a significant but poorly understood impact on soil nitrogen (N)
6 transformations and nitrous oxide (N₂O) emissions. For this reason, we undertook a
7 study of gross N transformations and related N₂O emissions in contrasting agricultural
8 soils from China and the UK. Seven Chinese and three UK agricultural soils were
9 collected for study using a ¹⁵N tracing approach. The soil pH ranged from 5.4 to 8.7,
10 with three acidic soils collected from Jinjing, Lishu and Boghall; one neutral soil
11 collected from Changshu, and the other six alkaline soils collected from Quzhou,
12 Zhangye, Changwu, Jinzhong, Boxworth and Stetchworth. Our results showed that
13 the main N transformation processes were oxidation of ammonium (NH₄⁺) to nitrate
14 (NO₃⁻) (ONH₄), and mineralization of organic N to NH₄⁺. The gross autotrophic
15 nitrification rates calculated in the three acidic soils were between 0.25 and 4.15 mg N
16 kg⁻¹d⁻¹, which were significantly lower (*p*<0.05) than those in the remaining neutral
17 and alkaline soils ranging from 6.94 to 14.43 mg N kg⁻¹d⁻¹. Generally, soil pH was
18 positively correlated (*p*<0.001) with gross autotrophic nitrification rate and
19 cumulative N₂O emissions, indicating that soil pH was an important factor regulating
20 autotrophic nitrification and N₂O emissions. There was also a significant positive
21 correlation between the gross autotrophic nitrification rate and cumulative N₂O
22 emissions, highlighting the importance of this process for producing N₂O emissions in

23 these agricultural soils under aerobic conditions. Gross NH_4^+ immobilization rates
24 were very low in most soils except for the Jinjing soil with the lowest pH. In
25 conclusion, the gross autotrophic nitrification rates and related N_2O emissions were
26 controlled by soil pH irrespectively of the soil's origin in these agricultural soils.

27

28 **Keywords:** Chinese agricultural soils; UK agricultural soils; gross N transformation;
29 N_2O emissions; ^{15}N tracing approach; soil pH

30

31 **1. Introduction**

32 Soil nitrogen (N) transformations (including processes and rates) play a crucial
33 role in regulating soil N availability and N losses (Galloway et al., 2008). While soil
34 N mineralization provides an important source of plant-available N (Wang et al.,
35 2015), the reverse process of immobilization transfers mineral N into the organic N
36 pools and is an important N sequestration mechanism in soils, mediated by microbial
37 activity linked to the humification processes (Ponge, 2013). Thus, soil N
38 mineralization-immobilization turnover (MIT) governs soil N availability (Murphy et
39 al., 2003; Huygens et al., 2007), which is regulated by soil pH, organic carbon (C), N
40 C/N ratios and microbial community when soil temperature and moisture conditions
41 do not limit microbial activity (Booth et al., 2005; Wang et al., 2015).

42 Nitrification including autotrophic and heterotrophic pathways produces nitrate
43 (NO_3^-) and regulates the ratio of ammonium (NH_4^+) to NO_3^- in soils. It also controls N
44 loss through leaching or as gaseous emissions e.g. nitrous oxide (N_2O) (Zhang et al.,
45 2018). Autotrophic nitrification is driven by chemo-autotrophic nitrifiers which
46 oxidize ammonia (NH_3) to NO_3^- and gain energy from this process, while
47 heterotrophic nitrification is carried out by heterotrophic nitrifiers or fungi, which can
48 oxidize either NH_4^+ or organic N to form NO_3^- , and utilize organic C for their energy
49 (Zhang et al., 2014; Liu et al., 2015). Because autotrophic nitrifiers are sensitive to
50 low pH, it is thought that heterotrophic nitrification may play a predominant role in
51 NO_3^- production in acidic soils (Huygens et al., 2007; Zhang et al., 2013a).

52 The N transformation processes that are capable of producing N₂O such as
53 autotrophic nitrification, heterotrophic nitrification, nitrifier-denitrification, coupled
54 nitrification-denitrification, denitrification and dissimilatory nitrate reduction to
55 ammonia (DNRA) can occur simultaneously in soils. These processes are regulated
56 by a range of soil properties, such as soil pH, texture, C and N content,
57 aerobic-anaerobic micro-sites and the microbial community structure which is
58 influenced by environmental factors (temperature, water content and oxygen)
59 (Butterbach-Bahl et al., 2013; Müller et al., 2014). Rates of N cycling are strongly
60 influenced by the magnitude of pools of soil NH₄⁺, organic N and NO₃⁻ which in turn
61 are strongly affected by N additions in the form of manufactured N fertilizer or
62 manure (Snyder et al., 2009). However, under aerobic conditions, N₂O production
63 from denitrification in agricultural soils with a high pH and low organic C content is
64 minor relative to autotrophic nitrification. For instance, N₂O emissions occur mainly
65 as a consequence of the ammonia oxidation process in intensively managed low
66 organic C (5-10 g kg⁻¹) calcareous (with a pH of between 7.2 - 8.5) agricultural soils
67 in the North China Plain (Wan et al., 2009; Ju and Zhang, 2017). By contrast, in
68 agricultural soils in the UK with high organic C (about 23 to 40 g kg⁻¹) (King et al.,
69 2005), high water contents together with relatively low soil pH (ranging from 5.0 to
70 7.5; Goulding, 2016), denitrification is a major contributor to overall N₂O emissions
71 (Clark et al., 2012).

72 The properties of agricultural soils in China and the UK are highly variable and
73 depend on parent material, climate, topography, vegetation and time, as well as land

74 use, and agricultural management practices (e.g. crop type, tillage, fertilization and
75 irrigation, etc.). For the purpose of this study we chose agricultural soils from
76 different parts of China and the UK that were representative of the agricultural
77 landscapes in these countries and therefore relevant to the research questions being
78 addressed by our study. The upland agricultural soils of Northern China are located in
79 a region to the north of a line connecting the Kunlun Mountains, the Qinling
80 Mountains and the Huaihe River. They have a temperate monsoon or continental
81 climate with dry cold winters and wet hot summers (Ju and Zhang, 2017). Based on
82 differences in soil properties, precipitation and agricultural management practices,
83 three major sub-regions, namely the Northeast Plain, the North China Plain and
84 Northwest China (including Loess Plateau), can be identified in this region of China
85 (Liu and Chen, 2005). The soils are characterized by high organic C contents ranging
86 from 8 to 44 g kg⁻¹ (Liu et al., 2006) and relatively low pH (5.5-7.2) (Shangguan et al.,
87 2013) in the Northeast Plain, but low organic C contents (5-10 g kg⁻¹) (Liao et al.,
88 2015) and high pH (7.2-9.8) (Shangguan et al, 2013) in the North China Plain and
89 Loess Plateau. However, the soils in the South China region (mainly paddy soils) are
90 located in a subtropical monsoon climate zone and are characterized by a low pH
91 ranging from 4.5 to 5.5 (with a few exceptions that are characterized by neutral pH)
92 (Shangguan et al., 2013). They are also highly weathered and have a poor buffering
93 capacity (Zhao et al., 2007). In addition, Chinese agricultural soils are often
94 intensively managed with frequent tillage, high N fertilization rates and a lack of

95 manure and crop residue inputs, which leads to soil acidification, low soil fertility and
96 reduced N retention (Ju et al., 2009; Guo et al., 2010).

97 Tilled agricultural soils in the UK are mainly located in the East of Scotland, and
98 the East and South of England (Brown and Shipley, 1982; Avery, 1990; Davies et al.,
99 1996), and are characterized by a temperate maritime climate, where the seasonal
100 temperature change is moderated by relatively cool summers and temperate winters.
101 The agricultural soils in the East of Scotland tend to have more organic C and be more
102 acidic than soils in the rest of the UK, with a texture that is predominantly sandy-loam
103 to loam, and often exhibit poor drainage (Hay et al., 2000). In the East and South of
104 England the proportion of alkaline soils is higher than in Scotland, and although
105 textures are typically heavier (clays and loams dominate large areas) (Dobbie and
106 Smith, 2003), the organic C concentrations are generally lower overall due to higher
107 temperatures and generally lower rainfall.

108 Previous research in the UK has shown that the delayed uptake of applied N in
109 wheat was associated with temporary net immobilization by soil microbes, and thus
110 could not temporarily supply mineral N in the rapid growth period of the plant (King
111 et al., 2001; Silgram and Chambers, 2002; Murphy et al., 2007). The low C and
112 calcareous soils of the North China Plain, by contrast, have a low potential for
113 immobilization, high rates of nitrate leaching and N₂O emissions are associated with
114 excessive net mineralization and nitrification (Ju and Zhang, 2017). Our hypothesis in
115 approaching this work was that the relatively high organic C contents associated with
116 UK soils would favour N immobilization. However, to date, differences in the N

117 transformation characteristics and related N₂O emissions from contrasting agricultural
118 soils in China and the UK, have not been compared. Such knowledge is a prerequisite
119 to evaluate novel techniques which aim to manipulate soil MIT, nitrification, and
120 denitrification rates to enhance the recovery of applied N and to provide information
121 on cropping systems, which can in turn help to optimize N fertilization and reduce N
122 losses.

123 Therefore, the objectives of this study were: 1) to quantify the gross N
124 transformation rates and N₂O emissions in a typical selection of Chinese and UK
125 agricultural soils; 2) to study factors controlling the N transformation process
126 responsible for N₂O emissions from these soils. Seven Chinese and three UK
127 agricultural soils were collected to study the above objectives.

128 **2. Materials and methods**

129 2.1 Site description and soil samples collection

130 Soils were sampled from seven typical Chinese agricultural sites and three UK
131 agricultural sites in 2016 (Fig. S1), which refers to dominant soil types and crop
132 systems in the main agricultural regions of China and UK. At the sampling sites
133 typical management practices for the local crops were adopted. Some important soil
134 and climatic properties, land use and crops for each site are summarized in Table 1.

135 Soil samples were collected after crop harvest between October and December
136 2016 from the plough layer (0-20 cm) at each site. Ten to fifteen soil cores were
137 randomly taken from each field site (approximately 400 m²) in a zigzag sampling
138 pattern using stainless steel augers (5 cm diameter) and mixed to produce one

139 composite sample (approximately 3 kg). After collection, soils were stored in a cool
140 box, before being transferred to the laboratory where they were sieved to 2 mm. Roots
141 and any visible organic substrates were removed by tweezers to reduce the
142 heterogeneity. Each soil sample was divided into two parts, and approximately 400 g
143 of the soil was air-dried for analysis of soil properties, and the remaining soil was
144 stored at 4.0 °C for the incubation experiment.

145 2.2 ¹⁵N tracing incubation experiment

146 Gross N transformation rates were quantified using a ¹⁵N tracing approach
147 (Kirkham and Bartholomew, 1954; Müller et al., 2007; Zhang et al., 2012a; Wang et
148 al., 2017). For each soil, there were two ¹⁵N-labelled treatments: ¹⁵N labelled NH₄⁺
149 (¹⁵NH₄NO₃ at 9.76 atom % ¹⁵N excess) and ¹⁵N labelled NO₃⁻ (NH₄¹⁵NO₃ at 9.74
150 atom % ¹⁵N excess). For each soil, 24 Erlenmeyer flasks were prepared (2 treatments,
151 3 replicates, and 4 extraction times) with 20 g (oven-dry basis) of fresh soil, which
152 were pre-incubated for 24 h at about 50% water holding capacity (WHC) and at 20°C.
153 Here the 50% of WHC referred to water holding capacity of the sieved soil, which
154 was higher than the WHC measured of well-structured soil *in situ*, but was an
155 appropriate water potential to promote microbial activity under aerobic conditions.
156 Two milliliters of ¹⁵NH₄NO₃ or NH₄¹⁵NO₃ solution were added to each Erlenmeyer
157 flask at a rate of 20 mg NH₄⁺-N kg⁻¹ or 20 mg NO₃⁻-N kg⁻¹ (equivalent to 40 mg N
158 kg⁻¹), respectively. The soils were adjusted to 60% water holding capacity (WHC) and
159 sealed with parafilm® with four pinholes to allow aeration, and incubated in the dark
160 for 48 h at 20°C. This represents a routine procedure for these kinds of studies (Müller

161 et al., 2007; Zhang et al., 2012a), and allows the investigation of fundamental
162 processes and rates of soil N transformation under conditions conducive to high rates
163 of microbial activity. The soil N transformations measured at the incubation
164 temperature of 20°C in this study would be close to conditions experienced at the
165 range of sampling sites. After incubation, soils were extracted with 2M KCl at 0.5, 12,
166 24 and 48 h after labelling to determine the concentrations and isotopic compositions
167 of NH₄⁺ and NO₃⁻.

168 Additionally, another 6 flasks of each soil (two ¹⁵N-labelled treatments and three
169 replicates) were prepared for measuring N₂O fluxes, which were treated in the same
170 way. Gas samples were collected at 12, 24 and 48 h after N application. Before
171 sampling, the flasks were flushed with ambient air using a multiport vacuum manifold.
172 The above procedure was repeated three times (each time for about 10 s). Thereafter,
173 the flasks were immediately sealed for 4 h using a silicone sealant and gas samples
174 were collected using a syringe fitted with a three-way stopcock and transferred to 18.5
175 ml pre-evacuated vial, to determine the N₂O concentration (using an Agilent 7890 GC,
176 USA). The N₂O flux was calculated as:

$$177 \quad F_{N_2O} = \{[M \times C \times V_g \times 273] / [W \times (273 + T) \times 22.4 / t]\} \times (28/44) \quad (1)$$

178 where F_{N_2O} ($\mu\text{g N kg}^{-1} \text{ h}^{-1}$) is the N₂O flux; M (g mol^{-1}) is the molar mass of N₂O; C
179 (ppm) is the concentration of N₂O; V_g (L) is the volume at the top of the flask; W (kg)
180 is the dry soil weight; T (°C) is mean air temperature during incubation; 22.4 (L mol^{-1})
181 is the molecular volume at 1,013 hPa and 273 K; t (h) is the time of flask closure;
182 28/44 is used for unit conversion. The fluxes of N₂O measured on each sampling date

183 were used to determine cumulative N₂O emissions by the trapezoidal integration
184 method.

185 2.3 Chemical analysis

186 Soil NH₄⁺ and NO₃⁻ were extracted with 2 M KCl at a soil / solution ratio of 1:5
187 (v/v) on a mechanical shaker for 1 h at 300 rpm at 20 °C. The extract was passed
188 through filter papers (Whatman No. 42 Cat No. 1442-055) and the concentrations of
189 NH₄⁺ and NO₃⁻ were analyzed using a continuous-flow analyzer (SA1000, Skalar, the
190 Netherlands). The isotopic composition of NH₄⁺ and NO₃⁻ was determined using the
191 modified micro-diffusion method (Zhang et al., 2017) and measured using a Delta V
192 plus isotope mass spectrometer (Thermo Fisher Scientific, Delta V plus, Germany).
193 Briefly, a portion of the extract was mixed with MgO to release NH₃ from the NH₄⁺
194 pool. Devarda's alloy was then added to the flask to reduce NO₃⁻ to NH₄⁺, and then to
195 NH₃. Liberated NH₃ was trapped using filter paper, which was acidified with 1 M
196 oxalic acid. After diffusion, filters were transferred to a free-ammonia environment
197 for drying, then dried filter papers were transferred to a tin capsule and wrapped to
198 enable the enrichment of ¹⁵N to be analysed. Soil pH was determined at a soil to water
199 ratio of 1:2.5 (w/v) using a pH meter (Mettler Toledo, Switzerland). Soil organic
200 carbon (SOC) and total N was determined with a C/N element analyser (Sample
201 preparation system, Europa EA-GSL).

202 2.4 ¹⁵N tracing model

203 The *Ntrace* model was used to quantify gross N transformations rates (Müller et
204 al., 2007). The measured concentrations and ¹⁵N enrichment values (mean ± standard

205 deviation) of NH_4^+ and NO_3^- from the triplicate measurements in the two ^{15}N
206 treatments were supplied to the model, and gross N transformation rates were
207 calculated by simultaneously optimizing the kinetic parameters for each individual
208 process by minimizing the misfit between modelled and observed concentrations of
209 NH_4^+ and NO_3^- and their respective ^{15}N enrichments. To obtain the most appropriate
210 model, several model modifications, which vary in the number of considered N pools
211 and processes, and kinetic settings, were tested (Rütting and Müller, 2008). The most
212 appropriate model was guided by the Aikake's information criterion (AIC), selecting
213 the minimum AIC value (Cox et al., 2006; Rütting and Müller, 2007; Wang et al.,
214 2016). In addition, the determination coefficient (R^2) can also be used to verify the
215 model; if $R^2 > 0.80$, the modelled result can be accepted (Quinn and Keough, 2002).
216 The numerical optimization model based on Markov Chain Monte Carlo Metropolis
217 algorithm, can provide reliable results for a large number of parameters. For more
218 detailed information on the model development and parameter optimization see
219 Müller et al. (2007).

220 Ten simultaneously occurring transformations were quantified (Müller et al.,
221 2007; McGeough et al., 2016): 1) M_{Nrec} , mineralization of recalcitrant organic-N to
222 NH_4^+ ; 2) M_{Nlab} , mineralization of labile organic-N to NH_4^+ ; 3) $I_{\text{NH}_4_Nlab}$,
223 immobilization of NH_4^+ to labile organic N; 4) $I_{\text{NH}_4_Nrec}$, immobilization of NH_4^+ to
224 recalcitrant organic N; 5) R_{NH_4ads} , release of adsorbed NH_4^+ ; 6) A_{NH_4} , absorption of
225 NH_4^+ on cation exchange sites; 7) O_{NH_4} , oxidation of NH_4^+ to NO_3^- ; 8) O_{rec} , oxidation
226 of recalcitrant organic N to NO_3^- ; 9) I_{NO_3} , immobilization of NO_3^- to recalcitrant

227 organic-N; and 10) D_{NO_3} , dissimilatory NO_3^- reduction to NH_4^+ . Gross transformation
228 rates were calculated by zero- or first-order kinetics. In this model, labile organic N
229 was assumed to be the maro-organic matter fraction and accounted for 1% of total
230 organic N according to Rütting and Müller (2008) and Chen et al. (2015). The
231 optimization procedure resulted in a probability density function for each parameter,
232 from which averages and standard deviations were calculated (Müller et al., 2007).
233 Each analysis run was carried out with three parallel sequences. Based on the kinetic
234 setting and the final parameters, average N transformation rates were calculated over
235 the initial 24 h incubation period (because the concentrations and abundances of NH_4^+
236 could not be reliably detected at 48 h in most soil samples) and expressed in units of
237 $mg\ N\ kg^{-1}\ dry\ soil\ d^{-1}$ (Table 2).

238 Net mineralization rates were calculated from the differences in NH_4^+ plus NO_3^-
239 concentration between two sampling dates divided by the number of days in the
240 sampling intervals, while net nitrification rate was calculated from the difference in
241 NO_3^- concentration (Table S1).

242 2.5 Statistical analyses

243 For gross rates of N transformation, most statistical tests are inappropriate for the
244 comparison of parameter results, because of the large number of iterations of the ^{15}N
245 tracing model (Yoccoz, 1991). Thus, parameter results based on the comparisons of
246 standard deviations and the 95% confidence intervals were used to distinguish three
247 cases: a) standard deviation overlap: the parameters are not different; b) standard
248 deviations do not overlap but 95% confidence intervals overlap, the parameters are

249 not significantly different but show a clear tendency to be different; c) 95%
250 confidence intervals do not overlap, parameters are significantly different (Müller et
251 al., 2011). A one-way analysis of variance (ANOVA) with a least significant
252 difference test ($p < 0.05$) was used to compare the cumulative emissions of N₂O in the
253 ten soils. Correlation and linear or nonlinear regression analyses were used to test
254 relationships between soil pH and total gross mineralization rate, autotrophic
255 nitrification rates and cumulative N₂O emissions, cumulative N₂O emissions and total
256 gross mineralization and autotrophic nitrification rate, and the relationship between
257 total gross mineralization and gross autotrophic nitrification. All the statistical
258 analyses were performed using SPSS software package 17.0 for windows (Inc., USA)
259 and correlations were analyzed using SigmaPlot 12.5 (Systat Software Inc., Erkrath,
260 Germany).

261

262 3. Results

263 3.1 Soil properties

264 Soil properties showed considerable variation in the ten soils used for this study
265 (Table 1). The soil pH ranged from 5.4 to 8.7, with pH values < 6.5 in the soils
266 collected from Jinjing, Lishu and Boghall (acidic soils); one soil collected from
267 Changshu had a pH value of 7.1 (neutral soil), while all the remaining soils had pH
268 values ≥ 7.6 (alkaline soils). Soil organic C ranged from 21.9 to 27.4 g kg⁻¹ in soils
269 from Jinjing, Changshu, Boghall, Boxworth and Stetchworth and these were
270 significantly higher ($p < 0.05$) than the other soils which ranged from 8.1 to 17.8 g kg⁻¹
271 (Lishu, Quzhou, Zhangye, Changwu and Jinzhong). The SOC contents of all three UK
272 soils (21.9 to 27.4 g kg⁻¹) and two paddy soils from Jinjing and Changshu (25 to 27.1
273 g kg⁻¹) was relatively high.

274 High SOC contents, were associated with high N contents in the two Chinese
275 paddy soils and the three UK soils ranging from 2.0 to 3.0 g N kg⁻¹, which was
276 significantly higher ($p < 0.05$) than the rest (ranging from 0.7 to 1.7 g N kg⁻¹). The C/N
277 ratio of all soils varied from 9.0 to 12.3, which were typical for such tilled agricultural
278 soils. The inorganic N was dominated by NO₃⁻ in all soils, ranging from 9.02 to 62.36
279 mg N kg⁻¹, and the minimum and maximum values were measured in the soils from
280 Changshu and Boghall, respectively. The NH₄⁺-N concentrations in soils from Jinjing
281 and Boxworth were 8.03 mg N kg⁻¹ and 4.70 mg N kg⁻¹, respectively, and were
282 substantially higher ($p < 0.01$) than the others, which ranged from 0.03 to 1.41 mg N

283 kg^{-1} . In addition, the $\text{NH}_4^+\text{-N}/\text{NO}_3^-\text{-N}$ ratio of all soils varied from 0 to 0.52, and the
284 maximum value were observed in Jinjing soil.

285 3.2 Concentrations and enrichment of NH_4^+ and NO_3^- pools

286 To reveal the relationship between NH_4^+ consumption and NO_3^- production
287 within different incubation periods, we calculated the net change of NH_4^+ (ΔNH_4^+)
288 and NO_3^- (ΔNO_3^-) concentrations (Table S1, Fig. S2), and found that a decline in
289 NH_4^+ concentration generally corresponded well with an increase of NO_3^-
290 concentration in all soils within different incubation periods, but the rate of NH_4^+
291 oxidation to NO_3^- during different incubation periods varied. During the period
292 between 0-0.5 h, it was found that $\Delta\text{NH}_4^+ < \Delta\text{NO}_3^-$ in all Chinese soils, but this was
293 reversed in the three UK soils (Fig. S2 a). However, during the periods 0.5-12 h,
294 12-24 h and 24-48 h, the ΔNH_4^+ was almost equal to the ΔNO_3^- in all soils. The value
295 of ΔNO_3^- was slightly higher than ΔNH_4^+ in most cases between 0.5-24 h and 0.5-48 h
296 of incubation, indicating a low net mineralization of soil organic N compared to a net
297 oxidation of NH_4^+ .

298 The dynamics of NH_4^+ and NO_3^- concentrations in the ten soils from $^{15}\text{NH}_4\text{NO}_3$
299 and $\text{NH}_4^{15}\text{NO}_3$ treatments were almost the same within the 48 h incubation period
300 (Fig. 1). There was a gradual decline in NH_4^+ concentrations, while NO_3^-
301 concentrations gradually increased during the incubation of the neutral and alkaline
302 soils (Fig. 1). By contrast, there was relatively little change in the NH_4^+ and NO_3^-
303 pools in the acidic soils, indicating a weaker ammonia oxidation ability in the acidic
304 soils. Model simulations of NH_4^+ and NO_3^- dynamics corresponded well with

305 measurements within the 24 h incubation period. Model evaluations were only
306 possible for the first 24 hours due to fast N transformation processes depleting the
307 mineral N pools so that the ^{15}N excess of the NH_4^+ and NO_3^- could not be determined
308 (see next section) (Table 2).

309 In the labelled $^{15}\text{NH}_4\text{NO}_3$ treatment, the atom % ^{15}N excess of NH_4^+ pool
310 decreased, while the atom % ^{15}N excess of NO_3^- pool increased during the incubation
311 in the neutral and alkaline soils, but not in the acidic soils (Fig. 2). This corresponded
312 well with the above changes of NH_4^+ and NO_3^- concentrations in different soil
313 situations, showing a high NH_4^+ oxidation activity in soils with a $\text{pH} > 7.0$. The
314 enrichment of NH_4^+ pools after 0.5 h incubation of the labelled $^{15}\text{NH}_4\text{NO}_3$ treatment
315 ranged between 7.87-8.15 in all soils (i.e. close to the applied 9.76 atom% ^{15}N excess),
316 except for soils from Jinjing (5.42 atom% ^{15}N excess) and Boxworth (7.65 atom% ^{15}N
317 excess) indicating that the ^{15}N dilution was low (Fig. S3 a). Simulated and observed
318 isotopic enrichments corresponded well within the 24 h incubation. The enrichment of
319 the NH_4^+ pool in the labelled $^{15}\text{NH}_4\text{NO}_3$ treatment decreased sharply between 24 and
320 48 h in the neutral and alkaline soils but not noticeably in the acidic soils (Fig. 2 a-j).
321 These situations could be also illustrated by the significant correlation between the
322 isotopic enrichment of NH_4^+ pools in the 24-48 h period and the NH_4^+ concentration
323 at 24 h, in which small changes in the acidic soils from Jingjing, Lishu and Boghall
324 corresponded with relatively high NH_4^+ concentrations after 24 h (Fig. S4).

325 In the labelled $\text{NH}_4^{15}\text{NO}_3$ treatment, the enrichment of the NO_3^- pool decreased
326 with the increasing incubation time in the neutral and alkaline soils, but not

327 noticeably in the acidic soils (Fig. 2 a'-j'). The enrichment of the NH_4^+ pool was
328 extremely low (ranging between 0.01-0.03 atom%) over the whole incubation period,
329 indicating that the NH_4^+ transformed from labelled NO_3^- pool was negligible. The
330 enrichment of the NO_3^- pool after 0.5 h in the labelled $\text{NH}_4^{15}\text{NO}_3$ treatment was in
331 the range of 2.25-5.58 atom% in all soils compared to the initial labelled $\text{NH}_4^{15}\text{NO}_3$
332 enrichment of 9.74 atom% ^{15}N excess of NO_3^- pool. Due to the relatively high initial
333 NO_3^- concentrations in all soils a ^{15}N dilution effect was observable. We also
334 observed a negative relationship ($p < 0.01$) between initial NO_3^- concentration and
335 atom % ^{15}N excess after 0.5 h in the NO_3^- pool in the labelled $\text{NH}_4^{15}\text{NO}_3$ treatment
336 (Fig. S3 b).

337 3.3 Gross N transformation rates

338 The main processes studied in these soils were oxidation of NH_4^+ to NO_3^- (O_{NH_4}),
339 and mineralization of labile (M_{Nlab}) and recalcitrant (M_{Nrec}) organic N to NH_4^+ , except
340 for the soil from Jinjing with the lowest pH (5.4) which was also characterized by
341 relatively high NH_4^+ immobilization to labile ($I_{\text{NH}_4_{\text{Nlab}}}$) and recalcitrant ($I_{\text{NH}_4_{\text{Nrec}}}$)
342 organic N. Total mineralization rates (M_{tot}) were calculated by: $M_{\text{tot}} = M_{\text{Nlab}} + M_{\text{Nrec}}$.

343 The maximum total gross mineralization rate was observed in the soil from
344 Jinjing (5.26 mg N $\text{kg}^{-1}\text{d}^{-1}$), followed by Stetchworth (3.43 mg N $\text{kg}^{-1}\text{d}^{-1}$), which was
345 significantly higher ($p < 0.05$) than that in other soils (1.03-3.24 mg N $\text{kg}^{-1}\text{d}^{-1}$). Gross
346 NH_4^+ immobilization ($I_{\text{NH}_4_{\text{tot}}} = I_{\text{NH}_4_{\text{Nlab}}} + I_{\text{NH}_4_{\text{Nrec}}}$) was observed in soil from Jinjing
347 (4.55 mg N $\text{kg}^{-1}\text{d}^{-1}$), but was negligible in the other soils.

348 Autotrophic nitrification (O_{NH_4}) was responsible for the largest flows of N in

349 these agricultural soils, except for the Jinjing soil with the lowest soil pH. The gross
350 autotrophic nitrification rates calculated in acidic soils from Jinjing, Lishu and
351 Boghall, were 0.25, 3.05 and 4.15 mg N kg⁻¹d⁻¹, respectively, which were significantly
352 lower ($p < 0.05$) than those in the remaining neutral and alkaline soils ranging from
353 6.94 to 14.43 mg N kg⁻¹d⁻¹. This indicates that autotrophic nitrification was favored by
354 high soil pH. Unexpectedly, the heterotrophic nitrification process (O_{Nrec}) was
355 observed in the soil from Lishu, but at a low rate of 0.80 mg N kg⁻¹d⁻¹. Compared to
356 NO₃⁻ production pathways, the NO₃⁻ consumption pathways, including gross NO₃⁻
357 immobilization and dissimilatory NO₃⁻ reduction to NH₄⁺ (DNRA), were negligible
358 for all studied soils (see Table 2).

359 Within the 0.5-12 h incubation period, gross mineralization was approximately
360 equal to the net mineralization in almost all soils (Fig. S6 a) and was similar in most
361 soils in the 0.5-24 h period, except for soils from Zhangye, Changwu and Jinzhong
362 which had a pH above 8.0 (Fig. S6 b). Within the 0.5-12 h period, the total gross
363 nitrification rates were higher than the net nitrification rates in soils from Lishu and
364 Boghall, while the opposite trend occurred in the other soils (Fig. S6 c). However, the
365 total gross nitrification rates were approximately equal to the net nitrification rates in
366 the soils within the 0.5-24 h incubation period, except for soils from Zhangye,
367 Changwu and Jinzhong with a pH above 8.0 (Fig. S6 d), in which there was lower
368 gross than net nitrification.

369 3.4 N₂O emissions during incubation

370 Generally, N₂O fluxes increased and reached a maximum after 24 h of incubation

371 except for soils from Changshu, Boxworth and Boghall, in which N₂O fluxes were
372 highest at 12 h of incubation, especially in the paddy soil from Changshu (Fig. S5 a).
373 After 24 h, N₂O fluxes decreased gradually and declined to a minimum after 48 h. We
374 calculated the cumulative N₂O emissions during the 24 and 48 h periods, and
375 observed a similar ranking of treatments (Fig. 3 and Fig. S5 b). To be consistent with
376 the period of model simulation, we used the cumulative N₂O emissions within the 24
377 h incubation to establish the relationships of N₂O emissions with the gross N
378 transformation rate and some soil properties as analyzed in following sections.

379 The cumulative N₂O emissions (ranged from 0.65 to 1.46 μg N kg⁻¹ dry soil) and
380 autotrophic nitrification in three acidic soils (pH<6.5) were both lower than those
381 from other soils with pH>7.0 (cumulative N₂O emissions ranging from 2.29 to 7.12
382 μg N kg⁻¹ dry soil), although the soil from Quzhou was an outlier (with a low
383 cumulative N₂O emission of <2 mg N kg⁻¹ dry soil) (Fig. 3). The cumulative N₂O
384 emissions of the remaining six soils were in the order of Changshu > Zhangye >
385 Jinzhong > Boxworth > Stetchworth > Changwu, although the Changshu and Zhangye
386 soils had significantly higher emissions ($p<0.01$) than the other soils.

387 3.5 Correlations between gross N rate, N₂O emissions and soil properties

388 Soil pH was positively correlated ($p<0.001$) with gross autotrophic nitrification
389 rates and cumulative N₂O emissions ($p<0.05$) (except for the paddy soil from
390 Changshu which had the highest cumulative N₂O emissions), confirming that soil pH
391 was an important driving factor regulating autotrophic nitrification and N₂O emissions
392 (Fig. 4). In addition, the autotrophic nitrification rate was significantly positively

393 correlated ($p < 0.01$) with cumulative N₂O emissions (except for the paddy soil from
394 Changshu), but was not correlated with total gross mineralization rates (Fig. 5). This
395 suggests that autotrophic nitrification plays an important role in producing N₂O in
396 these agricultural soils under aerobic conditions. A significant correlation ($p < 0.05$)
397 was found between the total gross mineralization rate and autotrophic nitrification rate
398 in all soils except soil from Jinjing (Fig. 6), suggesting that the availability of NH₄⁺
399 was a critical factor in limiting autotrophic nitrification. The total gross mineralization
400 and autotrophic nitrification rates were not significantly correlated with any other soil
401 properties (Fig. S7).

402 **4. Discussion**

403 4.1 Soil pH, autotrophic nitrification rate and N₂O emissions

404 Our results showed that autotrophic nitrification (O_{NH4}) was the dominant
405 process of N transformation and that soil pH was the key factor controlling both
406 autotrophic nitrification and N₂O emissions, which is consistent with previous studies
407 (Zhao et al., 2007; Zhang et al., 2013a). The gross autotrophic nitrification rates and
408 cumulative N₂O emissions in the acid soils were much lower than those in neutral or
409 alkaline soils. There was a significant positive correlation between gross autotrophic
410 nitrification and cumulative N₂O emissions ($p < 0.01$), highlighting the importance of
411 the nitrification as a source of N₂O emissions in these agricultural soils under aerobic
412 conditions. However, the soils in the field are not always aerobic, especially in UK.
413 Environments with higher labile C, higher moisture contents and relatively low soil
414 pH can promote denitrification and make a substantial contribution to N₂O emissions

415 (Dobbie et al., 1999; Dobbie and Smith, 2003). Similar situations also occur in the
416 low C, high pH and well-aerated loamy soils in North China where intense rainfall or
417 irrigation events occur, leading to microoxic or anaerobic conditions, which in turn
418 induce denitrification (Gao et al., 2014; Huang et al., 2014).

419 The role of pH in controlling overall N₂O emissions is complex, and depends
420 upon the balance of the nitrification process under aerobic conditions and
421 denitrification process under anaerobic conditions, (Butterbach-Bahl et al., 2013).
422 Studies have shown that decreasing soil pH could increase the N₂O/N₂ ratio by
423 denitrification as a consequence of the inhibition of N₂O reductase, resulting in
424 increasing N₂O emissions from denitrification under anaerobic conditions (Šimek and
425 Cooper, 2002; Saggar et al., 2013). However, under aerobic conditions, N₂O is the
426 major product of nitrification, and an increase in the soil pH can then stimulate N₂O
427 production (Signor and Cerri, 2013). This implies that different N₂O mitigation
428 strategies could be adopted under different soil conditions. For example, nitrification
429 inhibitors could be applied to neutral and alkaline soils to slow down the
430 ammonia-oxidizing process and reduce N₂O emissions (Ding et al., 2011; Huang et al.,
431 2014). In circumstances where well drained acidic soils are being used for crop
432 production reducing ammonium supply would be likely to be more effective for
433 reducing N₂O emissions than in wetter soils where denitrification would predominate
434 (Liu et al., 2017). The Changshu paddy soil exhibited higher N₂O emissions compared
435 to other soils, which were attributed to a high pH (7.1) and high SOC & TN providing
436 ideal conditions for producing large N₂O emissions under aerobic conditions

437 (Butterbach-Bahl et al., 2013). The crop rotation in Changshu was paddy rice
438 followed by winter wheat, and the aerobic incubation maintained in the current study
439 could reflect the characteristics of gross N transformation and N₂O emissions in the
440 wheat season.

441 In addition, autotrophic nitrification rates are also controlled by soil pH,
442 primarily through its effects on NH₃ availability (Nugroho et al., 2007; Cheng et al.,
443 2013). Ammonia is the substrate for ammonia-oxidizing microorganisms, and its
444 concentration declines exponentially with decreasing soil pH due to the ionization of
445 NH₃ to NH₄⁺, and NH₄⁺ uptake requires an active mechanism of transmembrane
446 transport and therefore an energy source which may also be limited at low pH
447 (Kemmitt et al., 2006; Baggs et al., 2010). Thus, low autotrophic nitrification rates in
448 acidic soils are thought to be caused by substrate limitations and a low number of
449 autotrophic nitrifiers (Jiang and Bakken 1999), with pH tolerances varying between
450 different species of ammonia-oxidising bacteria (De Boer et al., 1991). Low soil pH
451 suppresses NH₄⁺ oxidation, leading to relatively higher NH₄⁺ concentrations in acidic
452 soils, which causes a dilution effect of the ¹⁵NH₄⁺ pool at the initial time (0.5 h) under
453 the ¹⁵NH₄NO₃ treatment. There is increasing evidence that soil pH plays a central role
454 in shaping the communities of active ammonia oxidizers in response to soil conditions.
455 For example, autotrophic nitrification is driven by ammonia-oxidizing bacteria (AOB)
456 rather than ammonia-oxidizing archaea (AOA) in alkaline agricultural soils, while
457 AOA control autotrophic nitrification in acidic agricultural soils (Jiang et al., 2015;
458 Shi et al., 2016).

459 Heterotrophic nitrification was only observed in the Lishu soil with gross rates of
460 0.80 mg N kg⁻¹d⁻¹. Generally, heterotrophic nitrification is observed in forest and
461 grassland soils which are characterized by higher organic C and N, and is considered
462 to be negligible in agricultural soils (Nelissen et al., 2012; Zhang et al., 2015). This
463 was confirmed by a meta-study highlighting a positive relationship between
464 heterotrophic nitrification and SOC and C/N ratios but showed a negative correlation
465 with soil pH on a global scale (Chen et al., 2015). Based on previous studies, we infer
466 that high SOC, C/N and low pH are crucial factors in controlling heterotrophic
467 nitrification in the Lishu soil.

468 4.2 Mineralization and NH₄⁺-N & NO₃⁻-N immobilization

469 Mineralization of labile (M_{Nlab}) and recalcitrant (M_{Nrec}) organic N to inorganic N,
470 total gross mineralization and autotrophic nitrification were significantly positively
471 correlated in all soils except soil from Jinjing with the lowest soil pH, which also
472 showed a higher MIT. Zhang et al. (2018) reviewed the inherent connections between
473 soil N transformations and climate, and showed that gross rates of N mineralization
474 and immobilization (NH₄⁺-N & NO₃⁻-N immobilization) in acidic soils in humid
475 climate zones (mean annual precipitation > 800 mm) were significantly greater than
476 those in semi-humid climate zones (400 mm < mean annual precipitation < 800 mm)
477 and soils in semiarid and arid regions (< 400 mm), which were characterized by a
478 neutral or alkaline pH.

479 Gross NH₄⁺ immobilization rates were very low in most soils except for the
480 Jinjing soil with the lowest pH, which refutes our initial hypothesis that the UK soils

481 should have the highest capacity for NH_4^+ immobilization due to relatively high soil
482 organic C. So, NH_4^+ immobilization is also controlled by soil pH regardless of origin
483 or organic carbon concentration. However, during the period between 0-0.5 h, it was
484 found that the $\Delta\text{NH}_4^+ < \Delta\text{NO}_3^-$ in all Chinese soils, but this was reversed in the three
485 UK soils (Fig. S2 a), which indicated net immobilization of NH_4^+ by UK soils was
486 stronger than that in the Chinese soils at the very beginning of addition of NH_4NO_3 .
487 This is consistent with work by Silgram and Chambers (2002) who also found
488 temporary net NH_4^+ immobilization in UK soils. This may be because the NH_4^+
489 concentrations were sufficiently high at the early stage of incubation to support the N
490 demand of both the heterotrophs and the nitrifiers, but as the NH_4^+ concentrations
491 dropped, nitrifiers were more successful in competing for NH_4^+ with immobilizing
492 organisms in later incubation periods (Burger and Jackson, 2003).

493 Microbial mineralization in Jinjing and Stetchworth soils was significantly
494 higher ($p < 0.05$) than in other soils, which might have been caused by their
495 comparatively high total N and soil organic C contents. This is in line with previous
496 studies showing that gross mineralization rates were positively correlated with soil
497 organic C and total N concentrations, providing the energy and substrates for
498 microbial N cycling processes (Booth et al., 2005; Lang et al., 2010). However,
499 research across ten agricultural soils in the humid zones of China found no significant
500 relationship between gross N mineralization rates and individually measured soil
501 properties including soil organic C, total N and C/N ratio (Zhang et al., 2013a).

502 Nitrate (NO_3^-) immobilization was low in all of our agricultural soils which has

503 also been shown by others (Shi et al., 2004; Zhang et al., 2012b). Microbial
504 immobilization of NO_3^- requires more energy by comparison to NH_4^+ immobilization
505 and may be suppressed by NH_4^+ even at low concentrations (Templer et al., 2008). In
506 addition, the quantity and quality of soil organic C are also considered as key factors
507 affecting NO_3^- immobilization (Burger and Jackson, 2003; Häbteselassie et al., 2006).
508 Previous studies have reported that microbes need to immobilize inorganic N to
509 satisfy their metabolic needs during decomposition of organic matter with high C/N
510 ratios (Janssen, 1955; Zhang et al., 2013b), explaining why significant NO_3^-
511 immobilization is reported in forest, grassland and organically farmed soils that are
512 characterized by much higher organic C contents and high C/N ratios compared to
513 conventionally farmed agricultural soils (Bradley, 2001; Cheng et al., 2012), which
514 have relatively low organic matter inputs, high mineral N fertilization and regular
515 tillage leading to a decline in the ability to retain N. To restrict NO_3^- accumulation and
516 the associated risk of high N losses, it is important to develop management strategies
517 that promote organic C and N retention in cropland systems.

518 4.3 Model simulation of NH_4^+ and NO_3^- concentrations and their atom % ^{15}N excess

519 According to the *Ntrace* simulation results, we know that the methods for
520 determining gross soil N transformation rates using the ^{15}N labelling technique require
521 that concentrations and enrichments of the measured N change significantly over the
522 measurement period and that the applied ^{15}N pool is not rapidly consumed (Murphy et
523 al., 2003). It is difficult to accurately determine the gross N transformation rates if the
524 supplied ^{15}N (e.g. $^{15}\text{NH}_4^+$) is rapidly depleted (Watson et al., 2000; Stark and Schimel,

525 2001). In this study, we also found that the NH_4^+ concentration and its enrichment in
526 the labelled $^{15}\text{NH}_4\text{NO}_3$ treatment decreased dramatically after 24 h in the neutral and
527 alkaline soils, due to their high ability to transform NH_4^+ to NO_3^- (Fig. 2). Therefore,
528 for the neutral and alkaline soils with their high ability for ammonia oxidation, we
529 suggest that additional soil sampling should be performed at 6 h after ^{15}N labelling to
530 provide more robust data for model calculations of gross N transformation rates.
531

532 **5. Conclusions**

533 Our results showed that there were no intrinsic differences in soil N
534 transformations and N₂O emissions between Chinese and UK agricultural soils, but
535 that differences mainly resulted from differences in soil properties, specifically pH.
536 This leads to the inference that the dynamics of N transformations are globally
537 affected by similar conditions. Autotrophic nitrification dominated NO₃⁻ production
538 and was closely related to cumulative N₂O emissions under aerobic conditions, which
539 was significantly regulated by soil pH. This understanding of gross N transformation
540 processes and N₂O emission pathways will be valuable in helping to develop more
541 appropriate N management and N₂O mitigation strategies.

542

543 **Acknowledgments**

544 This work was supported by the National Natural Science Foundation of China
545 (41830751, 41471190), the Newton Funded N Circle project (Grant Ref.
546 BB/N013484/1) and the Scottish Government's Strategic Research Programme. We
547 greatly appreciate the soil sampling assistance from Chong Zhang, Jianlin Shen,
548 Guozhong Feng, Wei Zhou, Xinsheng Niu, Xiaodong Lv, Shengli Guo and Lijun Li.

549 **Appendix A. Supplementary material**

550 Supplementary tables and figures related to this article can be found in Appendix
551 A.

552 **References**

- 553 Avery, B.W., 1990. Soils of the British Isles. CAB International.
- 554 Baggs. E.M., Smales, C.L., Bateman, E.J., 2010. Changing pH shifts the microbial
555 source as well as the magnitude of N₂O emission from soil. *Biology and Fertility*
556 *of Soils* 46, 793-805.
- 557 Booth, M.S., Stark, J.M., Rastetter, E.B., 2005. Controls on nitrogen cycling in
558 terrestrial ecosystems: a synthetic analysis of literature data. *Ecological*
559 *Monographs* 75, 139-157.
- 560 Bradley, R.L., 2001. An alternative explanation for the post-disturbance NO₃⁻ flush in
561 some forest ecosystems. *Ecology Letters* 4, 412-416.
- 562 Brown, C.J., Shipley, B.M., 1982. South East Scotland: Soil and Land Capability.
563 Aberdeen: The Macaulay Institute for Soil Research.

564 Burger, M., Jackson, L.E., 2003. Microbial immobilization of ammonium and nitrate
565 in relation to ammonification and nitrification rates in organic and conventional
566 cropping systems. *Soil Biology and Biochemistry* 35, 29-36.

567 Butterbach-Bahl, K., Baggs, E.M., Dannenmann, M., Kiese, R.,
568 Zechmeister-Boltenstern, S., 2013. Nitrous oxide emissions from soil: how well
569 do we understand the processes and their controls? *Philosophical Transactions of*
570 *the Royal Society of London* 368, 20131022.

571 Chen, Z.M., Ding, W.X., Xu, Y.H., Müller, C., Rütting, T., Yu, H.Y., Fan, J.L.,
572 Zhang, J.B., Zhu, T.B., 2015. Importance of heterotrophic nitrification and
573 dissimilatory nitrate reduction to ammonium in a cropland soil: Evidences from a
574 ¹⁵N tracing study to literature synthesis. *Soil Biology and Biochemistry* 91,
575 65-75.

576 Cheng, Y., Cai, Z.C., Chang, S.X., 2012. Wheat straw and its biochar have contrasting
577 effects on inorganic N retention and N₂O production in a cultivated Black
578 Chernozem. *Biology and Fertility of Soils* 5, 941-946.

579 Cheng, Y., Wang, J., Mary, B., Zhang, J.B., Cai, Z.C., Chang, S.X., 2013. Soil pH has
580 contrasting effects on gross and net nitrogen mineralization in adjacent forest and
581 grassland soils in Central Alberta, Canada. *Soil Biology and Biochemistry* 57,
582 848-857.

583 Clark, I.M., Buchkina, N., Jhurrea, D., Goulding, K.W., Hirsch, P.R., 2012. Impacts
584 of nitrogen application rates on the activity and diversity of denitrifying bacteria

585 in the Broadbalk Wheat Experiment. *Philosophical Transactions of the Royal*
586 *Society of London* 367, 1235-1244.

587 Cox, G.M., Gibbons, J.M., Wood, A.T.A., Craigon, J., Ramsden, S.J., Crout, N.M.J.,
588 2006. Towards the systematic simplification of mechanistic models. *Ecological*
589 *Modelling* 198, 240-246.

590 Davies, D.B., Twd, G., Adh, R., 1996. Factors affecting nitrate leaching from a
591 calcareous loam in East Anglia. *Journal of Agricultural Science* 126, 75-86.

592 De Boer, W., Gunnewiek, PJAK, Veenhuis, M., Bock, E., Laanbroek, H.J., 1991.
593 Nitrification at low pH by aggregated chemolithotrophic bacteria. *Applied and*
594 *Environmental Microbiology* 57, 3600-3604.

595 Ding, W.X., Yu, H.Y., Cai, Z.C., 2011. Impact of urease and nitrification inhibitors
596 on nitrous oxide emissions from fluvo-aquic soil in the North China Plain.
597 *Biology and Fertility of Soils* 47, 91-99.

598 Dobbie, K.E., McTaggart, I.P., Smith, K.A., 1999. Nitrous oxide emissions from
599 intensive agricultural systems: variations between crops and seasons, key driving
600 variables, and mean emission factors. *Journal of Geophysical Research* 104,
601 26891-26899.

602 Dobbie, K.E., Smith, K.A., 2003. Nitrous oxide emission factors for agricultural soils
603 in Great Britain: the impact of soil water-filled pore space and other controlling
604 variables. *Global Change Biology* 9, 204-218.

605 Galloway, J.N., Townsend, A.R., Erisman, J.W., Bekunda, M., Cai, Z.C., Freney, J.R.,
606 Martinelli, L.A., Seitainger, S.P., Sutton, M.A., 2008. Transformation of the

607 nitrogen cycle: recent trends, questions, and potential solutions. *Science* 320,
608 889-892.

609 Gao, B., Ju, X.T., Su, F., Meng, Q.F., Oenema, O., Christie, P., Chen, X.P., Zhang,
610 F.S., 2014. Nitrous oxide and methane emissions from optimized and alternative
611 cereal cropping systems on the North China Plain: a two-year field study.
612 *Science of the Total Environment* 472, 112-124.

613 Goulding, K.W.T., 2016. Soil acidification and the importance of liming agricultural
614 soils with particular reference to the United Kingdom. *Soil Use and Management*
615 32, 390-399.

616 Guo, J.H., Liu, X.J., Zhang, Y., Shen, J.L., Han, W.X., Zhang, W.F., Christie, P.,
617 Goulding, K.W.T., Vitousek, P.M., Zhang, F.S., 2010. Significant acidification
618 in major Chinese croplands. *Science* 327, 1008-1010.

619 Häbteselassie, M.Y., Stark, J.M., Miller, B.E., Thacker, S.G., Norton, J.M., 2006.
620 Gross nitrogen transformations in an agricultural soil after repeated dairy-waste
621 application. *Soil Science Society of America Journal* 70, 1338-1348.

622 Hay, R.K.M., Russell, G., Edwards, T.E., 2000. Crop production in the East of
623 Scotland. Edinburgh, UK: Scottish Agricultural Science Agency.

624 Huang, T., Gao, B., Hu, X.K., Lu, X., Well, R., Christie, P., Bakken, L.R., Ju, X.T.,
625 2014. Ammonia-oxidation as an engine to generate nitrous oxide in an
626 intensively managed calcareous fluvo-aquic soil. *Scientific Reports* 4, 3950.

627 Huygens, D., Rütting, T., Boeckx, P., Van Cleemput, O., Godoy, R., Müller, C., 2007.
628 Soil nitrogen conservation mechanisms in a pristine south Chilean Nothofagus
629 forest ecosystem. *Soil Biology and Biochemistry* 39, 2448-2458.

630 Jansson, S.L., Hallam, M.J., Bartholomew, W.V., 1955. Preferential utilization of
631 ammonium over nitrate by micro-organisms in the decomposition of oat straw.
632 *Plant and Soil* 4, 382-390.

633 Jiang Q.Q., Bakken, L.R., 1999. Nitrous oxide production and methane oxidation by
634 different ammonia-oxidising bacteria. *Applied and Environmental Microbiology*
635 65, 2679-2684.

636 Jiang, X.J., Hou, X.Y., Zhou, X., Xin, X.P., Wright, A., Jia, Z.J., 2015. pH regulates
637 key players of nitrification in paddy soils. *Soil Biology and Biochemistry* 81,
638 9-16.

639 Ju, X.T., Xing, G.X., Chen, X.P., Zhang, S.L., Zhang, L.J., Liu, X.J., Cui, Z.L., Yin,
640 B., Christie, P., Zhu, Z.L., Zhang, F.S., 2009. Reducing environmental risk by
641 improving N management in intensive Chinese agricultural systems. *Proceedings*
642 *of the National Academy of Sciences of the United States of America* 106, 8077.

643 Ju, X.T., Zhang, C., 2017. Nitrogen cycling and environmental impacts in upland
644 agricultural soils in North China: a review. *Journal of Integrative Agriculture* 16,
645 60345-60347.

646 Kemmitt, S.J., Wright, D., Goulding, K.W., Jones, D.L., 2006. pH regulation of
647 carbon and nitrogen dynamics in two agricultural soils. *Soil Biology and*
648 *Biochemistry* 38, 898-911.

649 King, J.A., Sylvester-Bradley, R., Rochford, A.H., 2001. Availability of nitrogen after
650 fertilizer applications. *Journal of Agricultural Science* 136, 141-157.

651 King, J.A., Bradley, R.I., Harrison, R., 2005. Current trends of soil organic carbon in
652 English arable soils. *Soil Use and Management* 21, 189-195.

653 Kirkham, D., Bartholomew, W.V., 1954. Equations for following nutrient
654 transformations in soil utilizing tracer data. *Soil Science Society of America*
655 *Proceedings*, 18, 33-34.

656 Lang, M., Cai, Z.C., Mary, B., Hao, X.Y., Chang, S.X., 2010. Land-use type and
657 temperature affect gross nitrogen transformation rates in Chinese and Canadian
658 soils. *Plant and Soil* 334, 377-389.

659 Liao, Y., Wu, W.L., Meng, F.Q., Smith, P., Lal, R., 2015. Increase in soil organic
660 carbon by agricultural intensification in northeastern China. *Biogeosciences* 12,
661 1403-1413.

662 Liu, D.W., Wang, Z.M., Zhang, B., Song, K.S., Li, X.Y., Li, J.P., Li, F., Duan, H.T.,
663 2006. Spatial distribution of soil organic carbon and analysis of related factors in
664 croplands of the black soil region, Northeast China. *Agriculture Ecosystems and*
665 *Environment* 113, 73-81.

666 Liu, R., Suter, H., He, J.Z., Hayden, H., Chen, D.L., 2015. Influence of temperature
667 and moisture on the relative contributions of heterotrophic and autotrophic
668 nitrification to gross nitrification in an acid cropping soil. *Journal of Soils and*
669 *Sediments* 15, 2304-2309.

670 Liu, R., Hayden, H.L., Hu, H.W., He, J.Z., Suter, H., Chen, D.L., 2017. Effects of the
671 nitrification inhibitor acetylene on nitrous oxide emissions and
672 ammonia-oxidizing microorganisms of different agricultural soils under
673 laboratory incubation conditions. *Applied and Environmental Microbiology* 119,
674 80-90.

675 Liu, X.H., Chen, F., 2005. *Farming system in China*. China Agriculture Press, China.
676 (in Chinese)

677 McGeough, K.L., Watson, C.J., Muller, C., Laughlin, R.J., Chadwick, D.R., 2016.
678 Evidence that the efficacy of the nitrification inhibitor dicyandiamide (DCD) is
679 affected by soil properties in UK soils. *Soil Biology and Biochemistry* 94,
680 222-232.

681 Müller, C., Rütting, T., Kattge, J., Laughlin, R.J., Stevens, R.J., 2007. Estimation of
682 parameters in complex ¹⁵N tracing models via Monte Carlo sampling. *Soil*
683 *Biology and Biochemistry* 39, 715-726.

684 Müller, C., Laughlin, R.J., Christie, P., Watson, C.J., 2011. Effects of repeated
685 fertilizer and cattle slurry applications over 38 years on N dynamics in a
686 temperate grassland soil. *Soil Biology and Biochemistry* 43, 1362-1371.

687 Müller, C., Laughlin, R.J., Spott, O., Rütting, T., 2014. Quantification of N₂O
688 emission pathways via a ¹⁵N tracing model. *Soil Biology and Biochemistry* 72,
689 44-54.

690 Murphy, D.V., Recous, S., Stockdale, E.A., Fillery, I.R.P., Jensen, L.S., Hatch, D.J.,
691 Goulding, K.W.T., 2003. Gross nitrogen fluxes in soil: theory, measurement and
692 application of ^{15}N pool dilution techniques. *Advances in Agronomy* 79, 69-118.

693 Murphy, D.V., Stockdale, E.A., Poulton, P.R., Willison, T.W., Goulding, K.W.T.,
694 2007. Seasonal dynamics of carbon and nitrogen pools and fluxes under
695 continuous arable and ley-arable rotations in a temperate environment. *European*
696 *Journal of Soil Science* 58, 1410-1424.

697 Nelissen, V., Rütting, T., Huygens, D., Staelens, J., Ruyschaert, G., Boeckx, P., 2012.
698 Maize biochars accelerate short-term soil nitrogen dynamics in a loamy sand soil.
699 *Soil Biology and Biochemistry* 55, 20-27.

700 Nugroho, R.A., Röling, W.F.M., Laverman, A.M., Verhoef, H.A., 2007. Low
701 nitrification rates in acid Scots pine forest soils are due to pH-related factors.
702 *Microbial Ecology* 53, 89-97.

703 Ponge, J.F., 2013. Plant-soil feedbacks mediated by humus forms: A review. *Soil*
704 *Biology and Biochemistry* 57, 1048-1060.

705 Quinn, G.P., Keough, M.J., 2002. *Experimental Design and Data Analysis for*
706 *Biologists*. Cambridge University Press, Cambridge, U.K.

707 Rütting, T., Müller, C., 2007. ^{15}N tracing models with a Monte Carlo optimization
708 procedure provide new insights on gross N transformations in soils. *Soil Biology*
709 *and Biochemistry* 39, 2351-2361.

710 Rütting, T., Müller, C., 2008. Process-specific analysis of nitrite dynamics in a
711 permanent grassland soil by using a Monte Carlo sampling technique. *European*
712 *Journal of Soil Science* 59, 208-215.

713 Saggar, S., Jha, N., Deslippe, J., Bolan, N.S., Luo, J., Giltrap, D.L., Kim, D.G.,
714 Zaman, M., Tillman, R.W., 2013. Denitrification and N₂O:N₂ production in
715 temperate grasslands: processes, measurements, modelling and mitigating
716 negative impacts. *Science of the Total Environment* 465,173-195.

717 Shangguan, W., Dai, Y.J., Liu, B.Y., Zhu, A.X., Duan, Q.Y., Wu, L.Z., Ji, D.Y., Ye,
718 A.Z., Yuan, H., Zhang, Q., Chen, D.D., Chen, M., Chu, J.T., Dou, Y.J., Guo,
719 J.X., Li, H.Q., Li, J.J., Liang, L., Liang, X., Liu, H.P., Liu, S.Y., Miao, C.Y.,
720 Zhang, Y.Z., 2013. A China data set of soil properties for land surface modeling.
721 *Journal of Advances in Modeling Earth Systems* 5, 212-224.

722 Shi, W., Miller, B.E., Stark, J.M., Norton, J.M., 2004. Microbial nitrogen
723 transformations in response to treated dairy waste in agricultural soils. *Soil*
724 *Science Society of America Journal* 68, 1867-1874.

725 Shi, X.Z., Hu, H.W., Müller, C., He, J.Z., Chen, D.L., Suter, H.C., 2016. Effects of
726 the nitrification inhibitor 3,4-dimethylpyrazole phosphate on nitrification and
727 nitrifiers in two contrasting agricultural soils. *Applied and Environmental*
728 *Microbiology* 82, 5236-5248.

729 Signor, D., Cerri, C.E.P., 2013. Nitrous oxide emissions in agricultural soils: a review.
730 *Pesquisa Agropecuária Tropical* 43, 322-338.

731 Silgram, M., Chambers, B.J., 2002. Effects of long-term straw management and
732 fertilizer nitrogen additions on soil nitrogen supply and crop yields at two sites in
733 eastern England. *Journal of Agricultural Science* 139, 115-127.

734 Šimek, M., Cooper, J.E., 2002. The influence of soil pH on denitrification: progress
735 towards the understanding of this interaction over the last 50 years. *European*
736 *Journal of Soil Science* 53, 345-354.

737 Snyder, C.S., Bruulsema, T.W., Jensen, T.L., Fixen, P.E., 2009. Review of
738 greenhouse gas emissions from crop production systems and fertilizer
739 management effects. *Agriculture Ecosystems and Environment* 133, 247-266.

740 Stark, J., Schimel, J., 2001. Errors in “Overestimation of gross N transformation rates
741 in grassland soils due to non-uniform exploitation of applied and native pools”.
742 *Soil Biology and Biochemistry* 33, 1433-1435.

743 Templer, P.H., Silver, W.L., Pett-Ridge, J., DeAngelis, K.M., Firestone, M.K., 2008.
744 Plant and microbial controls on nitrogen retention and loss in a humid tropical
745 forest. *Ecology* 89, 3030-3040.

746 Wan, Y.J., Ju, X. T., Ingwersen, J., Stange, C.F., Zhang, F.S., Streck, T., 2009. Gross
747 Nitrogen Transformations and Related Nitrous Oxide Emissions in an Intensively
748 Used Calcareous Soil *Soil Science Society of America Journal* 73, 102-112.

749 Wang, J., Zhu, B., Zhang, J.B., Müller, C., Cai, Z.C., 2015. Mechanisms of soil N
750 dynamics following long-term application of organic fertilizers to subtropical
751 rain-fed purple soil in China. *Soil Biology and Biochemistry* 91, 222-231.

752 Wang, J., Wang, L., Feng, X.J., Hu, H., Cai, C.Z., Müller, C., Zhang, J.B., 2016. Soil
753 N transformations and its controlling factors in temperate grasslands in China: A
754 study from ¹⁵N tracing experiment to literature synthesis. *Journal of Geophysical*
755 *Research* 121, 2949-2959.

756 Wang, J., Cheng, Y., Jiang, Y.J., Sun, B., Fan, J.B., Zhang, J.B., Müller, C., Cai, Z.C.,
757 2017. Effects of 14 years of repeated pig manure applications on gross nitrogen
758 transformations in an upland soil. *Plant and Soil*, 415, 161-173.

759 Watson, C. J., Travers, G., Kilpatrick, D. J., Laidlaw, A. S., and O’Riordan, E., 2000.
760 Overestimation of gross N transformation rates in grassland soils due to
761 non-uniform exploitation of applied and native pools. *Soil Biology and*
762 *Biochemistry* 32, 2019-2030.

763 Yoccoz, N.G., 1991. Use, overuse, and misuse of significance tests in evolutionary
764 biology and ecology. *Bulletin of the Ecological Society of America* 72, 106-111.

765 Zhang, J.B., Zhu, T.B., Cai, Z.C., Qin, S.W., Müller, C., 2012a. Effects of long-term
766 repeated mineral and organic fertilizer applications on soil nitrogen
767 transformations. *European Journal of Soil Science*, 63, 75-85.

768 Zhang, J.B., Cai, Z.C., Yang, W.Y., Zhu, T.B., Yu, Y.J., Yan, X.Y., Jia, Z.J., 2012b.
769 Long-term field fertilization affects soil nitrogen transformations in a
770 rice-wheat-rotation cropping system. *Journal of Plant Nutrition and Soil Science*
771 175, 939-946.

772 Zhang, J.B., Zhu, T.B., Meng, T.Z., Zhang, Y.C., Yang, J.J., Yang, W.Y., Müller, C.,
773 Cai, Z.C., 2013a. Agricultural land use affects nitrate production and

774 conservation in humid subtropical soils in China. *Soil Biology and Biochemistry*
775 62, 107-114.

776 Zhang, J.B., Cai, Z.C., Zhu, T.B., Yang, W.Y., Müller, C., 2013b. Mechanisms for the
777 retention of inorganic N in acidic forest soils of southern China. *Scientific*
778 *Reports* 3, 2342.

779 Zhang, J.B., Sun, W.J., Zhong, W.H., Cai, Z.C., 2014. The substrate is an important
780 factor in controlling the significance of heterotrophic nitrification in acidic forest
781 soils. *Soil Biology and Biochemistry* 76, 143-148.

782 Zhang, J.B., Wang, J., Zhong, W.H., Cai, Z.C., 2015. Organic nitrogen stimulates the
783 heterotrophic nitrification rate in an acidic forest soil. *Soil Biology and*
784 *Biochemistry* 80, 293-295.

785 Zhang, J.B., Cai, Z.C., Müller, C., 2018. Terrestrial N cycling associated with climate
786 and plant-specific N preferences: a review. *European Journal of Soil Science*
787 DOI: 10.1111/ejss.12533.

788 Zhang, P.Y., Wen, T., Zhang, J.B., Cai, Z.C., 2017. On improving the diffusion
789 method for determination of $\delta^{15}\text{N-NH}_4^+$ and $\delta^{15}\text{N-NO}_3^-$ in soil extracts.
790 *Acta Pedologica Sinica*, 54, 948-957.

791 Zhao, W., Cai, Z.C., Xu, Z.H., 2007. Does ammonium-based N addition influence
792 nitrification and acidification in humid subtropical soils of China? *Plant Soil* 297,
793 213-221.

1 **Table 1** Some important soil-climatic properties of the top layer (0-20 cm) of ten studied agricultural soils [Mean (SD)]

Parameter	Jinjing	Lishu	Changshu	Quzhou	Zhangye	Changwu	Jinzhong	Boghall	Boxworth	Stetchworth
Latitude	28°33'N	43°17'N	31°32'N	36°52'N	37°96'N	35°12'N	37°32'N	55°52'N	52°14'N	52°13'N
Longitude	113°20'E	124°20'E	120°40'E	115°10'E	102°64'E	107°40'E	112°40'E	3°13'W	0°05'E	0°22'E
Soil type ^a	Haplic Acrisol	Haplic Phaeozems	Hydrgric Anthrosol	Calcaric Cambisol	Calcaric Fluvisol	Calcarid Regsol	Capcic Luvisol	Gleyic Cambisol	Calcaric Cambisols	Chromic Luvisols
Soil texture ^b	Loamy clay	Loamy clay	Silty clay	Sandy loam	Slity loam	Sandy loam	Silty loam	Sandy loam	Clay	Sandy loam
Clay (%)	27	30	26	3	18	22	17	18	45	17
Land use (crop) ^c	Rice-Rice (two harvest a year)	Spring maize monoculture (one harvest a year)	Rice-Wheat (two harvest a year)	Wheat-Maize (two harvest a year)	Spring maize Monoculture (one harvest a year)	Spring maize Monoculture (one harvest a year)	Spring maize Monoculture (one harvest a year)	Cereals-Oilseed Rotation (one harvest a year)	Cereals-Oilseed Rotation (one harvest a year)	Cereals-Oilseed Rotation (one harvest a year)
T (°C) ^d	17.5	6.5	15.0	13.2	7.6	9.3	9.7	8.8	12.0	12.0
P (mm) ^e	1330	650	1038	494	152	584	441	567	550	590
Climate	Subtropical monsoon	Temperate monsoon	Subtropical monsoon	Temperate monsoon	Temperate continental	Temperate continental	Temperate Continental	Temperate marine	Temperate marine	Temperate marine
pH	5.4 (0.0)	6.2 (0.0)	7.1 (0.0)	8.3 (0.0)	8.3 (0.0)	8.6 (0.1)	8.7 (0.1)	6.0 (0.0)	7.7 (0.1)	7.9 (0.0)
SOC (g kg ⁻¹) ^f	25.5 (0.1)	17.8 (0.1)	27.1 (0.2)	9.1 (0.2)	11.4 (0.8)	8.4(0.2)	8.1 (0.0)	24.2 (3.0)	21.9 (1.0)	27.4 (1.4)
TN (g kg ⁻¹) ^g	2.5 (0.0)	1.7 (0.1)	2.3 (0.0)	0.9(0.0)	1.0 (0.0)	0.9 (0.0)	0.7 (0.0)	1.8(0.2)	2.4 (0.1)	3.0 (0.2)
C/N	10.1 (0.0)	10.2 (0.1)	11.9 (0.1)	10.1 (0.4)	12.0 (0.0)	9.3 (0.3)	10.9 (0.2)	12.3(0.2)	9.0(0.2)	9.0 (0.2)
NH ₄ ⁺ -N (mg kg ⁻¹)	8.03 (0.07)	1.41 (0.17)	0.10 (0.00)	0.14 (0.08)	0.75 (0.07)	0.72 (0.03)	0.58 (0.17)	0.03 (0.01)	4.70 (0.07)	0.98 (0.10)
NO ₃ ⁻ -N (mg kg ⁻¹)	15.37 (0.64)	43.24 (0.67)	9.02 (0.85)	31.96 (0.11)	35.23 (0.67)	14.31 (0.25)	43.56 (4.23)	62.36 (1.34)	19.88 (2.88)	33.77 (1.13)
NH ₄ ⁺ /NO ₃ ⁻	0.52 (0.02)	0.04 (0.00)	0.01 (0.00)	0.00 (0.00)	0.02 (0.00)	0.05 (0.00)	0.01 (0.00)	0.00 (0.00)	0.24 (0.04)	0.03 (0.00)

2 ^aSoils are described according to the FAO/UNESO (1988) classification; ^bSoil texture are described according to the international grading standard
3 of soil texture; ^cRotation means changing the type of crops grown in the field each year (or changing from crops to fallow) and Monoculture means
4 growing only one crop year after year in the same field; ^dT is the 6-year (2011-2016) mean annual temperature; ^eP is the 6-year (2011-2016) mean
5 annual precipitation; ^fSOC is the soil organic carbon; ^gTN is the soil total nitrogen.

6 **Table 2** Gross N transformation rates [Mean (SD) /mg N kg⁻¹d⁻¹] based on ¹⁵N tracing model

Parameter	Jinjing ^a	Lishu	Changshu	Quzhou	Zhangye	Changwu	Jinzhong	Boghall	Boxworth	Stetchworth
M _{Nlab}	4.45 (1.08)	0.50 (0.11) ^d	1.77 (0.60) ^d	0.78 (0.47) ^d	3.20 (0.14) ^d	0.42 (0.06) ^d	2.00 (0.06) ^d	0.95 (0.09) ^d	2.44 (0.43) ^d	3.40 (0.09) ^b
M _{Nrec}	0.81 (0.85)	0.53 (0.11) ^b	0.85 (0.53) ^b	0.83 (0.44) ^b	0.04 (0.04) ^b	0.90 (0.10) ^b	0.02 (0.02) ^b	0.45 (0.09) ^d	0.47 (0.42) ^d	0.03 (0.04) ^d
I _{NH4-Nlab}	2.78 (0.84)	0.02 (0.01) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d
I _{NH4-Nrec}	1.77 (0.60)	0.02 (0.02) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d	0.00 (0.00) ^d
O _{Nrec}	0.00 (0.00)	0.80 (0.09) ^d	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b
O _{NH4}	0.25 (0.04)	3.05 (0.05) ^d	8.06 (0.09) ^d	11.04 (0.08) ^d	14.24 (0.20) ^d	6.94 (0.10) ^d	14.43 (0.17) ^d	4.15 (0.04) ^d	13.26 (0.19) ^d	11.29 (0.09) ^d
A _{NH4}	0.02 (0.00)	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b
R _{NH4}	0.00 (0.00)	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	1.33 (0.03) ^d	0.00 (0.00) ^b	0.00 (0.00) ^b
I _{NO3}	0.00 (0.00)	0.00 (0.00) ^b	0.07 (0.02) ^d	0.06 (0.02) ^d	0.14 (0.03) ^d	0.04 (0.02) ^d	0.07 (0.04) ^d	0.14 (0.08) ^d	0.09 (0.05) ^d	0.23 (0.16) ^d
D _{NO3}	0.00 (0.00)	0.00 (0.00) ^b	0.00 (0.00) ^b	0.00 (0.00) ^b	0.01 (0.00) ^b	0.01 (0.00) ^b	0.00 (0.00) ^b	0.03 (0.00) ^b	0.03 (0.00) ^b	0.04 (0.00) ^b

7 M_{tot}: M_{Nlab} (mineralization of labile organic N to NH₄⁺) and M_{Nrec} (mineralization of recalcitrant organic N to NH₄⁺);

8 I_{NH4tot}: I_{NH4_Nlab} (immobilization of NH₄⁺ to N_{lab}) and I_{NH4_Nrec} (immobilization of NH₄⁺ to N_{rec});

9 O_{rec}, oxidation of recalcitrant organic-N to NO₃⁻;

10 O_{NH4}, oxidation of NH₄⁺ to NO₃;

11 I_{NO3}, immobilization of NO₃⁻ to recalcitrant organic-N;

12 D_{NO3}, dissimilatory NO₃⁻ reduction to NH₄⁺;

13 R_{NH4}, release of adsorbed NH₄⁺;

14 A_{NH4}, adsorption of NH₄⁺ on cation exchange sites.

15 ^aThe differences in gross N transformation rates between Jinjing soil and other nine soils were compared based on the comparisons of standard
16 deviations and the 95% confidence intervals to distinguish following three cases:

17 ^bStandard deviations overlap: the parameters are not different;

18 ^cStandard deviations do not overlap but 95% confidence intervals overlap, the parameters are not significantly different but show a clear tendency
19 to be different;

20 ^d 95% confidence intervals do not overlap, parameters are significantly different.

21

1 **Figure captions**

2 **Fig. 1** Measured (*point*) and modeled (*line*) concentrations of the NH_4^+ pool and NO_3^-
3 pool in Jinjing (a), Lishu (b), Changshu (c), Quzhou (d), Zhangye (e), Changwu (f),
4 Jinzhong (g), Boghall (h), Boxworth (i) and Stetchworth (j). Error bars represent
5 standard deviation (n=6), some error bars are covered by symbols due to the low
6 standard deviation values.

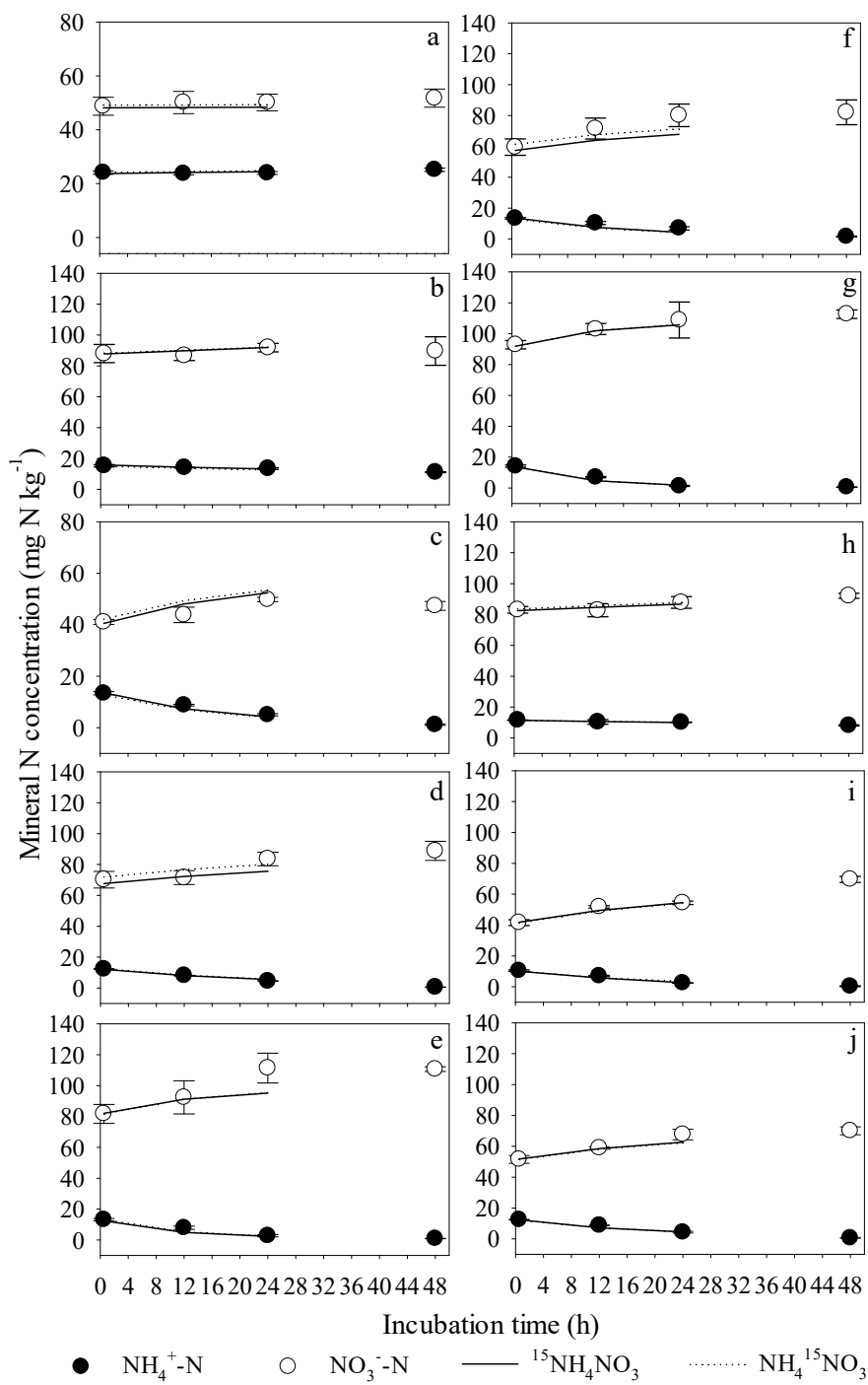
7 **Fig. 2** Measured (*point*) and modeled (*line*) atom % ^{15}N excess of the NH_4^+ pool and
8 NO_3^- pool in Jinjing (a and a'), Lishu (b and b'), Changshu (c and c'), Quzhou (d and
9 d'), Zhangye (e and e'), Changwu (f and f'), Jinzhong (g and g'), Boghall (h and h'),
10 Boxworth (i and i') and Stetchworth (j and j'). Error bars represent standard deviation
11 (n=3), some error bars are covered by symbols due to the low standard deviation values.

12 **Fig. 3** Cumulative N_2O -N emission in ten studied soils during the 24 h incubation.
13 Different letters indicate significant differences ($p<0.05$) among soils. Error bars
14 represent standard deviation (n=3).

15 **Fig. 4** Relationship between soil pH and total gross mineralization rate (a), gross
16 autotrophic nitrification rate (b) and cumulative N_2O -N emission (c). In Fig. 4c, the
17 linear relationship between pH and cumulative N_2O emission does not include
18 Changshu soil. Error bars represent standard deviation (n=3). Some error bars are
19 covered by symbols due to the low standard deviation values.

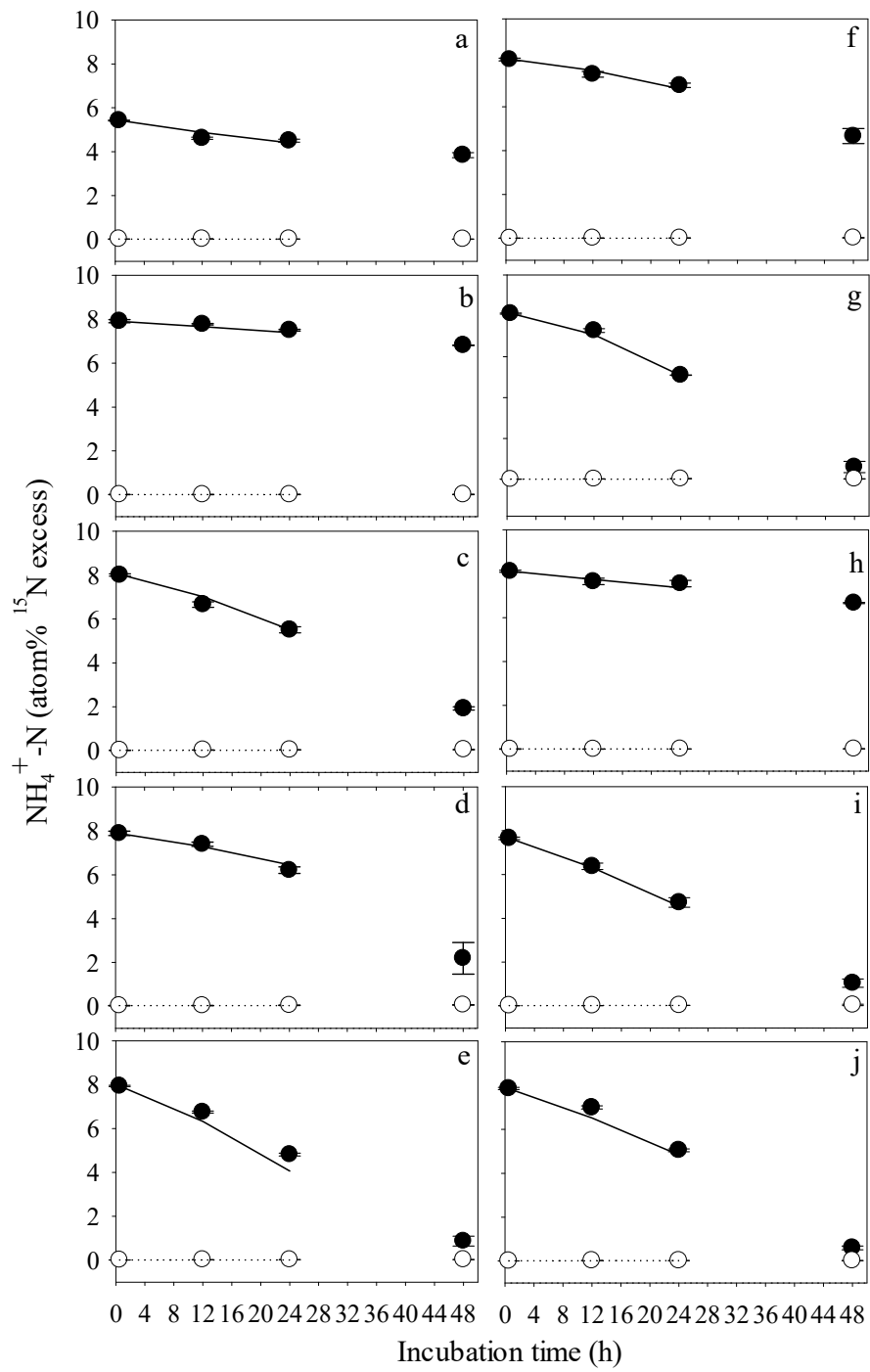
20 **Fig. 5** Relationship between cumulative N_2O -N emission and total gross mineralization
21 rate (a), autotrophic nitrification rate (b) in ten studied soils. In Fig. 5b, the linear
22 relationship between cumulative N_2O -N emission and autotrophic nitrification rate does
23 not include Changshu soil. Error bars represent standard deviation (n=3). Some error
24 bars are covered by symbols due to the low standard deviation values.

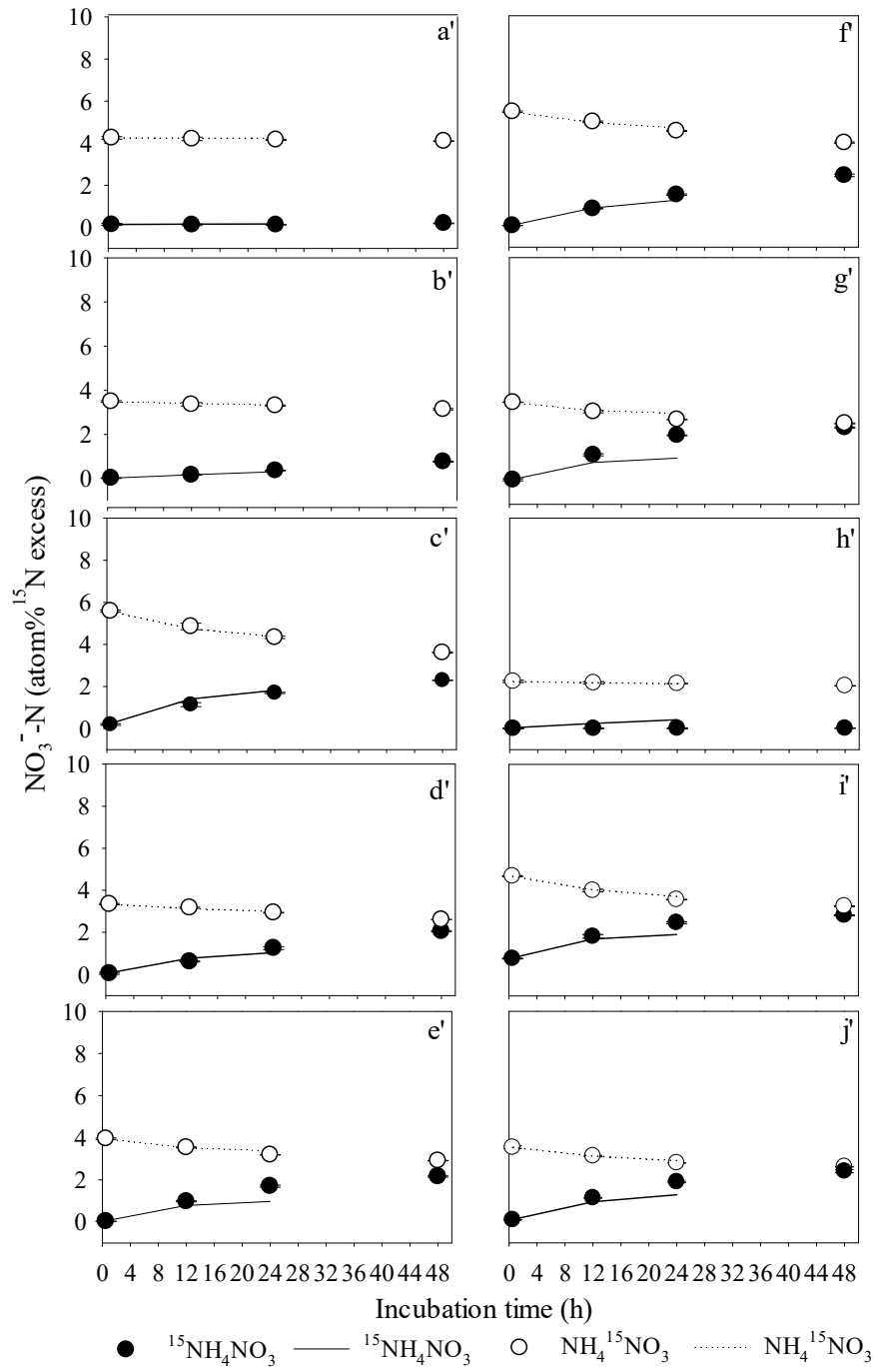
25 **Fig. 6** Relationship between total gross mineralization rate and autotrophic nitrification
26 rate excepting for Jinjing soil. Error bars represent standard deviation (n=3). Some error
27 bars are covered by symbols due to the low standard deviation values.



28

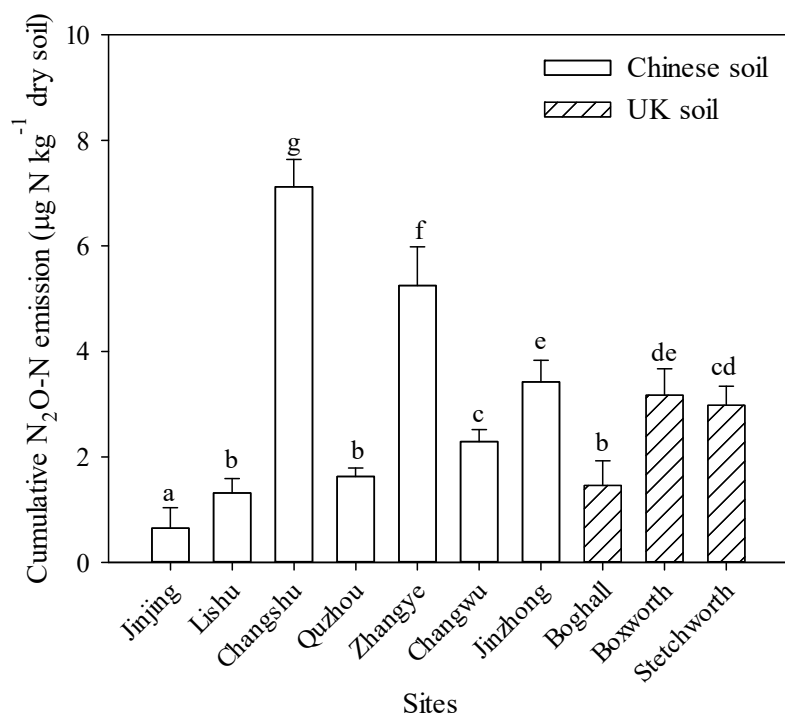
29 **Fig. 1 (Zhu GD)**





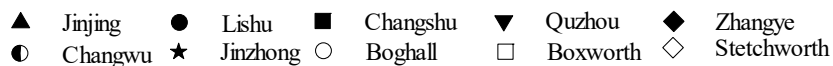
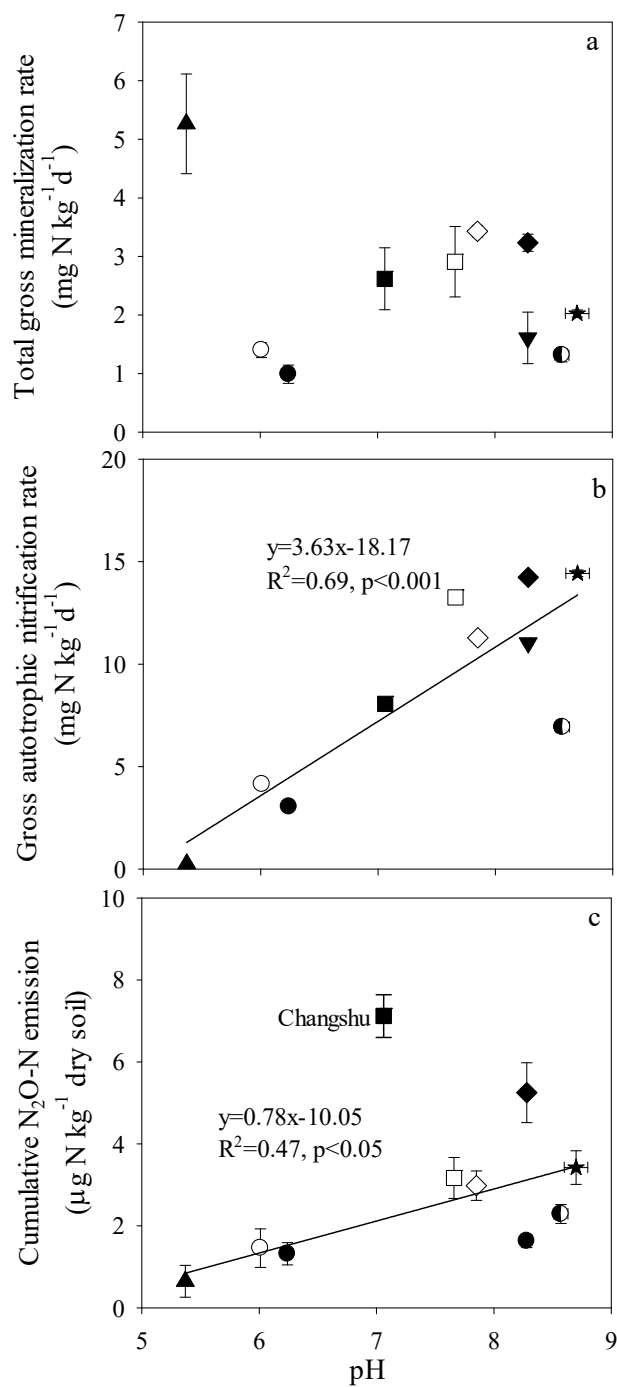
31

32 **Fig. 2 (Zhu GD)**



33

34 **Fig. 3 (Zhu GD)**



35

36 **Fig. 4 (Zhu GD)**

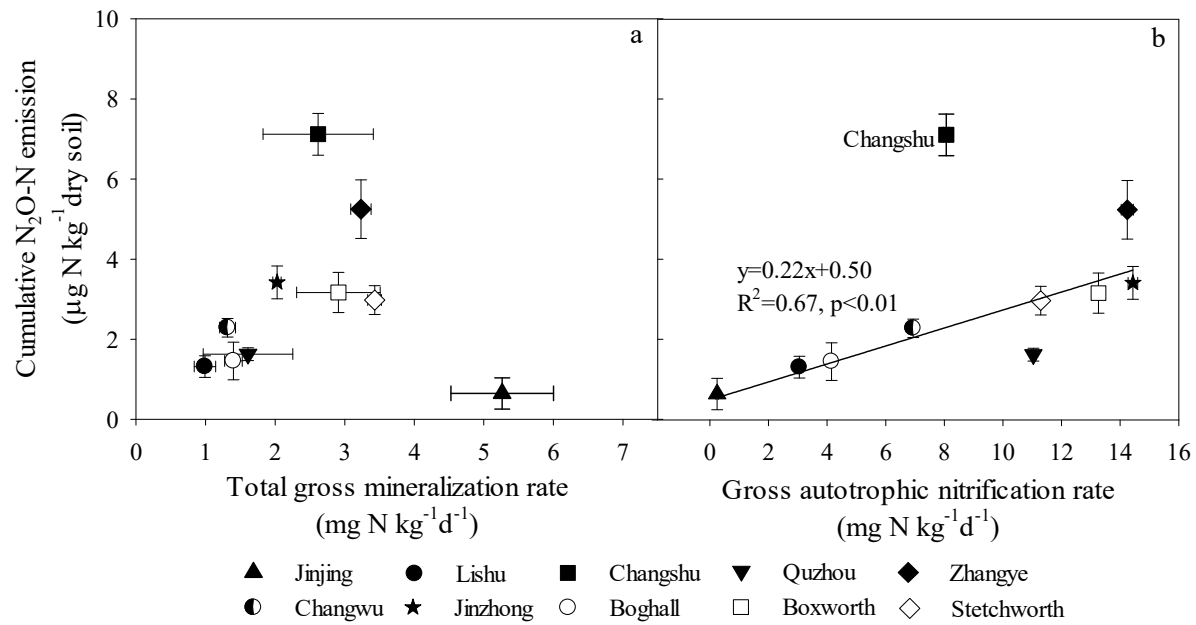
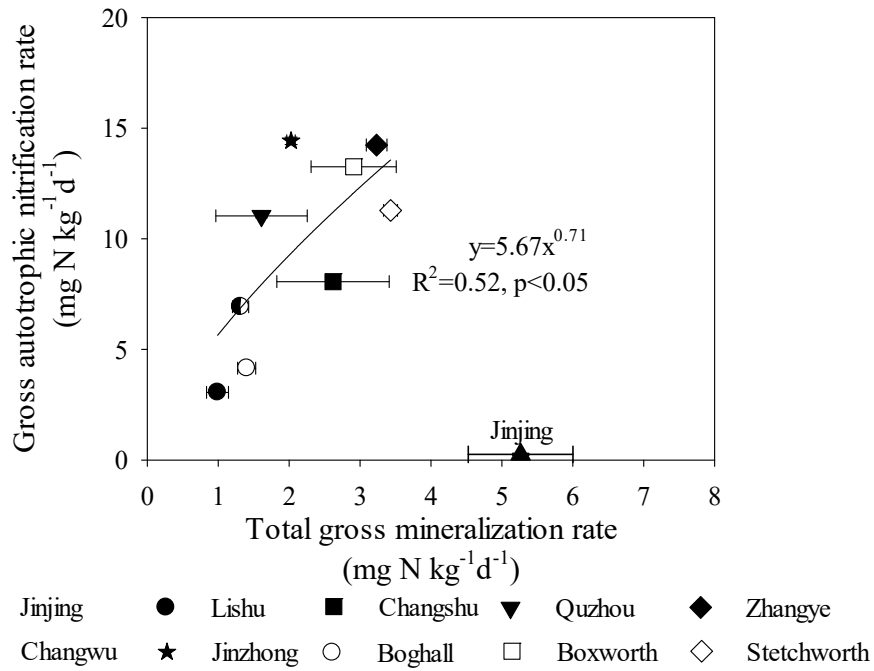


Fig. 5 (Zhu GD)



40

41

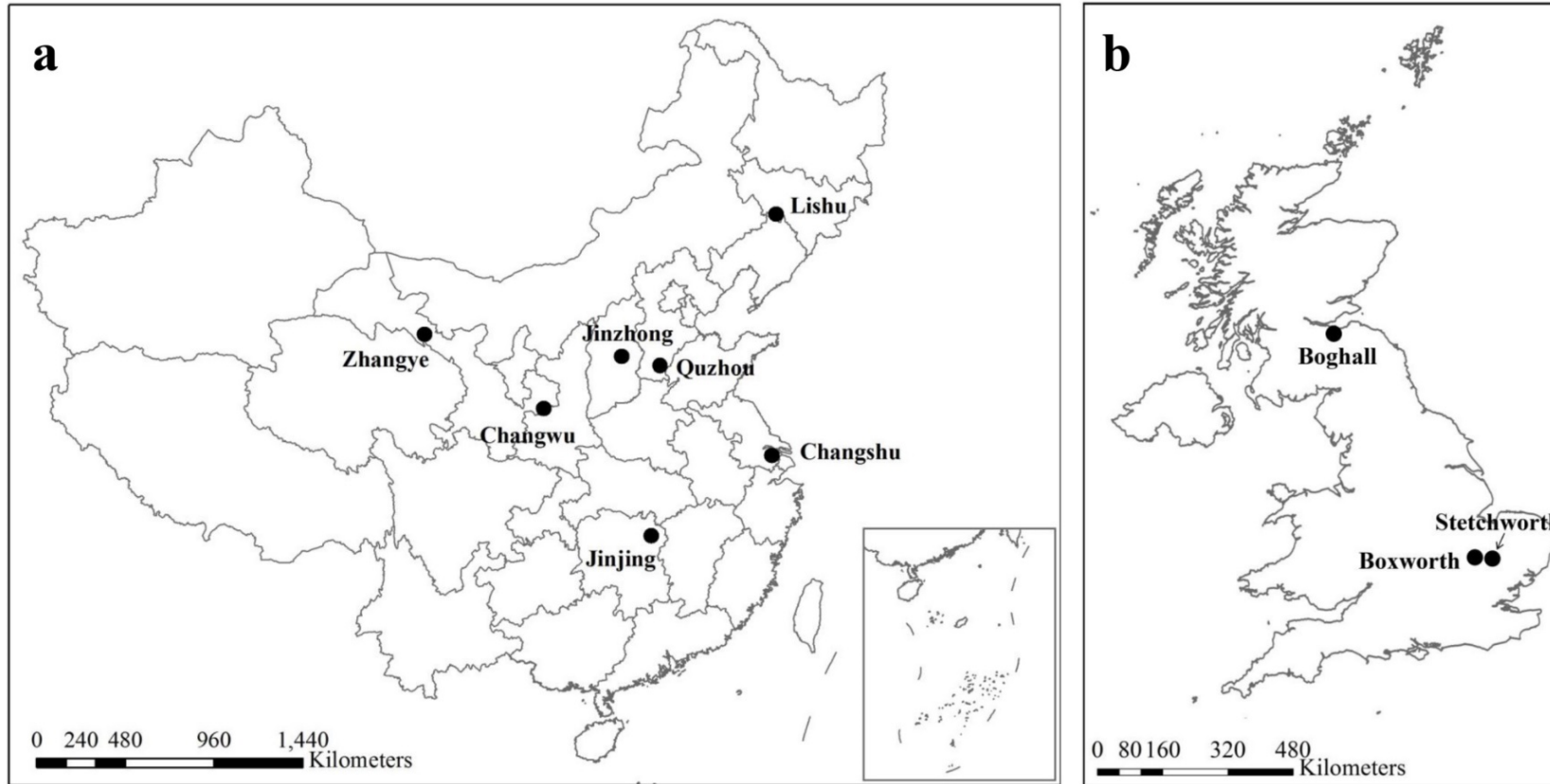
Fig. 6 (Zhu GD)

- 1 **Table S1** NH_4^+ and NO_3^- concentrations [Mean (SD) / mg N kg^{-1}] at different sampling times and net mineralization and nitrification rate (mg N
- 2 $\text{kg}^{-1}\text{d}^{-1}$) during different incubation periods in ten studied soils.

	Initial NH_4^+ in soil	Expected NH_4^+ after added NH_4NO_3 in zero time	NH_4^+				Initial NO_3^- in soil	Expected NO_3^- after added NH_4NO_3 in zero time	NO_3^-			
			at 0.5h	at 12h	at 24h	at 48h			at 0.5h	at 12 h	at 24 h	at 48 h
			A	B	C	D			E	F	G	H
Jinjing	8.03 (0.07)	28.03 (0.07)	24.28 (0.65)	23.78 (0.55)	23.94 (0.54)	25.94 (0.54)	15.37 (0.64)	35.37 (0.64)	48.73 (3.33)	50.57 (3.46)	50.15 (3.11)	51.73 (3.40)
Lishu	1.41 (0.17)	21.41 (0.17)	15.34 (0.70)	14.18 (0.25)	13.55 (0.54)	11.18 (0.25)	43.24 (0.67)	63.24 (0.67)	87.99 (5.84)	86.61 (3.13)	91.89 (2.77)	89.67 (9.22)
Changshu	0.10 (0.00)	20.10 (0.00)	13.40 (0.65)	8.78 (0.27)	5.03 (0.42)	1.15 (0.22)	9.02 (0.85)	29.02 (0.85)	41.07 (1.00)	43.86 (3.04)	49.82 (0.84)	47.35 (1.69)
Quzhou	0.14 (0.08)	20.14 (0.08)	12.27 (0.23)	8.08 (0.19)	4.47 (0.23)	0.48 (0.12)	31.96 (0.11)	51.96 (0.11)	70.14 (5.46)	74.55 (0.87)	83.71 (4.38)	88.90 (6.17)
Zhangye	0.75 (0.07)	20.75 (0.07)	13.09 (0.73)	7.84 (1.08)	2.38 (0.77)	0.89 (0.08)	35.23 (0.67)	55.23 (0.67)	81.65 (6.14)	86.83 (8.36)	111.36 (9.68)	110.66 (1.52)
Changwu	0.72 (0.03)	20.72 (0.03)	13.47 (0.47)	10.43 (1.04)	6.97 (1.01)	1.63 (0.35)	14.31 (0.25)	34.31 (0.25)	59.58 (5.30)	69.52 (5.13)	80.20 (7.27)	82.11 (8.02)
Jinzhong	0.58 (0.17)	20.58 (0.17)	14.26 (0.77)	7.13 (0.38)	1.42 (0.29)	0.60 (0.06)	43.56 (4.23)	63.56 (4.23)	92.87 (2.74)	103.04 (3.57)	115.86 (11.67)	112.70 (2.81)
Boghall	0.03 (0.01)	20.03 (0.01)	11.58 (0.12)	10.95 (0.26)	10.13 (0.22)	8.12 (0.40)	62.36 (1.34)	82.36 (1.34)	83.03 (2.12)	82.85 (1.66)	87.81 (3.80)	92.18 (1.63)
Boxworth	4.70 (0.07)	24.70 (0.07)	10.44 (0.46)	6.97 (0.42)	2.50 (0.02)	0.37 (0.25)	19.88 (2.88)	39.88 (2.88)	41.57 (1.86)	51.75 (0.76)	54.65 (1.12)	69.57 (2.03)
Stetchworth	0.98 (0.10)	20.98 (0.10)	12.43 (0.10)	8.81 (0.21)	4.33 (0.53)	0.66 (0.23)	33.77 (1.13)	53.77 (1.13)	51.48 (2.48)	58.86 (0.85)	67.62 (3.37)	70.03 (2.57)

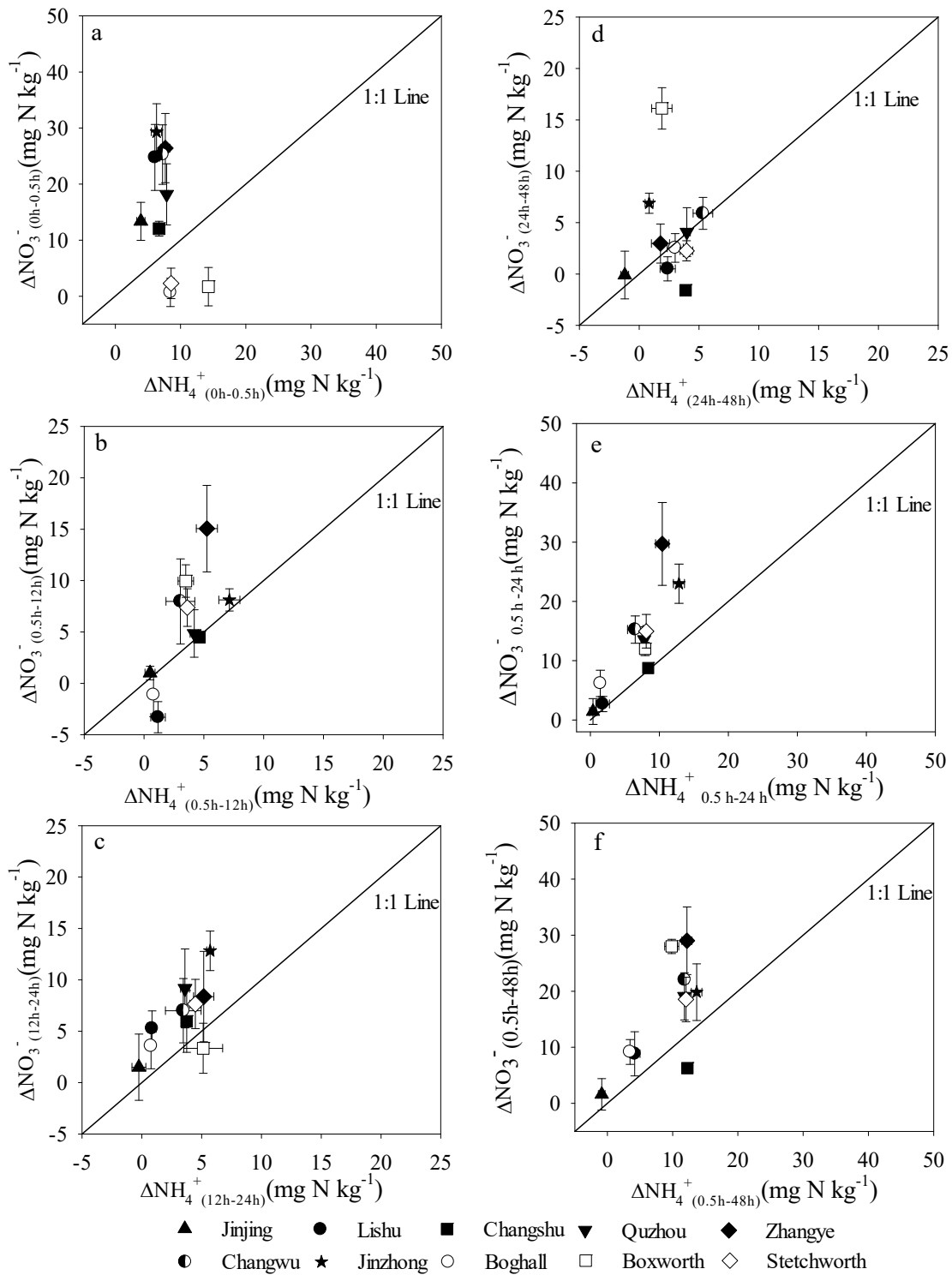
ΔNH_4^+ (0.5h- 12h)	ΔNH_4^+ (12h- 24h)	ΔNH_4^+ (24h- 48 h)	ΔNH_4^+ (0.5h- 24h)	ΔNH_4^+ (0.5h- 48 h)	ΔNO_3^- (0.5h- 12h)	ΔNO_3^- (12h- 24h)	ΔNO_3^- (24h- 48 h)	ΔNO_3^- (0.5h- 24h)	ΔNO_3^- (0.5h-48 h)	Net minerali zation (0.5h- 12h)	Net minerali zation (0.5h- 24h)	Net minerali zation (0.5h- 48h)	Net nitrificati on (0.5h- 12h)	Net nitrificat ion (0.5h- 24h)	Net nitrific ation (0.5h- 48h)
A-B	B-C	C-D	A-C	A-D	E-F	F-G	G-H	E-G	E-H						
0.50 (0.42)	-0.25 (0.58)	-1.21 (0.33)	0.38 (0.49)	-0.88 (0.52)	-1.00 (0.66)	-1.5 (3.22)	0.1 (2.32)	-1.42 (2.18)	-1.60 (2.79)	2.78 (3.65)	2.30 (1.81)	2.94 (1.49)	6.26 (1.38)	1.28 (0.90)	2.59 (1.74)
1.17 (0.61)	0.85 (0.34)	2.38 (0.64)	1.79 (0.98)	4.17 (0.82)	3.30 (1.52)	-5.27 (1.71)	-0.5 (1.17)	-2.69 (1.28)	-8.84 (3.94)	-7.87 (2.66)	1.84 (1.67)	3.04 (1.58)	-8.44 (0.71)	2.75 (1.31)	4.47 (1.90)
4.63 (0.44)	3.75 (0.35)	3.88 (0.45)	8.38 (0.34)	12.25 (0.68)	-4.50 (0.20)	-5.96 (3.01)	1.58 (0.29)	-8.75 (0.60)	-6.28 (0.93)	-3.30 (4.38)	0.57 (0.37)	-3.01 (0.74)	9.39 (0.43)	8.94 (0.68)	3.17 (0.47)
4.19 (0.34)	3.61 (0.37)	3.99 (0.29)	7.80 (0.16)	11.79 (0.28)	-4.85 (2.31)	-9.16 (3.84)	-4.08 (2.38)	-13.57 (2.17)	-19.09 (4.21)	0.44 (3.07)	5.21 (1.60)	3.70 (2.27)	16.30 (5.27)	13.85 (2.21)	9.64 (2.13)
5.25 (0.88)	5.17 (0.85)	1.78 (0.76)	10.42 (0.98)	12.20 (0.78)	-15.06 (4.21)	-8.37 (4.40)	-2.97 (1.90)	-29.71 (6.99)	-29.01 (6.04)	-0.15 (5.32)	19.42 (2.41)	9.33 (2.41)	31.42 (8.80)	28.18 (5.32)	14.66 (3.05)
3.04 (1.21)	3.46 (1.49)	5.33 (0.82)	6.5 (1.11)	11.83 (0.61)	-7.97 (4.15)	-7.0 (3.13)	-5.90 (1.55)	-15.25 (2.30)	-22.04 (0.31)	13.31 (8.76)	8.56 (3.06)	5.30 (0.27)	19.31 (7.24)	15.58 (2.34)	11.14 (0.15)
7.13 (0.88)	5.71 (0.29)	0.83 (0.34)	12.84 (0.82)	13.66 (0.80)	-8.12 (1.09)	-12.82 (1.93)	6.89 (0.98)	-22.99 (3.32)	-19.84 (5.05)	6.35 (2.61)	11.68 (3.01)	4.18 (1.43)	20.61 (2.29)	23.48 (3.39)	10.94 (1.37)
0.79 (0.09)	0.76 (0.13)	2.01 (0.67)	1.45 (0.25)	3.46 (0.47)	1.12 (2.12)	-3.57 (2.22)	-2.54 (1.40)	-6.17 (2.22)	-9.16 (2.21)	-1.61 (1.68)	4.90 (2.40)	3.24 (0.76)	-2.95 (4.58)	6.30 (2.26)	4.63 (1.12)
3.49 (0.65)	5.13 (1.62)	1.90 (0.85)	7.94 (0.47)	9.84 (1.07)	-9.95 (1.58)	-3.34 (2.44)	-16.12 (2.01)	-11.98 (1.12)	-28.0 (1.27)	13.49 (2.61)	4.13 (1.41)	9.18 (0.96)	20.77 (3.30)	13.36 (2.93)	14.15 (0.64)
3.62 (0.19)	4.48 (0.47)	3.95 (0.53)	8.10 (0.44)	12.04 (0.46)	-7.38 (1.83)	-7.65 (2.40)	-2.26 (0.98)	-14.97 (2.83)	-18.55 (3.97)	6.57 (2.17)	8.22 (3.71)	3.92 (1.39)	14.11 (2.38)	15.29 (2.90)	9.37 (2.00)

4 **Figure S1** Map showing the sampling sites in China (a) and UK (b).

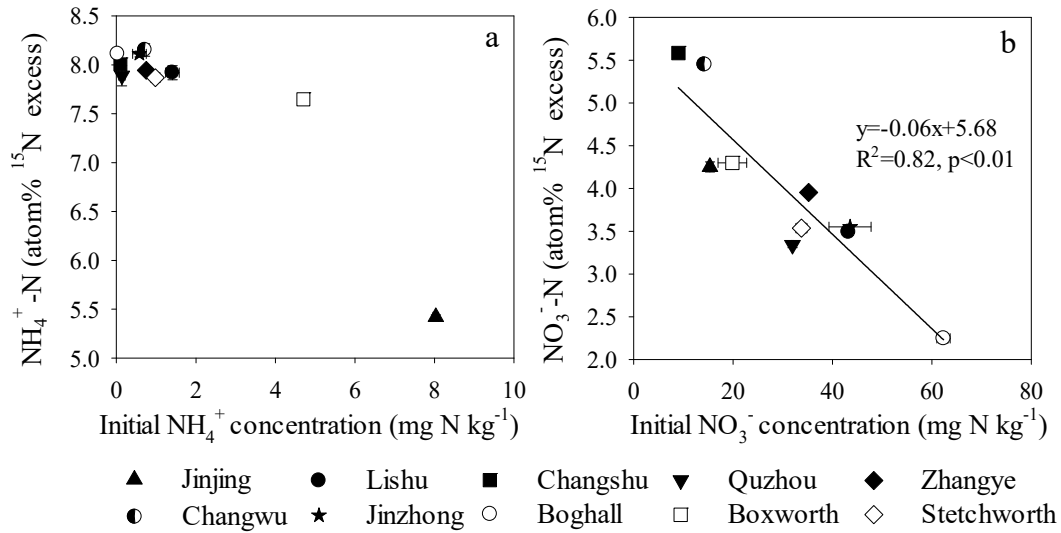


5
6

7 **Figure S2** Relationships between the change of NH_4^+ concentration (ΔNH_4^+) and
 8 change of NO_3^- concentration (ΔNO_3^-) during 0h-0.5h (a), 0.5h-12h (b), 12h-24h (c),
 9 24h-48h (d) and 0.5h-24h (e) and 0.5h-48h (f). Error bars represent standard deviation
 10 ($n=6$), some error bars are covered by symbols due to the low standard deviation values.

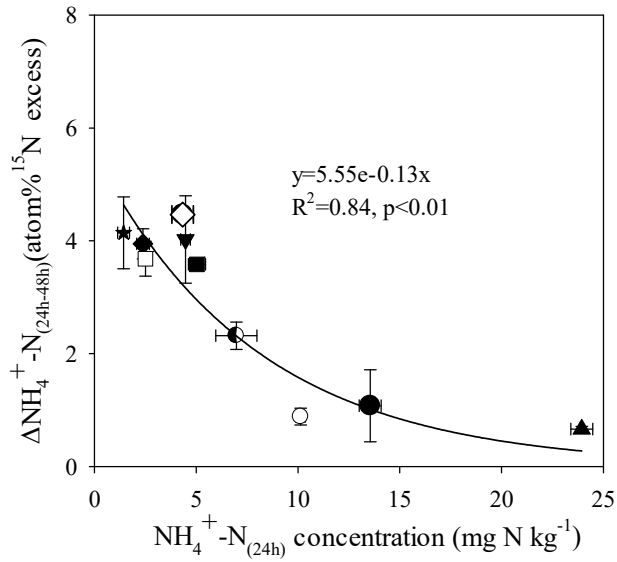


12 **Figure S3** Relationship between initial NH_4^+ concentration and atom% ^{15}N excess of
 13 the NH_4^+ pool (0.5 h) in $^{15}\text{NH}_4\text{NO}_3$ treatment (a), and initial NO_3^- concentration and
 14 atom% ^{15}N excess of the NO_3^- pool (0.5 h) in $\text{NH}_4^{15}\text{NO}_3$ treatment (b) in ten studied
 15 soils. Error bars represent standard deviation (n=3), some error bars are covered by
 16 symbols due to the low standard deviation values.



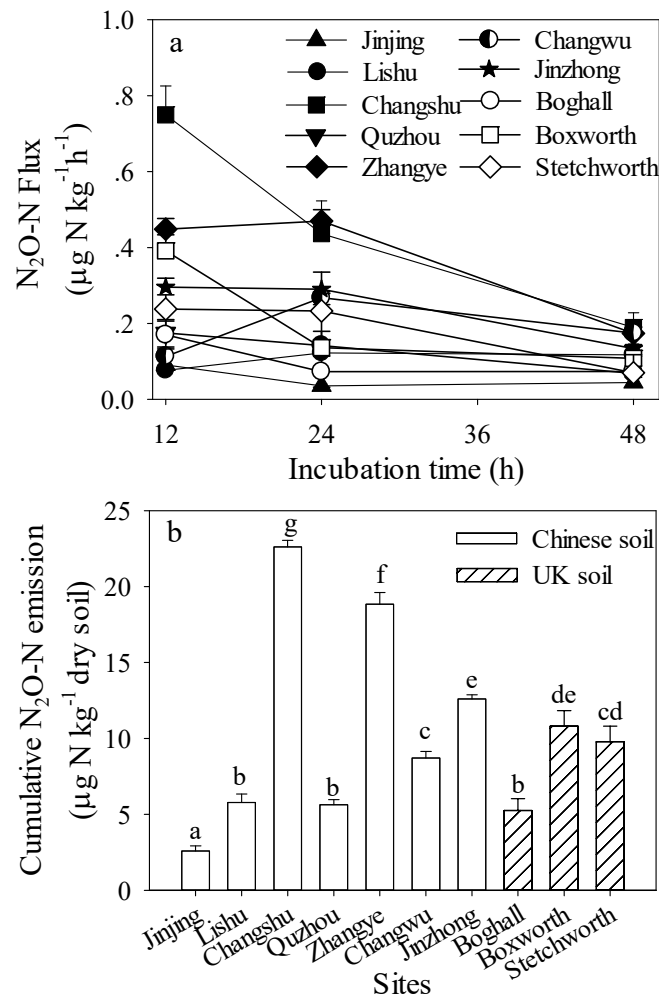
17

18 **Figure S4** Relationship between $\text{NH}_4^+\text{-N}$ concentration at 24h and the change of atom %
 19 ^{15}N excess of the NH_4^+ pool in the $^{15}\text{NH}_4\text{NO}_3$ treatment during 24h-48h incubation.

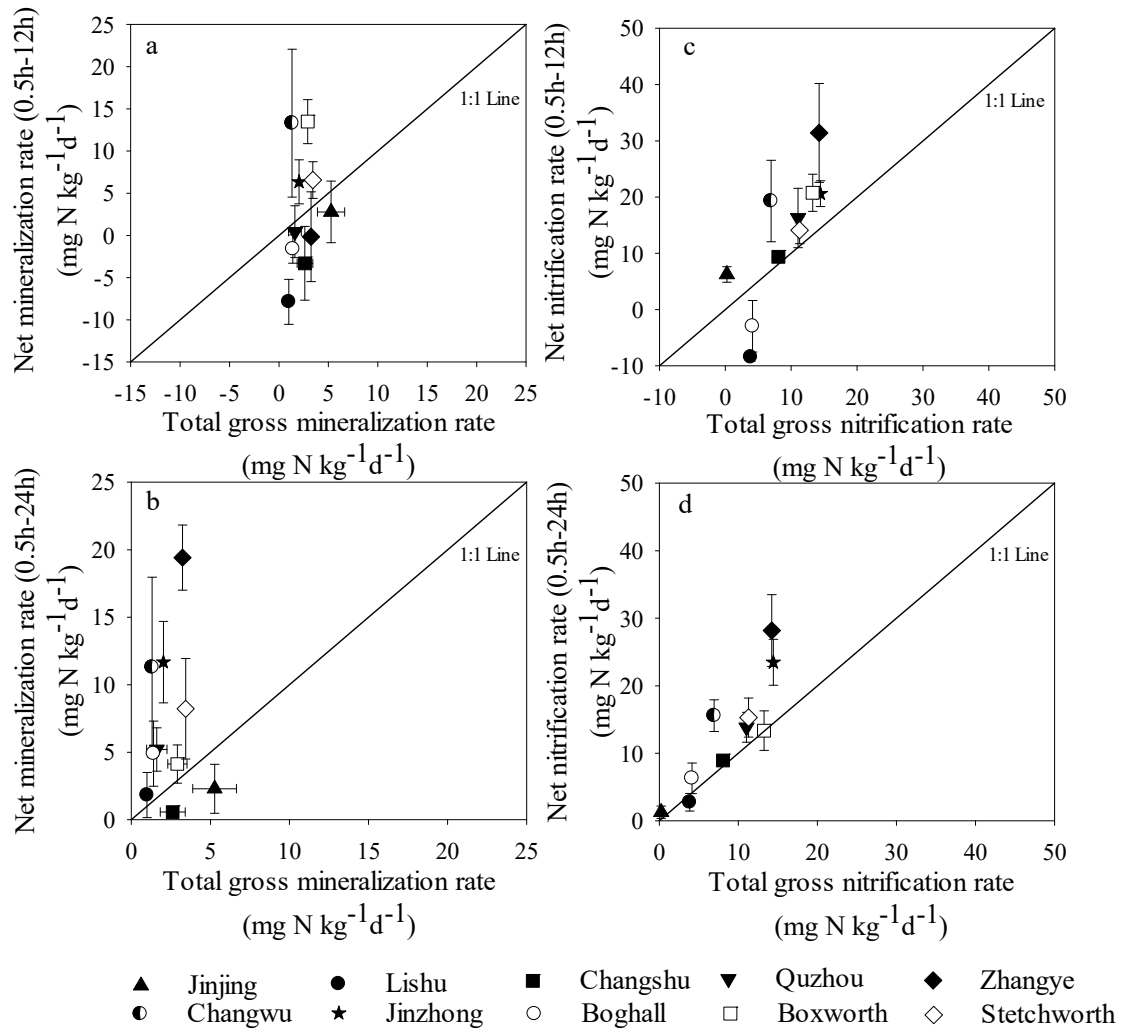


- 20
- ▲ Jinjing
 - Lishu
 - Changshu
 - ▼ Quzhou
 - ◆ Zhangye
 - Changwu
 - ★ Jinzhong
 - Boghall
 - Boxworth
 - ◇ Stetchworth

21 **Figure S5** N₂O flux (a), and cumulative N₂O emission (b) in the ten studied soils during
 22 the 48h incubation. Error bars represent standard deviation (n=3). Some error bars in
 23 Fig. S5a are covered by symbols due to the low standard deviation values. In Fig.S5b,
 24 different letters indicate significant differences ($p < 0.05$) among soils.



26 **Figure S6** Relationship between total gross mineralization rate and net mineralization
 27 rate during the 0.5h-12h (a) and 0.5h-24h (b), and total gross nitrification rate and net
 28 nitrification rate during the 0.5h-12h (c) and 0.5h-24h (d) incubation time in ten studied
 29 soils. Error bars represent standard deviation (horizontal error, n=3; vertical error, n=6),
 30 some error bars are covered by symbols due to the low standard deviation values.



32 **Figure S7** Relationship between total gross mineralization rate and SOC (a), TN (b) and C/N (c); between gross autotrophic nitrification rate and
 33 SOC (d), TN (e) and C/N (f) in ten studied soils. Error bars represent standard deviation (n=3), some error bars are covered by symbols due to the
 34 low standard deviation values.
 35

