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**Aggregate turnover in a dryland red soil after long-term application of chemical
4 fertilizers or manure**

4

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26 **Abstract**

Physical protection of carbon (C) in soil aggregates is an important mechanism affecting
28 soil organic carbon (SOC) stocks but there is little information on the turnover dynamics of
aggregates in dryland red soils after different long-term fertilization practices. Different
30 aggregate size classes in zero fertilizer (CK), chemical fertilizer (CF), and chemical fertilizer
combined with manure (MCF) treatments were examined. The Roth C model was used to
32 simulate C inputs based on SOC dynamics, the carbon, aggregation, and structure turnover
(CAST) model was used to evaluate soil structure and C sequestration at different aggregate
34 size classes with time. MCF treatment significantly ($P < 0.05$) increased total macroaggregate
C, and fractions of macroaggregate C compared with the other two treatments because C
36 inputs and C sequestration rate increased significantly ($P < 0.05$) according to Roth C model
simulation. The CAST model performed well in simulating the changes in soil structure and
38 organic C stocks in different aggregate size classes with time. The simulation results indicate
that particulate organic matter (POM) is the primary source of aggregate turnover, that free
40 silt-clay particles have a more rapid turnover than the silt-clay particles in other (larger)
aggregates, and that the disruption criterion for aggregate distribution in CAST model
42 parameters followed the sequence $MCF > CF > CK$ in the low pH and low SOC red soil.
Overall, the CAST model is a good tool for simulation of aggregate dynamics in red soil
44 under different fertilization treatments.

46 **Keywords:** soil aggregates; carbon stocks; fertilizer practices; model simulation; red soil

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Introduction

52 Climate warming induced by increasing atmospheric carbon dioxide concentrations is
widely accepted to be occurring and is an issue of increasing concern (Huesh et al., 2017;
54 Zhou et al., 2018; Jia et al., 2020). As a consequence, C storage and sequestration potential in
soils have attracted considerable interest (Carvalhais et al., 2014; Xiao et al., 2020) because
56 the amount of C stored in soils is about twice that stored in the atmosphere (Lal, 2004). The
protection of aggregate C by soil organic matter (SOM) is an important mechanism affecting
58 SOC stock (Six and Paustian, 2014; Garland et al., 2023). Moreover, soil aggregate structure
in agricultural land can be influenced by management practices such as tillage and
60 application of chemical fertilizers or exogenous C (Zhou et al., 2022; Liu et al., 2023; Dorji et
al., 2020). Understanding the mechanisms by which protected C pools respond to different
62 fertilization practices is crucial for maximizing SOC stocks.

Different fertilization practices can affect soil aggregate quantity and quality as well as
64 SOC and aggregate C concentrations (Lichter et al., 2008; Ramteke et al., 2024). Intensive
cropping systems with high C inputs from aboveground to belowground or exogenous C
66 application result in aggregate formation. Carbon accumulation in soils to which chemical
fertilizers are applied occurs mainly in free silt and clay particles. Further manure application
68 can increase the physically protected C and promote SOC accumulation (Glude et al., 2008;
Yu et al., 2012; Wen et al., 2021). Neff et al. (2002) report that nitrogen inputs promote the
70 decomposition of light fraction C (labile pools) and increase the stability of heavy fraction C
(stable pools). Besides, in agricultural systems frequent removal of aboveground production
72 and regular tillage may lead to complex changes in SOC induced by fertilization practices in
the short term, while the field located experiment with long-term fertilization practices can
74 effectively avoid the effect of tillage and other agronomic practices, and clearly demonstrate
the dynamics of SOC changes induced due to fertilization practices (Qiu et al., 2016; Schmidt

76 et al., 2011), therefore, the effects of fertilization practices on SOC and aggregates are well
illustrated by long-term fertilization experiment.

78 Aggregates are the structural units in soils that control SOC dynamics. The complex
structure of SOM is often arbitrarily divided into labile and stable pools (Brown et al., 2014;
80 Gulde et al., 2008). Labile SOM has a more rapid turnover than stable SOM. Particulate
organic matter (POM) is usually an important labile fraction that plays an important role in
82 short-term C cycling (Gulde et al., 2008), while stable SOC pools mainly determine the
magnitude of long-term C stocks (Zhou et al., 2017). In different soil aggregates the increase
84 in SOC is partly determined by the link between macroaggregate turnover, microaggregate
formation, and C stabilization within microaggregates (Six et al., 2000). It is therefore
86 necessary to explore the amount and the duration of C stocks in different aggregate size
classes in order to clarify the effects of different aggregate size classes on SOC stocks.

88 Mathematical models can effectively evaluate and predicate SOC dynamics (Coleman
and Jenkinson, 1999; Keating et al., 2003; Malamoud et al., 2009; Wang et al., 2013). Stamati
90 et al. (2013) recently developed a model of coupled C, aggregation, and structural turnover
(CAST) which integrates the advantages of the Struc-C model (Malamoud et al., 2009) and
92 the RothC model (Coleman and Jenkinson, 1999). The CAST model fully considers each
aggregate type as a single C pool and the input of exogenous organic materials and POM
94 derived from the decomposition of exogenous organic materials (Li et al., 2017; Stamati et al.
2013), and the parameters and C pools in RothC model are introduced into different size
96 aggregates in the CAST model. The CAST model also simulates the turnover rate of each
aggregate size and its feedbacks on SOC dynamics. Thus, the CAST model can better explore
98 the relationships among different aggregates in soils and the response of aggregate C
dynamics to management practices and other factors.

100 Red soils account for 22% of the total land area in China and are characterized by low

soil organic matter and available nutrient contents, poor soil structure, and high acidity and
102 aluminum toxicity. Currently, drylands with red soils are increasingly being cultivated to
meet the demand for forage maize or other high-income crops. In order to explore changes in
104 aggregate C in dryland red soils, a soil from a dryland maize cropping system under different
long-term fertilizer practices was selected to (1) explore changes in soil aggregate C under
106 long-term applications of chemical fertilizer and a combination of chemical fertilizer and
manure and (2) elucidate the turnover rate in different aggregate size classes under different
108 long-term fertilization practices using CAST model simulations.

110 **Materials and methods**

Site description

112 The long-term double maize cropping system began at the site (28°15'N, 116°20'E) in the
spring of 1986, and spring soybean or peanut was planted before 1986. The experimental site
114 is located at the Red Soil Research Institute in Jiangxi province, south China. This region has
a typical subtropical humid monsoon climate with an annual average temperature and
116 precipitation of 18.1 °C and 1620 mm, respectively, from 1986 to 2014. Red soils (Ferralic
Cambisols according to the FAO soil classification system) are the typical soil type. Local
118 climatic information during the experimental period is shown in Fig. 1 and has been obtained
from the meteorological station at our experimental sites in the China Meteorological Data
120 Sharing Service System (<http://www.cma.gov.cn>). Basic information of surface soil
properties (0-20 cm depth) at the initial of the located experiment was shown in Table 1.

122 **Table 1, Fig.1**

124 **Experimental design**

The field experiment comprised three treatments in triplicate with plots 22.2 m² in area in

126 a randomized complete block design. The treatments were one fertilizer treatment (CF), one
treatment combining manure and fertilizer (MCF) and unamended plots as a control (CK).
128 The fertilizer rate and cropping system are shown in Table 2. The chemical fertilizers applied
were urea, triple superphosphate or calcium superphosphate, and potassium chloride and the
130 manure used was pig manure. During the crop growth period the nitrogen (N) fertilizer
application was split twice as basal fertilizer and topdressing fertilizer and the phosphorus (P),
132 potassium (K), and manure were applied as basal fertilizers. Spring maize was sown at the
beginning of April and harvested in the middle of July and then summer maize was planted
134 and harvested at the beginning of November. The soil was ploughed after summer maize
harvested.

136 After the crop harvest in 2014, soil samples were collected from three points in each plot,
and then the fresh soil samples were immediately passed through a 5-mm sieve and air-dried.
138 The air-dried soil samples in 2014 and 1986 were isolated with wet sieving method to
analyze soil aggregates. In each treatment at the start (1986) and end (2014) of the
140 experiment, the determined data of mass and C stock in different size class aggregates and
SOC concentration with a total 24 data were input CAST model to simulate the dynamic of
142 soil aggregates. In order to simultaneously capture the regular curves of the dynamics of the
mass and C concentration in different size class aggregates, the CAST model simulation
144 originally ran with the mean monthly historical data from 1986 to 2014 for a period of 40
years.

146 **Table 2**

148 **Water-stable aggregate fractionation**

Soil aggregate separation was conducted with size density fractionation using wet sieving
150 according to Elliott (1986), and particulate organic matter was separated from the mixture of

POM and sand using heavy liquid according to Brown et al. (2014) and Gulde et al. (2008).
152 As described by Elliott (1986), water stable aggregates (WSA) were separated into three size
classes comprising macroaggregates (AC3) ($>250 \mu\text{m}$), free microaggregates (AC2) ($250 -$
154 $53 \mu\text{m}$), and free silt-clays (AC1) ($< 53 \mu\text{m}$) using a wet sieving method. Briefly, 80.0 g air
dried soil on the top of a $250 \mu\text{m}$ sieve was submerged in deionized water for 5 min at room
156 temperature and separated with 50 vertical movements of > 2 min. Floating material was
removed with a net during submergence. The efficiency of recovery during wet sieving
158 averaged 98.7% (range 98.2-100.7%).

The microaggregates within macroaggregates (AC2,3) ($250 - 53 \mu\text{m}$) were further
160 isolated based on the method of Six et al. (2000). Briefly, ≤ 15.0 g of oven-dried
macroaggregate subsample (AC3) was slaked for 15 min and transferred onto a $250\text{-}\mu\text{m}$ mesh
162 screen with a transparent plastic wall and the screen was shaken in running water with 50
glass beads (4 mm in diameter) until the water became clear. The $<250 \mu\text{m}$ soil slurry
164 continued to pass through a $53\text{-}\mu\text{m}$ mesh screen by wet sieving by water stable aggregate
fractionation. The remaining materials with size ranges $>250 \mu\text{m}$, $250 - 53 \mu\text{m}$ and $< 53 \mu\text{m}$
166 were coarse particulate organic matter plus sand (cPOM + sand), AC2,3, and silt-clays within
AC3 (AC1,3). The recovery efficiency was 99.4% (range 98.3-100.8%).

168 The AC2,3 particles comprised two parts, namely fine particulate organic matter (fPOM)
and silt-clays in AC2,3 (AC1,2,3). Firstly, the oven-dried AC2,3 was shaken for 12 h with 0.5%
170 sodium hexametaphosphate at a ratio of 1:3 (soil:liquid, w/v). The dispersed slurry was
passed through a $53\text{-}\mu\text{m}$ sieve, and the $> 53 \mu\text{m}$ and $< 53 \mu\text{m}$ particles were fPOM + sand in
172 AC2,3 and AC1,2,3 particles.

Finally, soil cPOM + sand and fPOM + sand in AC2,3 particles were isolated to remove
174 the sand with sodium polytungstate at a density of 2.3 g cm^{-3} at a ratio of 1:3 (soil:liquid,
w/v). Soil particles fractionated with sodium polytungstate were washed 7 -10 times with

176 deionized water on a 20- μ m mesh using a vacuum filtration device. The washed particles in
this step comprised cPOM and fPOM in AC2,3 (fPOM_inAC3).

178 At each of the fractionation steps above the separated soil particles were oven-dried at
60 °C and then weighed on a balance with 0.01 or 0.0001 g precision. After the oven-dried
180 soil samples were passed through a 0.15-mm sieve the C concentrations in the soil particles
were determined with a CN analyzer (Macrocube, Elementar, Hanau, Germany).

182 In addition, the quantity and type of soil clay minerals were determined with an x-ray
diffractometer (Rigaku D/MAX 2500, Tokyo, Japan). Soil texture was determined with the
184 classical sedimentation-based pipette method. The methods for the determination of other
commonly analyzed soil properties were those used by Qiu et al. (2016).

186

Model description

188 The CAST model divides soil aggregates into macroaggregates, microaggregates, and
silt-clays. Macroaggregates are formed by POM which is derived from the plant litter
190 fragment and then decomposed by microorganisms. The decomposed POM associated with
silt-clay sized aggregates and polymers of microbial origin which are responsible for “gluing”
192 the structural components of aggregates subsequently form the microaggregates within
macroaggregates. Microbial activity decreases gradually with POM biodegradation and the
194 macroaggregates become unstable because of the lack of microbially-derived polymers. New
macroaggregates form and the cycle of aggregation and disaggregation continues when fresh
196 plant residues enter the soil.

The C pools in the RothC model are adopted at each aggregate size class in the CAST
198 model. In RothC model, the C pools are decomposable plant material (DPM), resistant plant
material (RPM), microbial biomass (BIO), humified organic matter (HUM), and inert organic
200 matter (IOM). The turnover rate of IOM is from centuries to millennia (Powlson et al., 2011)

and IOM therefore acts as a resistor to decomposition in both the Roth C model and CAST
202 models. In the CAST model, fresh plant litter is split into DPM and RPM and these fractions
are fragmented to form the coarse fractions of DPM (DPMc) and RPM (RPMc) which further
204 decompose to form the fine fractions of DPM (DPMf) and RPM (RPMf). The
macroaggregates contain DPMc, RPMc, DPMf, RPMf, BIO, HUM, and IOM, the
206 microaggregates contain DPMf, RPMf, BIO, HUM, and IOM, and the silt and clay pools are
composed of HUM and IOM.

208 During the running of the CAST model, climatic conditions, basic soil properties, WSA
distribution and organic C distribution at each aggregate size, and C inputs were input as
210 initial parameters. The main climatic conditions, e.g. mean monthly temperature, total
monthly precipitation, and total monthly evaporation were used to drive the model. The basic
212 soil properties were silt and clay content, bulk density, soil depth and sand mass in AC3 and
AC2 fractions. The WSA distributions and their C contents at the beginning of the
214 experiment are required as the initial conditions of the aggregates and then the turnover rate
is tuned to make the measured and simulated data coincidence. In the CAST model each C
216 pool of aggregate types decomposes by a first-order process with a specific rate constant and
the decomposition rate of each C pool is determined by three climatic factors (e.g.
218 temperature, precipitation, and evaporation) through their effects on microbial activity as
described by the Roth C model. With respect to the input parameters in each size aggregates
220 at the initial condition, half of the C content in POM (POM-C) is partitioned into
decomposable and resistant plant material, respectively; 5 % of the silt-clay fraction C is
222 partitioned into BIO and the remaining silt-clay fraction C is partitioned into HUM (Stamati
et al. 2013).

224 We simulated the crop C input data using the RothC model (version 2.1 for Windows)
with changing SOC content during the experimental period. The simulated plant C input is

226 uniformly distributed each month during crop growth. The C inputs in the CAST model were
0.370, 0.480, and 0.375 t C ha⁻¹ month⁻¹ in the control, CF and MCF treatments from April to
228 October during crop growth, and a manure C input of 1602 kg C ha⁻¹ per season crop in the
MCF treatment was added in April and July as shown in Table 1. Further details of the CAST
230 model can be found in Li et al. (2017), Panakoulia et al. (2017), Stamati et al. (2013), and the
RothC model user guide (Coleman and Jenkinson, 1999).

232

CAST model adjustment

234 In the CAST model the calibrated parameters comprise those that control 1) the turnover
rates of the full cycle of C processing from plant material fragmentation to POM
236 decomposition in aggregate fractions, 2) the relative contribution of silt-clay units and POM
in the aggregate size fractions, and 3) the criteria including disruption or tilling effects on
238 aggregate distribution, and the correction factor for silt-clay mass flow which describes
silt-clay unit distribution in different aggregates.

240 The disruption and tilling criteria for aggregate distribution determine the starting point of
different aggregate size classes by CAST model simulation. The calibrated decomposition
242 rates of different aggregate size classes and fragmented plant materials capture the change in
soil structure and organic C stocks in different aggregate size classes in a chronosequence. A
244 correction factor is used to adjust silt-clay mass flow in each aggregate type (Stamati et al.,
2013). The reciprocal value of these calibrated rate constants for aggregate formation and
246 disaggregation are reported as turnover time (y) as shown in Table 5.

The disruption and tilling criteria for aggregate distribution were first calibrated to
248 account for the correct timing of change in the particle distribution between the measurement
and simulation time series. Secondly, the parameters for turnover rates and the proportional
250 contribution of the components in aggregation were calibrated for year 2014 data. The

calibrated parameters (turnover times) (Table 5) were the POM fractions in most cases.
252 Finally, the correction factor values were calibrated to fine tune the mass of water-stable
aggregates. The rate constants for formation of each aggregate size class from the constituent
254 particle fractions, disaggregation of each size class of aggregate, decomposition for each pool
of SOC and the values for tillage and silt-clay mass flow criteria were calibrated using a
256 step-wise process. The calibration steps proceeded from initial values taken from the RothC
handbook and literature values from published observations or model results, which were
258 individually varied around the initial values to test sensitivity before systematically varying
the value to minimize RMSE, and then testing sensitivity of RMSE to the individual
260 calibrated values once the calibrated parameter set was complete.

Root mean square error (RMSE) and normalized RMSE (nRMSE) were employed to
262 evaluate the performance of the CAST model using simulated values and measured values
which were the values of different aggregate size classes in 2007. Agreement is considered to
264 be good when $nRMSE \leq 20\%$. The equations are as follows:

$$RMSE = \sqrt{\frac{\sum_{i=1}^n (S_i - M_i)^2}{n}}$$

266 $nRMSE = \frac{RMSE}{\bar{M}} \times 100$

Where S_i is the simulated value, M_i is the measured value, n is the number of measured
268 values, and \bar{M} is the average of the measured values.

According to Jamieson et al. (1991) reported, the model shows an “excellent”
270 performance if the $nRMSE \leq 10\%$, “good” performance if $10\% < nRMSE \leq 20\%$, “fair”
performance if $20\% < nRMSE \leq 30\%$, and “poor” performance if the $nRMSE > 30\%$.

272

Statistical analysis

274 Statistical analysis was performed using the SPSS 16.0 for Windows software package.

Mean values of the variables in different aggregate size classes and SOC among the three
276 treatments were compared using least significant difference at the 5% level.

278 **Results**

Measured SOC contents in 2014, RothC simulated SOC contents in 2014, and SOC
280 sequestration rate were significantly different ($P < 0.05$) among the three treatments (Table 3).
C inputs and CO₂-C emissions in the MCF treatment significantly greater ($P < 0.05$) than in
282 the control or the chemical fertilizer treatment.

Table 3

284

Compared to the control in 2014 (Table 4), AC2 and AC1 masses significantly ($P < 0.05$)
286 decreased by 20.2 and 13.4% in MCF as well as 31.7 and 25.0% in CF treatments, AC3 and
AC1,3 masses significantly ($P < 0.05$) increased by 15.0 and 40.2% in MCF as well as 9.9
288 and 20.6% in CF treatments. Moreover, MCF treatment significantly ($P < 0.05$) increased
AC1,3, cPOM and fPOM_inAC3 masses by 16.3, 68.0 and 155.6% compared to the CF
290 treatment.

The C concentrations in different aggregate size classes are also shown in Table 4.
292 Compared to the control, the C concentration in AC3, AC2,3, AC1,3, AC1,2,3, and
fPOM_inAC3 particles significantly ($P < 0.05$) increased by 287.9, 80.9, 87.4, 48.5 and 191.3%
294 in MCF as well as 23.5, 30.1, 37.8, 40.9 and 17.4% in CF treatments, respectively. The MCF
treatment had significantly ($P < 0.05$) higher C concentrations in SOC, AC3, AC2,3, AC1,3,
296 AC1,2,3, cPOM_inAC3, and fPOM_inAC3 particles than the control or CF.

Table 4

298

The CAST model (Fig. 2) well simulated the data acquired on aggregate mass distribution

300 and carbon stocks in different aggregate size classes in the control, CF, and MCF plots over
the 29 years from the start of the field experiment in 1986 to 2014. According to the SOC
302 stock distribution among the three treatments (Fig. 2d-f), SOC stocks increased in tandem
with increases in AC3 mass and organic C in AC3 particles in the first three years and then
304 SOC stocks increased in tandem with increasing AC2 mass and organic C in AC2 particles, as
shown in the MCF treatment (Fig. 2 f). Conversely, the decrease in SOC stocks in the first
306 few years was mainly due to the disruption of AC3 in our low C content soil (Fig. 2d, e; Table
1). In the MCF treatment (Fig. 2 i) all of the OC stocks in AC3 fractions increased and the
308 changes in AC1,2,3 and AC1,3 were higher than those in cPOM and fPOM_inAC3.

Fig. 2

310

Aggregate C turnover (Table 5) occurred in most cases in the fragmentation,
312 macroaggregates, microaggregates, plant litter pools, RPMc, RPMf, DPMc, and DPMf in
AC3 aggregates, and AC1 aggregates. In the decomposition of different aggregate size
314 classes the turnover time of RPMc and RPMf in AC3 aggregate type followed the sequence
CK < CF < MCF and that of HUM_{inAC2,3} and HUM_{inAC2} was CK < CF < MCF, while the
316 turnover time of HUM_{inAC1} was CK > CF > MCF.

The correction factor (Table 5) for slit-clay mass flow was related to C content and
318 aggregate mass flow in different aggregates. The range was 0.6 to 1.3 in all parameters in the
three treatments. The correction factor for AC1 was lower than AC1,3. The order of the
320 disruption criterion for aggregate distribution changes was MCF > CF > CK.

Table 5

322

The evaluation of RMSE and nRMSE between measured and simulated values was better
324 when their values decreased (Table 6). The nRMSE value was < 20% and the RMSE value

was < 1.6. The CAST model therefore well predicted the soil organic C stock distribution in
326 different aggregates. On the whole, the evaluation of nRMSE for SOC, AC3, and AC2
between measured and simulated values was slight better than for AC1 in the three
328 treatments.

Table 6

330

Discussion

332 1. Impact of fertilization practices on soil aggregates

The differences in SOC among the three treatments were closely related to C inputs
334 (Table 3) as shown by the simulated C inputs using the RothC model (Table 3), sufficient C
input may increase SOC stocks and C sequestration (Gattinger et al., 2012; Zhang et al.,
336 2015). In our study the order of significance ($P < 0.05$) of SOC concentration in 2014 was
MCF > CF > CK and the SOC sequestration rates in the CK and CF treatments showed C
338 losses (negative) while that in the MCF treatment showed C accumulation (positive) (Table
3). The C loss in CF treatment was attributable that the C input from maize didn't reach the C
340 output because of N input limitation, the optimal N input didn't generally exceed 210 kg N/ha
(Qiu et al., 2015); the second reason was possible that soil acidification because of long term
342 chemical fertilizer application suppressed maize growth in the acidic soil (Guo et al., 2010).
The study soil has a low pH in the CF treatment, while the combination of manure and
344 chemical fertilizers can restrain soil acidification and increase soil pH, resulting in pH values
of 4.66, 4.42, and 5.68 in the CK, CF, and MCF treatments in 2014.

346 Carbon inputs from aboveground or from manure application can enhance or impede
SOC stability in aggregates, further affecting aggregate formation and altering soil structure
348 (Li et al., 2017), as shown the significantly ($P < 0.05$) increase of the mass and C
concentration in AC3 in MCF treatment than the control (Table 4). The POM in AC3 is

350 mainly derived from decomposed plant residues and responds sensitively to soil management
practices (Gulde et al., 2008) and therefore sufficient manure application (Table 2) can
352 increase POM. For instance, the study site had significantly higher cPOM and fPOM_inAC3
in the MCF treatment than in the control or CF treatment. A greater turnover rate of POM
354 (Gulde et al., 2008) results in a lower POM pool capacity compared to the other aggregate
size classes (Table 4). The mass and amount of C in microaggregates (AC2 and AC2,3) can
356 be affected by crop residues, rhizosphere deposited C and exogenous C, and furthermore,
microaggregates are regarded as an indicator of C sequestration, especially AC2,3 (Gulde et
358 al., 2008; Six et al, 2000), as shown the mass and C concentration in AC1,2,3 and AC2,3 in
the present study (Table 4). Organic substrates can contribute to the C in $< 53 \mu\text{m}$ soil
360 particles. For example, there were significant ($P < 0.05$) differences in masses and C
concentrations in AC1,3 and the C concentration in AC1,2,3 among the three treatments due
362 to the fact that $< 53 \mu\text{m}$ soil particles bound into macroaggregate after exogenous C input and
because of the different physical protection of AC1,2,3 and AC1,3 in macroaggregates (Gulde
364 et al., 2008; Six et al, 2000). In addition, the significantly ($P < 0.05$) lower masses and
non-significant C concentrations in AC1 in the fertilization treatments than in the control also
366 indicate an increase in C concentration in AC1 after the application of fertilizers and/or
manures (Table 4).

368

2. Adjustment of aggregate turnover and parameters

370 The simulated data in different aggregates size class in CAST model well fitted the
measured data on the whole in our study soil as shown the nRMSE values in Table 6
372 (Jamieson et al.,1991). Water-stable aggregate mass and C stocks in different aggregates
gradually approached steady state from the start of the experiment to the sampling date (Fig.
374 2, Table 6). POM turnover plays an important role in aggregate C turnover because the

adjusted parameters mainly occurred in POM fractions (Table 5). Zeller and Danbire (2011) reported that POM was the primary N source for plants in a natural system. In plant litter pool decomposition in the CAST model parameters (Table 5), the control treatment had an obviously low turnover time of DPM and the MCF treatment had a little higher turnover time of DPM, while the opposite phenomenon can be found in the turnover time of RPM, this was because that (1) POM and N decomposition is the only N source in the control treatment, (2) manure per se has plenty of liable and resistant decomposed organic materials, which is an important source of POM in the MCF treatment, (3) plenty of chemical fertilizer N in the CF and MCF treatments can promote POM decomposition (Neff et al., 2002). In contrast, the turnover time of AC1 type decomposition is shorter than AC2 and the particles of AC2,3, because AC1 has free status in soils with a higher surface area than the other size aggregates. The large surface area readily retains applied fertilizer N (Yan et al., 2012) and the associated N further released to meet crop demand by microbial metabolism or the exchange of C and N in the rhizosphere. Therefore, the AC1 aggregates was a more important source of C and N than the other silt-clay particles in macroaggregates. In macroaggregates among the three treatments the turnover time of each fraction showed an increasing trend with C input rate, especially for HUM in macroaggregates and this is consistent with the increase in C concentration in the different macroaggregate fractions. Neff et al. (2002) reported that application of N fertilizers increased the stability of < 53 μm soil particles. Abiotic factors also play an important role in regulating soil aggregate turnover (Conant et al., 2011). For example, the disruption criterion for aggregate distribution was different in the three treatments and this may be related to soil properties and fertilization practices.

Regarding the differences in correction factors (cf) in each treatment, Stamati et al. (2013) reported that a value close to 1 denoted a linear relationship between silt-clay mass flow and OC flow, a value < 1 indicates high OC concentration hotspots induced by microaggregates,

400 and a value > 1 indicates that substantial parts of mineral surfaces are not covered with
organic matter. For example, the lower cf value in AC1 than AC1,3 in the three treatments
402 indicates that mineral surfaces in AC1 more readily associated with organic matter than
AC1,3. The increase in cf value in AC1,3 from 0.8 in the control to 1.2 in the fertilization
404 treatments (Table 5) indicates that C inputs in the fertilization treatments promote the
association between organic matter and mineral surfaces, and this improves aggregate
406 structure and increases aggregations C content, resulting in an increase in cf value in AC1,3
in the fertilization treatments.

408 In addition, the chemical or physical forces between cations (e.g. Fe^{3+} , Al^{3+} , Ca^{2+}) and
organic compounds or between clays and SOM particles result in changes in the different size
410 aggregate C contents and aggregate mass (Bronick and Lal, 2005). The dominant cations in
our soil were Fe^{3+} and Al^{3+} , and application of manures with high ion concentrations can alter
412 soil properties and further affect the soil aggregates. Tillage can disrupt soil aggregate
(Bronick and Lal, 2005), however, the same parameters for tillage effect on POM in CAST
414 model (Table 5) indicated that tillage didn't affect the change of soil aggregate under different
fertilization management practices in our studied soil. In the initial phase of simulation, the
416 sharp changes in different size aggregate may be related to the C input rate (Fig. 2), for
example, the low C input rate in the CK and CF treatments decreased SOC concentration
418 from the initial to the sampling date (Table 1, 3), furthermore, the lack of C resulted in the
fragmentation of macroaggregate and the formation of $< 250 \mu\text{m}$ aggregates, subsequently
420 the dynamic of the mass and C concentration in aggregates gradually stabilized because of
the long-term unchanged C input rate in each fertilization practice (Fig. 2).

422

Conclusions

424 Differences in nutrient applications can result in differences in C inputs to soils and

sufficient C inputs contributes significantly ($P < 0.05$) to SOC stocks, as shown by the MCF
426 treatment which had significantly greater SOC content and SOC sequestration rate than the
control or the fertilizer treatment. The combination of different fertilization practices and
428 modeling tools well simulated soil organic carbon, aggregates, and structural turnover as time
proceeded. The simulation results indicate the effects of POM on soil aggregate turnover and
430 the response of different size aggregate classes to fertilization practices. Overall, the CAST
model is a good tool for simulating the response of aggregate dynamics to different
432 fertilization treatments in this dryland red soil.

434 **Abbreviations**

SOC: soil organic carbon, **CK:** control (no fertilizer or manure), **CF:** chemical fertilizers,
436 **MCF:** combination of manure and chemical fertilizers, **CAST:** Carbon, Aggregation, and
Structure Turnover, **WSA:** water stable aggregates, **AC3:** macroaggregates ($>250 \mu\text{m}$), **AC2:**
438 free microaggregates ($250 - 53 \mu\text{m}$), **AC1:** free silt and clays ($< 53 \mu\text{m}$), **AC2,3:**
microaggregates within macroaggregates (AC2 within AC3), **AC1,3:** AC1 in AC3, **AC1,2,3:**
440 AC1 in AC2 within AC3, **POM:** particulate organic matter, **cPOM:** coarse POM, **fPOM:** fine
POM, **DPM:** decomposable plant material, **RPM:** resistant plant material, **cDPM:** coarse
442 DPM, **cRPM:** coarse RPM, **fDPM:** fine DPM, **fRPM:** fine RPM, **BIO:** microbial biomass,
HUM: humified organic matter, **IOM:** inert organic matter

444

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Table 1 Basic soil properties in the top 20 cm of the soil profile at the start of the experiment in 1986.

Soil property	Soil property		
Soil organic C (g kg ⁻¹)	9.4	pH	6.0
Total N (g kg ⁻¹)	1.0	Patent material	Quaternary red clay
Total P (g kg ⁻¹)	1.4	Soil texture	
Total K (g kg ⁻¹)	15.8	Sand (%)	21.2
alkali-hydrolyzable N (mg kg ⁻¹)	60.3	Clay (%)	27.6
Olsen-P (mg kg ⁻¹)	12.9	Silt (%)	51.2
NH ₄ Ac-K (mg kg ⁻¹)	102.0		

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580 Table 2 Fertilizer rates in the control (CK), chemical fertilizer (CF), and combined manure and chemical fertilizer (MCF) treatments in each crop in the field experiment. Units: kg ha⁻¹

Crop	CK	CF	MCF		
	N:P:K	N:P:K*	N:P:K	Manure N	Manure C
Spring maize	0:0:0	60:13:50	60:13:50	141	1602
Summer maize	0:0:0	60:13:50	60:13:50	141	1602

582 *, Chemical fertilizer applied, not including manure nutrients.

584 Table 3 Measured SOC contents in 2014 and simulated SOC contents in 2014, simulated C
 inputs, simulated CO₂ emissions, and SOC sequestration rates by the RothC model in the
 586 control (CK), chemical fertilizer (CF), and combined manure and chemical fertilizer (MCF)
 treatments.

	Measured SOC (t ha ⁻¹)	Simulated SOC (t ha ⁻¹)	Simulated input (t C ha ⁻¹ yr ⁻¹)	C Simulated CO ₂ -C emission (t C ha ⁻¹ yr ⁻¹)	SOC sequestration rate* (kg ha ⁻¹ yr ⁻¹ C input ⁻¹)
CK	20.2±1.2c	20.3±1.2c	2.6±0.3b	0.74±0.08b	-78.9±14.0c
CF	23.2±0.4b	23.1±0.3b	3.4±0.1b	0.92±0.02b	-30.9±4.8b
MCF	33.2±1.5a	33.2±1.6a	5.8±0.4a	1.47±0.10a	41.4±5.8a

588 NB: SOC, soil organic carbon

Means followed by a different letter within a column at each site are different at P < 0.05.

590 * Sequestration rate was that the difference between measured SOC between 2014 and 1986
 divided the multiplication of simulated C input and experimental years.

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598 Table 4 Water stable aggregate (WSA) mass and concentration at 0-20 cm soil depth in control (CK), chemical fertilizer (CF), and combined manure and chemical fertilizer (MCF) treatments in 2014.

WSA	Treatment	AC1 [#]	AC2	AC3	AC2,3	AC1,3	AC1,2,3	cPOM	fPOM_inAC3
Mass	CK	6.0±0.6a	33.6±1.3a	59.3±2.1b	32.3±2.1a	20.9±0.5c	31.7±2.2a	0.60±0.08b	0.06±0.01b
(%)	CF	4.5±0.4b	29.1±1.1b	65.2±1.3a	34.3±1.4a	25.2±0.6b	33.8±1.4a	0.75±0.11b	0.09±0.02b
	MCF	4.1±0.4b	26.2±1.9b	68.2±2.2a	32.9±1.8a	29.3±1.3a	32.2±1.7a	1.26±0.15a	0.23±0.02a
C concentration	CK	0.44±0.04a	2.51±0.25a	4.42±0.31c	2.36±0.14c	1.27±0.08c	1.71±0.03c	0.62±0.06b	0.23±0.011c
(g kg ⁻¹)	CF	0.39±0.05a	2.37±0.09a	5.46±0.06b	3.07±0.12b	1.75±0.04b	2.41±0.06b	0.75±0.11b	0.27±0.008b
	MCF	0.41±0.06a	2.69±0.26a	8.26±0.26a	4.27±0.12a	2.38±0.13a	3.32±0.05a	1.29±a0.10	0.67±0.020a

600 NB:

[#]AC3, macroaggregates (> 250 µm); AC2, microaggregates (250-53 µm); AC, silt and clay (< 53 µm).

602 AC2,3, microaggregates in macroaggregates (250-53 µm); AC1,3, silt and clay in macroaggregates (< 53 µm).

AC1,2,3, silt and clay within microaggregates in macroaggregates (< 53 µm).

604 cPOM, coarse particulate organic matter; fPOM, fine particulate organic matter;

[†] Values are means of three replicates.

606 [‡] Means followed by a different letter within a column at each site are different at P < 0.05.

608 Table 5 Calibrated parameters (turnover time) of aggregate C at 0-20 cm soil depth in control (CK), chemical fertilizer (CF), and combined manure and chemical fertilizer (MCF) treatments using CAST model simulation.

	Parameter	CK	CF	MCF
<u>Turnover time* (y)</u>				
Fragmentation [#]	RPM	6.67	5.56	3.33
	RPM _{cinAC3}	5.00	10.0	10.0
	DPM _{cinAC3}	20.0	20.0	2.00
Macroaggregates [§]	RPM _c	2.00	10.0	16.7
	DPM _c	0.95	0.71	0.63
Microaggregates [¶]	RPM _{finAC2,3}	14.3	16.7	5.00
	DPM _{finAC2,3}	14.3	16.7	5.00
Plant litter pool decomposition	DPM	0.13	0.13	0.33
	RPM	322.6	3.28	3.28
	RPM _c	5.00	10.0	5.00
	RPM _f	3.28	3.28	3.28
AC3 aggregate type decomposition	RPM _{cinAC3}	3.33	6.67	10.0
	RPM _{finAC3}	3.33	6.67	10.0
	DPM _{cinAC3}	0.33	0.28	0.36
	DPM _{finAC3}	0.33	0.33	1.00
	BIO _{inAC1,3}	1.67	1.67	2.00
	HUM _{inAC1,3}	100.0	142.9	200.0
	BIO _{inAC2,3}	1.67	1.67	2.00
	HUM _{inAC2,3}	76.9	142.9	200.0
	RPM _{finAC2,3}	1.97	40.0	48.3
	DPM _{finAC2,3}	2.00	40.0	66.7
AC2 aggregate typedecomposition	BIO _{inAC2}	1.67	1.67	2.00
	HUM _{inAC2}	76.9	142.9	200.0
	RPM _{finAC2}	4.83	4.83	48.3
	DPM _{finAC2}	6.67	6.67	66.7
AC1 aggregate type decomposition	BIO _{inAC1}	1.67	1.67	1.00
	HUM _{inAC1}	5.00	2.50	2.00
<u>Proportional contribution of the components in aggregation (%)</u>				
Macroaggregates	RPM _c	30.5	35.0	27.0
	DPM _c	30.5	35.0	27.0
	AC1	23.0	18.0	22.0
	AC2	16.0	12.0	24.0
Microaggregates	RPM _{finAC3aggr}	23.4	23.4	40.0
	AC1 _{inAC3}	76.6	76.6	60.0
<u>Correction factor for silt-clay mass flow (fraction)</u>				
	AC1	0.60	0.65	0.90
	AC2	1.30	1.00	1.30
	AC1,3	0.80	1.20	1.20
<u>Disruption criterion for aggregate distribution (%)</u>				
	DPM within AC3	0.0015	0.002	0.01
	(DPM + RPM) inAC2,3	0.0015	0.002	0.01
	(DPM + RPM) in AC2	0.0015	0.002	0.01
<u>Tilling criterion for aggregate distribution (%)</u>				
	DPM within AC3	0.0035	0.0035	0.0035
	(DPM + RPM) inAC2,3	0.0035	0.0035	0.0035
	(DPM + RPM) in AC2	0.0035	0.0035	0.0035

610 Aggregate symbols: see the abbreviations indications.

NB: Aggregate symbols see the Abbreviations indications;

612 * Turnover time is calculated at the reciprocal value of the respective calibration rate constants (1/y) in CAST model;

Fragmentation refers to the decomposition of exogenous plant material;

614 § Macroaggregate is coarse plant material mass transfer for macroaggregate formation;

¶ Microaggregate is fine plant material mass transfer for microaggregate formation within macroaggregate.

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Table 6 RMSE and nRMSE of validation for soil organic C stock distribution between the
 618 measured value and simulated value using the CAST model in control (CK), chemical
 fertilizer (CF), and combined manure and chemical fertilizer (MCF) treatments in 2007.

WSA fraction	Treatment	Measured value	Simulated value	RMSE	nRMSE (%)
		(tC ha ⁻¹)	(tC ha ⁻¹)		
SOC	CK	21.9	20.3	1.6	7.4
	CF	24.4	23.2	1.2	4.8
	MCF	32.3	31.9	0.4	1.2
AC3	CK	12.1	11.9	0.3	2.3
	CF	16.0	15.4	0.6	3.8
	MCF	24.4	25.1	0.7	2.7
AC2	CK	8.7	7.4	1.3	14.6
	CF	7.2	6.8	0.4	5.1
	MCF	6.7	5.6	1.0	15.2
AC1	CK	32.3	31.9	0.1	11.7
	CF	1.3	1.0	0.2	17.2
	MCF	1.2	1.0	0.2	15.2

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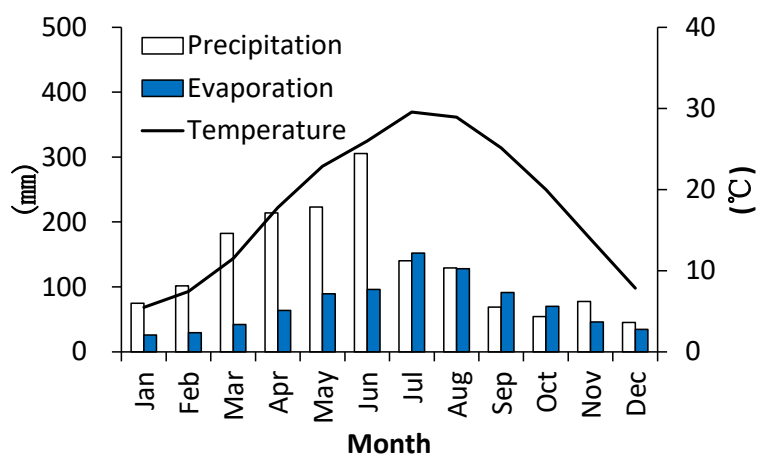
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634 Fig. 1 Averaged monthly temperature, total monthly precipitation, and total monthly
evaporation in Nanchang city, Henan province, during the period 1986-2014.

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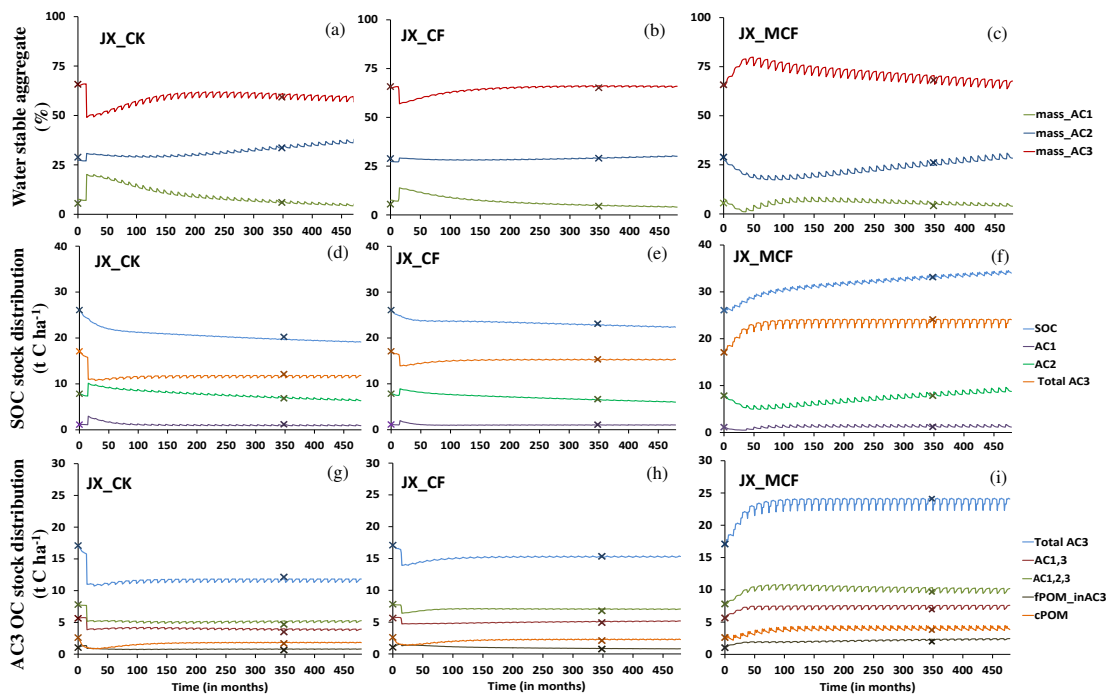
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660 Fig. 2 Water stable aggregates (%), soil organic carbon (SOC) stock distribution, and
 macroaggregate (AC3) OC stock distribution as affected by fertilization treatment in control
 662 (CK), chemical fertilizer (CF), and combined manure and chemical fertilizer (MCF)
 treatments in Jiangxi (JX) province using CAST model simulation.

664 Aggregate symbols in legend: see Table 3 footnote.

Crosses in Figure are the measured values of the samples at the start of the experiment and in
 666 2014.

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