- 1 Impacts of iron oxide nanoparticles on organic matter degradation and microbial
- 2 enzyme activities during agricultural waste composting
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### 11 Abstract

The effects of iron oxide nanoparticles (IONPs, including Fe<sub>2</sub>O<sub>3</sub> NPs and Fe<sub>3</sub>O<sub>4</sub> NPs) 12 on composting were investigated through evaluating their influences on organic matter 13 14 (OM) degradation, dehydrogenase (DHA) and urease (UA) activities, and quality of the final compost product. Results showed that composting amended with Fe<sub>2</sub>O<sub>3</sub> NPs was 15 more effective to facilitate OM degradation. At the end of composting, the total OM 16 loss in T-C, T-Fe<sub>2</sub>O<sub>3</sub> NPs and T-Fe<sub>3</sub>O<sub>4</sub> NPs was 66.19%, 75.53% and 61.31 17 respectively. DHA and UA were also improved on the whole by the amendme 18 19 IONPs, especially Fe<sub>2</sub>O<sub>3</sub> NPs. Although Relationships relationships between enzyme 20 activities and environmental variables were changed by different t eatme but temperature was the most influential to variations of both DHA and in all treatments, 21 which independently explained 75.1%, 34.7% and 38.4% 22 variations in the two enzyme activities in T-C, T-Fe<sub>2</sub>O<sub>3</sub> NPs and T-Fe<sub>3</sub>O<sub>4</sub> ectively. Compared with 23 res nen 24 DHA, UA was more closely related to the e al parameters. The germination index in T-C, T-Fe<sub>2</sub>O<sub>3</sub> NPs and T-Fe<sub>3</sub> 34.49%, 153.64% and 146.76%, and 25 va and 3.45 cm, respectively, indicating that 26 the average shoot length s y Fe<sub>2</sub>O<sub>3</sub> NPs, could promote seed germination and amendment of IONPs, especia 27 28 seedling growth. Therefore, composting amended with IONPs was a feasible and 29 promising method to improve composting performance, enzyme activities as well as 30 quality of the final compost product.

31 Keywords:

- 32 Composting; Iron oxide nanoparticles; Organic matter; Enzyme activity; Redundancy
- 33 analysis
- 34



### 35 1. Introduction

As one of the largest agricultural countries, China has produces produced more and 36 more crop straws in recent years which are regarded as waste materials. According to 37 38 Ministry of Agriculture of China (MOA, 2011), the annual output of straw residues in China was more than 600 million tons, ranking first in the world, and rice straw 39 accounted for the largest proportion of 32.3%. When it came to 2015, the total yield 40 exceeded 1.11 billion tons according to National Bureau of Statistics of China (N 41 2016). Traditionally, most of these straws are burned on-site, not only leading to s 42 air pollution and greenhouse gas emission, but also contributing tesoil fertility 43 degradation (Gong et al., 2009; Huang et al., 2018). Since the crop s 44 raw i ich of nutrient elements and organic matters (OM), exploring an approp thodology to 45 46 recycle it is a key way for sustainable crop production. Composting has been widely recommended as lendly, economical, and 47 ai eco-f practical way to recycle agricultural waste 8; Zhang et al., 2018b). 48 49 During the composting process, agri can be bio-decomposed into stable wa porganisms can also be eliminated, thereby 50 humus-like substances and oger mi 51 nvironmentally friendly soil conditioner, fertilizer and recycling the wasterecycled as remediation agent (Zhang et al., 2015). The readily available fractions of OM in 52 53 agricultural straw can be immediately utilized by composting microbes, while 54 polymeric organic compounds are dependent on the enzymatic activities to be degraded into directly available carbon and nitrogen sources (Jurado et al., 2015; He et al., 2018a). 55 56 Among the polymers, lignin and cellulose are the main components and the most

difficult to be degraded, thus inhibiting the composting process and reducing the final
compost quality. Many researches have tried to improve the OM degradation during
composting by various methods, such as by inoculation with exogenous
microorganisms (Jurado et al., 2015; Yang et al., 2018a), or by amendment of biochar,
zeolite, etc. (Hagemann et al., 2018; Yi et al., 2018).

62 The unique physical, electronic, optical, and biological properties of engineered 63 nanoparticles (ENPs) have resulted in a rapid increase in their application, including architecture, aerospace and airplanes, computer memory, catalysts, chem 64 environmental protection, etc. (Xu et al., 2012; Wang et al., 2018). With the production 65 and application of ENPs rapidly expanding, their environmental impact are a 66 acting more and more attentions. Iron is an essential nutrient for most of oc ganisms and 67 plants since it plays a critical role in cell growth. It also acts 68 a cofactor for many kinds of enzymes and is demanded for many biochem tions, such as respiration, 69 al rea 70 nitrogen fixation and DNA synthesis, etc. (L previous study suggested 71 that Fe<sub>2</sub>O<sub>3</sub> NPs could promote the gr pla such as mung bean (Vigna radiata) dehydrogenase (DHA) and urease activity 72 (Ren et al., 2011). It was also noi (UA) in soil positively responde to the treatments of iron oxide nanoparticles (IONPs, 73 74 including Fe<sub>2</sub>O<sub>3</sub> and Fe<sub>3</sub>O<sub>4</sub> NPs), in which Fe<sub>2</sub>O<sub>3</sub> NPs exhibited more significant effects, 75 as the changes of microbial community caused by IONPs could induce variations in enzyme activities (He et al., 2011; He et al., 2018b). DHA and UA are real reflections 76 of performance of biological process during composting in terms of OM degradation 77 78 and nitrogen cycle, and also provide information about maturity of the compost product

(Ye et al., 2017a; Ren et al., 2018b). However, as yet, few researches are concerned
about the enzyme activities and OM degradation during composting process with the
amendment of IONPs. Moreover, most of existing literatures-previous studies about
nanomaterials in composting systems were mainly focused on Ag or Ag-based NPs
(Gitipour et al., 2013; Stamou et al., 2016), so, additional kinds of NPs are also worth
studying.

Thus, the present study aimed to evaluate the OM degradation and enzyme activit 85 during composting which were amended with two types of IONPs (Fe<sub>2</sub>O<sub>3</sub> and 86 NPs) to improve and promote the process performance. Additionally, 87 the enzyme activities may be sensitive to the changes of environment, it is also neces 88 ary to aluate the relationships between enzyme activities and composting param rs and identify 89 the driving factors. The results of this study will provide a value 90 le basis for promoting the application of nanotechnology in the field of con 91 92 2. Materials and methods

93 2.1. IONPs preparation

Fe<sub>2</sub>O<sub>3</sub> NPs were was synthesized by forced hydrolysis of ferric nitrate solution according to Barton et al. (2014). Briefly, 60 mL of 1 M ferric nitrate solution was dripped into 750 mL of boiling ultrapure water. The boil was maintained using a hot plate with vigorous stirring during the whole process. After completion of ferric nitrate addition, the solution was removed from heat and cooled for 24 h. The synthetic NPs were purified with dialysis bag and stored at 4 °C in dark.

100 Chemical coprecipitation method was used for preparation of Fe<sub>3</sub>O<sub>4</sub> NPs according to

101 Yang et al. (2012) with a little modification. A mixture of iron solution was prepared by dissolving FeCl<sub>3</sub> 6H<sub>2</sub>O (12.3 g) and FeSO<sub>4</sub> 7H<sub>2</sub>O (8.5 g) in 200 mL of ultrapure 102 water, which was then heated to 90 °C with constant stirring in a 500 mL round-bottom 103 flask equipped with a reflux condenser. Then, 25% of ammonium hydroxide solution 104 (30 mL) was added into the solution rapidly and sequentially and stirred at 90 °C for 1 105 h. Finally, the solid products (Fe<sub>3</sub>O<sub>4</sub> NPs) were rinsed several times with ultrapure water 106 and were ready for use. 107 Particle size distribution and morphology of the two NPs were measured 108 transmission electron microscopy (TEM) using a field-emission transmission electron 109 microscope (Tecnai G2 F20, FEI). Results showed that the average dianeter 110 Fe<sub>2</sub>O<sub>3</sub> 111 NPs and Fe<sub>3</sub>O<sub>4</sub> NPs was 8.7 and 15.6 nm, respectively, and the sh globular (see 112 Fig. S1 in Supplementary material). 113 2.2. Composting raw materials preparation The components of raw materials were erring to our previous studies 114 115 (Zhang et al., 2017; Zhou et al., 2018 tray used as the typical agricultural waste ained from a suburb of Changsha, China. 116 which is difficult to be defrom a vegetable market and used to speed up the 117 Vegetable waste was collected 118 beginning of composingcomposting. Both rice straw and vegetable waste were air-dried 119 and cut into about 15 mm length. The soil, which was added to enrich microbial 120 populations and offer some necessary nutrients, was dug from Yuelu Mountain of Changsha, China, and sieved through 60-mesh screen to remove the coarse plant debris. 121 122 Bran, purchased from a farm of Henan province, China, was added to adjust the initial

- 123 carbon to nitrogen ratio (C/N) after air-dried. Characterizations of raw materials were
- 124 presented in Table S1 of Supplementary material.

125 2.3. Composting and sampling

60 days of lab-scale composting experiments with different treatments were conducted 126 127 in reactors with 65 L of capacity. Rice straw, soil, vegetable and bran were homogenized at a weight ratio of 30:27:8:5 to obtain an initial C/N ratio of about 30. The control 128 129 treated without any IONPs was marked as T-C, and treatments with Fe<sub>2</sub>O<sub>3</sub> NPs Fe<sub>3</sub>O<sub>4</sub> NPs at a concentration of 10 mg/kg compost were named as T- Fe<sub>2</sub>O<sub>3</sub> NF 130 T- Fe<sub>3</sub>O<sub>4</sub> NPs, respectively. Initial moisture content was adjusted to about 55%. To 131 ensure sufficient aeration, the composting piles were manually turned daily 132 in the 133 beginning 14 days, and on each sampling day afterwards. The ibiamples were collected and homogenized on day 0, 3, 5, 7, 17, 29, 43, 60. Th 134 e mixed samples were 135 then stored at 4 °C for further chemical physicochem and enzymatic analyses. 136 2.4. Physicochcemical Physicochemical det 137 The pile temperatures were determine ordi ary thermometer from three different 138 positions of the piles, and the erature was also recorded. Samples for pH nhie 139 determination were shaken in rapure water at a ratio of 1:10 (weight/volume, w/v) 140 and then the suspension was collected using ordinary filter paper. NH4+-N and NO3--N 141 were determined using flow injection analysis (AA3, Germany) after being extracted 142 by shaking the samples with 2 M KCl at a ratio of 1:50 (w/v) under 150 rpm for 1 h. The OM and ash content were determined using dry combustion in a muffle furnace at 143 550 °C for 6 h after the samples were firstly dried at 105 °C for 24 h and ground. The 144

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145 content of total organic carbon (TOC) and OM loss was were calculated on basis of ash content with reference to the following equations, respectively (Zhang et al., 2017; Qin 146 et al., 2018): 147 148 % TOC = (100 - % ash)/1.8-(1) The OM loss was computed according to the following equation: 149 150 %OM loss =  $100-100[X_1 (100-X_n)]/[X_n (100-X_1)]$ (2)151 Where  $X_1$  represented the initial ash content and  $X_n$  was the ash content on each sector. corresponding day. Water soluble carbon (WSC) was determined using Total Or 152 Carbon Analyzer (TOC-5000A, Shimadzu, Japan). Samples submerged in ultrapure 153 water at a ratio of 1:10 (weight/volume, w/v) were shaken at 200 rpm for 40 m 154 Then, 155 the supernatant was collected via filter filtering using ordinary piper and then centrifuged at 12,000 rpm for 10 min, and further cleared us 156 g 0.45 µm membrane filter. Total nitrogen (TN) was determined by elementa er (Elementar, Vario Max 157 analy CN, Germany). C/N equals to the ratio of T 158 rad h seed according to Wu et al. (2019) 159 The germination index (GI) was teste 160 with a little modification. compost was shaken with ultrapure water (1:10, w/v) for 1 h and the mixtu e was filtered to collect the aqueous extracts. Secondly, 161 162 Twenty twenty seeds were placed into 9 cm Petri dishes which were lined with 163 Whatman filter paper and contained 10 mL of extracts. Ultrapure water was used as 164 control. Then the above dishes Petri dishes were incubated at 25 °C in dark for 3 days. Finally, seed germination, root length, shoot length were determined, and relative seed 165 166 germination (SG), relative root elongation (RE) and germination index (GI) were

167 calculated according to Wu et al. (2019).

168 2.5. Enzyme activity determination

DHA was analyzed according to the transformation of 1,3,5-triphenyltetrazolium 169 170 chloride (TTC) to triphenylformazan (TPF) by method of Liu et al. (2014). Firstly, 2 g of fresh compost was mixed with 10 mL of 0.5% TTC solution. Then, the mixture was 171 172 incubated at 37 °C for 8 h in dark. The blank control was processed by replacing TTC with the same volume of ultrapure water. After incubation, the formed TPF 173 extracted by shaking the mixture with 100 mL of methanol at 300 rpm for 1 h 174 supernatant was cleared and collected through centrifugation. Finally, the concentration 175 of TPF was determined using spectrophotometry at 485 nm and th 176 resu were reported as mg TPF g<sup>-1</sup> dry compost (8 h)<sup>-1</sup>. 177 UA was calculated based on the NH4+-N produced after incuba 178 n of compost samples with urea solution as described by Guti érrez et al. **0**). mL of 40 mM aqueous 179 tronwas added with 10 mL of urea solution was added to 1 g of compost 180 ultrapure water. After incubation at 3 0 mL of ultrapure water (control: 10 181 6 h mL of 40 mM urea solution a KCl-HCl (1 M KCl; 0.01 M HCl) were 182 added to extract the produced  $H_4^+$ -N by shaking the mixture at 150 rpm for 40 min. 183 The suspension for NH4+-N determination by spectrophotometry at 490 nm was filtered 184 185 successively using filter paper and 0.45 µm membrane filter. The enzyme activity was expressed as mg NH4+-N g-1 dry compost (6 h)-1. 186

187 2.6. Statistical analysis

188 The first-order kinetic model was used to assess OM evolution and loss during

composting via fitting the data of OM content or OM loss as a function of composting 189

(3)

(4)

- 190 time which were expressed as follows (He et al., 2018c; Xiong et al., 2018):
- OM (%) =  $A \times e^{-kt}$ 191
- 192 OM loss (%) =  $A (1-e^{-kt})$

In which A was the initial OM content (%) for equation (3) or the maximum OM loss 193 (%) for equation (4), k was the rate constant (day<sup>-1</sup>) and t was the composting time in 194 days. The residual mean square (RMS) and F-values were used to verify 195 significance of the curve fitting. 196 197 Mean values with standard deviation (n = 3) were presented on basis of day weight (DW) for all determinations. The significance of difference between mean values of 198 ferent samples-which were collected on the same day from different tre was assessed 199 by one -way analysis of variance (ANOVA) using SPSS 200 <u>which was</u>. All the significance tests were based on a 95% confidence Iultivariate relationships 201 vel. between enzyme activities and physicocher araı 202 were tested with Canoco eter al., 2018b). Pearson correlations 203 4.5 as described in our previous ang ntal variables were evaluated using SPSS 204 between enzyme activities nm 19.0. 205 3. Results and discussion 206 207

- 3.1. Changes of physicochemical parameterstemperature
- 208 Temperature is an important parameter affecting the performance of organic solid waste aerobic composting and can reflect the biological activity in composting system (Ren 209 210 et al., 2018a). As shown in Fig. 1A, the maximum and minimum ambient temperature

in all the three treatments showed a rapid increase in the first 6 days, and the ma	ximum
213 value was greater than 65 °C in each composting treatment. This rapid increases	ease of
temperature might be attributed to the decomposition of easily degradable of	organic
215 matters by microorganisms which released a lot of heat. Though the ma	ximum
216 temperatures in all treatments were not significantly different, the durat	tion of
217 temperature above 55 °C in IONPs treatments was longer than that in the control	ol. With
the degradation of simple organic matters, the temperature in all treatments be	egarte
219 decrease until the end of composting process. Intriguingly, the temperature in T	C-C was
significant higher ( $P < 0.05$ ) than IONPs treatments during day 20 to 4 espe	ally T-
221 Fe <sub>2</sub> O <sub>3</sub> NPs, this might be caused by the more slowly degradation of wailab	ble OM
during earlier stage in T-C, which consequently maintained higher temperature	in T-C
223 during this phase.	
224 In addition, the pH of the three composting treat ents experienced the similar p	<u>pattern,</u>
225 <u>showing significant fluctuation in the sine 17 days due to the intensive active</u>	ities of
226 microorganisms during this same, and they it changed slowly (Fig. 1B). The fi	<u>inal pH</u>
227 of all treatments maintained is 8~8.2, indicating that the final compost r	reached
228 maturation (Zhang et al., 2018a). The higher pH in IONPs treatments during the	<u>e 10<sup>th</sup> to</u> 带格式
229 $43^{rd}_{2}$ day might be attributed to higher NH <sub>4</sub> <sup>+</sup> -N in the two treatments (Fig. 1C). As	shown 带格式
230 in Fig. 1C, the $NH_{4^+}$ -N contents in all treatments reached peak value on the $\frac{1}{2}$	5 <sup>th</sup> day, 带格式
231 which might be caused by the ammonification of organic N, and then decreased by	Decause 带格式
232 <u>of nitrification, <math>NH_3</math> emission, etc. At the end of composting, the <math>NH_4^+</math>-N in T</u>	F-Fe2O3 带格式 带格式

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233	NPs was significant nigher than that of 1-C, while the NO <sub>3</sub> -N in 1-Fe <sub>2</sub> O <sub>3</sub> NPs was
234	significant lower than T-C (Fig. 1D). The final mineral nitrogen in T-C, T-Fe <sub>2</sub> O <sub>3</sub> NPs
235	and T-Fe <sub>3</sub> O <sub>4</sub> NPs was 0.78, 0.92 and 0.78 g/kg DW compost, respectively, indicating

that Fe<sub>2</sub>O<sub>3</sub> NPs was more beneficial to improve the nitrogen utilization efficiency of

237	final	compost	produc
237	rinal	compost	produ

238 3.2. Changes of organic matter

Organic matter (OM) is an important indicator to evaluate the transformation a 239 mineralization of organic nutrients during composting process (Chan et al., 2016 240 et al., 2018b). As shown in Fig. 2A, OM content was constantly decreased in all 241 treatments, which was generally attributed to the utilization of OM and enission of CO<sub>2</sub> 242 by microorganisms (Awasthi et al., 2016b; Tang et al., 2018). Over eating, the OM 243 was decreased from 64.38, 63.64 and 61.95% to 37.93, 29.9 nd 38.65% in T-C, T-244 Fe<sub>2</sub>O<sub>3</sub> NPs and T-Fe<sub>3</sub>O<sub>4</sub> NPs, respectively. The OM t intensively mineralized 245 s mo re within the first 17 days in all treatments ulter in higher temperatures. 246 247 Afterwards, it was decreased more s the, bnsumption of easily degradable OM int to be degraded. The OM loss rates were 248 and the remaining compone as r 51.29, 52.82 and 52.29% within the first 17 days, respectively, accounting for 77.49, 249 69.93 and 85.29% of the total OM loss. The higher OM loss and lower proportion of 250 251 OM loss in the first 17 days to total OM loss in T-Fe<sub>2</sub>O<sub>3</sub> NPs indicated that more OM 252 was degraded not only in the first 17 days of composting but also afterwards, suggesting that the amendment of Fe<sub>2</sub>O<sub>3</sub> NPs was beneficial to the degradation of recalcitrant OM. 253 254 At the end of composting, the total OM loss in T-Fe<sub>2</sub>O<sub>3</sub> NPs was significant higher than

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other two treatments with values of 66.19, 75.53 and 61.31% in T-C, T-Fe<sub>2</sub>O<sub>3</sub> NPs and T-Fe<sub>3</sub>O<sub>4</sub> NPs (Fig. 2B), respectively, and this was confirmed by the ANOVA test (P < 0.001).

To assess the organic biological degradation, the kinetics of OM degradation and loss 258 were calculated. In all treatments, the OM evolution and loss profiles followed first-259 order kinetics along with the proceeding of composting. The parameters obtained from 260 261 curve fitting of experimental statistics data showed that all OM evolution and lo kinetics well fitted with the equations (P < 0.05) (Table 1). The rates of OM evol 262 and loss in T-Fe<sub>2</sub>O<sub>3</sub> NPs were more rapid than T-C and T-Fe<sub>3</sub>O<sub>4</sub> NPs, as shown by the 263 higher k value. Greater OM loss was also detected in T-Fe<sub>2</sub>O<sub>3</sub> NPs (Fig. (B). h 264 wever, Ben-Moshe et al. (2013) suggested that OM in soil was not chan, vit the addition 265 of nanoparticles, and the underlying explanation was that 266 given experimental condition (i.e., short exposure time) did not support e mineralization of OM. 267 omple The conditions of composting in present st ve of microbial activities 268 269 to degrade OM and thus caused diffe n th three treatments. The changes of TOC during cess were presented in Fig. 2C. Generally, 270 nno TOC contents decreased during the whole process of composting. More rapid loss of 271 272 TOC contents during the first 17 days might be attributable to the utilization of easily 273 degradable OM by microorganisms. Moreover, Awasthi et al (2016b) demonstrated that 274 during the early phase of composting, a part of organic carbon in the compost is transformed to H<sub>2</sub>O, CO<sub>2</sub> and energy, while the rest is continuously converted to more 275

stable humus like substances. As shown in Fig. 2C, the TOC content in T-Fe<sub>2</sub>O<sub>3</sub> NPs

277	was relatively lower than the other two treatments on the whole during composting. At
278	the end of composting, the TOC in T-Fe <sub>2</sub> O <sub>3</sub> NPs was significantly lower than T-C and
279	T-Fe <sub>3</sub> O <sub>4</sub> NPs which was verified by the ANOVA results ( $P < 0.001$ ). There were no
280	significant differences in the TOC content as well as OM between T-C and T-Fe <sub>3</sub> O <sub>4</sub> NPs
281	(P > 0.05) in the end, since the positive effect of iron in Fe <sub>3</sub> O <sub>4</sub> NPs as a nutrient might
282	be impaired by the release of Fe <sup>2+</sup> which could generate reactive oxygen species (ROS)
283	(Auffan et al., 2008). The final TOC contents were 21.1%, 16.7% and 21.5%, and the
284	TOC reduction compared with initial contents were observed with 41.1%, 52.9% and
285	37.6% in T-C, T-Fe <sub>2</sub> O <sub>3</sub> NPs and T-Fe <sub>3</sub> O <sub>4</sub> NPs, respectively.
286	WSC is always used as one of the most biologically active parameters o evaluate the
287	compost stability. It can be easily and directly utilized or degraded by mitroorganisms,
288	as it mainly consists of low-molecular weight organic acids, sug is, phenolic substances,
289	amino acids, hemicellulose and other easily biodegradable ompounds (Zhang et al.,
290	2018b; Ye et al., 2017b). In this study, the VSC oncentration in all treatments
291	increased rapidly within the early 7 ders (v. 21), because the production of WSC by
292	solubilization of some simple regard matters or by microbial activities and growth was
293	greater than their consumption and degradation by microorganisms (Zhang et al., 2017).
294	Afterwards, the WSC showed constant drop until the end of composting. The final WSC
295	in T-C was significantly higher than that in T-Fe <sub>2</sub> O <sub>3</sub> NPs and T-Fe <sub>3</sub> O <sub>4</sub> NPs ( $P < 0.01$ ),
296	indicating that the further degradation or utilization of WSC in T-C was not greater than
297	the other two treatments. As shown in Fig. 2A and 2D, both OM and WSC degradation
298	in T-Fe <sub>2</sub> O <sub>3</sub> NPs were greater than T-Fe <sub>3</sub> O <sub>4</sub> NPs, indicating that the amendment of Fe <sub>2</sub> O <sub>3</sub>

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299 NPs was more effective to improve the composting performance.

300 *3.3. Enzyme activities* 

301	Dehydrogenase is an intracellular enzyme that catalyzes metabolic reactions for OM	
302	degradation to provide ATP. It has been confirmed that DHA can reflect the rate of	
303	biochemical reaction during composting and has been generally regarded as a reliable	
304	indicator for overall microbial activity in the composting system because it directly	
305	participates in the respiratory chain (Li et al., 2015). The dynamics of DHA in the three	5
306	treatments were shown in Fig. 3A. It increased rapidly in all treatments during the trav	
307	stage of composting until the maximum value was achieved in thermosphilic phases,	
308	which was attributed to the oxidation of easily degradable OM catalyzed by the azyme	
309	(Li et al., 2015), and then decreased until the end of composting. Vir natiobservations	
310	were also reported by <u>a previous study</u> (Nikaeen et al., 2015) but found DHA reached	
311	the maximum in the initial stage and then decreased an ug will the composting process.	
312	Maximum DHA occurred on day 7 with values (9.1), 1184, 10.38 mg TPF g <sup>-1</sup> dry	
313	compost (8h) <sup>-1</sup> in T-C, T-Fe <sub>2</sub> O <sub>3</sub> and T-Fe <sub>3</sub> (NF), respectively. As shown in Fig. 3A,	
314	DHA was higher in T-Fe <sub>2</sub> O <sub>3</sub> and T-F $Q_4$ DPs than the control on the whole, especially	
315	in the treatment with addition of Fe <sub>2</sub> O <sub>3</sub> NPs during the $3^{rd}_{rd}$ to $17^{th}_{rd}$ days ( $P < 0.05$ ),	带格 带格
316	indicating that the addition of IONPs; especially Fe <sub>2</sub> O <sub>3</sub> NPs facilitated DHA and rates	
317	of biochemical reactions, and this corresponded to the greater degradation of OM and	
318	WSC in T-Fe <sub>2</sub> O <sub>3</sub> NPs as-which were shown in Fig. 2A and 2D.	
319	Urease, as an intracellular enzyme, can catalyze the transformation of amides into	
320	ammonium. Its activity has been considered as an enzymatic indicator of N	

**带格式的:**上标 **带格式的:**上标 321 mineralization (Wong et al., 2000). Similar to DHA, the UA in all treatments increased rapidly in the first 7 days, and then decreased until the end of composting (Fig. 3B), 322 which was also observed in other previous study (Li et al., 2015). The production and 323 accumulation of substrates related to nitrogen by other enzymatic activities might be 324 the possible reason for the increase of UA, while the decrease of UA might be caused 325 by the depletion of readily biodegradable OM which supports microbial activity 326 (Sudkolai et al., 2017; Ren et al., 2018c). Compared with T-C, the treatments v 327 IONPs enhanced the average UA in order of T-Fe<sub>2</sub>O<sub>3</sub> NPs > T-Fe<sub>3</sub>O<sub>4</sub> NPs, as w 328 DHA. Similar results were reported by He et al. (2011) who found that IONPs had 329 NPs positive effects on the soil UA and DHA, and they also found that Fe<sub>2</sub>O 330 dition had greater impacts on the enzyme activities. 331 332 It has been suggested that iron-based NPs are toxic because the reactive oxygen species (ROS) (Auffan et al., 2008). Therefore, the p ffect of Fe<sub>3</sub>O<sub>4</sub> NPs as an 333 itive iron nutrient may be counteracted by th , resulting in a weaker 334 Ps consist of fully oxidized crystals 335 improvement of microbial activity, y 0 336 which are very stable in en showing a lower capacity to generate ROS mei nt of microbial activity (He et al., 2011). Overall, the 337 and inducing greater improven amendment of IONPs, especially Fe2O3 NPs, could enhance the enzyme activities and 338 promote the OM degradation in this study. 339 3.4. Relationships between physicochemical parameters and enzyme activities 340

According to the results of detrended correspondence analysis (DCA), the length of the

342 first axis in all treatments was less than 3, which indicated linear relationships between

enzyme activities and environmental variables during composting. Therefore, RDA was 343 performed to evaluate the multivariate relationship between physico-chemical 344 parameters and enzyme activities. Totally, eight environmental variables were selected 345 in RDA model, including temperature, pH, C/N, TOC, WSC, NH4<sup>+</sup>-N, NO<sub>3</sub><sup>-</sup>-N and TN. 346 347 In addition, Pearson correlations were conducted to support these analyses. To identify the driving factors of enzyme activities, manual forward selection was performed. 348 Accordingly, temperature was the most influential factor (P < 0.01) in all treatment 349 and it was the only significant factor in T-C. Another driving factor in T-Fe<sub>2</sub>O<sub>3</sub> N 350 pH (P < 0.05) and that in T-Fe<sub>3</sub>O<sub>4</sub> NPs was WSC (P < 0.05). Partial RD showed that 351 temperature solely explained 75.1% (P = 0.002), 34.7% (P = 0.016) a d 38. 352 (P =353 0.044) of the variation in enzyme activities in T-C, T-Fe<sub>2</sub>O<sub>3</sub> N -Fe<sub>3</sub>O<sub>4</sub> NPs, 354 respectively (Table 2). The importance of temperature fluctuat ns for the evolution of enzyme activities has been highlighted by a previo (Wang et al., 2011). In 355 stud agreement, it was also found that both temp DFA activity showed a rapid 356 ar increase during the early days of con sthi et al., 2018), indicating impacts 357 on enzyme activities by ter 358 ionally, as an important parameter during atur composting, pH significantly a counted for 11% (P = 0.05) of the variation in enzyme 359 360 activities in T-Fe<sub>2</sub>O<sub>3</sub> NPs<del>, and the The shared contribution of temperature and pH was</del> 361 40.1%, indicating that the strong inter-correlation of the two factors also contributed to 362 the changes of enzyme activities. These were also supported by the Pearson correlation between environmental variables in which temperature and pH were closely correlated 363 364 with each other (P < 0.05) (see Table S3 of Supplementary materials). For T-Fe<sub>3</sub>O<sub>4</sub>, 365 WSC, another significant explanatory variable tested by manual forward selection, solely explained 4.1% of the variation in enzyme activities, which was not significant 366 (P = 0.512), while the total explanation of temperature and WSC was 76.3% (P = 0.01)367 (Table 2). This suggested that WSC contributed to the changes of enzyme activities 368 mainly via cooperating with temperature (see Table S3 of Supplementary materials). 369 However, these results did not imply that other environmental variables were not 370 371 influential to the changes of enzyme activities, it was only deduced that the parameters contributed to the variations of enzyme activities mainly through the 372 correlation with other significant factors, as proved by Pearson correlation analysis 373 between different variables (see Table S2-S4 of Supplementary materia 374 To further evaluate the relationships between enzyme activitie vironmental 375 et al., (2018), t<u>T</u>he 376 variables, ordination triplots were created. As introduced in 2 length and orientation of arrows represent the 377 aport nce of parameters and associations between axis and parameters. Moreover, the angles between 378 /el two parameters < 90° reflect positiv while that  $> 90^{\circ}$  reflect negative 379 onsb 380 relationships. As shown in e activities were negatively correlated with th pH and TN, and shared positi relationship with other parameters in all treatments 381 except weak negative correlations between NO3-N and UA in T-C. Longer arrow line 382 of temperature, NH4+-N and TN indicated that these parameters played more important 383 roles in explaining variations of enzyme activities in T-C (Fig. 4A). According to 384 Pearson correlation (Table 3), both DHA and UA were positively correlated with 385 386 temperature in all treatments (P < 0.01). This finding was in accordance with a previous

387 study which found that DHA-was increased in the thermophilic stage and then rapidly decreased when the temperature dropped (Wang et al., 2011). Moreover, there was a 388 significant (P < 0.05) positive relationship between UA and NH<sub>4</sub><sup>+</sup>-N, and negative 389 correlation with TN in T-C (Table 3). From Fig. 4B, temperature, pH, WSC and NH4+-390 391 N played relatively more important roles in variations of enzyme activities than other parameters, as supported by Table 3 showing an extremely significant negative 392 correlation (P < 0.01) between UA and pH, extremely significant positive correlation 393 between the enzyme activities and temperature (P < 0.01), and significant point 394 correlation between WSC, NH<sub>4</sub><sup>+</sup>-N and enzyme activities (P < 0.05) in Te<sub>2</sub>O<sub>3</sub> NPs. A 395 Ы () previous study also reported that the UA was negatively influenced by 396 et al., 2017). pH not only has an influence on the decomposition and min 397 at on of organic 398 macromolecules, species and activities of microorganisms, but so directly affects the rate of enzymes involved in biochemical processes ( 2000; Lai et al., 2016). k et 399 As for T-Fe<sub>3</sub>O<sub>4</sub> NPs which was shown in F ngth of temperature, pH, C/N, 400 ch DHA was significantly correlated 401 TOC, WSC, NH4+-N and TN were la 402 with temperature, C/N, W. d UA shared significant correlation with nd  $NH_4^+-N$  (P < 0.05) (Table 3). In T-Fe<sub>2</sub>O<sub>3</sub> NPs and Ttemperature, pH, C/N, TOC an 403 Fe<sub>3</sub>O<sub>4</sub> NPs, WSC played an important role as well, similar with a previous study 404 (Calvarro et al., 2014). As implied by Pearson correlation, UA was more closely related 405 to environmental factors than DHA in all treatments. The different relationships 406 between enzyme activities and environmental variables in three treatments suggested 407 408 that the amendment of IONPs changed their correlations.

# *3.5. Maturity of the final compost product*

410	Generally, C/N ratio is one of the most commonly used indicators to evaluate the rate
411	of composting as well as final compost maturity (Awasthi et al., 2016a). Another
412	important parameter is seed germination index (GI) which is widely used to evaluate
413	the phytotoxicity and maturity of compost, since it directly reflects the effect of compost
414	on the seed germination and seedling growth (Chan et al., 2016). The acceptable final
415	value of C/N for a successful composting is lower than 25 (Chan et al., 2016), and that
416	of GI is above 80% (Wu et al., 2019). As shown in Fig. 5, both C/N and GI me the
417	above requirements, indicating that the end product can be recycled for agricultural
418	application (Chan et al., 2016; Wu et al., 2019). The GI in IONPs treatments, execually
419	that in T-Fe <sub>2</sub> O <sub>3</sub> NPs was significant higher than T-C ( $P < 0.05$ ), a done overage shoot
420	length in T-C, T-Fe <sub>2</sub> O <sub>3</sub> NPs and T-Fe <sub>3</sub> O <sub>4</sub> NPs was 3.16, 3.87(3.45 cm, respectively.
421	This suggested that amendment of IONPs into composing could promote seed
422	germination and seedling growth, a similar finding was reported by Ren et al. (2011)
423	who found that Fe <sub>2</sub> O <sub>3</sub> NPs could stimulate the growth of mung bean ( <i>Vigna radiata</i> ).
424	4. Conclusion
425	In present study, Fe <sub>2</sub> O <sub>3</sub> NPs and Fe <sub>3</sub> O <sub>4</sub> NPs were added at a concentration of 10 mg/kg
426	compost to investigate the effects of IONPs on composting performance. With the
427	addition of IONPs, especially Fe <sub>2</sub> O <sub>3</sub> NPs, the following aspects were changed: (i) the
428	OM degradation was promoted. (ii) DHA and UA were enhanced and the relationships
429	between enzyme activities and environmental variables were changed. (iii) The seed

germination and seedling growth were improved. Therefore, composting amended with

- 431 IONPs, especially Fe<sub>2</sub>O<sub>3</sub> NPs, is a promising approach to improve the performance of
- 432 composting process and quality of final compost product.

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437 Appendix A. Supplementary data

- 438 The E-Supplementary data of this work can be found on online.
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624	▼	

### 625 Figure captions:

- 626 **Figure 1.** Changes of (A) temperature; (B) pH; (C) NH<sub>4</sub>+-N and (D) NO<sub>3</sub>--Ntemperature
- 627 during composting. Mean values were shown and the error bars represented the 628 standard deviations (n = 3).
- 629 Figure 2. Temporal courses of (A) organic matter (OM) degradation; (B) OM loss; (C)
- 630 TOC, total organic carbon; (D) WSC, water soluble carbon during composting process.
- 631 T-C: treatment without Fe<sub>2</sub>O<sub>3</sub> NPs or Fe<sub>3</sub>O<sub>4</sub> NPs as control; T- Fe<sub>2</sub>O<sub>3</sub> NPs: treatme
- with  $Fe_2O_3$  NPs; T-  $Fe_3O_4$  NPs: treatment with  $Fe_3O_4$  NPs. Data were calculated by
- basis of dry weight compost. Mean values were shown and the error bas represented

634 the standard deviations (n = 3).

Figure 3. Evolutions of enzyme activities (A) dehydrogenase (DHA); (B) 635 urease activity (UA) during composting process. T-C: treatment 636 without Fe<sub>2</sub>O<sub>3</sub> NPs or D<sub>3</sub> NPs; T- Fe<sub>3</sub>O<sub>4</sub> NPs: Fe<sub>3</sub>O<sub>4</sub> NPs as control; T- Fe<sub>2</sub>O<sub>3</sub> NPs: treatment 637 h Fe treatment with Fe<sub>3</sub>O<sub>4</sub> NPs. Data were cal of dry weight compost. 638 on bas. ented the standard deviations (n = 3). 639 Mean values were shown and the err epre me activities and environmental variables. 640 Figure 4. RDA triplot analy etw (A) T-C: treatment without Fe  $O_3$  NPs or Fe<sub>3</sub>O<sub>4</sub> NPs as control; (B) T- Fe<sub>2</sub>O<sub>3</sub> NPs: 641 treatment with Fe<sub>2</sub>O<sub>3</sub> NPs; (C) T- Fe<sub>3</sub>O<sub>4</sub> NPs: treatment with Fe<sub>3</sub>O<sub>4</sub> NPs. Significant 642 643 variables were denoted using red solid lines, and supplementary variables were denoted with red dotted lines. Enzyme activities were presented using black solid lines. The 644 circles indicated samples and the numbers around were the sampling day. 645

**Figure 5.** Maturity indices of the final compost product. GI: germination index; C/N:

- 647 total organic carbon/total nitrogen. T-C: treatment without Fe<sub>2</sub>O<sub>3</sub> NPs or Fe<sub>3</sub>O<sub>4</sub> NPs as
- 648 control; T- Fe<sub>2</sub>O<sub>3</sub> NPs: treatment with Fe<sub>2</sub>O<sub>3</sub> NPs; T- Fe<sub>3</sub>O<sub>4</sub> NPs: treatment with Fe<sub>3</sub>O<sub>4</sub>
- 649 NPs. Mean values were shown and the error bars represented the standard deviations (n
- 650 = 3)**.**
- 651 Table legends:
- 652 Table 1. Parameter values of the first-order equation kinetics obtained from curve
- 653 fitting of OM degradation and OM loss.
- 654 Table 2. Partial RDA results indicating the influence of the significant variable
- 655 enzyme activities.
- Table 3. Pearson correlations between enzyme activities and environmental variables.

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658 Table 1. Parameter values of the first-order equation kinetics obtained from curve

Treatment	A (%)	k	RMS	F		
OM degradatio	on					
T-C	58.69 (2.01)	0.0095 (0.0016)	13.76	42.38		
T-Fe <sub>2</sub> O <sub>3</sub> NPs	58.39 (2.43)	0.0123 (0.0021)	19.32	43.47		
T-Fe <sub>3</sub> O <sub>4</sub> NPs	58.40 (2.08)	0.0085 (0.0016)	14.94	32.24		
OM loss						
T-C	63.82 (2.20)	0.1335 (0.0157)	13.85	265.44		
T-Fe <sub>2</sub> O <sub>3</sub> NPs	68.51 (4.53)	0.1379 (0.0286)	57.04	82.33		
T-Fe <sub>3</sub> O <sub>4</sub> NPs	59.87 (2.27)	0.0906 (0.0108)	10.30	868.43		
A: the initial or	ganic matter (OM)	) content (%) for Q	M degn Mei	on kinetic or the		
maximum OM l	oss (%) for OM loss	s kinetic. <i>k</i> : rate con	tant (d <sup>-1</sup> ). RM	MS: residual mean		
square. F: factor	significant at $P < 0$	.05. The value in bea	ckets: standa	ard deviation. T-C:		
treatment without	ut Fe <sub>2</sub> O <sub>3</sub> NPs or Fe	04 NFX ac control;	T- Fe <sub>2</sub> O <sub>3</sub> NI	Ps: treatment with		
Fe <sub>2</sub> O <sub>3</sub> NPs; T- F	e <sub>3</sub> O <sub>4</sub> NPs: treatmen	t with fe <sub>3</sub> O <sub>4</sub> NPs.				

659 fitting of OM degradation and OM loss

## 666 Table 2. Partial RDA results indicating the influence of the significant variables on

Treatments	Variables	Eigenvelues	Sole explanation	F	Р	
Treatments	tested in model	Eigenvalues	(%)	value	value	
T-C	Temperature	0.751	75.1	18.087	0.002	
	Temperature	0.347	34.7	12.242	0.016	
T-Fe <sub>2</sub> O <sub>3</sub> NPs	pH	0.110	11.0	3.896	0.050	
	Together	0.858	85.8	15.150	0.0	
	Temperature	0.384	38.4	8.0, 0	0.044	•
T-Fe <sub>3</sub> O <sub>4</sub> NPs	WSC	0.041	4.1	0 65	512	
	Together	0.763	768	8.0 6	0.010	

667 enzyme activities.

668 WSC: water soluble carbon; T-C: treatment without Fe 5, VPs or 1, 304 NPs as control;

669 T- Fe<sub>2</sub>O<sub>3</sub> NPs: treatment with Fe<sub>2</sub>O<sub>3</sub> NPs; T- Fe<sub>2</sub>O<sub>4</sub> NPs. Comment with Fe<sub>3</sub>O<sub>4</sub> NPs.

Treatments	Enzyme activities	Temperature	рН	C/N	TOC	WSC	NH4 <sup>+</sup> -N	NO <sub>3</sub> N	TN
TC	DHA	0.992**	-0.519	0.557	0.540	0.520	0.583	0.189	-0.550
I-C	UA	0.838**	-0.677	0.732*	0.669	0.337	$0.726^{*}$	-0.126	-0.707*
	DHA	0.953**	-0.591	0.384	0.409	0.827*	0.542	0.420	-0.481
1-Fe <sub>2</sub> O <sub>3</sub> NPS	UA	0.895**	-0.949**	0.498	0.568	0.730*	0.820*	0.253	-0.508
TE O ND	DHA	0.920**	-0.542	0.712*	0.557	0.809*	0.578	0.210	0.860*
I-Fe <sub>3</sub> O <sub>4</sub> NPs	UA	0.878**	-0.905**	0.771*	0.747*	0.571	0.807*	0.117	0.700

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 Table 3. Pearson correlations between enzyme activities and environmental variables.

C/N: total organic carbon/total nitrogen; TOC: total organic carbon 671 water

soluble carbon; TN: total nitrogen. T-C: treatment without Fe<sub>2</sub>O<sub>3</sub> 672 NPs as

control; T- Fe<sub>2</sub>O<sub>3</sub> NPs: treatment with Fe<sub>2</sub>O<sub>3</sub> NPs; T- Fe<sub>3</sub>O<sub>4</sub>, Ps: treatment with Fe<sub>3</sub>O<sub>4</sub> 673

NPs. \* Correlation is significant at 0.05 level (2-taile). 674 prrelation is significant at *k*ce,

0.01 level (2-tailed). 675





- calculated on a basis of dry weight omport. Mean values were shown and the error
- bars represented the standard levitions (n = 3).



activity (UA) during composting process. T-C: tratment without  $Fe_2O_3$  NPs or  $Fe_3O_4$ 

692 NPs as control; T-  $Fe_2O_3$  NPs: treatment with Fe  $O_3$  NPs; T-  $Fe_3O_4$  NPs: treatment

with Fe<sub>3</sub>O<sub>4</sub> NPs. Data were signated on 2 basis of dry weight compost. Mean values

694 were shown and the error bars is presented the standard deviations (n = 3).



**Fig. 4.** RDA triplot analysis between enzyme activities and environmental variables.

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700 (A) T-C: treatment without  $Fe_2O_3$  NPs or  $Fe_3O_4$  NPs as control; (B) T-  $Fe_2O_3$  NPs:

rot treatment with Fe<sub>2</sub>O<sub>3</sub> NPs; (C) T- Fe<sub>3</sub>O<sub>4</sub> NPs: treatment with Fe<sub>3</sub>O<sub>4</sub> NPs. Significant

variables were denoted using red solid lines, and supplementary variables were

703 denoted with red dotted lines. Enzyme activities were presented using black solid

lines. The circles indicated samples and the numbers around were the sampling day.39



705

706 Fig.5. Maturity indices of the final compost product. GI: germination index; C/N: total

707 organic carbon/total nitrogen. T-C: treatment without Fe<sub>2</sub>O<sub>3</sub> NPs or Fe<sub>3</sub>O<sub>4</sub> NPs a

control; T- Fe<sub>2</sub>O<sub>3</sub> NPs: treatment with Fe<sub>2</sub>O<sub>3</sub> NPs; T- Fe<sub>3</sub>O<sub>4</sub> NPs: treatment with Fe<sub>3</sub>O<sub>4</sub>

NPs. Mean values were shown and the error bars represented the standard  $\frac{1}{2}$  viations (n

710 = 3).

