1 **Fertilizer potential of liquid product from hydrothermal treatment of**

2 **swine manure**

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Abstract

Compared with composting, hydrothermal treatment (HTT) technology can dramatically shorten the duration for manure waste treatment. This study firstly investigated the effect of HTT on solubilization of N, P and organics from swine manure, and then evaluated the phytotoxicity of liquid product from hydrothermally treated manure by seed germination test. Results show that 98% of N in manure could be converted into soluble form after HTT at 200°C for 60 min. Soluble P in hydrothermally treated manure (at 150°C for 60 min) was 2.7 times that in raw manure. The germination indices (GI) were all greater than 100% when the liquid product (from HTT at 150°C for 60 min) or its diluted samples being used. Results from this study suggest that HTT could be a promising technology for producing safe and value-added liquid fertilizers from swine manure.

Keywords: Hydrothermal treatment; Liquid fertilizer; Swine manure

1. Introduction

Nutrients are essential for plant growth. To insure enough food production from the limited farmland on Earth for the increasing global population, fertilizers play an important role in the increase of plant yield and remediation of soil depletion resulted from intensive farming activities. However, long-term application of chemical fertilizers shows negative effect on soil's N and C storage capacities and agronomic efficiency (Khan et al., 2007; Mulvaney et al., 2009). In addition, the production of synthetic fertilizer requires a large amount of gases, energy (Haber-Bosch process for N) and natural mineral resources (like phosphate rock) (Teenstra et al., 2014).

Nutrients recovery from organic wastes can become a prospective option if the process's environmental friendliness and sustainability could be guaranteed. As a result of the increasing global population, one important sector of food production, animal husbandry has also been rapidly developed, generating far more manure than the amount needed by local farmers for land application (He et al., 2016). Composting is a typical method widely used to convert manure waste to fertilizer that can be safely used, since the process can stabilize organics, sterilize pathogens and weed seeds in raw manure (Eghball and Lesoing, 2000). However, in terms of manure waste treatment and resources recovery, composting might be less efficient, since after long time processing, the major beneficial and finished product is compost only. During the long period of composting process, around half of the C and N sources would be lost and emitted as harmful and greenhouse gases (GHGs) (Hao et al., 2004), demonstrating its low environmental friendliness and sustainability.

In order to reduce nutrients loss and develop a highly efficient process to convert

manure waste into safe fertilizers, hydrothermal treatment (HTT) technology was

attempted in this study, since the high temperature used (above 100°C) can release high

content of nutrients and sterilize pathogens (Barber, 2016). The solid residue is expected

to produce biogas via dry anaerobic digestion (AD) process. Most recently, Huang et al.

(2017) pointed out the possibility of using the water extract from hydrothermally treated

manure as liquid fertilizer, and the solid fraction can achieve 51% increase in CH_4 yield

by dry AD. In addition, the resultant dry AD residue can be further used as solid

fertilizer. HTT as a treatment method has been tried on sewage sludge for enhanced

sludge dewaterability and improved subsequent AD (Li et al., 2017). In recent years,

increasing attention has been paid to recovery of valuable compounds from solid wastes

by using HTT (Suárez-Iglesias et al., 2017; Aida et al., 2017). For instance, when

microalgae were hydrothermally treated at 175-350°C for 10-90 min, 38-100% of

nitrogen (N) and 57-99% of phosphorus (P) could be recovered in the water-soluble

fraction (Aida et al., 2017). They also found that when HTT was performed at lower

temperature than 200°C, organic N was the predominant N form in the water-soluble

fraction. The high nutrients recovery rate with high organic N content reflects the great

potential of HTT technology for nutrients recovery and liquid fertilizer production from

manure waste. Idowu et al. (2017) also mentioned the fertilizer potential of process

water from food waste after hydrothermal process. Restated, although previous research

works have pointed out the fertilizer potential of liquid product from sludge, chicken

feathers and empty fruit bunch by using HTT (Gollakota et al., 2018; Sun et al., 2014;

Nurdiawati et al., 2015; Nurdiawati et al., 2018), the said liquid fertilizer potential has

rarely been explored. In addition, limited information could be found on the feasibility

of liquid fertilizer production from manure waste by using HTT, let alone the nutrients

variation of treated manure at lower temperature than 200°C.

This study aimed to investigate the fertilizer potential of liquid product from HTT of

swine manure. The effects of treatment temperature (110-200°C) and retention time (0-

60 min) on soluble N and P, soluble organics (carbohydrate, protein and volatile fatty

acids (VFAs)) were clarified. The properties of liquid product were further

demonstrated by seed germination test and its pH, electric conductivity (EC), and metal

ions.

2. Material and methods

2.1. Swine manure

Swine manure used in this study was sampled from a local farm in Tsukuba, Ibaraki,

Japan. The pigs were raised in traditional pig houses with cement floor. The manure was

collected as solid state and stored at 4°C before use. The main characteristics of swine

manure were as follows: total solids (TS) 26.9±0.53% of fresh weight, volatile solids

(VS) 20.1±0.42% of fresh weight, total nitrogen (TN) 27.7±1.02 g N/kg-VS, soluble N

11.1±0.33 g N/kg-VS, total phosphorus (TP) 25.8±0.50 g P/kg-TS, and soluble P

1.7±0.08 g P/kg-TS.

2.2. Hydrothermal treatment

Hydrothermal treatment (HTT) trials were conducted in an enclosed stainless reactor

equipped with a propeller stirrer (OM Lab-tech MMJ-200, Japan). The reactor has a

working volume of 200 ml and maximum temperature of 300°C. These trials, each in

triplicate, were performed at different temperature (110, 150, 180, or 200°C) for

different retention time (0, 10, 30, or 60 min) with agitation (60 rpm). For each trial, 80

5

gram of raw swine manure was loaded into the reactor. After maintaining at different

temperature for a designated retention time, the reactor was cooled to room temperature.

Then, the treated swine manure was collected for further analysis. The schematic of

experimental setup is illustrated in Fig. 1.

Soluble organic carbon (SOC), proteins, carbohydrates, volatile fatty acids (VFAs),

ammonia nitrogen, orthophosphate, total solids (TS) and volatile solids (VS) contents,

and pH of each sample were determined according to the procedures and methods

described elsewhere (Yuan et al., 2017). Soluble N was detected with alkaline potassium

persulfate digestion and UV spectrophotometric method. Soluble P was determined

according to the phosphomolybdenum blue method after potassium persulfate digestion

(APHA, 2012). Metal ions were analyzed by ICP-OES (Perkin Elmer Optima 7300DV)

after the sample being completely digested with the mixture of HNO_3 and H_2O_2 (1:1,

v/v) at 98°C.

2.3. Seed germination test

Seed germination was conducted to evaluate the phytotoxicity of swine manure after

HTT. The seeds of Komatsuna were chosen for this germination test. The filtrates of

treated manure after HTT at 150°C for 60 min (150-60), 180°C for 30 min (180-30),

200°C for 60 min (200-60) were diluted correspondingly to a SOC concentration of

500, 1000, or 2000 mg/L. For each test, 1.5 ml filtrate or distilled water was added on

the filter paper in a petri dish where 15 seeds were placed. Three petri dishes were

prepared for each condition. Then, the germination test was conducted at 30°C and 60%

of humidity. After 72 hours' incubation, the number of viable seeds and the root length

in each test were measured and recorded. Besides, the electric conductivity (EC) and pH

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of each filtrate used for germination test were measured. Germination index (GI) was

calculated using the following equation (Tiquia and Tam, 1998).

$$\text{Germination index (\%)} = \frac{n_s \times \text{Root length in filtrate}}{n_c \times \text{Root length in control}} \times 100$$

where n_s is the number of germinated seed in the tested filtrate sample, and n_c is the

number of germinated seed in the control (distilled water).

2.4. Statistical analysis

The analysis of variance (ANOVA) was applied to analyze the statistical difference of

N, P and organics solubilization at different HTT conditions (temperature and retention

time). Pearson's correlation coefficients were calculated to show the relationship

between GI and chemical properties of liquid products at different SOC levels. All the

statistical analyses were conducted by using IBM SPSS Statistics 20.0.

3. Results and discussion

3.1. Effect of HTT on solubilization

3.1.2. Organics solubilization

The soluble organic carbon (SOC) concentration and resultant pH in swine manure

after HTT are shown in Fig. 2. As seen, the highest SOC concentration achieved after

HTT at 180°C for 30 min was 4.1 times that in the raw swine manure, in agreement

with the statement by Barber et al. (2016) who claimed that the optimal temperature and

duration for thermal hydrolysis were around 160-180°C and 20-40 min, respectively.

When HTT was conducted at higher temperatures (180°C or 200°C), the SOC

concentration in the treated manure began to decrease when the treatment lasted for

longer than 30 min or 10 min, respectively.

141   The concentrations of VFAs, soluble carbohydrates and proteins varied with HTT

142   conditions (treatment temperature and retention time). VFAs concentration in the treated

143   manure did not show much change after HTT at 110°C and 150°C. When HTT

144   temperature increased to 180°C or 200°C, the VFAs concentration increased with the

145   prolongation of retention time, and it was about 2 times that in the raw swine manure

146   after treatment for 60 min. As for soluble carbohydrates, its concentration decreased

147   obviously at higher temperatures (180°C and 200°C) for a longer retention time. The

148   carbohydrates may be hydrolyzed into monosaccharide at lower temperature and further

149   generate larger molecular compounds at higher temperature for a longer retention time

150   (Gai et al., 2015). For instance, Maillard reaction might occur between reducing sugar

151   and amino acids in the temperature range of 140-165°C (Barber et al., 2016). The

152   highest soluble carbohydrate was 13.7 g C/kg-VS (about 4.4 times that in the raw swine

153   manure) after HTT at 150°C for 60 min. Soluble proteins occupied a large portion of

154   SOC in the treated manure, about 15.3 times that in the raw swine manure after HTT at

155   180°C for 30 min. However, under the highest temperature (200°C) tested in this study,

156   a longer retention time exhibited decrease effect on soluble protein concentration,

157   possibly attributable to its decomposition into peptide or amino acids under this

158   condition. Except for VFAs, carbohydrates and proteins, large amount of other soluble

159   organics were also dissolved in the liquid phase. The pH generally decreased along with

160   the HTT duration and the increase in temperature. However, the increase of VFAs

161   production was milder compared to the drastic decrease of pH. This might be due to

162   more efficient decomposition or hydrolysis of organics and production of other acidic

163   compounds under higher HTT temperature conditions. As Gollakota et al. (2016)

pointed out, pyrolysis liquids generally have an acidic character.

3.1.2. N and P solubilization

Fig. 3a illustrates the ammonia N and organic N in the soluble fraction of swine manure after HTT at different temperature for a designated retention time. The highest soluble N concentration was achieved at 200°C for 60 min. 98% of TN in the manure was dissolved and transferred to the soluble fraction (Fig. 3b). It was found that the soluble N in swine manure increased with the increase in HTT temperature, and became relatively stable when the temperature > 180°C. Seen from Fig. 3b, the N dissolution ratio gradually increased with the retention time, from 71.3% (0 min) to 85.0% (30 min), then slightly decreased (80.9%) when the retention time was further prolonged to 60 min at 180°C. Aida et al. (2017) observed a similar phenomenon that N yield in the water-soluble fraction decreased when HTT at 200°C lasted longer than 30 min. In addition, ammonia N concentration showed almost a similar increase trend with soluble N when HTT was conducted at temperature lower than 180°C. However, ammonia N was noticed to significantly decrease with the prolongation of retention time at 200°C. The decreased ammonia N might replace the hydroxyl groups in the long-chain fatty acids to form aliphatic amine compounds or react with propanoic acid to produce propionic amide at 200°C for a longer retention time (Gai et al., 2015). The organic N in liquid phase may also further repolymerize and transfer to oil phase or other insoluble form at higher HTT temperature (> 200 °C), which favors the production of bio-oil (Zheng et al., 2015). Although many researchers studied the possible pathways during HTT under different temperatures (Gai et al., 2015; Gollakota et al., 2018), more detailed investigations are still necessary since much more complex reactions are

187 involved in HTT due to the different components in different substrates.

188   The solubilization of P in swine manure after HTT was also investigated (Fig. S1).

189 The multi-factor analysis of variance (ANOVA) showed that the soluble P after HTT at

190 150°C was significantly higher than those conducted at other temperatures, while the

191 retention time did not have significant effect on P dissolution ratio. The soluble P

192 increased by 2.8 times after HTT at 150°C for 60 min or 200°C for 0 min, achieving the

193 highest P dissolution ratio of around 13% (Fig. S1), which agrees with the finding by

194 Ekpo et al. (2016) who obtained water extractable < 15% of P from swine manure after

195 being treated at 170°C for 1 h. HTT at higher temperatures (180°C or 200°C) resulted in

196 decreased P dissolution ratio. Similar results could be found in previous researches

197 (Aida et al., 2017; Ekpo et al., 2016), possibly due to the fact that precipitation of

198 phosphate with multivalent metal ions became more easily at higher temperatures. The

199 results of metal ions analysis by using ICP-OES in this study indicated that calcium

200 concentration in the liquid decreased with the increase in temperature and prolongation

201 of retention time, while increase in magnesium concentration was detectable in the

202 liquid (Table 1). Thus, the formation of $Ca_3(PO_4)_2$ might be the major reason for the

203 decreased soluble P (Aida et al., 2017). However, higher P dissolution was also found at

204 200°C for 0 min of retention time, probably due to a shorter retention time at higher

205 temperature. Depolymerization is the dominant reaction during the initial stage of HTT,

206 while repolymerization is usually active at later stage (Zheng et al., 2015). This

207 observation is also in agreement with Aida et al. (2017) who obtained a higher P

208 concentration in the water-soluble fraction after HTT at 250°C and 350°C for 10 min.

209 **3.2. Seed germination test**

In order to evaluate the phytotoxicity of the liquid product from hydrothermally

treated swine manure, the filtrates of swine manure after HTT at 150°C for 60 min

(L150-60), 180°C for 30 min (L180-30) and 200°C for 60 min (L200-60) were chosen

to conduct seed germination test, due to their higher contents of soluble N, P, and

organics. To make the results more comparable, the filtrates were diluted according to

their SOC concentrations. The results of seed germination test are presented as

germination index (GI). According to the Compost Maturity Index (CCQC, 2001), a GI

higher than 90% indicates the phytotoxic-free property of the tested substrate which can

be safely applied for soil. In this study, the liquid products from HTT at lower

temperatures showed better behavior in the seed germination test when comparing their

GI values at a same SOC level. The GI values by using either diluted or undiluted L150-

60 were all greater than 100% (Fig. 4), implying that it can be safely used for plant

growth. Besides, the pH and EC of the filtrates (Table 1) were within the threshold

values, which are 6.0-8.5 for pH and 4 mS/cm for EC, respectively (Crohn, 2016;

Nurdiawati et al., 2015), again reflecting that the filtrates can be safely used for soil

application and plant growth.

3.3. Fertilizer potential of liquid product from hydrothermally treated swine manure

Except for macronutrients (N, P and K), small amounts of micronutrients are also

required by plants for their growth. However, a higher concentration of micronutrients

would inhibit plant growth. In this study, HTT promoted the solubilization of N, P, K

and Mg, while Al and Cu in the liquid products were found to decrease when compared

with those in raw manure (Table 1). Although the concentrations of Fe and Zn increased

233   to some extent after HTT, only 0.78 and 1.33 mg/L were detected in L150-60,

234   respectively, which are much lower than the reported limit values that inhibit plant

235   growth and the national effluent standards in Japan (Arif et al., 2016; Ko et al., 2008;

236   MOE, 2015).

237   Correlation coefficients were computed to show the relationship between the

238   physicochemical properties of liquid products and the GI value, in order to clarify the

239   factors that may contribute to their phytotoxicity (Table S1). Ammonia N and Cu were

240   found to be well correlated with the GI value, while the correlation coefficient of

241   soluble N (ammonia N + organic N) and GI was much lower than that of ammonia N

242   and GI. During seed germination period, the seeds adsorb water to rupture the coat, then

243   the radicles can elongation. Inorganic N, especially ammonia can be easily absorbed

244   along with water (Hirel et al., 2011). Organic N can be slowly decomposed by

245   microorganisms in soil after application, improving the nitrogen use efficiency of plants

246   compared with chemical fertilizers due to the reduced N loss through ammonia

247   volatilization or nitrate leaching (Franklin et al., 2017). Negative correlations were

248   noticed for most of the metal ions, except for Cu and Ca. Ca plays many roles in

249   regulating plant system functions like respiration and cell division, both of which are

250   the predominant activities during seed germination. Two aspects are possibly associated

251   with the negative correlations: 1) metal ions contribute to the high EC value, which

252   would prevent the nutrients uptake by plants if higher than a threshold value (4 mS/cm);

253   and 2) Komatsuna seeds are sensitive to the toxicity of metal ions. Since the EC values

254   in all the three liquid products were detected to be lower than 4 mS/cm and exhibit a

255   positive correlation with GI, the sensitivity of Komatsuna seeds should be considered as

256   the major reason resulting in the negative correlations. This result agrees with the report

257   by Tam and Tiquia (1994). Besides, the levels of metal ions that are beneficial for plant

258   growth also depend on different plant species. As the seeds mainly use the nutrients

259   stored in cotyledons during seed germination, plant growth assay should be conducted

260   to further explore the effect of metal ions on plant growth. The liquid product with a

261   relatively lower metal ions content or after appropriate dilution can be used as a high-

262   quality liquid fertilizer.

263   **4.   Conclusions**

264   The fertilizer potential of liquid product from hydrothermally treated swine manure

265   was firstly evaluated by investigating the solubilization of N, P and organics in swine

266   manure after HTT at different conditions and subsequently conducting seed germination

267   test. HTT technology can significantly enhance the solubilization of nutrients from

268   swine manure, and the resultant liquid product at 150ºC for 60 min achieved higher than

269   100% GI. Considering the results from GI test together with the pH and EC of the liquid

270   product, HTT could be proposed as a promising technology for producing safe and

271   value-added liquid fertilizers from swine manure.

272   **Supplementary materials**

273   Supplementary data of this work can be found in online version of the paper.

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278   **References**

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371 **Table 1** Characteristics of liquid products used for seed germination test.

	pH	EC (mS/cm)	SOC (mg/L)	Soluble N (mg N/L)	Soluble P (mg P/L)	K (mg/L)	Mg (mg/L)	Ca (mg/L)	Fe (mg/L)	Al (mg/L)	Zn (mg/L)	Cu (mg/L)
Raw manure	7.93	1.25	973.8	223.2	46.4	50.9	23.7	39.34	N.D.	2.15	N.D.	3.92
L150-60[a]	7.80	1.70	2130.8	409.2	128.8	59.7	68.1	28.92	0.78	0.31	1.33	0.54
L180-30[a]	7.57	1.78	3963.0	529.5	107.4	224.5	98.1	14.81	4.66	1.23	3.04	0.10
L200-60[a]	7.27	2.00	2626.0	547.3	108.4	247.8	157.2	15.43	5.05	1.46	3.39	n.d.

372 *EC, electrical conductivity; SOC, soluble organic carbon. N.D., not detectable.

373 [a] Lx-y denotes the liquid product from HTT at temperature of x (°C) for y min. n.d.-not detectable.

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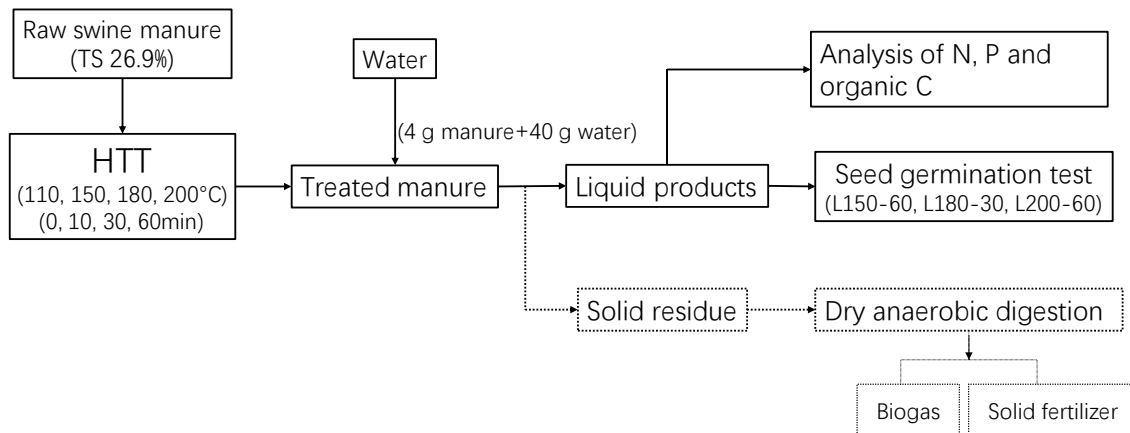
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Figures

Fig. 1 Schematic of experimental setup in this study. HTT, hydrothermal treatment; Lx-y, the liquid products from HTT at temperature of x (°C) for y min. Solid line, the experiments did in this study; Dash line, the suggested treatment method for solid residue after water extraction.
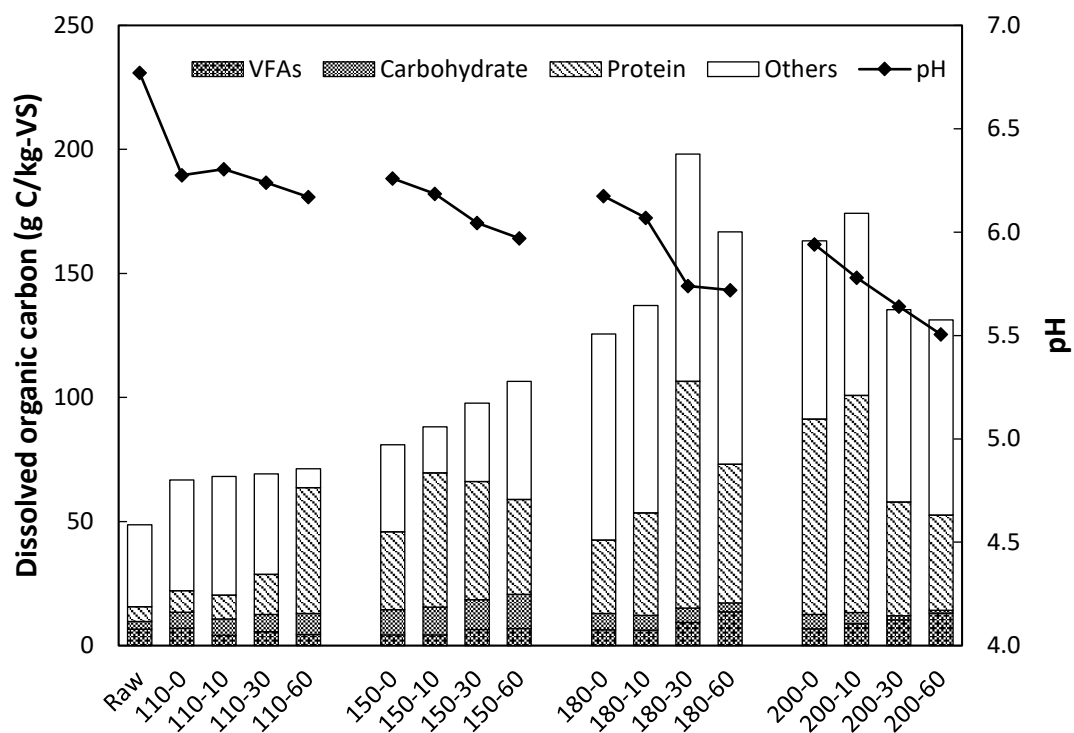
Fig. 2 Effect of hydrothermal treatment (HTT) on organic carbon dissolution from swine manure and the resultant pH of treated swine manure. 'x-y' denotes the conditions of HTT at x (°C) for y min. Raw-raw swine manure without HTT.
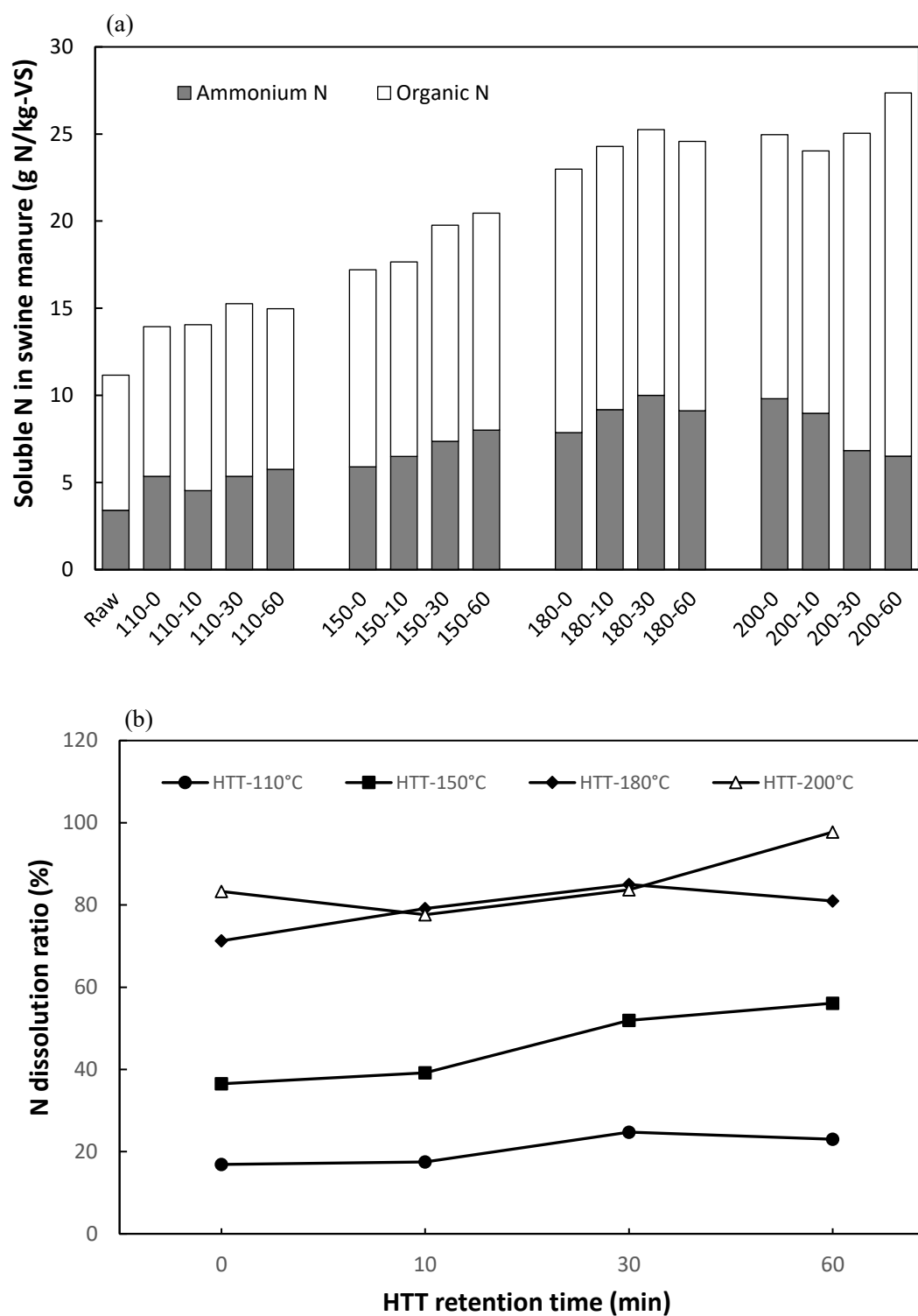
Fig. 3 Effect of hydrothermal treatment (HTT) on N dissolution from swine manure. (a)

Soluble N in swine manure, and (b) N dissolution ratio. 'x-y' denotes the conditions of

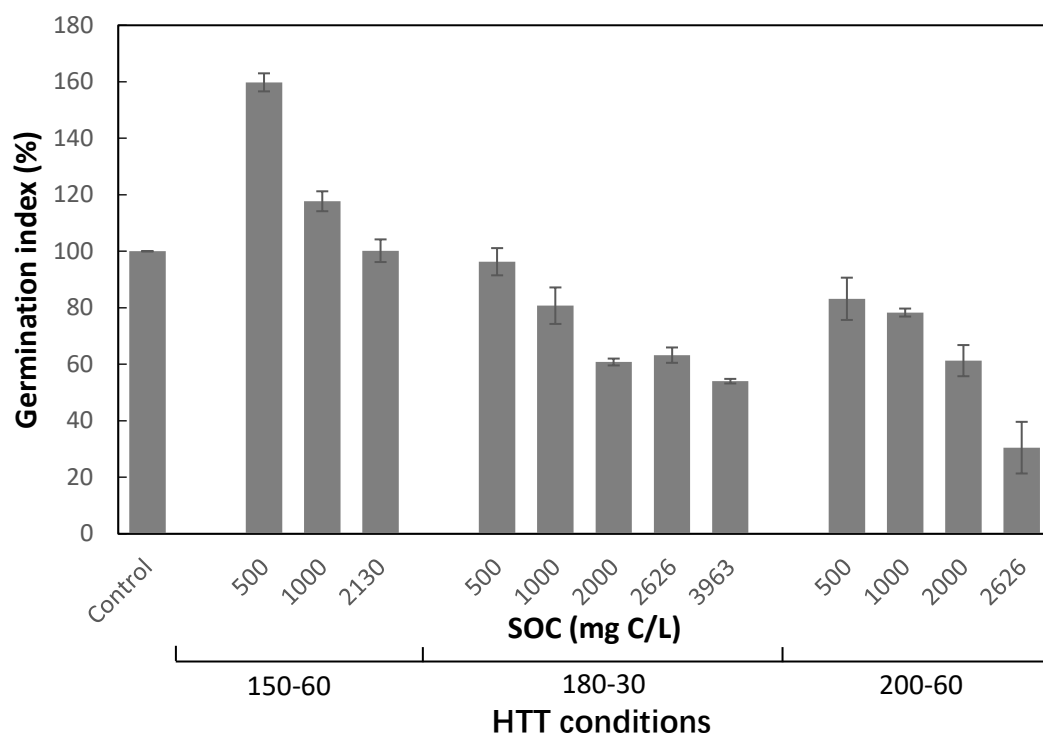HTT at x (°C) for y min. Raw-raw swine manure without HTT.

Fig. 4 Germination test by using the liquid products from hydrothermally treated swine

manure.

Supplementary Materials

Fertilizer potential of liquid product from hydrothermal treatment of swine manure

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Table S1 Correlation coefficients between germination index and related parameters at different SOC levels.

SOC500, SOC1000, and SOC2000 denote the soluble organic concentration of 500, 1000 and 2000 mg C/L, respectively.

Parameters	Germination index (GI)		
	SOC500	SOC1000	SOC2000
Electric conductivity (EC)	0.766	0.751	0.679
Ammonia N	0.991	1.000*	0.999*
Soluble N	0.360	0.457	0.514
Ortho-P	0.803	0.862	0.893
Soluble P	0.835	0.888	0.916
K	-0.896	-0.844	-0.807
Mg	-0.413	-0.315	-0.252
Ca	0.939	0.970	0.984
Fe	-0.940	-0.899	-0.868
Al	-0.881	-0.827	-0.788
Zn	-0.743	-0.668	-0.617
Cu	0.997*	1.000*	0.995

The data were analyzed by Pearson's correlation by SPSS 20.0. * indicates significance at $p < 0.05$.
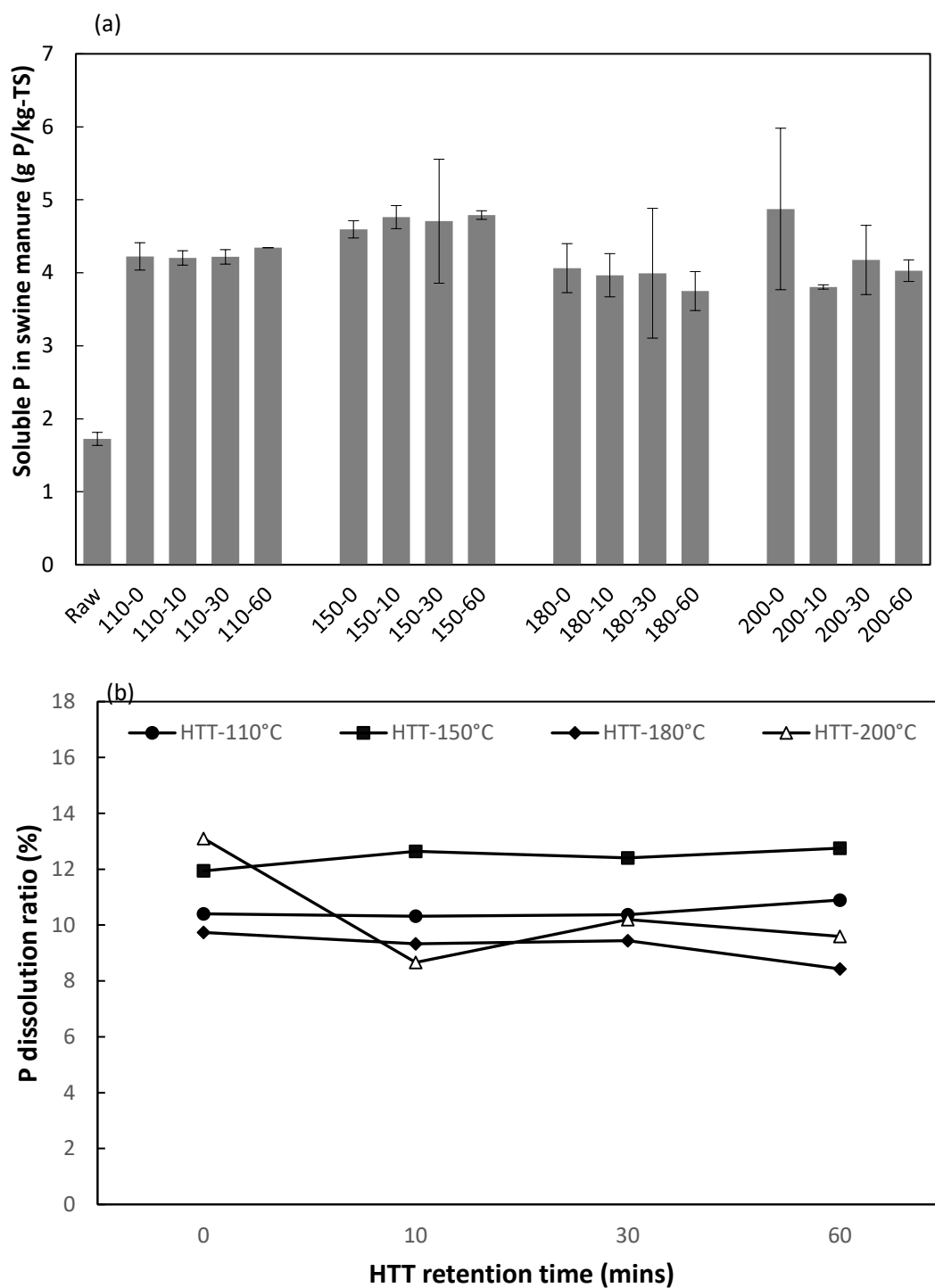
Fig. S1 Effect of hydrothermal treatment (HTT) on P dissolution from swine manure. (a) Soluble P in swine manure and (b) P dissolution ratio. 'x-y' denotes the conditions of HTT at x (°C) for y min. Raw-raw swine manure without HTT.