



# Effects of Initial Moisture Content on Phenanthrene Degradation Behaviour During Sludge Composting

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## ABSTRACT

The influence of different initial moisture content on the degradation of phenanthrene was studied during sewage sludge composting under the same aerobic fermentation conditions. Wood chips were used as amendment and the moisture content of mixture was adjusted to 56.21% (treatment 1) or 60.89% (treatment 2). The results showed that initial moisture content has a significant effect on sewage sludge composting, the composting of two treatments all lasted 12 days. In addition, variations in moisture content led to different temperature tendencies, with the temperature of treatment 1 increasing faster than that of treatment 2, while the thermophilic stage of treatment 2 lasted for longer than that of treatment 1. The overall oxygen content, moisture content, volatile solids, pH, and total nitrogen content of the two treatments were basically the same. Microbial degradation of phenanthrene during sewage sludge composting primarily occurred in the rapid heating stage and thermophilic temperature stage, and a portion of the 4-5 ring polycyclic aromatic hydrocarbons would concentrate relatively. The phenanthrene degradation rate of treatment 1 and treatment 2 were 52.37% and 45.64%, respectively. Based on the phenanthrene degradation efficiency, an initial moisture water content of 56.21% should be used during the sewage sludge composting.

## INTRODUCTION

By the end of 2015, the processing capacity of sewage treatment plants in China reached 140 million m<sup>3</sup> per day, and the annual total volume of sewage sludge produced was 41.03 billion m<sup>3</sup>, which was more than 30 million tons (moisture content 80%). Accordingly, there are severe challenges to disposal of this sewage sludge. Sewage sludge is rich in N, P and other nutrients (organic matter, etc.), has a very high agricultural value (Smith 2009), and is therefore a potential resource capable of improving soil structure, increasing soil organic matter and promoting plant growth (Debosz et al. 2002, Albiaha et al. 2001). However, most of the heavy metals and polycyclic aromatic hydrocarbons (PAHs) in wastewater and other organic pollutants can be transferred into sludge, which can lead to secondary pollution if the sludge is not disposed of properly. This, in turn, can lead to contamination of the food chain and eventually to adverse effects on human health. Polycyclic aromatic hydrocarbons (PAHs) are common persistent organic pollutants in sludge that are carcinogenic, teratogenic and mutagenic; accordingly, they are also one of the most important organic pollutants in developed countries (Petrasek et al. 1983, Smith 1996, Wild et al. 1995). For example, Australia stipulates that PAHs content in

composting products cannot exceed 6 mg·kg<sup>-1</sup>, while Luxembourg has limited PAHs content in composting products to 10 mg·kg<sup>-1</sup> (Hogget et al. 2002). Many scholars have investigated the PAHs degradation process in soil and sludge (Li et al. 2008, Ambrosoli et al. 2005, Hamdi et al. 2007, Wild & Jones 1995, Yuan et al. 2000), but there is relatively little research regarding PAHs degradation during sewage sludge composting available, especially in the Philippines. Therefore, this study investigated the influence of initial moisture content on degradation behavior of phenanthrene during sludge composting to improve the quality of sludge compost and control the contamination risk of PAHs.

## MATERIALS AND METHODS

**Materials:** The sludge of the Wulongkou wastewater treatment plant was collected as test material. The sludge moisture content was 80% and the volatile solids (VS) content was 38.2%. Wood chips with a size of 2cm × 2cm × 2cm were selected as an amendment and the moisture content was adjusted to 7%. Wood chips were selected instead of other traditional amendments, because they are of uniform size, easier to screen, recyclable, and do not participate in composting reaction; therefore, they can be used to adjust only the moisture content and porosity of the pile.

Two treatments were designed according to the different mass ratio of sludge and wood chips. The mixtures were then adjusted to moisture contents of 56.21% and 60.89% for treatment 1 and 2, respectively (Table 1), after which phenanthrene was mixed into to give final levels of 37.0 mg·kg<sup>-1</sup> and 36.3 mg·kg<sup>-1</sup>, respectively. It should be noted that the phenanthrene content in the initial mixtures differed slightly, which was likely due to phenanthrene volatilization during the process of mixing, sampling and testing.

Joint control of temperature-time and intermittent mandatory ventilation were adopted in the fermentation process for static aerobic fermentation, with 2 min of ventilation being provided at intervals of 15 min.

**Methods:** The experiment was conducted in semi-open composting fermentation tanks that were 140 cm high and had an inner diameter of 60 cm. The tanks were constructed of PVC and wrapped with a 5 cm thick cotton insulation layer to prevent fermentation tank heat loss. The bottom of the fermentation tanks contained a screen that functioned as a uniform air breather to prevent the infiltration liquid from being blocked and ensure uniform ventilation via an air blast blower at the bottom of the tanks. The composting fermentation tanks were equipped with an exhaust system, and the tail gas produced during the composting process could remove emissions from the system. The central axis of the fermentation tank was put on temperature and oxygen feeler levers, which had several probes connected to a computer to enable real-time monitoring of temperature and oxygen content during the composting process and feedback control of the blast blower. The two treatments adopted the same ventilation strategy, which was applied simultaneously to the two fermentation tanks.

The samples were collected after 0, 3, 6, 9 and 12 days of composting from 20 cm below the surface of the pile body, which could be divided into three layers. Samples were collected using a multipoint sampling method, then mixed evenly and stored at 4°C until analysis. Additionally, some fresh samples were immediately analysed for moisture content, pH, volatile solids (VS) content, nitrate nitrogen and ammonia nitrogen. Samples were dried naturally by air, then sieved with 80 mesh for analysis of total nitrogen (TN) in

the sludge feed, while samples were freeze-dried and screened through 100 mesh sieve for determination of the phenanthrene content in sludge.

The moisture content was measured by drying, the VS content by high temperature calcination, the pH using a pH meter, the TN content by the Kjeldahl method, and PAHs by using GC/MS.

## RESULTS AND DISCUSSION

**Temperature changes during composting:** Composting is a self-heating process, and temperature is one of the most important indicators of the efficiency of the composting (Patrik 2006). Sludge aerobic fermentation continued for 12 days, during which time both the treatments underwent a rapid heating stage, thermophilic temperature stage (above 50°C) and a cooling stage. However, the temperature development trend differed among treatments (Fig. 1). Initially, the treatment temperatures were basically same; however, once the rapid heating stage started, the temperature of treatment 1 rose more quickly than that of treatment 2. On day one of composting, the temperature of treatment 1 was more than 50°C and entered into a thermophilic temperature stage, while that of treatment 2 did not exceed 50°C until day 2. This was because the initial moisture content of treatment 2 (60.89%) was higher, so the temperature rose slowly.

**Oxygen content changes during composting:** For both the treatments, the oxygen content first decreased, then gradually increased, because microorganism activity reduced due to decrease in both, the amount of biodegradable organic matter and temperature (Zhou et al. 2013). In the early stage of composting, microorganisms were very active and consumed more oxygen and the oxygen content in the fermentation tanks decreased. As composting proceeded, the oxygen content changed with the oxygen consumption rate, increasing in the interim and later-stages of composting. The moisture content of treatment 1 was lower than that of treatment 2, the composting pile density was small, the pile body structure was loose, and the oxygen storage capacity was stronger. As a result, the oxygen content of treatment 1 was higher than that of treatment 2 throughout the composting experiment.

Table 1: Physico-chemical properties of composting materials.

Treatment	Materials	Mass ratio	Total weight (kg)	Moisture content (%)	Phenanthrene concentration (mg·kg <sup>-1</sup> )
Treatment 1	sludge + wood chips	2:1	135.2	56.21%	37.0
Treatment 2	sludge + wood chips	3:1	175.3	60.89%	36.3

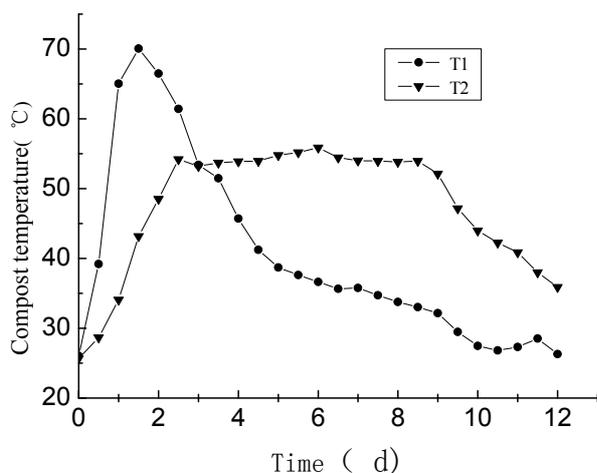


Fig. 1: Temperature changes during sewage sludge composting.

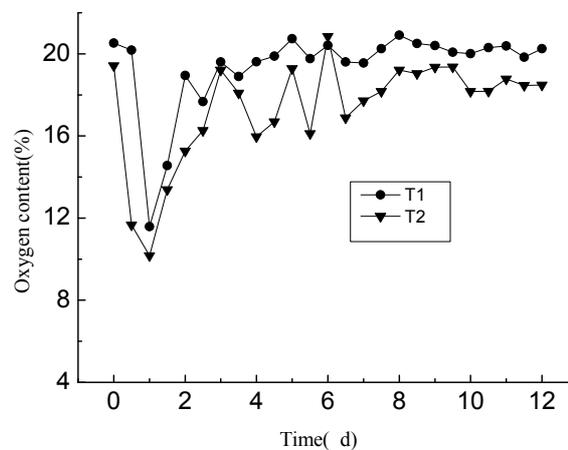


Fig. 2: Oxygen content changes during sewage sludge composting.

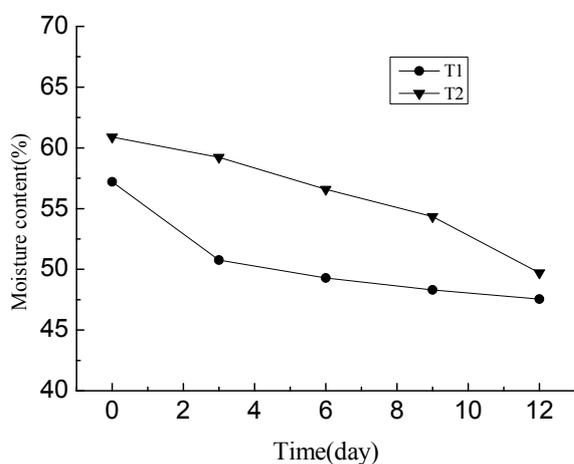


Fig. 3: Water content changes during sewage sludge composting.

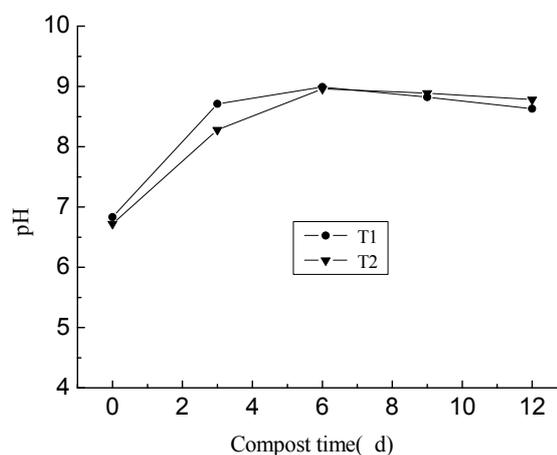


Fig. 4: Changes in pH during sewage sludge composting

**Changes in moisture content during composting:** Water is essential to microbial activity and organic matter decomposition, and changes in moisture content during the composting process are closely related to pile temperature and porosity. Sludge composting was almost a drying process, where the moisture content change was mainly caused by microorganisms, which decompose organic matter, generate heat, and make the water to evaporate (Zhou et al. 2013). As shown in Fig. 3, the moisture content of the two treatments after 12 d of composting has changed obviously, among which the moisture content of treatment 1 decreased by 9.66%, while that of treatment 2 decreased by 11.19%. Due to the rapid heating speed of treatment 1, after it entered into the thermophilic temperature stage, a large amount of water evaporated until day 3, when the temperature dropped to 53.4°C and the moisture content decreased by 6.54%. In the subsequent composting period, the tempera-

ture decreased and water evaporation became more difficult, so the moisture content decreased slowly. The rapid heating stage of treatment 2 was longer, continuing from day 3 to 9. In addition, the thermophilic temperature stage was longer, and the moisture content decrease was more noticeable during this period.

**Changes in pH during composting:** pH is an important factor that influences microbial enzymes in piles, phenanthrene biodegradation is affected by changes in pH value, it is generally believed that the optimal pH for composting is 6-9 (Yuan et al. 2000). As shown in Fig. 4, after composting, the pH of the two treatments was increased. This was because the pH of the composting material was relatively low, and the small rate of ventilation caused hypoxia. Moreover, the nitrogenous organic matter produced large amounts of  $\text{NH}_3$  under the effect of mineralization and ammonification because of the action of microorganisms, causing the pH to

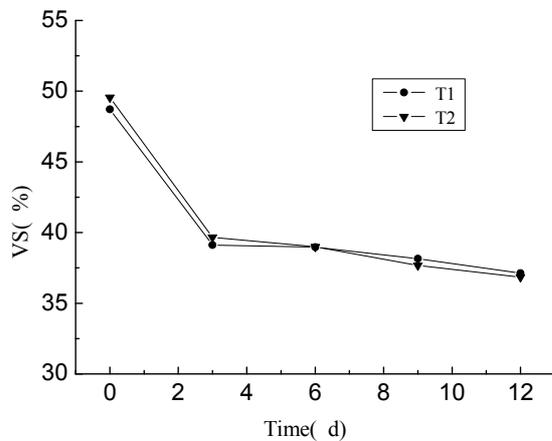


Fig. 5: Changes in volatile solids during sewage sludge composting.

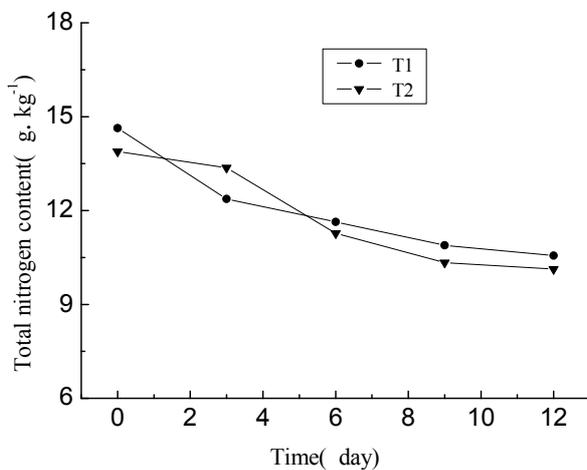


Fig. 6: Changes in total nitrogen content during sewage sludge composting.

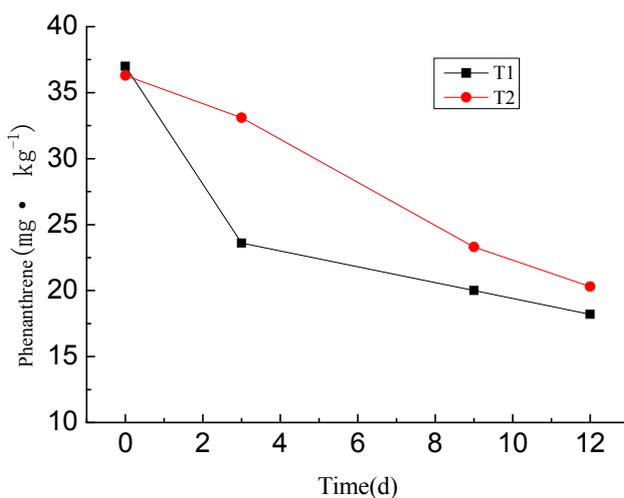


Fig. 7: Changes in phenanthrene concentration during sewage sludge composting.

rise. The pH value had declined slightly after six days, what was due to temperature reducing, enhanced nitrification of ammonium nitrogen into nitrate nitrogen, and at the same time continuous blast also reduced the amount of ammonia gas existing in the pile body, and material pH dropped. At the end of the compost period, the pH of treatment 1 and treatment 2 was 8.82 and 8.63, respectively, both of which were greater than 7, showing that composting achieved was complete.

#### Changes in volatile solids (VS) content during composting:

The initial VS values of treatment 1 and treatment 2 were 48.71% and 49.56%, respectively. After composting was initiated, the microbial activity began and the organic matter was degraded, with the greatest degradation occurring during the rapid heating stage and the thermophilic temperature stage (Fig. 5), which could be due to the intense microbial activity (Wang et al. 2017). After composting, the VS values of treatment 1 and treatment 2 were 37.13% and 36.85%, respectively. These findings indicate that both the treatments had achieved the purpose of reducing VS.

#### Changes in total nitrogen content during composting:

Nitrogen loss generally existed during the process of composting. The majority of ammonia emission occurred accompanied by the rapid decomposition of nitrogenous organic materials during the early thermophilic phase and 65% TKN could be lost in the form of  $\text{NH}_3$  (Wong et al. 2009). Nitrogen loss not only reduced the nutrient content of compost, but also led to pollution of the atmosphere. The total nitrogen content of treatment 1 dropped by 27.83%, while that of treatment 2 fell by 27.08%, indicating that the loss ratio of the two treatments was basically same. As shown in Fig. 6, both treatments showed a downward trend in nitrogen loss during composting, which also showed that the nitrogen losses had occurred during the composting process. At the beginning of the composting period, because the microorganisms grew rapidly in the period of heating and high temperature, organic nitrogen was decomposed rapidly, producing ammonia that later volatilized and was reduced in the alkaline environment. As the composting process continued, the organic matter was decomposed into carbon dioxide and water, which evaporated, making the pile body smaller and causing the total nitrogen to change slowly.

#### Changes in phenanthrene content during composting:

Sewage sludge composting is an interesting optimisation method to achieve the best possible properties in terms of its content of polycyclic aromatic hydrocarbons, where composting clearly lowered the content of PAHs (Petryk 2006). The composting experiment adopted wood as amendment, the content of phenanthrene was less than the detec-

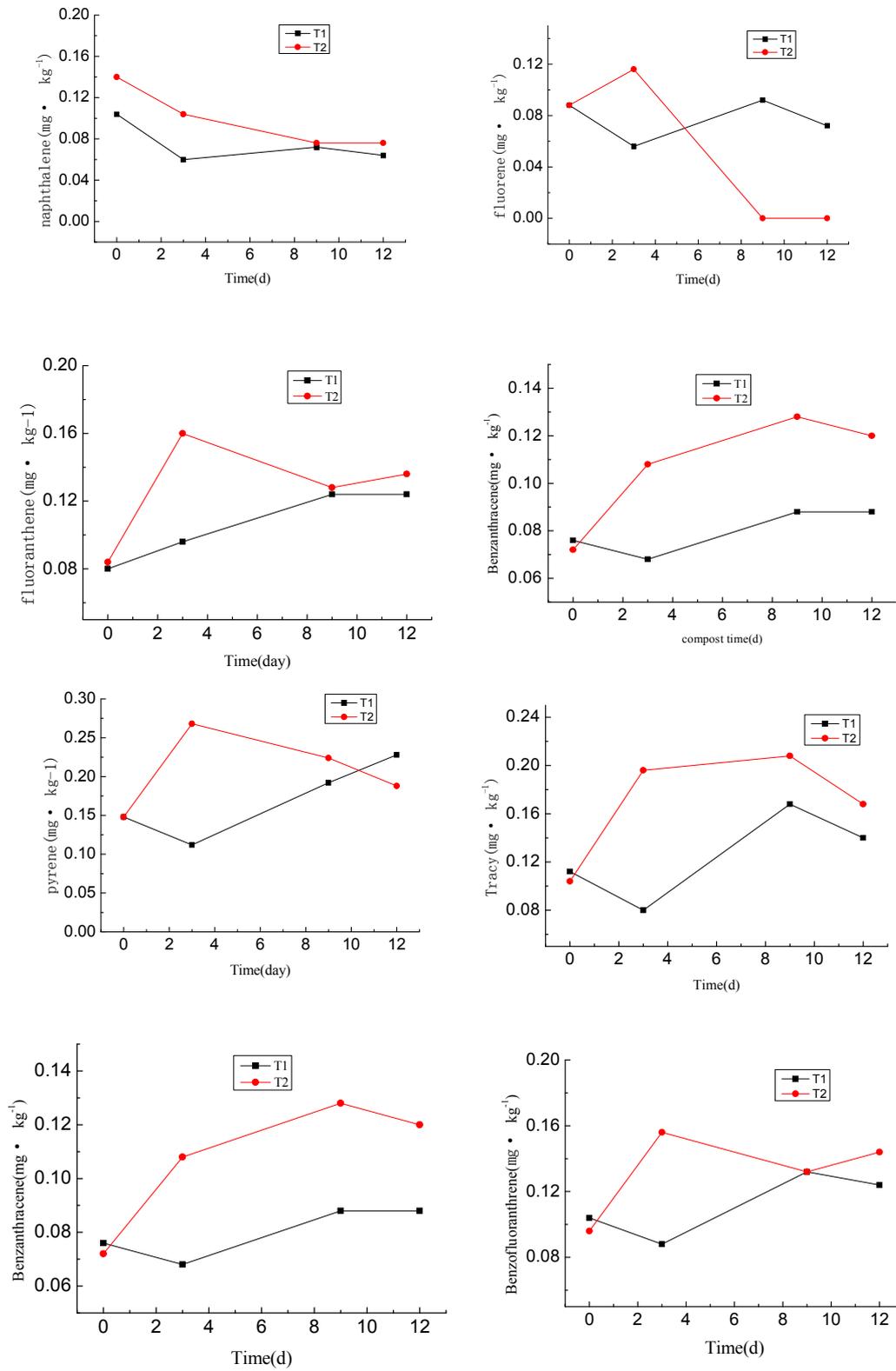


Fig. 8: Changes in concentrations of other polycyclic aromatic hydrocarbons during sewage sludge composting.

tion limit, two treatments used different quality ratio of sludge and amendment to mixtures, and tried to mix evenly, adjusting the mixture water content to 56.21% and 60.89%, respectively. The composting occurred over 12 days, after which the degradation of phenanthrene in the sludge had been to some degree.

The degradation of phenanthrene in two treatments differed, with the rate of phenanthrene degradation of treatment 1 occurring more rapidly than that of treatment 2 in the first 3 days, then becoming slower than that of treatment 2 (Fig. 7). The speed of phenanthrene biodegradation in the rapid heating stage and early thermophilic temperature stage of treatment 1 was faster, while the degradation quantity of phenanthrene in the mid-to late thermophilic temperature stage and the cooling stage was lower. It indicates that heating was helpful to the decomposition of organic matter, but the excessive temperatures would inhibit microbial activity influencing the degradation of phenanthrene; and the last stage of composting was characterised by no significant statistical changes (Patrik 2006).

By the end of the composting period, the total weight of treatment 1 and treatment 2 were 109.3 kg and 132.5 kg, respectively, during the process of composting. There was a high rate of organic matter degradation, which reduced the treatment weights significantly. It needs to eliminate the influence of the quality change in calculation of phenanthrene degradation rate. The phenanthrene content of treatment 1 decreased from 37.0 mg·kg<sup>-1</sup> to 18.2 mg·kg<sup>-1</sup>, while that of treatment 2 decreased from 36.3 mg·kg<sup>-1</sup> to 20.3 mg·kg<sup>-1</sup>. Additionally, the phenanthrene degradation rates of treatment 1 and treatment 2 were 52.37% and 45.64%, respectively.

Overall, these results indicate that the degradation of phenanthrene in the sludge mainly occurred during the heating stage and the thermophilic temperature stage. Additionally, the degradation of phenanthrene was better in treatment 1 than treatment 2.

Changes in the concentration of 15 other kinds of polycyclic aromatic hydrocarbons in the sludge during composting were also tested. The concentration of acenaphthylene, acenaphthene, anthracene, benzo[k]fluoranthene, indene, [1, 2, 3 -c, d] pyrene and diphenyl(a, h)anthracene in the sludge of the two treatments were all below the detection limit (0.01 mg kg<sup>-1</sup>) after 12 days. Additionally, the concentration of naphthalene, fluorene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo(b)fluoranthene and benzo(a)pyrene were all below 0.2 mg·kg<sup>-1</sup> after 12 days, while the levels of 4-5 ring polycyclic aromatic hydrocarbons (fluoranthene, pyrene and benzo[a] anthracene, chrysene, benzo(b)fluoranthene, benzo(a)pyrene in-

creased at the end of the composting period (Fig. 8). This may result from lower evaporation rates or decreased biological decomposition because of stronger bonding to the solid surfaces (Carlstrom & Tuovinen 2003, Smith et al. 1997) for higher molecular weight compounds. And, organic matter was degraded by microorganisms as composting proceeded, resulting in reduced weight, but little change in the quality of the high molecular weight compounds; therefore, their concentrations rose because of a concentration effect (Ozaki et al. 2017), part of the organic matter was degraded during the composting process (Fig. 5).

## CONCLUSION

The temperature of treatment 1 with an initial moisture content of 56.21% increased faster than that of treatment 2, which had an initial moisture content of 60.89%. The thermophilic stage of treatment 2 lasted for longer than that of treatment 1, but the overall trends in oxygen content, moisture content, volatile solids, pH, and total nitrogen content of the two treatments were basically the same.

Microbial degradation of phenanthrene during sewage sludge composting occurred mainly in the rapid heating stage and thermophilic temperature stage. Additionally, composting resulted in increased levels of 4-5 ring polycyclic aromatic hydrocarbons due to concentration. The initial moisture content had a significant impact on degradation of phenanthrene sewage sludge composting. Specifically, the degradation of phenanthrene in treatment 1 was better than that of treatment 2. Considering the phenanthrene degradation efficiency, adjusting the initial moisture content of mixing sludge to 56.21% before composting is recommended.

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