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Substrate Degradation and Gases Emission during Co-composting of Chicken Manure Digestion

Huang Guangqun　Huang Jing　Zhang Yang　Han Lujia
(*College of Engineering*, *China Agricultural University*, *Beijing* 100083, *China*)

Abstract: A laboratory scale experiment of chicken manure digestion co-composting was carried out using a reactor system with oxygen monitoring and feedback control. The physiochemical indexes, including temperature, oxygen concentration, volatile solid content (VS), moisture, C/N, pH value, EC, cellulose, hemicellulose, lignin, biological index, and main gas emissions (CH_4, NH_3 and N_2O) were monitored and analyzed, respectively. Physical, chemical and biological indicators presented a good dynamic corresponding relationship, and the degradation of cellulose materials mainly appeared at the middle and later periods of aerobic co-composting. Temperature and oxygen concentration had an effect on gas emissions: high temperature promoted the emissions of NH_3 and N_2O with a significantly positive correlation ($r = 0.378$; $r = 0.695$), high temperature and oxygen concentration decreased the emission of CH_4 with a significantly negative correlation ($r = -0.470$; $r = -0.494$). Gas emissions can be controlled through adjusting the levels of composing oxygen concentration and temperature. Multi-parameters characterization and correlation analyses can provide theoretical and methodological basis for process optimization of chicken manure and digestion co-composting. However, characterization and the coordination mechanism should be explored in depth on fermentation performance and gas emissions combining with comprehensive multi-indexes during and digestion co-composting in future studies.

Key words: chicken manure digestion; composting; substrate degradation; gas emission; correlation analysis

0　Introduction

In recent years, the rapid development of large-scale livestock and poultry industry promotes the rapid growth of biogas project in China, and anaerobic fermentation technology has been widely applied, besides biogas production, meanwhile it also generates a mass of fermentation residues-digestate in solid fraction and biogas slurry. The resource utilization of these residues is one of the important factors to restrict the sustainable development of biogas project[1-2]. At present, biogas slurry has been mainly used for soaking seed, feed additives, foliage dressing, insect disease prevention and other aspects, with favorable utilization value[3]. As one of solid waste, digestate has relatively high improvable element content including nitrogen, phosphorus, potassium and other elements, which is the favorable alternative to inorganic fertilizer[4]. However, digestate often contains some biodegradable organic matters, which can easily lead to "secondary fermentation". Moreover, digestate for direct application can easily lead high organic matter, nitrogen, phosphorus and potentially pathogenic microorganisms into the environment and hinder its efficient and safe recycling[5]. Aerobic composting treatment can be an effetive way for digastate disposal, which can not only ensure its biological stability, but greatly reduce the potential risk of direct application[6].

Digestate co-composting with animal manure is the process of aerobic composting using digestate with animal manure, crop straw as well as other high organic matter content wastes by reasonable ratio. During this process, the superiority of digestate and other adding materials can complement each other, which not only drives the secondary fermentation of digestate with the help of high organic matter content animal manure, but takes digestate as excellent mixing

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Corresponding author: Han Lujia, Professor. E-mail: hanlj@cau.edu.cn. Tel: +86-010-62736313.

material and conditioner, conducive to the progress of aerobic composting[2, 7], consequently at the same time of achieving efficient and safe resource utilization, obtaining organic fertilizer products with more excellent quality and value in agricultural production[8].

Researches show that, the fermentation effect of digestate and animal manure co-composting have otherness due to different ratios of raw materials, bulking agents and oxygen-supplying modes[6, 9-11]. In addition, as an approach of secondary treatment, digestate co-composting with animan manure should also pay attention to energy saving and emission reduction, otherwise it will easily cause secondary pollution and form ecological safety loophole under dual background of global warming and engineering benefits[2]. At present, researches about aerobic composting substrate degradation and gases emission regularity based on multi-parameter characterization and correlation analysis method have been rarely reported.

Therefore, in this study, chicken manure digestate was used as research object and the aerobic co-composting method was used to study on physicochemical and biological key parameters as well as dynamic changes of gas emission based on time series and correlation analysis, in order to provide theoretical and methodological support for digestate co-composting process optimization.

1 Materials and methods

1.1 Raw materials

Chicken manure digestate was collected from Biogas Station of Liuminying Village, Daxing District, Beijing; Anaerobic reactor was up flow solid anaerobic reactor (800 m^3/d), and the digestate was the solid part of solid-liquid separation after anaerobic fermentation for 18 d in normal temperature. The pig manure was taken from the farm of Chinese Academy of Agricultural Sciences, Changping District, Beijing; Mushroom residue was taken from DuoDuoXian Ecological Mushrooms Park, Tongzhou District, Beijing; Wheat straw, which was cut short to 3 ~ 5 cm for backup, was taken from one farm of Shandong Province.

The mass ratio of chicken manure digestate, pig manure, mushroom residue and wheat straw was 10∶6∶3∶1 with total weight of 45 kg to control the initial moisture content and the carbon to nitrogen (C/N) ratio close to 60% and 20, respectively[12-13].

Basic physicochemical properties of compost raw materials and initial mixture are listed in Tab. 1. The volatile solid (VS) content and C/N ratio of chicken manure digestate were 62.45% and 29.98 respectively. In contrast, the VS content of pig manure was relatively high as 80.75%, while C/N ratio was relatively low as 11.67, so both serving as main raw materials mixing could play complementary role. At the same time, adding high VS content of mushroom residue and wheat straw as conditioners helped maintain composting porous medium structure[2].

Tab. 1 Main physicochemical properties of the raw materials and initial mixture

Raw materials	Moisture/%	VS/% *	C/N *
Chicken manure digestate	68.55 ± 0.07	62.45 ± 0.32	29.98 ± 0.23
Pig manure	70.03 ± 0.06	80.75 ± 0.14	11.67 ± 0.02
Mushroom residue	50.20 ± 0.34	91.25 ± 0.04	25.06 ± 0.16
wheat straw	8.41 ± 0.32	93.22 ± 0.04	54.91 ± 1.51
Mixture	61.81 ± 0.14	73.96 ± 0.01	19.86 ± 0.08

Note: * dry matter basis.

1.2 Aerobic composting reactor system

The main structure diagram of aerobic composting reactor system used in experiment is shown as Fig. 1[14]。

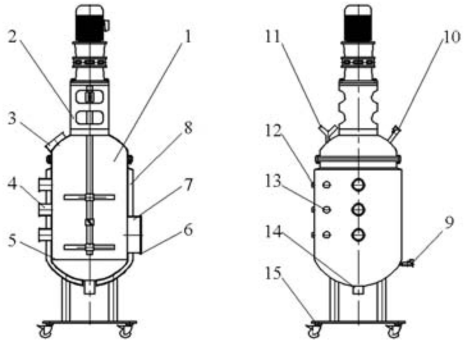

Fig. 1 Main structure of aerobic composting reactor

1. Composting bin 2. Vertical mixing system 3. Material inlet 4. Sampling opening 5. Screen mesh for air supply 6. Blind packaged plate 7. Material outlet 8. Insulation layer 9. Gas inlet 10. Water supplement 11. Gas outlet 12. Built-in temperature sensor 13. Built-in oxygen sensor 14. Leachate outlet 15. Movable support

Reaction tank is stainless steel double-layer structure, and intermediate thermal insulation layer is formed by polyurethane foaming, while the heat in upper connecting and top arc-shaped cover is relatively easy to dissipate, with effective volume of 85 L. The tank is designed with three sampling ports, three oxygen concentration sensors (City Technology 40X-V, England) and three temperature sensors (Heraeus 32208551, Germany) distributed vertically and uniformly. The real-time monitoring and feedback control of temperature and oxygen concentration can be realized by PLC control, while the composting oxygen concentration control can be realized through adjusting ventilation rate and time according to experimental requirements.

1.3 Aerobic composting experiment

Studies have shown that, the secondary aerobic fermentation of digestate needs less oxygen content than traditional aerobic composting using animal manure as raw materials[5]. Therefore, this study postulates adequate oxygen supply at the early stage (0 ~ 3 d) of composting, in order to meet the needs of vigorous activity for aerobic microorganisms in the early stage[15], and the oxygen concentration is controlled within 5% ~ 15% by automatic feedback in the rest time.

On days 0, 3, 6, 9, 12, 15, 18, 21, equal samples (about 150 g) were taken from the upper, middle and lower sampling port of the reactor, then mixed evenly and divided into two parts, one of which was air-dried and sieved, the other one was stored at −4℃ for backup.

At the end of each sampling, the mixing system started, in order to reduce composting spatial difference among the upper, middle and lower layers.

1.4 Assaying and analysis method

Fresh samples were oven-dried to a constant weight at 105℃ to determine the moisture content[16]; The VS content of samples was analyzed by the dry combustion method at 540℃[17]; Scanning electron microscopy (S−3400N, Hitachi, Japan) was used to acquire compost particle images. The particles were prepared to sieve the air-dried samples after 20 meshes.

Water extracts of the fresh compost samples were prepared by shaking the fresh sample with distilled water at 1∶10 (w/v), after which the pH value and electrical conductivity (EC) of the fresh extract were measured using pH meter (SC8231, Japan) and conductivity meter (SC8221, Japan). The total carbon and total nitrogen content were measured using an auto elemental analyzer (Vario EL, CHNOS, Germany) and then employed to calculate the C/N ratio. The cellulose, hemicellulose and lignin contents were determined using a standard detergent method called Van Soest method[18]. The measuring method of seed germination index (GI) referred to Practical Handbook for Compost Engineering[19].

Portable pump suction gas detection analyzer was used to monitor the emission concentration changes of CH_4, N_2O, NH_3 during composting process, while the gas detection analyzer sensors were CH_4 sensor (Premier, Dynament, England), N_2O sensor (NAP−100A, Nemoto, Japan), NH_3 sensor ($4NH_3$−1000, Solidsens, Germany) respectively. A plastic tube was used to connect the port of gas detector with the gas outlet to monitor and record the gas composition and concentration discharged through the air outlet from upper chamber of reactor.

1.5 Statistical analysis

Data statistical analysis and drawing were conducted using SPSS Statistics V.17.0 (IBM, USA) and OriginPro V.8.5 SR1 (OriginLab Corporation, USA) platform respectively.

2 Results and discussion

2.1 Multi-parameter dynamic changes during composting process

2.1.1 Dynamic changes of sensory index and particle morphology

Fig. 2 shows sensory changes in the mixtures and corresponding sample particle microstructure during the preliminary, interim (Day 9) and final stages (Day 21) of composting. During the preliminary phase, the mixture was yellowish-brown and had a pungent odor, with uneven granularity. As the reaction proceeded, the smell and color of the mixtures became stronger and darker during the interim stage.

Subsequently, the smell gradually became weaker, the color of the pile became blackish brown, as well as the particle became smaller diameter and loose granular structure. From the microscopic point of view, with the change of composting time, the pore of compost particles increased obviously, which was caused by the decomposition of organic matter during composting process. At the end of the reaction, the structure of compost particles was loose with an irregular shape and

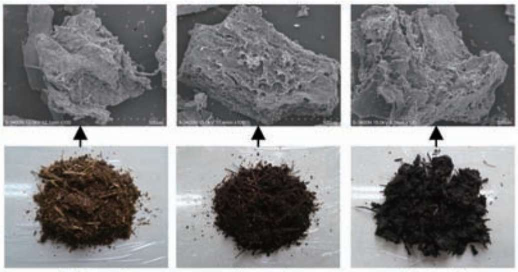
(a) Day 0 (b) Day 9 (c) Day21

Fig. 2 Pictures of mixture and scanning electron microscopic images of oven-dried compost particles at a magnification of 100 during the preliminary (Day 0), interim (Day 9) and final (Day 21) composting

higher porosity.

2.1.2 Dynamic changes of temperature and oxygen concentration

Fig. 3 is the changing scatterplot of average oxygen concentration during composting process, which is basically consistent with the setting value. Fig. 4 is the changing curve of average temperature during composting and ambient. The average environmental temperature remained at around 20℃ throughout the composting process.

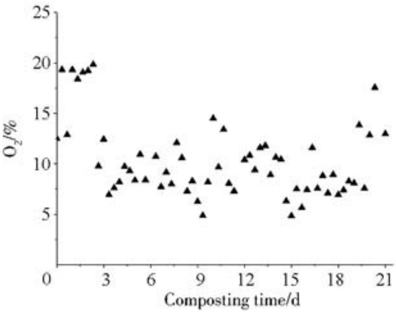

Fig. 3 Changing of oxygen concentration during composting

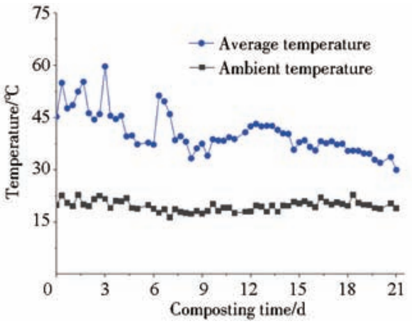

Fig. 4 Changing curve of average temperature during composting and ambient

The number of days that the compost temperature reached above 50℃ was in accordance with hygienic requirements for harmless disposal of night soil in China[20]. In the early stage of composting, microbial decomposition utilized composting organic matter to conduct own metabolism and reproduction, which intensively released lots of heat, and then the composting temperature quickly rose to over 50℃. At this time, large oxygen was needed, and sufficient oxygen was correspondingly provided, as shown in Fig. 3, to promote the activities of aerobic microorganisms.

On day 6, the temperature showed an obvious increase up to 51.2℃, which could be the result of turning, making material more fully mixed and causing the reactivation of composting process. In the later stage of composting, temperature showed a relatively moderate secondary heating and cooling period. At this time, microbial metabolic activity was low, and the temperature decreased slowly, until the whole compost gradually tended to be stable and close to the ambient temperature[6, 21-22].

2.1.3 Dynamic changes of moisture content, VS content and C/N ratio

As shown in Fig. 5, the moisture content of compost continuously increased from 61.81% to 71.81%, showing an overall uptrend, and this was because the moisture generated by microbial metabolism exceeded evaporated moisture during aerobic fermentation process[21]. Comparing with open composting, the evaporation and cooling of reactor composting were limited, while moisture was not easier to lose.

During the composting process, the overall change of VS content showed a decreasing trend because of microorganism degradation of organic matter, while VS content mainly contains biological volatile solid, and non-biological volatile solid, namely the low biodegradability part. Since the non-biological degradation volatile solid has small change during the composting process, the VS content could reflect the change of composting organic matter degradation degree.

As shown in Fig. 6, the C/N ratio showed an overall downward trend during composting process, from initial value 19.86 to the end 15.33, which was consistent with the C/N ratio dynamic change of pig manure aerobic composting studied by QIN LI et al[22].

2.1.4 Dynamic changes of pH and EC values

Fig. 7 shows the changing curves of pH value and EC value during composting process. Compost microorganisms usually have higher activity when the pH value ranges from 6.7 to 9.0[23]. As shown in the figure, the pH value basically remained stable around 9.0 during composting process. In the later period of

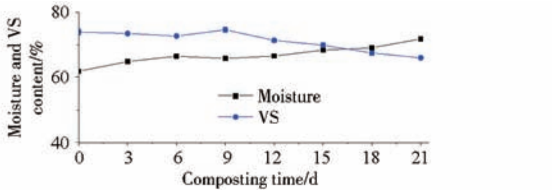

Fig. 5 Changing curves of moisture and VS content during composting

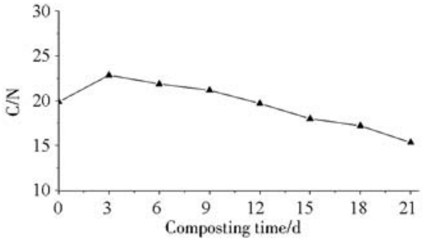

Fig. 6 Changing curve of C/N during composting

composting, pH value declined slightly. This was because the ammonia emission decreased, furthermore, there were many available energy substances, so microbial reproduction was accelerated, and the organic acid generated in microbial activities had neutralization, making the pH value decreased. At the end of composting, the pH value was 8.69, which met the requirement.

The initial EC value gradually rose from 1.16 mS/cm to 3.03 mS/cm (Day 12), and then gradually declined to be stable. At the end of the reaction, the EC value was 1.51 mS/cm. The EC value was less than 4 mS/cm during the whole composting process, which met the requirement of crop safe growth[19].

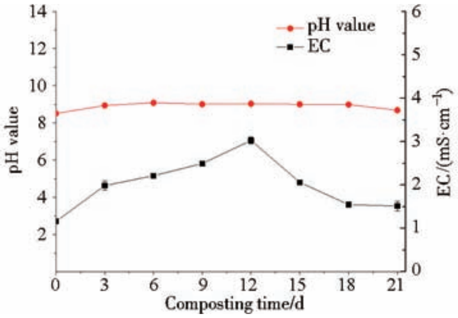

Fig. 7 Changing curves of pH value and EC value during composting

2.1.5 Dynamic changes of cellulose, hemicellulose and lignin

Tab. 2 shows the degradation of cellulose, hemicellulose and lignin during composting process, while the initial values were 21.59%, 29.66% and 7.81% respectively. In the early stage of composting, the relative content of cellulose remained stable, with less degradation. Until the reaction later stage (Day 15), the relative content of cellulose gradually declined and became 17.45% at the end of reaction. The hemicellulose decomposition was quite slow in the initial reaction, while its relative content increased slowly. After Day 12, the hemicellulose decomposition went rapid, became 24.43% until the end of the reaction. In the whole composting process, lignin content was relatively low, and remained relatively stable. In later stage, its relative content increased slightly and then remained stable until the end of reaction. This indicated that lignin was less decomposed during digestate co-composting, which was consistent with existed research of aerobic composting[24].

Tab. 2 Changes of concentration of cellulose, hemicellulose and lignin during composting %

Composting time/d	Cellulose	Hemicellulose	Lignin
0	21.59 ±0.07	29.66 ±0.52	7.81 ±0.08
3	21.76 ±0.15	29.56 ±1.11	8.72 ±0.30
6	21.88 ±0.31	33.05 ±0.44	8.73 ±0.13
9	21.71 ±0.16	32.96 ±0.81	8.84 ±0.43
12	21.05 ±0.42	34.55 ±0.76	8.72 ±0.47
15	23.54 ±0.15	28.57 ±0.96	11.14 ±0.88
18	20.62 ±0.28	21.29 ±1.18	11.62 ±0.15
21	17.45 ±0.43	24.43 ±0.87	11.60 ±0.33

Microorganisms mainly decompose VS during composting process, while cellulose, hemicellulose and lignin are essential part of VS. In this composting experiment process, the main degradation part of VS was hemicellulose, followed by cellulose, and lignin degradation was very limited. Cellulose type material is difficult to degrade, because its decomposition requires the synergistic effect of a variety of microorganisms, which needs oxygen during this process, while cellulolytic enzymes need to create local anaerobic environment during decomposing cellulose[25]. In the middle and late period of digestate aerobic composting, the temperature dropped, cellulose type materials and other low-decomposability compounds slowly decomposed in relatively anaerobic condition during secondary fermentation process, then the composing tended to be stable and went into complete maturity[26].

In general, the decomposition of cellulose and hemicellulose were mainly concentrated in the middle and late period of digestate aerobic co-composting process. This may be caused by the declining of composting temperature and moisture content during middle and late stage, resulting in fungi and actinomycetes proliferation, and then cellulose type material could be gradually decomposed to generate heat. This phenomenon was consistent with the rising temperature curve after Day 12 shown as Fig. 4.

KOMILIS et al[27] showed that the ratio between cellulose and lignin (CEL/LIGN) could be used as one of indexes to distinguish the fresh and thoroughly decomposed waste. During this digestate composting process, the initial value 2.7 of CEL/LIGN fell to 1.5, showing a favorable fermentation effect of digestate composting.

2.1.6 Dynamic change of GI

GI is one of the most sensitive indexes to evaluate the toxicity of material, not well-composted material contains toxic substances, which plays inhibitory effect on seed germination and plant growth[19]. However, different types of compost (material) have different GI value[16]. Under normal circumstances, it is believed that the compost (material) with GI over 50% nearly have no toxicity for plant seed, and completely non-toxic is believed when GI is over 85%[19].

Fig. 8 shows the GI value for original chicken manure digastate and mixture during co-composting. As shown in the figure, GI value of original chicken manure digastate was 72.04%, showing no strong toxicity to plant. This was because digestate was the by products of anaerobic fermentation, moreover, the dry matter content of digestate was relatively high after solid-liquid separation, to some extent, it had lower phytotoxicity than unfermented animal manure and other raw materials[5], however when it acts as high-quality organic modifying agent, it still has certain potential phytotoxicity[4].

In the process of chicken manure digestate co-composting, GI value showed an overall upward trend. Among them, on Day 9, GI value decreased, probably caused by enhanced mineralization degree, increasing conductivity, partial anaerobism and other integrated factors during composting process; at the end of composting, GI value was 102.99%, which indicated the compost was completely decomposed and no toxicity[19, 28]. From the perspective of lowering phytotoxicity and ensuring application safety, aerobic composting of chicken manure digastate for secondary fermentation is an effective way.

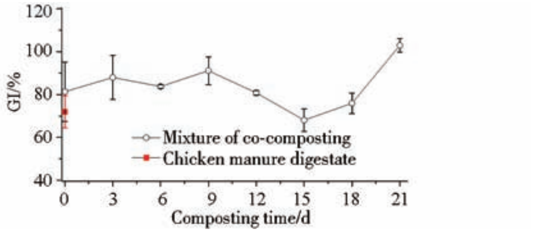

Fig. 8 Germination index changing curve during composting

2.1.7 Dynamic changes of NH_3, CH_4 and N_2O

As Fig. 9 shown, NH_3 concentration (volume fraction) was basically remained below 0.05% in the whole composting process, except two concentration peak values of 0.1% appear on Day 3 and Day 6. The volatile peak period of NH_3 mainly occurred during the high temperature period. Corresponding with temperature changing curve, there are also two peaks on Day 3 and Day 6. Upon comprehensive analysis, NH_3 volatile peak value may be caused by high temperature and enhanced alkaline environment[11]. In general, the NH_3 concentration in this study was higher than the composting condition with less cyclical turning[29], but similar to the composting conditions with forced ventilation and daily turning[30-31].

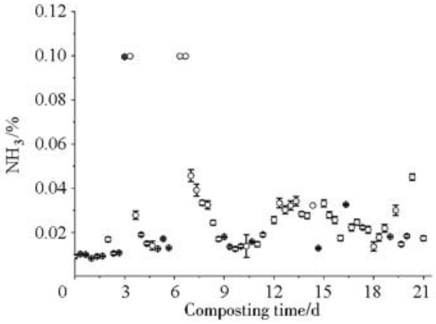

Fig. 9 Scatterplot of dynamic changes of NH_3 during composting

In theory, favorable aerobic composting environment is not suitable to produce greenhouse gases such as CH_4, N_2O and so on. For example, CH_4 is produced when methanogen deoxidizes CO_2 or hydrocarbons under anaerobic conditions[32]. In this study, the dynamic changes of above gases content were monitored and shown in Fig. 10 and Fig. 11. In the early stage of composting, the CH_4 concentration (volume fraction) was relatively low and stable because of sufficient oxygen supplying. In the intermediate stage of composting with high temperature, the CH_4 concentration of the compost showed periodic fluctuation. In the late stage, the CH_4 concentration was high with periodic fluctuation as a whole, and reached the highest value of 3.45% on Day 15; from the late period of reaction to the end, CH_4 concentration fell to a lower level. During composting process, the N_2O concentration (volume fraction) fluctuated greatly, but maintained at a low level, and show overall declining trend. At the end of the reaction, it tended to be zero. In this study, the CH_4 and N_2O emission level were consistent with the condition of pig manure straw composting studied by SOMMER[33], but lower than the condition of pig manure and straw composting with passive ventilation[29].

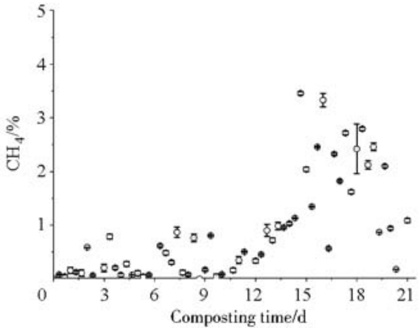

Fig. 10 Scatterplot of dynamic changes of CH_4 during composting

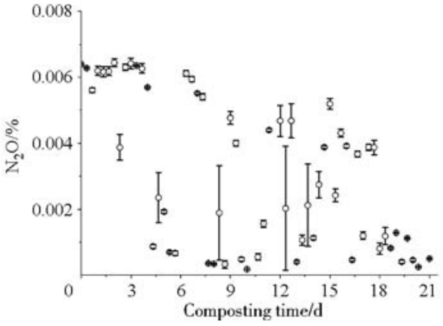

Fig. 11 Scatterplot of dynamic changes of N_2O during composting

2.2 Gas emission changes based on correlation analysis

Tab. 3 shows the correlation analysis between gas emission concentration with average oxygen concentration and temperature of composting. As shown in Tab. 3, CH_4 had good negative correlation with oxygen concentration, which indicated that the lower oxygen concentration in the compost, the more rapidly the methane bacteria proliferated, the higher level of producing CH_4[11]. For the reasons, moisture content can influence the overall oxygen supply efficiency of the compost. In this study, moisture content gradually increased in the late stage, which caused less pores and more aggregation of composting pile; moreover, decreasing GI value in late stage also further indicated, there were more partial anaerobic zones in late stage of composting corresponding with GI curve. In addition, from the micro perspective, GE, et al.[15] quantitatively characterized the aerobic layer thickness of pig manure particles during composting based on Fourier transform infrared microspectroscopy. Each composting particle had its own anaerobic core and aerobic layer, which meant more anaerobic core with thinner aerobic layer could produce more anaerobic zones, and easier to produce more CH_4.

Tab. 3 Correlation result between gas emissions and average oxygen/temperature during composting

	CH_4	N_2O	NH_3
Average oxygen	−0.494**	0.110	−0.144
Average temperature	−0.470**	0.695**	0.378**

Note: ** Significant correlation at 0.01 level.

In addition, the CH_4 concentration also had good negative correlation with temperature, which indicated when the aerobic reaction accounted for the absolute proportion under adequate oxygen condition, and generated heat accumulation, the compost would remain high temperature, playing inhibitory effect on CH_4 production and emission.

The emission concentration of NH_3 and N_2O were positively correlated with temperature, which indicated higher temperature could accelerate the production and emission of NH_3 and N_2O; especially a large number of NH_3 was produced during organic matter degradation process which generated heat, and NH_3 volatilization in high temperature was the main cause of nitrogen loss[32].

Therefore, studying emission gas components and dynamic changes during digastate composting are significant for optimizing composting process, energy saving and emission reduction, and reducing secondary pollution to the environment just as other organic wastes composting.

3 Conclusions

(1) Based on key parameters of physicochemical and biological indexes as well as dynamic changes of gas emission, the multi-characterization analysis shows that aerobic composting with reasonable ratio of raw

materials and moderate oxygen supplying can be an effective method to handle chicken manure digastate, and ensure its safety and qualify for utilization to some extent .

(2) It can be seen from correlation analysis and multi-characterization analysis, in chicken manure digastate co-composting process, the degradation of cellulose type material mainly concentrates in late stage of reaction, and each index has good dynamic corresponding relationship.

(3) During co-composting process, CH_4 emission concentration has good negative correlation with temperature and oxygen, while NH_3 and N_2O emission concentration show positive correlation with temperature; therefore, oxygen concentration and temperature of composting can be adjusted to control gas emission. Based on correlation analysis, studying emission gas composition and dynamic changes is significant for optimizing composting process and reducing emissions.

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鸡粪沼渣联合好氧堆肥基质降解与气体排放研究

黄光群　黄　晶　张　阳　韩鲁佳

（中国农业大学工学院，北京 100083）

摘要：利用好氧堆肥反应器系统开展鸡粪沼渣联合好氧堆肥试验，进行物理、化学、生物学指标和甲烷（CH_4）、氨气（NH_3）及氧化亚氮（N_2O）等排放气体的多元动态表征，并开展主要排放气体与堆体氧浓度和温度相关性分析。研究结果表明，通过合理物料配比、适度供氧的联合好氧堆肥可更有效地实现鸡粪沼渣安全、优质资源化利用。鸡粪沼渣联合好氧堆肥过程中，物理、化学、生物学指标动态变化具有良好的动态对应关系，纤维素类物质的降解主要集中在反应中后期。堆肥过程中CH_4与堆体氧浓度和温度均呈现良好的负相关关系，NH_3和N_2O与温度均呈正相关关系；可通过控制堆体氧浓度和温度调控气体排放。综上，基于多元参数表征、相关性分析可为鸡粪沼渣联合好氧堆肥工艺优化提供理论和方法学支撑。

关键词：鸡粪沼渣；好氧堆肥；基质降解；气体排放；相关性分析

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Substrate Degradation and Gases Emission during Co-composting of Chicken Manure Digestion

Huang Guangqun　Huang Jing　Zhang Yang　Han Lujia

(*College of Engineering*, *China Agricultural University*, *Beijing* 100083, *China*)

Abstract: A laboratory scale experiment of chicken manure digestion co-composting was carried out using a reactor system with oxygen monitoring and feedback control. The physiochemical indexes, including temperature, oxygen concentration, volatile solid content (VS), moisture, C/N, pH value, EC, cellulose, hemicellulose, lignin, biological index, and main gas emissions (CH_4, NH_3 and N_2O) were monitored and analyzed, respectively. Physical, chemical and biological indicators presented a good dynamic corresponding relationship, and the degradation of cellulose materials mainly appeared at the middle and later periods of aerobic co-composting. Temperature and oxygen concentration had an effect on gas emissions: high temperature promoted the emissions of NH_3 and N_2O with a significantly positive correlation ($r = 0.378$; $r = 0.695$), high temperature and oxygen concentration decreased the emission of CH_4 with a significantly negative correlation ($r = -0.470$; $r = -0.494$). Gas emissions can be controlled through adjusting the levels of composing oxygen concentration and temperature. Multi-parameters characterization and correlation analyses can provide theoretical and methodological basis for process optimization of chicken manure and digestion co-composting. However, characterization and the coordination mechanism should be explored in depth on fermentation performance and gas emissions combining with comprehensive multi-indexes during and digestion co-composting in future studies.

Key words: chicken manure digestion; composting; substrate degradation; gas emission; correlation analysis

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作者简介：黄光群（1979—），男，副教授，博士生导师，主要从事生物质资源与利用研究，E-mail：huangguangqun@126.com

通信作者：韩鲁佳（1964—），女，教授，博士生导师，主要从事生物质资源与利用研究，E-mail：hanlj@cau.edu.cn

引言

近年来，规模化畜禽养殖业的快速发展促进了我国沼气工程的快速增长，厌氧发酵处理技术得到了广泛应用，除生产沼气外，同时也产生了大量的发酵残余物——沼渣和沼液。而沼渣、沼液的资源化利用是制约沼气工程可持续发展的重要因素之一[1-2]。目前，沼液的资源化利用主要包括浸种、饲料添加剂、叶面施肥、防治病虫害等方面，具有良好的利用价值[3]。作为固体废弃物的沼渣，其可利用的氮、磷、钾等元素含量相对较高，是良好的无机肥替代品[4]。然而，沼渣往往含有部分可生物降解有机物，除易产生“二次发酵”外，若直接施用于土壤极易导致高有机质、氮、磷及潜在病原微生物进入环境，阻碍其高效、安全循环利用[5]。沼渣经好氧堆肥化处理，可有效确保其生物稳定性，大大降低直接施用的潜在风险[6]。

沼渣与畜禽粪便联合好氧堆肥是将沼渣与畜禽粪便等高有机质含量的有机废弃物、农作物秸秆等通过合理配比进行好氧堆肥，利用沼渣和加入物料各自优势进行互补，既能借助高有机质含量的畜禽粪便带动沼渣的二次发酵，又能将沼渣作为优良掺混料和调理剂，有利于好氧堆肥过程的进行[2,7]，从而在实现高效、安全资源化利用的同时，获得农业生产上更具优良品质和价值的有机肥产品[8]。

研究表明，沼渣与畜禽粪便联合好氧堆肥的发酵效果因物料配比、填充剂及通风供氧方式的不同而具有差异性[6,9-11]，此外，作为“二次处理”途径，在全球变暖和工程比较效益双重背景下，沼渣联合好氧堆肥应同时注重节能和减排两个方面，否则极易造成“二次污染”，形成生态安全隐患[2]。目前，基于多参数表征和相关性分析方法研究沼渣联合好氧堆肥基质降解及气体排放规律的研究鲜见报道。

因此，本文以鸡粪沼渣为研究对象，通过联合好氧堆肥方法，开展基于时间序列的理化和生物学关键指标及气体排放的动态变化规律研究，并进行相关性分析，以期为沼渣联合堆肥工艺优化提供理论和方法学支撑。

1　材料与方法

1.1　堆肥原料

鸡粪沼渣取自北京市大兴区留民营村沼气站，厌氧反应器为升流式固体厌氧反应器（800 m^3/d），所取鸡粪沼渣为中温厌氧发酵 18 d 后固液分离的固体部分；猪粪取自北京市昌平区中国农业科学院养殖场；蘑菇渣取自北京市通州区朵朵鲜生态蘑菇园区；麦秸取自山东省某农场，切短至 3 ~ 5 cm 备用。

将鸡粪沼渣、猪粪、蘑菇渣与麦秸按质量比 10∶6∶3∶1进行均匀混合，总质量为 45 kg；使得初始混合物料含水率和碳氮比分别控制在 60% 和 20 左右[12-13]。

堆肥原料及其初始混合物料基本理化特性如表 1。鸡粪沼渣挥发性固体（VS）质量分数和碳氮比分别为 62.45% 和 29.98。猪粪 VS 质量分数相对较高，为 80.75%，碳氮比相对较低，为 11.67，两者作为主要原料混合可以起到互补作用。同时，添加高 VS 含量蘑菇渣和麦秸作为调理剂，有助于维持堆体多孔介质架构[2]。

表 1　堆肥原料及初始混合物料基本理化特性

Tab. 1　Main physicochemical properties of raw materials and initial mixture

堆肥原料	含水率/%	VS 质量分数/% *	碳氮比 *
鸡粪沼渣	68.55 ± 0.07	62.45 ± 0.32	29.98 ± 0.23
猪粪	70.03 ± 0.06	80.75 ± 0.14	11.67 ± 0.02
蘑菇渣	50.20 ± 0.34	91.25 ± 0.04	25.06 ± 0.16
麦秸	8.41 ± 0.32	93.22 ± 0.04	54.91 ± 1.51
混合物料	61.81 ± 0.14	73.96 ± 0.01	19.86 ± 0.08

* 干基。

1.2　好氧堆肥反应器系统

试验所用好氧堆肥反应器系统的主罐体结构示意图如图 1 所示[14]。

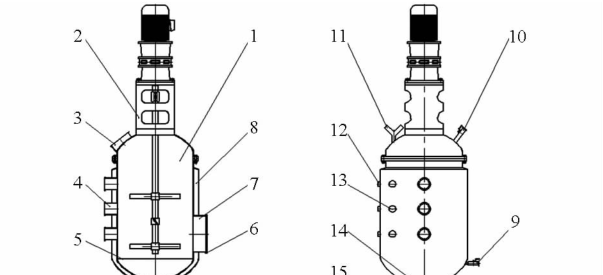

图 1　好氧堆肥反应器系统主罐体结构示意图

Fig. 1　Main structure of aerobic composting reactor

1. 物料舱　2. 搅拌系统　3. 入料口　4. 取样口　5. 布气筛网　6. 快装盲板　7. 出料口　8. 保温层　9. 进气口　10. 补水口　11. 出气口　12. 温度传感器通道　13. 氧浓度传感器通道　14. 沥出液收集口　15. 可移动式支撑底座

反应器罐体为不锈钢双层结构，中间经聚氨酯发泡形成保温层，其中上层连接处和圆弧形顶盖部分热量相对易散失，其有效容积为 85 L。罐体设有 3 个垂直均匀分布的取样口、3 个垂直均匀分布的氧浓度传感器（City Technology 40X − V 型，英国）及 3 个垂直均匀分布的温度传感器（Heraeus 32208551

型,德国)。利用PLC控制实现温度、氧浓度的实时监测和反馈控制,可根据试验需求通过调节通风速率和时间实现堆体氧浓度水平控制。

1.3　好氧堆肥试验

研究表明,沼渣二次好氧发酵对供氧量的需求少于传统的以畜禽粪便为原料的好氧堆肥[5]。因此,本研究设定堆肥前期(0~3 d)足量供氧,以满足前期好氧微生物剧烈活动的需要[15],其余时间经自动反馈控制在5%~15%范围内。

分别在第0、3、6、9、12、15、18、21天从反应器上、中、下层取样口取等量样品(约150 g),混合均匀后分为2部分,一部分经热风干燥粉碎备用,另一部分置于-4℃保存备用。每次取样结束后,启动反应器系统搅拌装置对堆体搅拌混合均匀,减少上、中、下层堆体空间差异性。

1.4　测定分析方法

在105℃下干燥鲜样至恒质量,计算样品含水率[16];540℃下将干燥样品置于马弗炉中烧至恒质量,计算VS含量[17];干燥样品过20目筛,利用扫描电子显微镜(S-3400N型,日立,日本)获取堆肥样品微观图像。将鲜样按固液比(质量体积比)1∶10浸提过滤取其上清液,分别利用pH计(SC8231型,日本)和电导率仪(SC8221型,日本)测定pH值和电导率。利用元素分析仪(Vario EL CHNOS型,德国)测定总碳、总氮含量并计算碳氮比。利用Van Soest法测定纤维素、半纤维及木质素含量[18]。种子发芽指数(GI)的测定方法参照《堆肥工程实用手册》[19]。

利用便携泵吸式气体检测分析仪监测堆肥过程中CH_4、N_2O、NH_3的排放浓度动态变化情况,所用气体检测分析仪传感器分别为CH_4传感器(Premier型,Dynament,英国)、N_2O传感器(NAP-100A型,Nemoto,日本)和NH_3传感器(4NH_3-1000型,Solidsens,德国)。利用塑料软管连接反应器上部的出气口及气体检测分析仪入口,监测并记录反应器堆体上腔经出气口排出的气体组成及浓度。

1.5　数据处理与分析方法

数据统计分析及绘图分别利用SPSS Statistics V.17.0(IBM,美国)和OriginPro V.8.5 SR1(OriginLab Corporation,美国)软件平台完成。

2　结果与讨论

2.1　堆肥过程多元参数动态变化

2.1.1　堆体形貌特征动态变化

图2所示为堆肥初始、第9天及第21天堆体物料感观变化情况及其颗粒水平下扫描电镜图像。堆肥初期物料呈黄褐色,臭味浓,粒度较大且不均匀;随着基质的不断降解,堆体散发出的气味较浓,颜色逐渐变深;堆肥后期,气味逐渐变淡,堆体颜色呈灰黑褐色,颗粒粒径变小,且呈现疏松的团粒结构。从微观角度来看,随着堆肥时间的变化,堆体颗粒的孔隙明显增多,这是由堆肥过程中有机物的分解引起的。反应结束时,堆体颗粒结构疏松,形状不规则,有较多孔隙。

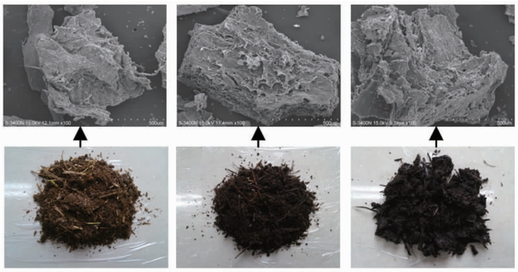
(a) 堆肥初始　(b) 第9天　(c) 第21天

图2　堆肥初始、第9天、第21天堆体感观变化图及其烘干样品的100倍电子扫描电镜图

Fig.2　Pictures of mixture and scanning electron microscopic images of oven-dried compost particles at a magnification of 100 during the preliminary (Day 0), interim (Day 9) and final (Day 21) composting

2.1.2　温度和氧浓度动态变化

图3为堆肥过程中堆体平均氧浓度(体积分数)变化,与所设值基本一致。图4为堆体平均温度变化曲线。在整个堆肥过程中,外界环境温度一直保持在20℃左右。堆体温度达到50℃以上的天数符合我国粪便无害化卫生标准[20]。堆肥初期,微生物分解利用堆料有机物进行自身代谢与繁殖,集中释放大量热量,堆体温度快速升高至50℃以上。此时需氧量大,对应提供足量氧气,如图3所示,促进好氧微生物的活动。

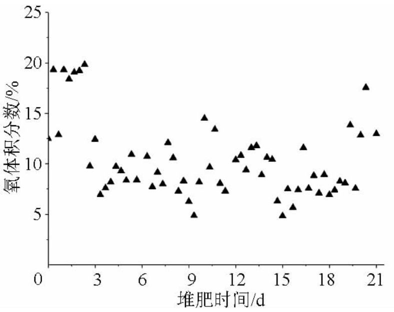

图3　堆体氧浓度动态变化

Fig.3　Changing of oxygen concentration during composting

堆肥中期第6天,温度又有所升高,达到51.2℃,这是由于此时进行机械搅拌后,物料混合更为充分,产生“二次发酵”现象。堆肥后期,堆体温度出现较为缓和的二次升温、降温期;此时,微生物代谢活性低,温度缓慢降低,直至整个堆体逐渐趋于

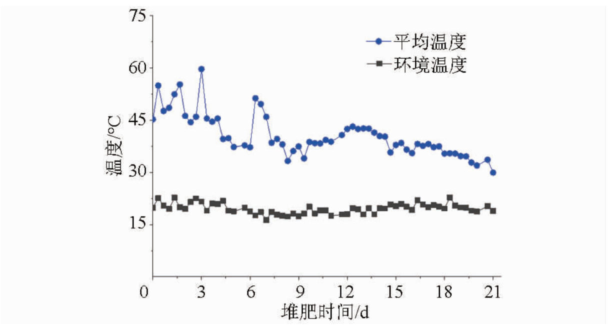

图 4　堆体平均温度与环境温度动态变化曲线

Fig. 4　Changing curve of average temperature during composting and ambient

稳定并接近环境温度[6,21-22]。

2.1.3　含水率、VS 含量及碳氮比动态变化

如图 5 所示，堆体含水率从 61.81% 逐渐增加到 71.81%，整体呈上升趋势，是由于好氧发酵过程中微生物新陈代谢产生的水分超过了蒸发的水分[21]。相比于开放式堆肥，反应器式堆肥蒸发散热受到限制，水分更加不易散失。

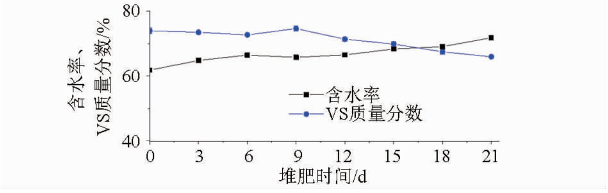

图 5　堆体含水率与 VS 质量分数动态变化曲线

Fig. 5　Changing curves of moisture and VS content during composting

堆肥过程中由于微生物降解有机质，VS 含量总体变化呈减少趋势，VS 主要包括生物挥发固体和非生物挥发固体即生物难降解部分。因堆肥过程非生物降解挥发性固体变化很小，VS 含量变化可反映堆肥有机物降解程度的变化。

在堆肥反应过程中，如图 6 所示，碳氮比整体呈下降趋势，从初始值 19.86 到结束时降至 15.33，与秦莉等[22]研究的反应器式猪粪好氧堆肥碳氮比动态变化一致。

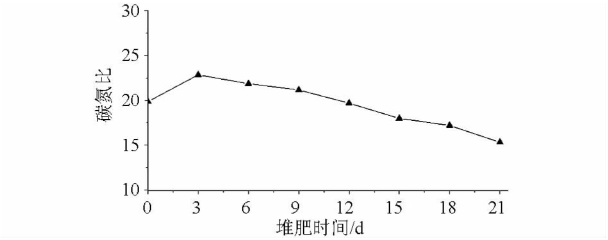

图 6　堆体碳氮比动态变化曲线

Fig. 6　Changing curve of C/N during composting

2.1.4　pH 值和电导率动态变化

图 7 为堆肥过程中 pH 值和电导率变化曲线。通常 pH 值在 6.7 ~ 9.0 范围内堆肥微生物有较高的活性[23]。如图所示，在整个堆肥过程中，pH 值基本保持平稳，维持在 9.0 左右。在堆肥后期，pH 值略有所降低，这是因为氨气释放量减少，加之可利用的能量物质较多，微生物繁殖加快，其活动产生的有机酸有中和作用，使 pH 值降低，堆肥结束时，pH 值为 8.69，满足需求。

电导率从初始值 1.16 mS/cm 逐渐增加到 3.03 mS/cm（第 12 天），随后不断降低趋于平稳，反应结束时，电导率为 1.51 mS/cm。在整个堆肥过程中，电导率均小于 4 mS/cm，满足作物生长安全的要求[19]。

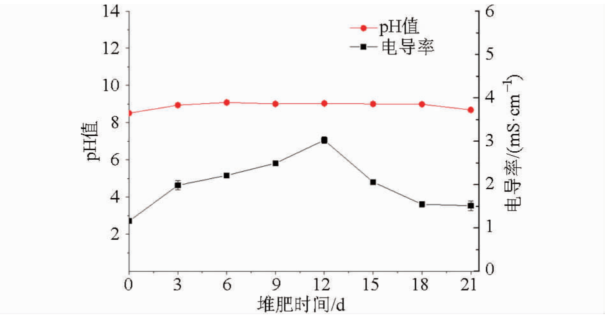

图 7　堆体 pH 值与电导率动态变化曲线

Fig. 7　Changing curves of pH value and electrical conductivity during composting

2.1.5　纤维素、半纤维素和木质素动态变化

表 2 为堆肥过程中纤维素、半纤维素及木质素相对含量的变化情况，其初始值分别为 21.59%、29.66%、7.81%。在堆肥前期，纤维素相对含量基本保持稳定，分解较少，直至反应后期第 15 天后逐渐下降，到反应结束时为 17.45%。半纤维素在反应初期分解缓慢，其相对含量缓慢增加，在第 12 天后，半纤维素快速分解，直至反应结束时为 24.43%。在整个堆肥过程中，木质素含量整体较低，且保持相对稳定，后期其相对含量有所增加后又保持稳定直至反应结束，表明木质素在本沼渣堆肥试验过程中，较少分解，与已有好氧堆肥研究趋势一致[24]。

表 2　堆体纤维素、半纤维素及木质素相对含量动态变化

Tab. 2　Change of concentration of cellulose, hemicellulose and lignin during composting　　%

堆肥时间/d	纤维素	半纤维素	木质素
0	21.59 ± 0.07	29.66 ± 0.52	7.81 ± 0.08
3	21.76 ± 0.15	29.56 ± 1.11	8.72 ± 0.30
6	21.88 ± 0.31	33.05 ± 0.44	8.73 ± 0.13
9	21.71 ± 0.16	32.96 ± 0.81	8.84 ± 0.43
12	21.05 ± 0.42	34.55 ± 0.76	8.72 ± 0.47
15	23.54 ± 0.15	28.57 ± 0.96	11.14 ± 0.88
18	20.62 ± 0.28	21.29 ± 1.18	11.62 ± 0.15
21	17.45 ± 0.43	24.43 ± 0.87	11.60 ± 0.33

堆肥过程中微生物分解有机质，主要分解 VS，纤维素、半纤维素和木质素是其重要组成部分。该堆肥过程 VS 主要降解部分为半纤维素，其次是纤维素，木质素降解非常有限。纤维素类物质分解较难，其分解需要多种微生物的协同作用，此过程需要氧气，而分解纤维素的酶在分解纤维素时则需要创造局部的厌氧环境[25]，在沼渣好氧堆肥的中后期，堆温回落，在相对厌氧条件下纤维素等较难分解的化合物缓慢分解，经过二次发酵，堆体趋于稳定，进入完熟[26]。

总体来说，沼渣联合好氧堆肥过程中，纤维素与半纤维素的分解主要集中在堆肥中后期，这可能是由于在堆肥中后期，随着堆体温度与水分含量的降低，导致真菌与放线菌增殖，纤维素类物质逐渐被分解，且产生热量，这与图 4 中温度曲线第 12 天后堆体温度升高一致。

KOMILIS 等[27]认为用纤维素与木质素相对含量比值（CEL/LIGN）可作为区分新鲜与腐熟废弃物的指标。在沼渣堆肥过程中，CEL/LIGN 从初始值 2.7 降至 1.5，表明沼渣堆肥发酵效果良好。

2.1.6　种子发芽指数动态变化

种子发芽指数（GI）是评价物料毒性最敏感的指标之一，未腐熟的堆肥（物料）含有毒性物质，对种子萌发、植物生长产生抑制作用[19]。不同种类的堆肥（物料）其 GI 也不尽相同[16]，一般情况下，认为 GI 大于 50% 的堆肥（物料）对植物种子基本无毒性，GI 大于 85% 可认为完全无毒性[19]。

图 8 为鸡粪沼渣原样及沼渣堆肥过程中 GI 的变化情况，如图所示，鸡粪沼渣原样 GI 为 72.04%，未表现出强烈的植物毒性。这是因为沼渣是经过厌氧发酵后的副产物，且经固液分离处理干物质含量较高，一定程度上比未发酵畜禽粪便等原始物料具有较低的植物毒性[5]，但作为高品质有机改良剂仍具有一定的潜在植物毒性[4]。

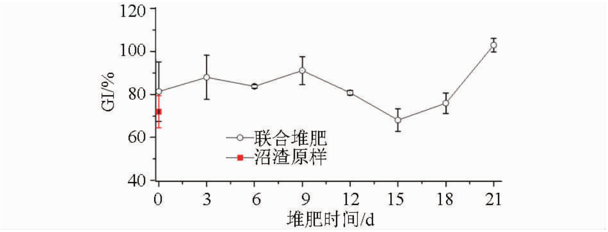

图 8　堆体种子发芽指数动态变化曲线

Fig. 8　Germination index changing curves during composting

在鸡粪沼渣堆肥过程中，GI 整体呈上升趋势。其中，第 9 天后，GI 有所降低，可能是由于堆肥过程矿质化程度增强、电导率增大和局部厌氧等综合因素所致；堆肥结束时，GI 为 102.99%，表明堆肥完全腐熟且无毒性[19, 28]。从降低植物毒性、确保施用安全角度，鸡粪沼渣的二次好氧堆肥发酵是有效途径。

2.1.7　NH_3、CH_4与 N_2O 动态变化

由图 9 可知，整个堆肥过程 NH_3浓度（体积分数）基本维持在 0.05% 以下水平，但在第 3 天和第 6 天产生了 2 个体积分数为 0.1% 的峰值，NH_3挥发高峰期主要在高温期。对应温度动态变化曲线，在第 3 天和第 6 天的温度也出现了 2 个高峰，综合分析，NH_3挥发高峰值可能是由于堆体温度升高和碱性环境增强等所致[11]。整体而言，本研究中 NH_3浓度水平高于周期性翻堆次数较低的堆肥工况[29]，但与强制通风及每日翻堆堆肥工况水平相近[30-31]。

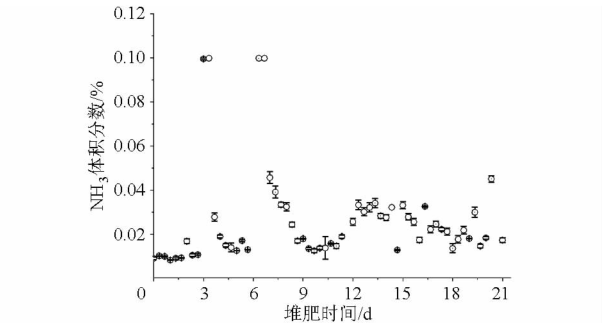

图 9　堆体 NH_3 动态变化

Fig. 9　Scatterplot of dynamic changes of NH_3 during composting

理论上，良好的好氧堆肥环境不宜产生 CH_4、N_2O等温室气体，如有产生是由于产甲烷菌在厌氧条件下还原 CO_2或碳氢化合物所致[32]。该研究中，实际监测的上述气体含量动态变化情况如图 10 和图 11所示。在堆肥前期，堆体氧气充足，CH_4浓度（体积分数）较低且相对稳定。堆肥中期高温阶段，堆体 CH_4呈现周期性波动。在堆肥中后期，CH_4整体偏高，呈现周期性波动，在第 15 天达到最高值 3.45%；反应后期至结束时，CH_4浓度下降至较低水平。堆肥过程，N_2O 的浓度（体积分数）波动较大，但一直维持在较低水平，且整体呈现下降趋势。反应结束时，趋近于零。本研究中 CH_4和 N_2O 浓度的变化情况与 SOMMER 等[33]研究猪粪秸秆堆肥中排放水平较一致，但低于被动通风式猪粪秸秆堆肥浓度水平[29]。

2.2　基于相关性分析的气体排放规律

表 3 为排放气体浓度与堆体平均氧浓度及平均温度的相关性分析。如表 3 所示，CH_4与堆肥氧浓度呈现良好的负相关关系，表明堆体氧浓度越低，甲烷菌繁衍越迅速，产 CH_4水平越高[11]。究其原因，含水率会影响堆肥整体的供氧效率，堆体后期含水率逐渐升高会造成堆体孔隙的减少及团聚现象的增加；从对应 GI 曲线来看，后期 GI 的降低也可进一步

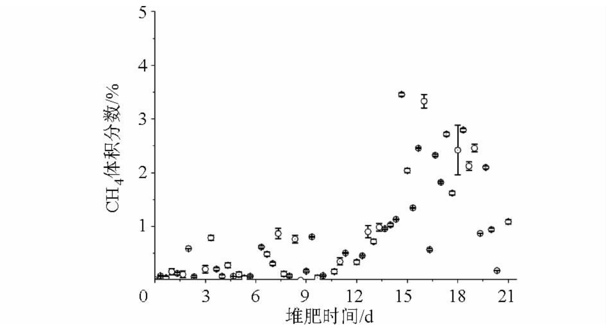

图 10　堆体 CH_4动态变化

Fig. 10　Scatterplot of dynamic changes of CH_4 during composting

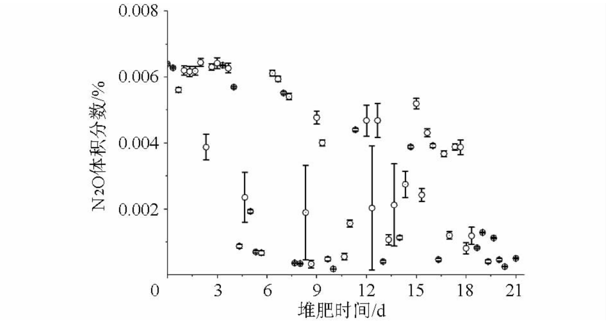

图 11　堆体 N_2O 动态变化

Fig. 11　Scatterplot of dynamic changes of N_2O during composting

表明，堆体后期存在较多的局部厌氧区。GE 等[15]通过显微红外成像方法定量表征分析了猪粪堆肥颗粒“好氧层”和“厌氧层”，颗粒内部厌氧层越厚，表明其厌氧区域越多，易产生较多的 CH_4，进一步从微观机理角度阐释了其原因。

此外，堆体 CH_4浓度与温度也呈现良好的负相关关系，表明氧气相对充足，好氧反应占绝对比重，产生的热量累积使堆体处于高温，对 CH_4产排有抑制作用。NH_3 和 N_2O 的排放浓度与温度均呈正相关，表明温度越高，将加速其产排；尤其因堆肥中有机物降解过程产生大量 NH_3，其随高温挥发是堆肥氮素损失主要原因[32]。因此，沼渣为原料的好氧堆肥过程同其他有机固废原料好氧堆肥一样，研究其排放气体组成及动态变化对于优化堆肥工艺、节能减排，减少对环境的二次污染具有重要意义。

表 3　排放气体浓度与堆体平均氧浓度及温度相关性结果

Tab. 3　Correlation result between gas emissions and average oxygen concentration/temperature during composting

参数	CH_4	N_2O	NH_3
平均氧浓度	-0.494**	0.110	-0.144
平均温度	-0.470**	0.695**	0.378**

** 表示显著性水平为 0.01，即 $P<0.01$ 水平。

3　结论

（1）基于物理、化学、生物学关键指标和排放气体动态变化的多元表征分析表明，通过合理物料配比、适度供氧的好氧堆肥处理，一定程度上更有效地确保鸡粪沼渣安全优质利用。

（2）通过多元表征和相关性分析发现，在鸡粪沼渣联合好氧堆肥过程中，纤维素类物质的降解主要集中在反应中后期，且各指标间具有良好的动态对应关系。

（3）堆肥过程 CH_4的排放浓度与堆体氧浓度和温度呈现良好的负相关关系，NH_3和 N_2O 与温度均呈现正相关关系；可通过控制堆体氧浓度和温度调控气体排放。基于相关性分析研究排放气体组成及动态变化对于优化堆肥工艺、减少排放具有重要意义。

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