

### 堆肥过程中抗生素和耐药基因消减研究进展

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# 堆肥过程中抗生素和耐药基因消减研究进展

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**摘要:**堆肥是常用的有机固废处理技术,能够快速实现有机固废资源化利用,但传统工业化堆肥对于抗生素及其耐药基因(Antibiotic resistance genes, ARGs)消减重视不足。事实上,近年来畜禽粪污等堆肥原料中抗生素及ARGs残留问题日益凸显,抗生素及ARGs消减已经成为堆肥过程中不容忽视的部分和亟待解决的问题。本文综述了近年来抗生素及ARGs在堆肥过程中消减的研究现状和一般特点,以及影响消减效果的理化和生物学因素、相关优化技术措施的效果和瓶颈等,以期为最大限度降低ARGs传播风险提供参考。

**关键词:**堆肥;抗生素耐药基因;消减

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## Progress of the degradation of antibiotics and the elimination of antibiotic resistance genes

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**Abstract:** Composting is a technology commonly used for the treatment of organic solid wastes. Although it can quickly utilize such wastes, traditional industrial composting does not give sufficient concern to the degradation of antibiotics and antibiotic resistance genes (ARGs). Common raw composting materials, such as livestock manure, poultry litter, food waste, and sewage sludge, act as reservoirs of antibiotic contaminants, which is an urgent problem during composting and cannot be ignored. This review, based on recent studies, pointed out the characteristics, biological and physiochemical factors, enhancement measures, and main bottlenecks of the degradation of antibiotics and ARGs during composting, thereby offering advice for minimizing the risk of ARGs dissemination.

**Keywords:** composting; antibiotic resistance genes; degradation

迄今为止,抗生素仍是人类对抗细菌性感染的重要手段。由于其不当使用等问题,环境中的抗生素残留及相关抗生素耐药基因(Antibiotic resistance genes, ARGs)已经成为威胁公共健康安全的环境污染物,世界上每年有多达70万人因耐药性问题死亡<sup>[1]</sup>。抗生素耐药性问题是世界各国面临的共同挑战,我国的耐药性问题也不容忽视。我国是目前世界上最大的抗生素原料药生产国和抗生素产品消费国,供应全球90%的抗生素原料药市场<sup>[2]</sup>,年产量超过

100万t<sup>[3]</sup>,年消费量约为世界总量的12.5%<sup>[4]</sup>,抗生素生产过程中的制药废弃物<sup>[5]</sup>、人和动物给药后的粪尿<sup>[6-7]</sup>以及食物垃圾<sup>[8]</sup>和污泥<sup>[9]</sup>等都已经成为事实上的抗生素及ARGs储藏库。堆肥是一种常用的有机固废处理技术,在抗生素降解及其ARGs消减方面也有重要作用<sup>[7,10]</sup>,充分了解堆肥过程中抗生素及其ARGs消减的规律、影响因素及优化技术,不仅对我国有机固废利用具有重要意义,也为最大限度减少抗生素及ARGs扩散风险提供重要参考。

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## 1 研究现状分析方法

在Web of Science数据库中以TS=“compost”AND “antibiotic”以及TI=“compost” and (TS=“antibiotic” OR “antimicrobial” OR “pharmaceutical” OR “drug” OR “substance” OR “ARG” OR “ARB” OR “resistance” OR “security” OR “emerging contaminants”),分两次筛选文献(截至2021年6月25日),筛选条件为:(1)堆肥最高温度达到50℃及以上;(2)有完整的升温和降温过程;(3)至少报告了一种或一类或总抗生素/ARGs在堆肥前后的浓度或丰度变化;(4)报告了堆肥原料和堆肥方式。共筛选到132篇文献。

抗生素及ARGs在堆肥过程中的消减规律研究已成为近年来(特别是最近5 a)的热点问题(图1),通过堆肥技术降低ARGs的传播风险逐渐引发关注。

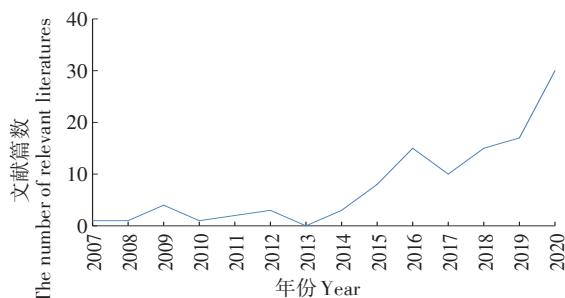


图1 相关文献发表年度变化

Figure 1 The number of relevant literatures in recent years

我国既是抗生素生产大国<sup>[2]</sup>,也是抗生素产品消费大国<sup>[4]</sup>,面临的ARGs污染风险尤为突出,对于堆肥过程中抗生素及ARGs消减规律关注度更高,其次是美国和加拿大,其他国家在这一领域的聚焦相对较少(图2)。相关研究在我国分布区域并不均衡,以北京(19.6%)和陕西(18.7%)最为集中,其次为黑龙江(13.1%),其他地区相对较少。

粪污、污泥和制药废弃物是抗生素及ARGs在堆肥中消减规律研究中涉及的主要堆肥原料,也是主要的抗生素及ARGs污染风险来源,其中粪污是最主要的研究原料,76.5%的相关研究完全或部分以粪污为主要堆肥原料(图3),17.4%的相关研究完全或部分以污泥为主要堆肥原料,15.2%的相关研究完全或部分以制药废弃物为主要堆肥原料。

## 2 堆肥过程中抗生素及ARGs消减情况

### 2.1 堆肥过程中抗生素降解情况

堆肥能够有效降解多种抗生素(表1)。其中,磺

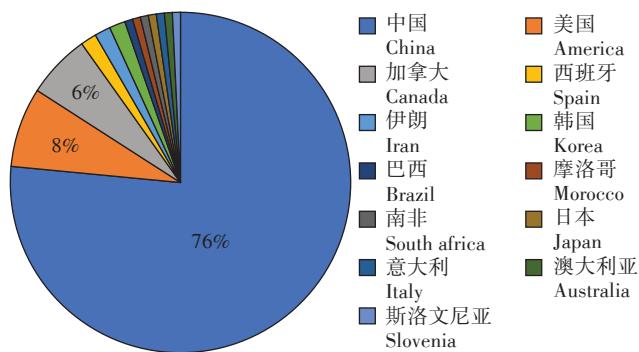


图2 相关文献国际分布情况

Figure 2 Distribution of nations and areas of relevant studies

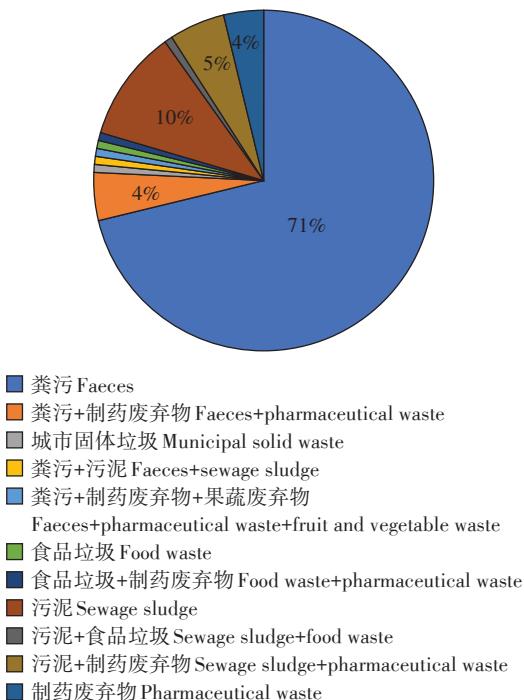


图3 相关文献堆肥主要原料

Figure 3 Distribution of main raw materials of relevant literatures

胺类<sup>[20]</sup>、喹诺酮类<sup>[23]</sup>、四环素类<sup>[33]</sup>、大环内酯类<sup>[63]</sup>、 $\beta$ -内酰胺类<sup>[57]</sup>抗生素甚至可以降至检测限以下,甲硝唑<sup>[52]</sup>、盐霉素<sup>[70]</sup>、三氯卡班<sup>[29]</sup>、氟苯尼考<sup>[47]</sup>、拉沙菌素<sup>[68]</sup>、达托霉素<sup>[5]</sup>和林可酰胺<sup>[20]</sup>等抗生素也能得到有效降解。

有些堆肥试验中抗生素降解率较低,如火鸡粪便堆肥中,磺胺类抗生素未能降解<sup>[40]</sup>,污泥堆肥<sup>[19]</sup>和商业化畜禽粪污堆肥<sup>[34]</sup>中,氟喹诺酮类抗生素几乎没有变化,四环素类抗生素在阉公猪粪便堆肥中降解率仅为27.33%<sup>[35]</sup>,牛粪堆肥中泰乐菌素基本没有降解<sup>[20]</sup>,头孢菌素C菌渣和污泥共堆肥中头孢菌素C的降解率仅为6.58%<sup>[24]</sup>。

总体来说,大部分抗生素均能在适当的堆肥处理

表1 抗生素在堆肥中消减情况  
Table 1 The degradation of antibiotics during composting

| 类别<br>Class | 抗生素<br>Antibiotic  | 国家<br>Nation | 堆肥原料<br>Raw material | 初始浓度<br>Initial concentration/<br>(ng·g <sup>-1</sup> ) | 降解率<br>Degradation<br>rate/% | 参考文献<br>Reference          |
|-------------|--|--------------|----------------------|---|------------------------------|----------------------------|
| 四环素类        | 四环素 Tetracycline   | 中国           | 粪污                   | 6.03~150 000.00   | 37.77~100.00                 | [11~18]                    |
|             |  | 摩洛哥          | 污泥                   | 200   | 35                           | [19]                       |
|             |  | 美国           | 粪污                   | 81.6~452.0  | *~72                         | [20~21]                    |
|             | 脱水四环素 Anhydrotetracycline                                | 美国           | 粪污                   | 7.2   | *                            | [21]                       |
|             | 4-表位四环素 4-epitetracycline                                | 美国           | 粪污                   | 100   | *                            | [21]                       |
|             | 盐酸四环素<br>Tetracycline hydrochloride                      | 伊朗           | 粪污                   | 20 000~100 000  | 99                           | [22]                       |
|             | 土霉素<br>Oxytetracycline                                   | 中国           | 粪污                   | 38.14~200 000.00  | 38.21~100.00                 | [4, 12~18, 23~28]          |
|             |  | 中国           | 污泥                   | 1 508.4~1 520.6   | 65.7~82.8                    | [29]                       |
|             |  | 美国           | 粪污                   | nd~115 000  | *~100                        | [21, 30~31]                |
|             |  | 伊朗           | 粪污                   | 20 000~100 000  | 99.5                         | [22]                       |
|             |  | 南非           | 粪污                   | 35 200~123 300  | 29.12~77.34                  | [32]                       |
|             | 金霉素<br>Chlortetracycline                                 | 中国           | 粪污                   | 25.42~879 600   | 27.33~100.00                 | [12~15, 17~18, 33~39]      |
|             |  | 美国           | 粪污                   | 675~1 500   | 71~99                        | [20, 40]                   |
|             |  | 伊朗           | 粪污                   | 20 000~100 000  | 99.3                         | [22]                       |
|             |  | 加拿大          | 粪污                   | 51.5~3 678.5  | 84.65~99.38                  | [41~42]                    |
| 磺胺类         | 4-表位金霉素<br>4-epi-chlortetracycline                       | 美国           | 粪污                   | 4.3   | 100                          | [21]                       |
|             | 金霉素/4-表位金霉素<br>Chlortetracycline/4-epi-chlortetracycline | 加拿大          | 粪污                   | 142 000~902 000   | 95.73~99.53                  | [43]                       |
|             | 烯醇/酮基金霉素<br>Enol/keto-chlortetracycline                  | 美国           | 粪污                   | 107 646~113 000   | 32.79~100.00                 | [30, 44]                   |
|             | 异金霉素 Iso-chlortetracycline                               | 加拿大          | 粪污                   | nd~104 000  | 97.85~99.00                  | [43]                       |
|             | 强力霉素 Doxycycline   | 中国           | 粪污                   | 18 000~85 630   | 40.15~100.00                 | [30, 44]                   |
|             |  | 西班牙          | 粪污                   | 17.2~120 400.0  | 39.70~100.00                 | [12~13, 16~17, 23, 35, 45] |
|             | 甲基苯并嘧啶<br>Methylbenzopyrimidine                          | 中国           | 粪污                   | 4.49  | 100                          | [13]                       |
|             | 磺胺醋酰胺 Sulfacetamide                                      | 中国           | 粪污                   | 7.48  | 100                          | [13]                       |
|             | 磺胺氯吡啶 Sulfachlorpyridazine                               | 中国           | 粪污                   | 2.2~484.0   | *~100                        | [12, 15, 23]               |
|             | 磺胺氯嗪 Sulfaclozine  | 中国           | 粪污                   | 0.98~1.96   | *                            | [17]                       |
| 磺胺类         | 磺胺嘧啶 Sulfadiazine  | 中国           | 粪污                   | 0.96~274.99   | *~86.81                      | [12~13, 15, 17]            |
|             |  | 中国           | 污泥+食品垃圾              | 577~5 860   | 35.0~68.4                    | [9]                        |
|             | 磺胺二甲氧嘧啶 Sulfadimethoxine                                 | 中国           | 粪污                   | 20.8  | 49.52                        | [15]                       |
|             |  | 美国           | 粪污+污泥                | 410~510   | 98                           | [47]                       |
|             | 磺胺多辛 Sulfadimoxine                                       | 中国           | 污泥+食品垃圾              | 2.49~887.00   | 16.20~100.00                 | [9]                        |
|             | 磺胺甲嘧啶 Sulfamerazine                                      | 中国           | 粪污                   | 3.92~10 000.00  | 0~100                        | [4, 12, 17, 26, 39]        |
|             | 磺胺间甲嘧啶 Sulfamethazine                                    | 中国           | 粪污                   | 226.19~32 000.00  | 68.98~100.00                 | [12, 15, 27, 48]           |
|             |  | 美国           | 粪污                   | 992~10 800  | 97~100                       | [20, 40]                   |
|             |  | 美国           | 粪污+污泥                | 2 200~4 200   | 97~99                        | [47]                       |
|             |  | 加拿大          | 粪污                   | 1.84~299 000.00   | *~99                         | [41~43]                    |
| 磺胺类         | 磺胺甲基噻唑 Sulfamethizole                                    | 中国           | 粪污                   | 3.5~6.6   | 100                          | [15]                       |
|             | 磺胺甲恶唑 Sulfamethoxazole                                   | 中国           | 粪污                   | 4.9~23 206.0  | *~100                        | [15, 17, 48~49]            |
|             | 磺胺甲氧基哒嗪<br>Sulfamethoxypyridazine                        | 中国           | 粪污                   | 15.3  | 100                          | [15]                       |
|             | 磺胺甲基嘧啶 Sulfamethyldiazine                                | 中国           | 粪污                   | 1.4   | *                            | [15]                       |

续表1 抗生素在堆肥中消减情况

Continued table 1 The degradation of antibiotics during composting

| 类别<br>Class   | 抗生素<br>Antibiotic                    | 国家<br>Nation | 堆肥原料<br>Raw material | 初始浓度<br>Initial concentration/<br>(ng·g <sup>-1</sup> ) | 降解率<br>Degradation<br>rate/% | 参考文献<br>Reference           |
|---------------|--------------------------------------|--------------|----------------------|---|------------------------------|-----------------------------|
|               | 磺胺对甲氧嘧啶<br>Sulfamethoxydiazine       | 中国           | 粪污                   | 8.6   | 45.12                        | [13]                        |
|               | 磺胺间甲氧嘧啶<br>Sulfamonomethoxine        | 中国           | 粪污                   | 89.85~255.56  | 64.07~73.29                  | [12,23]                     |
|               | 磺胺毗啶 Sulfapyridine                   | 中国           | 粪污                   | 12  | 69.17                        | [15]                        |
|               | 磺胺喹啉 Sulfaquinoxaline                | 中国           | 粪污                   | 3.81~48.10  | 62.79~100.00                 | [13,15]                     |
|               | 磺胺噻唑 Sulfathiazole                   | 中国           | 粪污                   | 9.3~14.7  | 34.41~61.03                  | [13,15]                     |
|               | 磺胺异恶唑 Sulfoxazole                    | 中国           | 粪污                   | 10.9  | 100                          | [15]                        |
|               | 磺胺二甲氧嗪 Sulphadimethoxine             | 中国           | 粪污                   | 10.84   | 100                          | [13]                        |
|               | 磺胺二甲嘧啶 Sulphamethazine               | 中国           | 粪污                   | 63.43~25 000.00   | 76.12                        | [23,50]                     |
| 氟喹诺酮类         | 甲氧苄啶 Trimethoprim                    | 摩洛哥          | 污泥                   | 200   | 35                           | [19]                        |
|               | 环丙沙星 Ciprofloxacin                   | 中国           | 粪污                   | 9.21~53 000.00  | *~100                        | [4,12~13,15,17,23,26,37,51] |
|               |                                      | 中国           | 污泥+食品垃圾              | 23~40 200   | 29.3~99.1                    | [9]                         |
|               |                                      | 伊朗           | 粪污+制药废弃物+果蔬废弃物       | 20 000~100 000  | 100                          | [52]                        |
|               |                                      | 西班牙          | 粪污                   | 4 724   | 93.61                        | [46]                        |
|               |                                      | 摩洛哥          | 污泥                   | 5 820   | *                            | [19]                        |
|               | 恩诺沙星 Enrofloxacin                    | 中国           | 粪污                   | 29.24~67 680.00   | *~100                        | [12~13,15~17,23,39,51,53]   |
|               |                                      | 西班牙          | 粪污                   | 5 186   | 72.6                         | [46]                        |
|               | 诺氟沙星 Norfloxacin                     | 中国           | 粪污                   | 16.95~54 200.00   | *~100                        | [13,15,17,23,37,51]         |
|               |                                      | 中国           | 污泥                   | 680~1 457   | 97.65~98.76                  | [54]                        |
|               |                                      | 中国           | 污泥+食品垃圾              | 105.6~57 200.0  | 74.5~99.2                    | [9]                         |
|               | 氧氟沙星 Ofloxacin                       | 中国           | 粪污                   | 2.94~59.17  | *~100                        | [12,15,17]                  |
|               |                                      | 中国           | 污泥                   | 684~1 225   | 97.22~98.59                  | [54]                        |
|               |                                      | 摩洛哥          | 污泥                   | 4 140   | *                            | [19]                        |
|               | 氟罗沙星 Fleroxacin                      | 中国           | 粪污                   | 9.00~64.31  | 22.59~66.67                  | [13,23]                     |
| $\beta$ -内酰胺类 | 盐酸二氟沙星<br>Difloxacin hydrochloride   | 中国           | 粪污                   | 20.2~66.7   | *~43.78                      | [13,15]                     |
|               | 培氟沙星 Pefloxacin                      | 中国           | 粪污                   | 1.91  | 47.74                        | [12]                        |
|               | 甲磺酸培氟沙星<br>Pefloxacin mesylate       | 中国           | 粪污                   | 20.19   | 59.29                        | [13]                        |
|               | 洛美沙星 Lomefloxacin                    | 中国           | 粪污                   | 2 100~55 000  | 45.3~61.6                    | [51]                        |
|               | 沙拉沙星 Sarafloxacin                    | 中国           | 粪污                   | 2 350~56 900  | 61.5~68.6                    | [51]                        |
|               | 羟氨苄青霉素 Amoxicillin                   | 中国           | 粪污                   | 1 190.43  | 97.95                        | [13]                        |
|               | 氨苄青霉素 Ampicillin                     | 摩洛哥          | 污泥                   | 2 400   | 46                           | [19]                        |
|               | 苄甲青霉素 Benzylpenicillin               | 西班牙          | 污泥                   | 1 243   | 100                          | [55]                        |
|               | 青霉素 Penicillin                       | 中国           | 粪污+制药废弃物             | 898 510   | 100                          | [56]                        |
|               |                                      | 中国           | 污泥+制药废弃物             | 1 065 200~1 325 600                                     | 76.90~100.00                 | [57~59]                     |
| 大环内酯类         |                                      | 中国           | 制药废弃物                | 3 228.3~3 651.9   | 100                          | [60]                        |
|               | 青霉素 G Penicillin G                   | 中国           | 污泥+制药废弃物             | 1 100 000   | 100                          | [61]                        |
|               | 青霉素 G 钠盐<br>Penicillin G sodium salt | 中国           | 粪污                   | 973.95  | 80.27                        | [13]                        |
|               | 头孢菌素 C Cephalosporin C               | 中国           | 污泥+制药废弃物             | 1 180 400   | 6.58                         | [24]                        |
|               | 阿奇霉素 Azithromycin                    | 西班牙          | 污泥                   | 665~2 673   | 72.25~97.51                  | [55]                        |
|               | 克拉霉素 Clarithromycin                  | 中国           | 污泥+食品垃圾              | 24.9~74 100.0   | 90.6~97.6                    | [9]                         |
|               |                                      | 摩洛哥          | 污泥                   | 2 400   | 40                           | [19]                        |

续表1 抗生素在堆肥中消减情况

Continued table 1 The degradation of antibiotics during composting

| 类别<br>Class | 抗生素<br>Antibiotic  | 国家<br>Nation    | 堆肥原料<br>Raw material | 初始浓度<br>Initial concentration/<br>(ng·g <sup>-1</sup> ) | 降解率<br>Degradation<br>rate/% | 参考文献<br>Reference |      |
|-------------|--------------------|-----------------|----------------------|---|------------------------------|-------------------|------|
| 大环内酯类       | 红霉素 Erythromycin   | 中国              | 粪污+制药废弃物             | 18 430~136 230  | 99.87~99.96                  | [62]              |      |
|             |                    | 中国              | 粪污                   | 1 149~1 318   | 100                          | [39]              |      |
|             | 吉他霉素 Kitasamycin   | 中国              | 粪污+制药废弃物             | 815~2 320   | 95.26~100.00                 | [63]              |      |
|             | 罗红霉素 Roxithromycin | 中国              | 污泥+食品垃圾              | 38~63 100   | 67.5~95.9                    | [9]               |      |
|             | 替米考星 Tilmicosin    | 中国              | 粪污                   | 1 052.38~2 109.20                                       | 71.82~93.22                  | [12, 23]          |      |
|             | 泰乐菌素 Tylosin       | 中国              | 粪污                   | 769.45  | 89.05                        | [12]              |      |
|             |                    | 中国              | 制药废弃物                | 84.47   | 95.96                        | [64]              |      |
|             |                    | 中国              | 污泥+制药废弃物             | 1 029 000~1 031 000                                     | 99.52~99.89                  | [65]              |      |
|             |                    | 中国              | 食品垃圾+制药废弃物           | 87 200~3 008 600  | 99.6~100.0                   | [66]              |      |
|             |                    | 美国              | 粪污                   | 36.1~3 700.0  | 54~85                        | [20, 40]          |      |
| 林可酰胺类       |                    | 美国              | 粪污+污泥                | 2 300~2 800   | 96~98                        | [47]              |      |
|             |                    | 日本              | 粪污                   | 35~250  | 87.84~100.00                 | [67]              |      |
|             |                    | 加拿大             | 粪污                   | 11.76~40 900.00   | 66.67~99.00                  | [41~43]           |      |
|             | 克林霉素 Clindamycin   | 中国              | 粪污                   | 44.87   | 70.11                        | [13]              |      |
|             | 林可霉素 Lincomycin    | 中国              | 粪污                   | 3 428.5   | 93.75                        | [34]              |      |
|             |                    | 摩洛哥             | 污泥                   | 1 200   | 36                           | [19]              |      |
|             | 多肽类                | 拉沙菌素 Lasalocid  | 斯洛文尼亚                | 粪污  | 10 600                       | 100               | [68] |
|             |                    | 莫能菌素 Monensin   | 美国                   | 粪污  | 11 900                       | 54~76             | [40] |
|             |                    | 盐霉素 Salinomycin | 巴西                   | 粪污  | 3 000                        | 60~100            | [69] |
|             |                    |                 | 加拿大                  | 粪污  | 22 000                       | 99.8              | [70] |
| 硝基咪唑类       | 甲硝唑 Metronidazole  | 伊朗              | 粪污+制药废弃物+果蔬废弃物       | 16 021~95 631   | 100                          | [52]              |      |
| 苯尼考类        | 氟苯尼考 Florfénicol   | 美国              | 粪污+污泥                | 230~240   | 95~99                        | [47]              |      |
| 氨基糖苷类       | 庆大霉素 Gentamicin    | 中国              | 制药废弃物                |   | 92.0~97.2                    | [71]              |      |
| 环脂肽类        | 达托霉素 Daptomycin    | 意大利             | 制药废弃物                | 2 400   | >99.6                        | [5]               |      |
| 其他          | 三氯生 Triclosan      | 中国              | 污泥                   | 763.4~820.6   | 45.45~59.93                  | [72]              |      |

注:nd代表低于检测限;\*代表未降解。

Note: nd indicates nondetectable; \* indicates not degraded.

中得到有效降解,但相同堆肥处理中不同类别的抗生素以及同类抗生素在不同堆肥处理中的降解率差别很大,特别是部分试验处理中出现抗生素降解率低甚至基本没有降解的情况,这种差异可能是受到堆肥原�理化性质、抗生素浓度差异、堆体温度变化等因素影响。

## 2.2 堆肥过程中 ARGs 及移动基因元件(Mobile genetic elements, MGEs)消减情况

堆肥可以大幅降低沼渣<sup>[11]</sup>和城市固体垃圾<sup>[53]</sup>以及大部分畜禽粪污<sup>[6, 46]</sup>等有机固废中的 ARGs,但部分 ARGs,特别是 *sull* 在畜禽废弃物<sup>[73]</sup>和城市污泥<sup>[74]</sup>等多种有机固废堆肥中都很难清除,且堆肥腐熟期往往发生反弹<sup>[75]</sup>,导致消减效率较低甚至在堆肥后不降反升等问题<sup>[76]</sup>。

ARGs 大部分位于 MGEs 中<sup>[77]</sup>,其丰度变化受到 MGEs 的强烈影响<sup>[78]</sup>,*intI* 与 ARGs 丰度变化正相关<sup>[10]</sup>。在城市固体垃圾<sup>[53]</sup>、畜禽粪污<sup>[6, 46, 79]</sup>堆肥过程中,*intI* 的丰度均出现不同程度上升或始终保持在较高水平。堆肥原料及堆肥处理等因素对 *intI* 的消减都有影响,例如,鸡粪堆肥不能有效降低 *intI* 丰度<sup>[46]</sup>,但在鸡粪与中药渣共堆肥过程中 *intI* 相对丰度显著下降<sup>[80]</sup>。

*sull* 和 *intI* 被普遍认为是人源耐药性和降解效果的重要监测指标<sup>[6]</sup>,且存在共轭传播<sup>[12]</sup>,在各类堆肥中广泛存在,应予以重点关注。

## 3 堆肥过程中抗生素及 ARGs 消减的影响因素

### 3.1 堆肥原料

堆肥原料中的抗生素甚至制药废弃物<sup>[24, 60]</sup>中残

留的超高浓度抗生素往往并不影响堆肥腐熟,但会影响堆体菌群结构并导致 ARGs 丰度上升。如:青霉素制药废弃物堆肥过程中各理化指标均能达到腐熟标准,青霉素浓度也降低至检测限以下<sup>[60]</sup>,但堆肥后 ARGs 丰度明显上升<sup>[61]</sup>。加热预处理能够有效降低  $\beta$ -内酰胺类<sup>[81]</sup>、四环素类<sup>[82]</sup>、大环内酯类<sup>[83]</sup>等抗生素制药废弃物堆肥过程中的 ARGs 丰度<sup>[66]</sup>。

堆肥填充料对抗生素<sup>[58]</sup>及 ARGs<sup>[84]</sup>消减的作用较小。例如,木屑在堆肥中只起到改善堆体通透性的作用,基本不参与微生物作用<sup>[60]</sup>,堆肥填充料是否来自转基因作物对 ARGs 消减也没有明显影响<sup>[85]</sup>。但需要注意其本身存在的 ARGs 传播风险。如:将含有耐药菌(Antibiotic resistance bacterial, ARB)的玉米秸秆用作堆肥填充料,最终堆肥成品中 ARB 检出率甚至高于静态堆放处理<sup>[86]</sup>。

### 3.2 菌群结构

微生物菌群结构是影响堆肥中抗生素及其 ARGs 消减的主要因素<sup>[67,76]</sup>。厚壁菌门、变形菌门、拟杆菌门、放线菌门是最重要的 ARGs 潜在宿主菌门<sup>[12-13,87]</sup>,此外还有部分古菌(如嗜盐菌属<sup>[88]</sup>)。在具体堆肥过程中,潜在宿主菌除受原料及堆肥时期的影响外,在堆体不同深度的分布也有差异<sup>[89]</sup>。

### 3.3 理化性质

堆肥中适当增加碳(C)投入有利于 ARGs 消减<sup>[76]</sup>,C/N<sup>[90]</sup>、可溶性 S<sup>[53]</sup>、pH<sup>[53]</sup>对 ARGs 宿主菌都具有重要影响。pH 还会影响抗生素降解过程<sup>[52]</sup>,如青霉素在碱性堆肥中更易降解<sup>[91]</sup>。

堆肥产生的高温能够明显促进抗生素<sup>[14,47]</sup>及 ARGs 消减<sup>[52]</sup>,但温度并不是影响抗生素降解的直接因素,而是通过改变微生物菌群结构影响抗生素降解<sup>[67]</sup>。超高温堆肥去除 ARGs 的机理与传统堆肥不同,能够去除几乎全部的 MGEs,从而限制 ARGs 的水平转移,相比于传统堆肥更利于 ARGs 消减<sup>[77]</sup>。

总体来说,堆体理化性质,如 C、N、S 等元素含量及 pH、温度等都会影响堆肥过程中抗生素及 ARGs 消减,但这些因素的影响往往是间接的,通过改变堆肥过程中的微生物菌群发挥作用。

### 3.4 重金属

高浓度 Cu 含量会抑制磺胺类<sup>[48]</sup>、四环素类<sup>[36]</sup>抗生素降解,对于泰乐菌素<sup>[65]</sup>的降解影响较小。高浓度 Zn 则会导致堆体温度峰值降低和滞后<sup>[50]</sup>。

生物有效态 Cu、Zn 与 ARGs 丰度存在共轭性<sup>[76,92]</sup>,可能是因为 ARGs 和重金属耐药基因(Heavy

metal resistance genes, MRGs)有共同的宿主微生物<sup>[93]</sup>,高浓度的生物有效态 Cu、Zn 会对微生物的抗生素耐药性产生共同的选择压力<sup>[94]</sup>,引起部分 ARGs 丰度上升<sup>[42,58]</sup>,重金属钝化则利于 ARGs 消减<sup>[95]</sup>。

### 3.5 其他

有关抗生素在堆肥过程中消减的研究中,往往在堆肥材料(如畜禽粪污等)中人为添加抗生素以形成处理之间更高的浓度差,但人工添加至畜禽粪污中的抗生素与畜禽饲喂过程中给药后残留在粪污中的抗生素在堆肥过程中的降解规律及其对 ARGs 的影响并不完全一致。人工添加的磺胺类和大环内酯类抗生素在堆肥过程中降解更快,四环素类抗生素则在动物给药后排泄的粪污堆肥中降解更快<sup>[41]</sup>。另外,畜禽给药方式使 ARGs 宿主菌群受到选择压力时间更长,ARGs 也更不易消减<sup>[42]</sup>。总体来讲,人工添加抗生素的试验处理方式并不能准确反映生产条件下畜禽粪污堆肥过程,还应当进一步予以印证。

此外,堆肥时间<sup>[52]</sup>、含水率及翻堆频次<sup>[34]</sup>、堆肥标准化程度<sup>[15]</sup>都会对抗生素降解及 ARGs 消减产生显著影响。堆体不同部位的 ARGs 消减率也有所不同,堆体中心优于顶层,底层的消减率最低<sup>[7]</sup>,这可能是堆体不同部位的透气性、保温性等方面差异导致的。堆肥厂的空气环境同样显著影响腐熟堆肥中的 ARGs 丰度变化<sup>[96]</sup>,这可能与堆肥过程中抗生素挥发导致空气环境中存在一定的抗生素及 ARGs 有关<sup>[54]</sup>。

## 4 堆肥优化工艺对抗生素及其 ARGs 消减的影响

传统堆肥在抗生素降解(特别是其 ARGs 消减)方面仍然存在诸多局限性,在传统堆肥基础上进行优化和强化很有必要。常见的堆肥优化工艺见表 2。

### 4.1 堆肥管理

合理优化的堆肥工艺可以有效提高畜禽粪污处理效率,就抗生素降解效果来看,发酵罐堆肥的抗生素消减效果优于控制通风的条垛堆肥和静态堆制堆肥,可以在一周内达到商业堆肥标准<sup>[115]</sup>。持续通风优于间断通风和不通风<sup>[54]</sup>,负压通气方式比正压通气方式更利于 ARGs 和 *intI1* 的消除<sup>[117]</sup>,这可能与负压通气方式更利于减少引入空气中的抗生素及 ARGs 有关。半透膜覆膜堆肥也能够有效阻遏空气环境对腐熟堆肥中 ARGs 丰度反弹的促进作用,提升 ARGs 消减效果<sup>[96]</sup>。

### 4.2 添加外源微生物

在堆肥中加入外源微生物的主要方式有添加堆

表2 堆肥优化工艺

Table 2 Enhancing methods of composting

| 堆肥优化技术<br>Enhancing method | 优化措施<br>Enhancing measure | 国家<br>Nation         | 堆肥原料<br>Raw material                 | 参考文献<br>Reference  |
|----------------------------|---------------------------|----------------------|--------------------------------------|--|
| 外源微生物                      | 堆肥成品                      | 中国                   | 粪污<br>污泥                             | [37, 73]<br>[11, 77]                                       |
|                            | 酵熟填充料                     | 中国                   | 污泥                                   | [74]   |
|                            | 菌剂                        | 中国                   | 粪污<br>食品垃圾+制药废弃物<br>制药废弃物<br>污泥+食品垃圾 | [7, 16, 18, 34, 45, 88, 97-100]<br>[66]<br>[71, 78]<br>[9] |
| 添加剂                        | 生物炭                       | 中国                   | 粪污<br>污泥                             | [10, 17, 37, 75, 92, 101-104]<br>[95]                      |
|                            | 沸石                        | 中国                   | 粪污<br>污泥                             | [10, 101, 105]<br>[106]                                    |
|                            | 泥炭                        | 中国                   | 污泥                                   | [95]   |
|                            | 褐煤                        | 澳大利亚                 | 粪污                                   | [107]  |
|                            | 粉煤灰                       | 中国                   | 粪污                                   | [101]  |
|                            | 石灰                        | 中国                   | 粪污                                   | [33]   |
|                            | 建筑垃圾                      | 中国                   | 粪污                                   | [76]   |
|                            | 建筑垃圾(木制品和石膏)              | 加拿大                  | 粪污                                   | [89]   |
|                            | 红泥                        | 中国                   | 粪污                                   | [108]  |
|                            | 竹醋                        | 中国                   | 粪污                                   | [75]   |
|                            | 糖浆                        | 中国                   | 粪污                                   | [16]   |
|                            | 葡萄糖、蔗糖                    | 中国                   | 制药废弃物                                | [60]   |
|                            | 生物质粉                      | 中国                   | 粪污                                   | [109-110]  |
|                            | 大孔吸附树脂                    | 中国                   | 粪污                                   | [111]  |
|                            | 聚丙烯酸钠                     | 中国                   | 粪污                                   | [112]  |
|                            | 鼠李糖脂                      | 中国                   | 粪污                                   | [113]  |
|                            | 吐温 80                     | 中国                   | 粪污                                   | [113]  |
|                            | 纳米零铁                      | 中国                   | 粪污                                   | [114]  |
|                            | 过磷酸钙                      | 中国                   | 粪污                                   | [105]  |
|                            | 3,4-二甲基吡唑磷酸盐              | 中国                   | 污泥                                   | [106]  |
| 堆肥管理                       | 堆肥方式                      | 中国                   | 粪污                                   | [115]  |
|                            | 含水率                       | 伊朗<br>美国             | 粪污+制药废弃物+果蔬废弃物<br>粪污                 | [52]<br>[40]   |
|                            | 高温氧化预处理                   | 中国                   | 食品垃圾+制药废弃物                           | [66]   |
|                            | 加热预处理                     | 中国                   | 粪污+制药废弃物<br>污泥+制药废弃物                 | [81]<br>[116]  |
|                            | 超高温                       | 中国                   | 制药废弃物                                | [64]   |
|                            | 通气                        | 美国<br>中国             | 粪污<br>粪污<br>污泥                       | [20, 40]<br>[79, 117]<br>[54, 72, 77]                      |
|                            | 半透膜覆盖                     | 中国                   | 污泥                                   | [59]   |
|                            | 覆草                        | 美国                   | 粪污                                   | [96]   |
|                            | 原料配比                      | 伊朗<br>南非<br>美国<br>中国 | 粪污<br>粪污<br>粪污<br>粪污                 | [30]<br>[22]<br>[32]<br>[30]                               |
|                            |                           | 中国                   | 粪污                                   | [84, 93, 118]  |
|                            |                           | 中国                   | 污泥+制药废弃物                             | [58]   |
|                            |                           | 中国                   | 制药废弃物                                | [60]   |
|                            |                           | 中国                   | 污泥                                   | [74]   |
|                            |                           | 中国                   | 粪污+制药废弃物                             | [81-82]  |

肥成品<sup>[73]</sup>、添加配制菌剂等。引入外源菌剂能够改变堆体微生物群落结构,提高堆肥温度峰值,延长堆肥高温期<sup>[7,73]</sup>。研究证实,适当添加外源菌剂对于抗生素及ARGs的消减具有明显促进作用<sup>[97,16]</sup>,在堆肥过程中采用的菌种有黄孢原毛平革菌、地衣芽孢杆菌、黑曲霉<sup>[16]</sup>、土曲霉<sup>[71]</sup>、枯草芽孢杆菌<sup>[98]</sup>、木质素降解菌<sup>[99]</sup>等,复合菌剂施用效果往往优于单一菌剂<sup>[45]</sup>。

#### 4.3 堆肥添加剂

堆肥添加剂对于ARGs消减的影响已经成为目前堆肥优化技术研究热点之一,前人对生物炭和沸石<sup>[10]</sup>、泥炭<sup>[95]</sup>、虾壳粉<sup>[109]</sup>、玉米穗轴<sup>[76]</sup>、褐煤<sup>[107]</sup>、竹醋<sup>[75]</sup>、纳米零价铁<sup>[114]</sup>、铁基材料和溶磷剂<sup>[100]</sup>、生物表面活性剂(鼠李糖脂)和化学表面活性剂(吐温80)<sup>[113]</sup>、过磷酸钙<sup>[105]</sup>、砖粒<sup>[76]</sup>、红泥<sup>[108]</sup>、3,4-二甲基吡唑磷酸盐(DMPP)等不同材料对堆肥过程中ARGs消减的影响进行了研究。除砖粒、红泥和DMPP外,大部分材料对ARGs消减有一定积极意义。

生物炭类添加剂虽然广受关注,但其对于ARGs消减的作用尚不明确。大部分研究认为添加生物炭(如竹炭<sup>[37]</sup>)对于堆肥过程中ARGs的消减有促进作用<sup>[75,101]</sup>,但有研究发现,稻草生物炭会促进部分畜禽粪污<sup>[92,102]</sup>堆肥中ARGs丰度增加,蘑菇生物炭能降低鸡粪<sup>[92]</sup>和猪粪<sup>[102]</sup>堆肥中ARGs丰度,但会促进鸭粪<sup>[102]</sup>堆肥中ARGs丰度增加。也有研究认为,生物炭添加比例是影响堆肥过程ARGs消减的主要因素,而非生物炭的种类<sup>[103]</sup>。

不可逆吸附是抗生素在堆肥过程中消减的重要途径之一<sup>[54]</sup>,在堆体中添加适量的大孔吸附树脂<sup>[111]</sup>、沸石<sup>[106]</sup>等多孔结构材料,有利于增强堆体物理吸附能力,促进ARGs和MGEs的消减。

### 5 展望

#### 5.1 筛选堆肥复合菌剂

目前,已有部分研究对堆肥菌剂进行了探索,并在促进抗生素及其ARGs消减方面取得一定成果,但相关菌种资源挖掘还比较有限,今后的研究中应当进一步筛选相关高效功能菌,明确不同菌种之间的协同及拮抗作用,探究能够同时有效去除多种抗生素及ARGs的复合菌剂。

#### 5.2 重视规模堆肥研究

目前,抗生素及其ARGs在堆肥过程中的消减规律研究以实验室水平及中试水平堆肥试验为主,多以人工添加抗生素的方式形成处理间的浓度梯度,但抗

生素及ARGs在工业水平堆肥中的消减规律与模拟堆肥及小规模堆肥试验中并不完全一致,往往受到更多因素影响,人工添加到粪污等堆肥材料中的抗生素与动物给药后残留在粪污中的抗生素消减规律及对ARGs的诱导效应也有差异,今后的研究应在实验室及中试试验基础上,更加重视在工业水平堆肥中对相关规律的验证,提高相关成果对于生产实践的指导意义。

#### 5.3 优化堆肥技术

目前,堆肥对于病原微生物的杀灭作用及为作物提供稳定养分等方面的作用已经得到普遍认同,但在降低耐药风险方面的研究尚不成熟,也未能形成完整的技术体系。已经有研究分析了理化指标、微生物菌群结构、重金属、堆肥管理、空气环境及添加剂等诸多因素对堆肥过程中抗生素及ARGs的影响,今后的研究中还应加强对各类影响因素的整合分析,逐步形成针对不同原料的堆肥技术体系。

#### 5.4 深挖分子机理

目前,关于抗生素及ARGs在堆肥过程中消减规律的研究大多是对其在具体试验处理中的消减特征进行表述,并通过相关性数据分析理化指标、微生物群落结构、重金属等因素对抗生素浓度及ARGs丰度变化的贡献度,但不同堆肥试验中抗生素及ARGs消减效果往往差别较大,其具体分子机理尚不明确,今后的研究中还应该进一步深挖相关分子机理并逐渐完善理论基础,提高相关研究的针对性。

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