

Progress in Additives That Promote Humification During Agricultural Waste Composting

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Abstract

Aerobic composting converts agricultural waste into stable, humus-rich products, and the application of exogenous additives is an efficient strategy to enhance humification. This systematic review synthesized studies published between 2023 and 2026 on additives that promote humification during agricultural waste composting. Based on the distribution of the retrieved literature, additives are categorized into inorganic additives, organic additives, biological strategies, and composite systems, and the effects and mechanisms of each category are systematically discussed. Iron-based additives achieve the highest humic acid (HA) increases of 82–267% through Fenton-like redox catalysis. Clay minerals and biochar produce moderate HA enhancements of 25–163% via physical structuring and surface adsorption with broader applicability. Small-molecule precursors and exogenous humic substances achieve HA gains exceeding 100% at sub-percent doses. Biological strategies provide self-sustaining catalytic activity but are sensitive to environmental conditions. Composite additives, the largest category, generally outperform single additives through functional complementarity, though antagonistic effects have also been documented. Cross-study patterns suggest that different feedstocks respond preferentially to distinct additive types, though systematic experimental validation is lacking. Critical gaps between laboratory findings and practical application are identified, including the predominance of small-scale studies, the absence of techno-economic analysis, and the unassessed environmental fate of metal-based additives. Future research priorities include pilot-scale validation under industrial conditions, the establishment of standardized humification metrics, and long-term field monitoring.

Keywords: aerobic composting; additives; humification; agricultural waste; humic acid



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1. Introduction

Intensified agricultural production has led to the massive generation of agricultural waste, including crop straw and livestock manure [1]. Global annual livestock manure production exceeds 125 million tons, while lignocellulosic waste totals ~140 billion tons, accounting 30–40% of total solid waste [2]. Improper disposal of these wastes poses substantial environmental risks, including groundwater contamination, greenhouse gas emissions, and pathogen proliferation. Current treatment methods for agricultural waste include direct return to soil, anaerobic digestion, feed utilization, and aerobic composting. Among these technologies, aerobic composting is a cost-effective biological treatment

technology that relies on microorganisms to convert organic matter into stable, sanitized humus [3]. This process reduces waste volume and associated hazards while generating humus that improves soil structure, enhances soil fertility, immobilizes heavy metals, and delivers significant ecological and economic benefits.

Humification refers to the process by which labile organic matter is transformed into stable, chemically complex humic substances (HSs) via biotic and abiotic pathways [4]. HSs are primarily composed of humic acid (HA), fulvic acid (FA), and humin. HA is soluble under alkaline but not acidic conditions ($\text{pH} < 2$), and it possesses a high molecular weight and aromatic core. It is enriched in oxygen-containing functional groups such as carboxyl, phenolic hydroxyl, and quinone moieties, which confer cation exchange capacity and metal-chelating properties [3]. FA is soluble across the entire pH spectrum and has a lower molecular weight, serving as an intermediate in the humification process [5]. Humin is insoluble at any pH and constitutes the most recalcitrant HS fraction. The content and structural characteristics of HSs, particularly the HA/FA ratio and the degree of aromatic condensation, are core indicators of compost maturity and quality [6]. Consequently, enhancing the conversion of organic matter into HSs is therefore of central importance for improving composting efficiency and end-product value.

However, conventional composting is often constrained by prolonged processing time, low humification efficiency, and insufficient product maturity, which restrict large-scale deployment and limit commercial viability. Numerous studies have demonstrated that exogenous additives can optimize the composting process and improve product quality. For instance, attapulgit at 5–7.5% increased humic acid content by 163% in cabbage waste composting [7]. Bamboo biochar applied at 10% elevated average humic acid by 93% relative to the control in pig manure composting [8]. Magnetite at 5% enhanced humic acid content by 267% while reducing fulvic acid by 64% in pig manure–wolfberry branch composting [9]. Inoculation of *Phanerochaete chrysosporium* at the cooling stage promoted humic substances accumulation by 82% during corn straw–canola residue co-composting [10]. Catechol amendment at 0.5% boosted humic substances by 123% and humic acid by 100% in chicken manure–corn straw co-composting [11].

Several reviews have previously addressed the role of additives in composting. Chen et al. [12] provided a comprehensive overview of inorganic, organic, biological, and composite additives for humification enhancement, synthesizing studies published up to 2022. Other reviews have focused on specific additive subcategories, including iron-based additives [13], biochar [14], and amendments for green waste composting [15]. Since 2023, however, the field has undergone rapid advances across five key areas: (1) application of synthetic microbial communities for targeted lignocellulose degradation and humus synthesis [16]; (2) use of metabolic regulators such as malonic acid to redirect carbon flux toward humus formation rather than mineralization [17]; (3) elucidation of free-radical-mediated humification pathways driven by Fenton-like reactions [9]; (4) development of novel composite additives integrating minerals, biochar, and microbial inoculants with synergistic effects [18]; and (5) emergence of pilot- and field-scale validation studies offering actionable insights for full-scale composting operations [19]. Notably, these recent developments, particularly studies published between 2023 and 2026, have not yet been systematically synthesized in a dedicated review focused exclusively on agricultural waste composting.

Accordingly, this systematic review aims to undertake the following: (1) provide a PRISMA-guided synthesis of recent advances (2023–2026) in the application of inorganic, organic, and biological additives to promote humification during agricultural waste composting; (2) critically compare additive performance using normalized indicators and identify factors underlying divergent outcomes across studies, including feedstock

type, additive dosage, and process conditions; (3) establish a feedstock-additive matching framework to guide practical additive selection across diverse composting scenarios; and (4) evaluate the practical applicability of various additives in terms of cost-effectiveness, scalability, environmental fate, and agronomic performance. By addressing these objectives, this review provides timely, actionable insights that transcend previous surveys and support the rational design of additive strategies to enhance compost humification.

2. Methods

2.1. Search Strategy and Eligibility Criteria

This systematic review was conducted and reported in accordance with the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) 2020 guidelines [20]. A systematic literature search was performed across Web of Science Core Collection and PubMed databases for English-language articles published between January 2023 and June 2026. The following search string was applied: (compost OR composting) AND (humification OR “humic acid” OR “humic substances” OR humus) AND (additive OR additives OR addition OR amendment) AND (“agricultural waste” OR straw OR manure OR “crop residue” OR lignocellulosic). The complete PRISMA 2020 checklist is provided in Supplementary Material S1.

Studies were eligible for inclusion if they met the following criteria: (i) investigated aerobic composting of agricultural waste (crop straw, livestock manure, or their mixtures); (ii) examined the application of one or more exogenous additives (inorganic, organic, or biological) during composting; and (iii) reported quantitative humification-related indicators, including humic acid (HA), fulvic acid (FA), humus content, the HA/FA ratio, or the humification index. Studies were excluded if they met the following criteria: (i) focused on non-agricultural feedstocks (e.g., municipal solid waste, sewage sludge, food waste, or industrial waste) as the primary substrate; (ii) involved anaerobic digestion, vermicomposting, or other non-aerobic processes; (iii) lacked quantitative humification data or mechanistic analysis; (iv) examined only process parameters (e.g., aeration, moisture, C/N ratio) without additive intervention; or (v) were review articles, conference abstracts, retracted publications, or duplicate records.

2.2. Study Selection and Data Extraction

The initial search yielded 1139 unique records (630 from Web of Science and 509 from PubMed). Following the removal of 138 duplicates, 1001 records were screened by title and abstract, of which 584 were excluded based on the predefined eligibility criteria. Full texts of the remaining 417 records were retrieved for detailed evaluation; seven records were inaccessible through available subscriptions and were excluded. Of the 410 full-text articles assessed, 279 were excluded for the following reasons: non-target agricultural waste substrate or pollution-remediation focus ($n = 113$); humification or humic substance formation was not the main research focus ($n = 105$); no additive, amendment, pretreatment, or bioaugmentation intervention ($n = 27$); non-composting process or primary focus on compost/soil application and crop response ($n = 26$); non-aerobic or non-traditional composting process ($n = 5$); and ineligible record type including duplicates, retraction notices, or review articles ($n = 3$). Ultimately, 131 studies were included in the final synthesis. The study selection process is summarized in Figure 1 (PRISMA flow diagram).

Two independent reviewers (Q.Z. and J.L.) screened titles, abstracts, and full texts. Disagreements were resolved through discussion with a third senior reviewer (J.L.). Data extracted from each included study comprised: additive type and dosage; composting feedstock and scale (laboratory, pilot, or field); composting duration; quantitative humification indicators (HA, FA, HS content, HA/FA ratio); and key mechanistic findings.

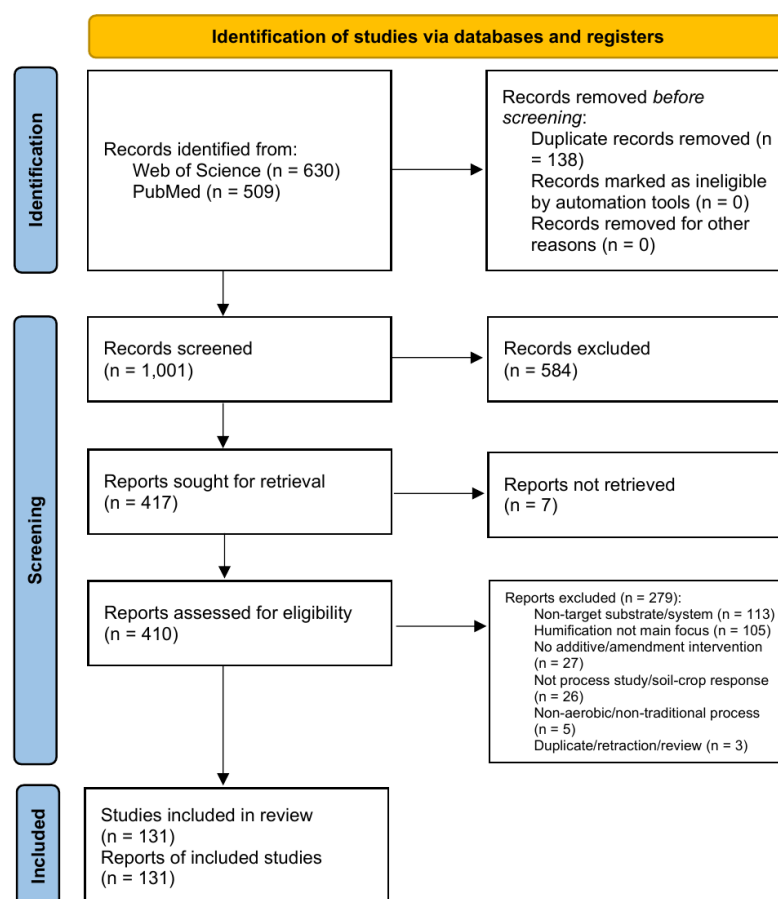


Figure 1. PRISMA flow diagram.

2.3. Quality Assessment

Given the substantial heterogeneity among the included studies—spanning diverse feedstocks, additive types and dosages, composting scales, and operational conditions—no standardized risk-of-bias assessment tool was directly applicable. Instead, methodological heterogeneity was addressed qualitatively throughout the synthesis by explicitly considering differences in study scale (laboratory, pilot, or field), additive dosage, composting duration, and analytical methods when comparing results across studies. This approach ensures that the conclusions drawn account for the inherent variability among the reviewed experiments.

3. Inorganic Additives

3.1. Iron-Based Additives

Iron-based additives represent a substantial proportion of inorganic strategies and include metallic iron (Fe(0)), iron oxides (Fe₂O₃, Fe₃O₄), nano-zero-valent iron (nZVI), and iron-based composite materials (FeS₂/palygorskite, magnetic LDO beads). These materials promote humification through two principal mechanisms: redox catalysis with free radical generation, and iron–organic matter complexation with adsorptive stabilization. Figure 2 illustrates the Fe(II)/Fe(III)-driven Fenton-like cascade: Fe²⁺ reacts with H₂O₂ generated during composting to produce ·OH, which attacks lignocellulose and releases phenolic humus precursors that subsequently condense into humic acid.

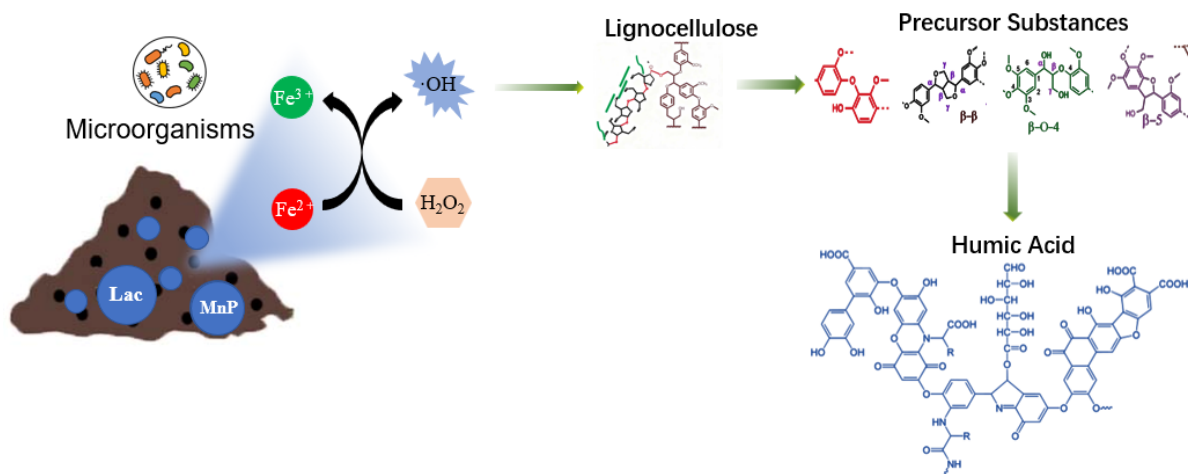


Figure 2. Iron-catalyzed humification via Fenton reactions.

Iron-based additives can promote humification through redox-catalyzed free radical generation. Fe_2O_3 , Fe_3O_4 , and related species exploit $\text{Fe(II)}/\text{Fe(III)}$ valence cycling to catalyze Fenton-like reactions that drive $\cdot\text{OH}$ production, attack lignocellulosic structures, release phenolic humus precursors, and facilitate aromatic polymerization. In pig manure–wolfberry branch composting, inoculation with 5% magnetite increased HA by 267%, decreased FA by 64%, and yielded an HA/FA ratio of 2.25; the mixed $\text{Fe(II)}/\text{Fe(III)}$ sites in its spinel structure sustain continuous electron cycling [9]. In the same feedstock, inoculation with 5% nZVI increased HA by only 82%, likely because nZVI oxidation consumes protons and shifts pH away from the optimal range for Fenton chemistry [21]. In cattle manure and food waste systems, inoculation with 1% $\alpha\text{-Fe}_2\text{O}_3$ increased HS by 9–10% via $\text{Fe}^{3+}/\text{Fe}^{2+}$ cycling that catalyzes aromatic polymerization [22]. In terms of composite materials, the combined application of 2% palygorskite-loaded FeS_2 and 2% glutathione in dairy manure–corn straw composting exploited the enzyme-like activity of FeS_2 to catalyze electron transfer, raising the humification degree (DP) from 0.79 to 1.18 and achieving a germination index of 136% [23]. Redox catalysis also exerts downstream effects on microbial communities: both magnetite and $\alpha\text{-Fe}_2\text{O}_3$ treatments enriched *Bacillus*, *Pseudomonas*, and *Actinobacteria*—functional groups associated with aromatic condensation [9,22]. A comparative study of Fe(0) , Fe_2O_3 , and Fe_3O_4 further demonstrated that Fe_2O_3 treatment increased HA by approximately 14.9% with enhanced PARAFAC humic acid-like components, showing the best overall performance among the three iron species [24].

Furthermore, iron ions in these additives can complex or co-precipitate with carboxyl and phenolic hydroxyl groups on humus surfaces, forming a protective layer on humus molecules that enhances their resistance to microbial degradation while simultaneously achieving heavy metal passivation. In chicken manure–grape pomace composting, Fe(0) , Fe_2O_3 , and Fe_3O_4 at 1% equivalent iron loading not only increased HA by approximately 15% but also promoted synergistic Cu/Zn passivation through HA–metal complexation [25]. Magnetic LDO beads improve humus formation and DOM aromatization by providing adsorption and metal fixation sites [26].

The outstanding advantage of iron-based additives lies in the effect magnitude driven by chemical catalysis: magnetite at its optimal dose achieved a 267% HA increase, the highest in this dataset. However, attention must be paid to optimal dosing: at 7.5%, the HA enhancement of magnetite dropped from 267% to 89%, and that of nZVI fell from 82% to 81%, indicating that oxidative stress from excess iron counteracts catalytic gains [9,21]. Moreover, all seven studies were conducted at laboratory scale, with no pilot validation

or cost analysis available, limiting the translation of iron-based additives from laboratory findings to practical composting operations.

3.2. Clay Minerals

The porous structure of clay minerals enhances compost porosity and oxygen diffusion, promoting aerobic microbial metabolism and organic matter degradation. In mushroom waste–cattle manure composting, the addition of 9% medical stone—a sponge-like porous silicate—increased HA from 40.5 g/kg to 48.6 g/kg and raised HA/FA from 1.58 to 2.16 [27].

The high cation exchange capacity and swelling properties of clay minerals enable them to adsorb and retain water, NH_4^+ , and dissolved organic precursors within the compost matrix, reducing volatilization and leaching losses. In a direct comparison of vermiculite, zeolite, medical stone, bentonite, and magnetite at 10% each in swine manure composting, bentonite achieved the largest HA increase (45%) and the highest HA/FA improvement (94%), attributed to its superior water absorption, swelling capacity, and nutrient retention. Zeolite, with its high cation exchange capacity and molecular-sieving micropores, preferentially adsorbs NH_4^+ and preserves nitrogenous precursors for humus synthesis, though its overall humification performance was less pronounced than that of bentonite [28]. Sepiolite and palygorskite, both magnesium-rich chain silicates, were evaluated individually and in combination at 5% in chicken manure–rice straw composting; both minerals enhanced humus aromaticity and stability, as confirmed by ^{13}C NMR, FTIR, and EEM fluorescence spectroscopy, with combined application producing additive effects and additional heavy metal passivation through surface complexation [29]. The germination index concurrently improved from 82% to 97%, indicating that enhanced aeration benefits both humification and overall compost maturity. In cabbage waste–corn straw composting, attapulgite at 5–7.5% increased HA by 130–163% and raised the humification index from 9.7 to 11.1; HA continued to rise with dose up to 7.5% [7].

Furthermore, the porous layered surfaces of clay minerals provide attachment sites and protective microhabitats for microorganisms, supporting the proliferation of functional communities involved in lignocellulose degradation and humification. The addition of 3–9% medical stone enriched Firmicutes, Chloroflexi, and *Streptomyces*—populations directly associated with lignocellulose decomposition and FA-to-HA conversion [27]. The multi-mineral comparison study found that bentonite amendment enriched cellulolytic bacteria [28]. Sepiolite and palygorskite altered DOM spectral properties through the combined effects of adsorption and microbial community modulation [29].

Clay minerals offer several practical advantages: they are widely available, low in cost, effective across diverse feedstock types, and free of the oxidative stress risks associated with redox-active metal additives. Their principal limitation is the moderate effect magnitude compared with catalytic additives—HA increases of 30–163% versus 82–267% for iron—reflecting the intrinsic ceiling of their indirect mechanism. Optimal doses cluster at 5–10%, higher than those of iron-based additives, which may increase material handling requirements at large scale.

3.3. Other Inorganic Amendments

Beyond iron-based catalysts and clay minerals, various other inorganic materials have been evaluated for promoting humification during composting, including phosphate-based salts, industrial byproducts, and natural materials.

Phosphate-based additives promote humification through dual mechanisms of nutrient supplementation and abiotic condensation. In pig manure–rice straw composting, the addition of a $\text{Ca}(\text{H}_2\text{PO}_4)_2$ and MgSO_4 composite retained nitrogenous humus precursors through $\text{Mg}^{2+}/\text{PO}_4^{3-}$ -mediated struvite formation, while enriching *Thermomyces*

and *Aspergillus* fungi to enhance lignocellulose degradation and HA formation, with a 19.6% increase in 3D-EEM Region V [30]. In chicken manure–corn straw composting, a comparative study of superphosphate and fly ash at 10% each demonstrated that fly ash outperformed superphosphate, increasing HA by 83% and raising HI from 1.0 to 2.9, doing so through enhanced microbial diversity and Chloroflexi enrichment that promoted the conversion of protein-like components into humic acid-like substances; superphosphate acted primarily through Ca^{2+} -mediated abiotic precursor condensation [31].

Industrial byproducts offer the combined advantages of low cost and waste valorization. High-carbon coal gangue applied at 10% in cattle–sheep manure composting with furfural residue achieved an HS content of 133 g/kg and an HA/FA ratio of 5.6, with *Planifilum* identified as the keystone genus coupling microbial activity to HA formation [32]. In terms of natural materials, the addition of 10% soil (three soil types) to wheat straw–cattle manure composting increased HA by 28–36%, attributed to improved aeration and the enrichment of lignocellulose-degrading bacteria such as *Corynebacterium*, *Acinetobacter*, and *Chaetomium*, although the 10% loading increases total compost mass and transport costs [33]. Selenium, applied as Na_2SeO_3 , Na_2SeO_4 , or endogenous organic Se in spent mushroom substrate composting, enriched Chloroflexi, enhanced DOM–bacterial interactions and aromatic structure formation, with 66–73% of total Se ultimately incorporated into the HA fraction [34].

3.4. Comparative Synthesis of Inorganic Additives

The mechanisms by which inorganic additives promote humification can be categorized into three types. Chemical catalysis is exemplified by the redox reactions of iron-based additives, which drive Fenton-like reactions through Fe(II)/Fe(III) valence cycling, achieving the strongest effects but requiring precise dosage ranges. Clay minerals act through physical mechanisms via their porous layered structures, indirectly promoting humification by improving aeration, retaining water, and providing microbial habitats, offering broad applicability but yielding moderate effects. Nutrient supplementation and abiotic condensation, represented by phosphate-based salts, drive humus formation through nitrogen retention, ion complexation, and Ca^{2+} -mediated precursor condensation. Notably, these mechanisms are not mutually exclusive. Iron-based additives simultaneously contribute to heavy metal passivation through iron–organic matter complexation while exerting their catalytic function, and clay minerals alter DOM transformation pathways through surface adsorption beyond their physical regulation role.

Effect magnitude is closely tied to mechanism type. Iron-based additives achieve the highest HA increases, ranging from 82% to 267%, with the lowest optimal doses of 1% to 5%, though they carry oxidative stress risks at elevated doses—both magnetite and nZVI showed marked decline in efficacy at 7.5% [9,21]. Clay minerals produce more moderate HA increases, between 30% and 163% [7,28], requiring higher optimal doses of 5% to 10%, yet they exhibit no dose inhibition and greater feedstock versatility, with attapulgite performing notably in cabbage waste and bentonite excelling in swine manure systems [7,28]. Phosphate-based salts and industrial byproducts occupy an intermediate position: fly ash increased HA by 83% [31], while coal gangue achieved an HS content of 133 g/kg and an HA/FA ratio of 5.6 [32], highlighting the potential of waste valorization in compost humification.

Two notable shortcomings characterize the current literature. All studies were conducted at laboratory scale, and the performance of optimized doses and proposed mechanisms under industrial-scale composting conditions remains unvalidated. Furthermore, no study has systematically varied feedstock type while holding additive type constant. The current understanding that iron-based additives are better suited to lignocellulose-rich

substrates and clay minerals to protein-rich manure rests on cross-study inference rather than direct experimental evidence.

4. Organic Additives

4.1. Biochar and Carbon-Based Materials

Biochar is a carbon-rich, porous material produced by the pyrolysis of biomass under oxygen-limited conditions, and is the most extensively studied organic additive. Lignite and hydrochar, as biochar-like carbon-based materials, also exhibit humification-promoting effects to a certain degree. These materials promote humification primarily through two mechanisms: physical structure optimization and surface chemistry with adsorption.

The porous structure and large specific surface area of biochar improve compost aeration and water retention, while providing attachment surfaces and microhabitats for microorganisms. In pig manure–corn straw co-composting, 10% corn stalk biochar increased HA/FA by 67% over the control, attributed to its larger specific surface area and pore volume, while its carboxyl-rich surface facilitated FA degradation and conversion to HA [35]. Bamboo biochar increased average HA by 93% over the control in pig manure composting, outperforming wheat straw biochar and attributed to richer surface functional groups [8]. An interaction exists between biochar source and composting substrate. In sheep manure composting, walnut shell biochar most effectively preserved organic matter by adsorbing and stabilizing small-molecule precursors, while in Chinese herb residue composting, corn cob biochar was more effective at stimulating microbial conversion of macromolecular organics into HA due to its larger total pore volume [36]. Regarding dosage, 10% biochar addition produced the optimal effect, while 20% caused suppression accompanied by a shift in dominance from Firmicutes to Proteobacteria [37,38]. Improvements in physical structure also influence microbial community changes. Biochar treatment enriched lignocellulose-degrading fungi (e.g., Basidiomycota, *Mycothermus*) and bacteria (e.g., *Chryseolinea*), enhanced cellulase and urease activities, and improved bacterial–fungal cross-kingdom network stability [39,40].

Functional groups on biochar surfaces mediate the adsorption and retention of precursors and nutrients. Oxygen-containing functional groups ($-OH$, $-COOH$, $C-O$) can adsorb NH_4^+ and dissolved organic precursors, reducing volatilization and leaching losses while participating in humus polymerization reactions. In a 500 kg (dry weight) pig manure–mulberry branch composting trial, 6% biochar increased FA and HA production efficiency by 17% and 39%, respectively, through the selective regulation of fungal communities and CAZyme gene expression, enhancing quinone biosynthesis and aromatic amino acid metabolism [41]. Microbial necromass carbon (MNC) is a quantitatively important contributor to humus formation. In chicken manure–mushroom residue composting, MNC accounted for 6–21% of TOC, and biochar addition increased MNC accumulation by 29%, with HA carbon significantly and positively correlated with both bacterial ($R^2 = 0.72$) and fungal ($R^2 = 0.51$) necromass carbon [42]. In terms of phosphorus cycling, biochar promoted the formation of humic acid–metal–phosphate complexes, increasing available phosphorus by 60% [43].

Lignite and hydrochar are two notable biochar-like carbon-based materials. Lignite contains approximately 14% native HA, serving dual functions as both a humus precursor source and a microbial carrier. In chicken manure–rice husk composting, 5–15% lignite increased HA by up to 45% and shifted the dominant humification pathway from sugar–amino acid condensation toward a polyphenol-driven route, mediated by enhanced laccase and polyphenol oxidase activity [44,45]. In goat manure–corn straw composting, 10% lignite increased the germination index by 68% and enhanced bacterial network complexity by 47% [46]. Hydrochar, produced by hydrothermal carbonization at 220 °C, carries acidic

oxygen-containing functional groups that lower compost pH and adsorb NH_4^+ . In chicken manure–corn straw composting, 10% hydrochar reduced NH_3 and N_2O emissions by 55% and 45%, respectively, increased HS by 11%, and accelerated the time to GI exceeding 80% by 10 days, with nitrogen retention effects associated with *amoA* upregulation and downregulation of denitrification genes (*nirK*, *nirS*, *nosZ*) [47].

4.2. Agricultural Byproducts and Organic Co-Substrates

Beyond purpose-produced additives such as biochar, a wide range of agricultural byproducts and organic residues characterized by on-farm availability and low cost have been evaluated as humification-enhancing co-substrates. These materials serve dual functions of physical structure improvement and chemical precursor supply.

Certain byproducts participate in humus synthesis by directly supplying humification precursors such as amino acids and polysaccharides. Monosodium glutamate residue, rich in amino acids and peptides, applied at 15% (dry weight) in chicken manure–spent mushroom substrate composting, increased HA to 89.1 g/kg and enhanced protease, urease, cellulase, and peroxidase activities, with *Pelagibacter*, *Caldibacillus*, and *Nitrateductor* identified as key functional genera [48]. Waste chicken feathers pretreated with *Chryseobacterium indologenes* and added at 1% released free amino acids, yielding terminal HA of 71.0 mg/g and enriching *Bacillus* and *Paenibacillus* [49]. When mushroom residues were co-composted with nitrogen-rich supplements such as chicken manure, the humification level exceeded that achieved with cattle manure or urea, with HS reaching 35.4% and HI reaching 22.2%; the nitrogen-rich sources promoted the conversion of protein-like and polysaccharide-like DOM into humic substances by increasing TN/NO_3^- [50]. Replacing 30% of spent mushroom substrate with cattle manure (7:3 *w/w*) doubled terminal HA (49.5 vs. 28.3 g/kg), directly demonstrating that cattle manure-derived polysaccharides serve as quantitatively important humus precursors [51]. Flax retting liquid, used as a moisture-regulating supplement, improved the HA/FA ratio (1.28 vs. 1.05 in the control) [52].

Another category of byproducts indirectly enhances humification by improving the physical structure of the compost matrix and providing microbial inoculum sources. The addition of 10% (wet weight) cooling-stage compost to cattle manure–corn straw composting increased HA to 71.5 g/kg and DP to 3.64, outperforming compost harvested at the thermophilic or maturation stages [53]. Mature compost (15%) combined with semi-permeable membrane covering increased HS by 45%, with *Myceliophthora*, *Kernia*, and *Mycothermus* identified as keystone fungi [54]. Agricultural and forestry wastes, used as structural conditioners at pilot scale, shifted organic matter transformation from labile components toward stabilized lignin and humus [55]. The addition of 25% corn straw or 40% sheep manure to vegetable waste composting increased HA by 32–49% by improving aeration and supplying lignocellulose-degrading microbiota [56]. Garden waste applied at 15% (fresh weight) increased HA by 35%, with 52% of the total HA increase occurring within the first 10 days [57].

In addition, certain specialized byproducts exhibit distinctive modes of action. The *hydroxylapatite* in cow bone powder promoted humic acid-like component enrichment through adsorption, ion exchange, and pH buffering, reducing Cu and Zn mobility by 10% and 19%, respectively, and eliminating metal resistance gene abundance [58]. Tobacco powder at 15% (wet weight) enhanced humification and reduced greenhouse gas emissions by more than 75%, although doses exceeding 22.5% increased salinity and phytotoxicity [59]. Compared with purpose-produced additives, the agricultural byproducts described above are low in cost and available on-farm, yet quantitative cost-effectiveness comparisons remain absent from the literature.

4.3. Humification Precursors and Small Organic Molecules

The direct addition of reactive small molecules that participate in humification reactions or regulate metabolic pathways represents a targeted strategy for enhancing humification. These compounds can either directly enter humification condensation pathways as reaction participants, or indirectly promote humus accumulation by altering microbial carbon flow direction or community function. Figure 3 depicts the two principal humus formation pathways. The lignin–protein pathway proceeds through the depolymerization of lignin and proteinaceous macromolecules into reactive intermediates that recombine into humic substances, while the Maillard pathway condenses reducing sugars with amino compounds. Both pathways are modulated by microbial activity and composting parameters including feedstock type, the C/N ratio, pH, temperature, and aeration, all of which can be influenced by additive addition.

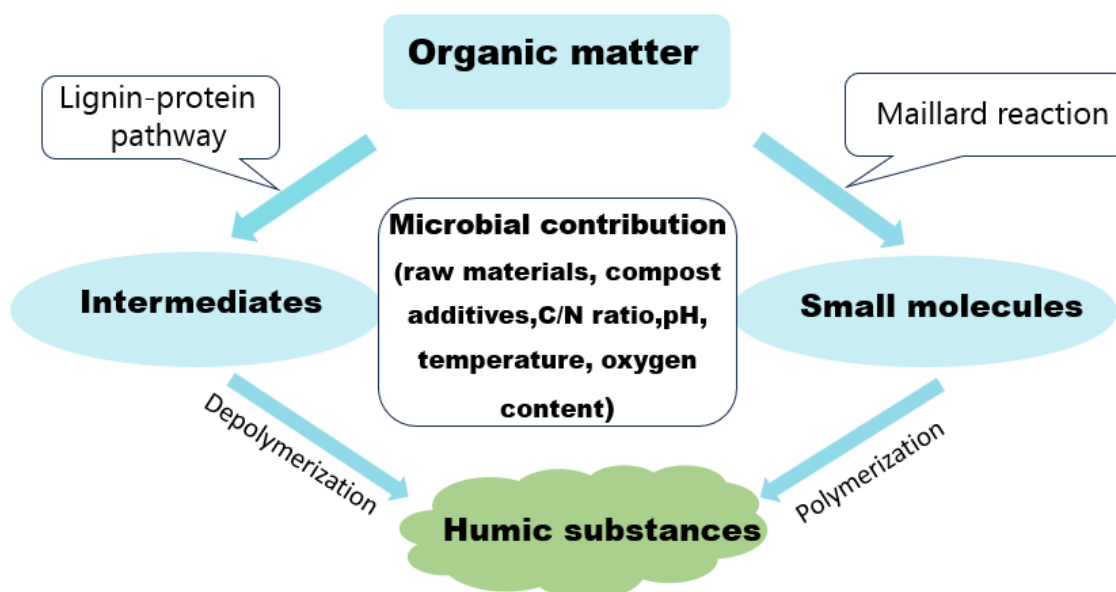


Figure 3. Dual pathways of humus formation.

Catechol, gallic acid, maleic anhydride, wood vinegar, and hydrothermal carbonization aqueous phase directly enter the humification reaction network as reaction participants. Catechol (0.5% dry weight, added on day 15) produced the most pronounced effect among precursor additives, increasing HS by 123% and HA by 100% in chicken manure–corn straw composting; it functioned not merely as a carbon skeleton but reshaped the microbial functional community, converting polyphenols and inorganic nitrogen from limiting factors into humification drivers [11]. Gallic acid (0.5% dry weight) enhanced lignin and cellulose degradation by 42% and 21%, respectively, and increased the synthesis of highly humified HA components by 22% in cow manure–soil composting, achieving the coupling of lignocellulose degradation and humus synthesis by directly stimulating microbial decomposition while enhancing polyphenol utilization [60]. Maleic anhydride (0.038 M/kg dry weight) in cattle manure–hay composting directly participated in precursor condensation reactions through its anhydride and vinyl groups, while simultaneously enhancing laccase and tyrosinase activities to catalyze the polymerization of polyphenols, carboxylic acids, amino acids, polysaccharides, and reducing sugars into humic substances [61]. Wood vinegar (0.65–1.75% *v/w*) in swine manure–rice straw composting promoted the conversion of stable humus fractions into more highly aromatized active humic acid components through its constituent organic acids and phenolic compounds, with 1.75% being more suitable for low-temperature conditions [62]. Hydrothermal carbonization aqueous phase, applied at

10% in split additions to chicken manure composting, provided small-molecule organics as microbial substrates and humification precursors, raising the HA/FA ratio from 1.70 to 2.87 and increasing HA by 32% over the control [63].

Malonic acid and chitosan indirectly promote humus accumulation as metabolic regulators. Malonic acid, a competitive inhibitor of succinate dehydrogenase in the tricarboxylic acid cycle, was applied at 0.5% dry weight at three composting stages in chicken manure composting; by partially blocking the TCA cycle, it redirected carbon flux from complete mineralization (CO_2 release) toward humus synthesis, while simplifying the bacterial interaction network and promoting Firmicutes dominance [17]. Chitosan (1% fresh weight) in chicken manure–corn straw composting increased HS by 9.3–12.9 mg/g over the control and preserved 6% more organic matter; its distinctive mechanism lies not in actively promoting humus formation but in protecting already-formed humic substances from microbial decomposition during the maturation phase by suppressing 18 bacterial genera possessing aromatic carbon degradation capacity [64]. The TCA cycle and its regulatory role in carbon partitioning are illustrated in Figure 4. Malonic acid competitively inhibits succinate dehydrogenase, blocking the oxidation of succinate to fumarate. This partial TCA cycle suppression redirects carbon flux away from complete mineralization as CO_2 and toward humus synthesis.

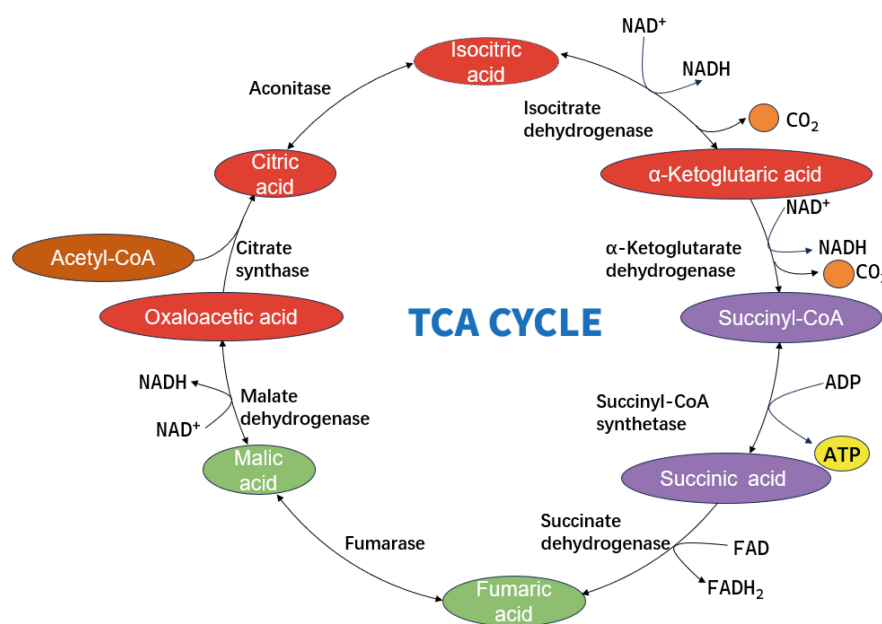


Figure 4. Tricarboxylic acid cycle.

Compared with bulk organic co-substrates, small-molecule additives can achieve pronounced effects at sub-percent doses—catechol and gallic acid at 0.5% realized HA increases exceeding 100%. However, their high unit cost and the requirement for precise dosage control limit their direct application in farm-scale composting, and all studies to date have been conducted at laboratory scale.

4.4. Exogenous Humic Substances

The direct addition of humic substances represents a strategy distinct from precursor supply and metabolic regulation. Exogenous humus can accelerate the humification process by providing aromatic carbon skeletons, reactive functional groups, and associated microorganisms. The types of humic substances applied fall into two categories: commercial humic acid and compost-derived humus.

Commercial humic acid (purity $\geq 90\%$) was systematically investigated at a 6% dose in swine manure–corn straw composting. HA increased by 143%, HS increased by 41%, and FA decreased to 12.8 g/kg, with carbohydrate and amino acid metabolism upregulated and multiple ARG classes significantly reduced [3]. Concurrently, HA addition increased final TN by 25% and reduced denitrification gene abundance by 30%, primarily achieved by suppressing Proteobacteria (57% reduction) and targeting denitrifying communities [5]. Furthermore, HA promoted the removal of aminoglycoside antibiotics and reduced the expression of T4SS secretion system and oxidative stress genes, coupling humification enhancement with improved biosafety [6]. Taken together, commercial humic acid provides a condensation template for nascent HA through its aromatic carbon skeleton, participates in precursor complexation via carboxyl and phenolic hydroxyl groups, and modulates functional microbial communities by altering carbon and amino acid metabolism, ultimately achieving the multiple benefits of accelerated humification, enhanced nitrogen retention, and contaminant mitigation.

Compost-derived humus, extracted from a prior batch of corn stalk–cattle manure compost, embodies a circular composting strategy. Its addition raised the initial HS of a new batch from 13.5 to 36.6 g/kg, extended the thermophilic phase by 8 days, increased humus content by 28.0 g/kg, and subsequently enhanced maize plant nitrogen content by 5.1 g/kg [65]. Proteobacteria and Bacteroidota were enriched during the thermophilic phase, accelerating cellulose and lignin decomposition, indicating that compost-derived humus simultaneously improves the chemical environment and provides an adapted microbial inoculum.

4.5. Comparative Synthesis of Organic Strategies

The promotion mechanisms of organic additives are more diverse than those in inorganic systems. Biochar and agricultural byproducts promote humification primarily through physical structure optimization and surface adsorption, whereas small-molecule precursors and exogenous humic substances directly enter condensation reaction networks, and malonic acid and chitosan indirectly influence the humification process through metabolic regulation or community protection. Effective doses span over thirty-fold: catechol at 0.5% increased HS by 123%, while agricultural byproducts require 10–15%, a difference rooted in the former participating in stoichiometrically defined reactions and the latter relying on bulk physical improvement.

The upper limit of effect magnitude for organic additives is lower than that of iron-based catalytic types, with catechol and commercial humic acid achieving HA increases of 100–143%, compared with 267% for magnetite. Most organic additives provide ancillary benefits beyond humification enhancement, such as greenhouse gas emission reduction by biochar, ARG reduction by humic acid, and heavy metal passivation by cow bone powder, reflecting the inherent multifunctionality of organic materials.

Certain organic additives are functionally close to inorganic mechanisms. Cow bone powder achieves adsorption and ion exchange through hydroxylapatite, while lignite, containing approximately 14% native HA, serves both as a precursor source and a microbial carrier, indicating that the organic/inorganic classification is not absolute at the functional level and offering instructive insights for composite additive design.

Current research on organic additives faces limitations similar to those in the inorganic domain. The vast majority of studies have been conducted at laboratory scale, cost-effectiveness comparisons between agricultural byproducts and purpose-produced additives are absent, and the synergistic and antagonistic effects among organic additives remain systematically unexplored.

5. Biological Strategies

5.1. Fungal Inoculation

Fungal inoculation promotes humification by secreting lignin-degrading enzymes (laccase, lignin peroxidase, and manganese peroxidase) that depolymerize lignin, releasing phenolic humus precursors [10], making it an ideal choice for compost bioaugmentation.

Inoculation conditions and strain selection are key factors determining efficacy. *Phanerochaete chrysosporium* inoculated at 5% (v/w) during the cooling stage of corn straw–canola residue composting, with pH pre-adjusted to approximately 6.5, increased HA to 95 g/kg and HS from 78 to 142 g/kg, an 82% increase, while LiP activity increased by 238%; pH adjustment resulted in a 3.6-fold higher strain abundance compared with the unadjusted treatment [10]. *Aspergillus fumigatus* Z1 inoculated at 1% dry weight in rice straw composting pretreated with Fenton reagents ($\text{FeCl}_2 + \text{H}_2\text{O}_2$), combined with rhamnolipids, resulted in Basidiomycota comprising 63% of the fungal community [66]. Metabolomic analysis further demonstrated that inoculation upregulated the shikimate pathway and phenylpropanoid metabolism, channeling aromatic metabolites toward humus polymerization [67].

Multi-strain combinations can achieve synergistic enhancement through functional complementarity. Combined inoculation of *Trichoderma harzianum* and *P. chrysosporium* at 0.05% (v/w) each in chicken manure–rice husk composting increased HA to 22.7 g/kg and GI to 111%, outperforming single-strain inoculation, attributable to the functional complementarity of *Trichoderma* preferentially targeting cellulose and *P. chrysosporium* preferentially targeting lignin [68]. The fungi-static bacterium *Cellulomonas iranensis* ZJW-6 inoculated at 1% in cattle manure–corn straw composting increased HA content by 336%; its mechanism does not involve direct lignocellulose degradation but rather the indirect promotion of Actinobacteriota proliferation through the suppression of competing fungi [69].

5.2. Bacterial and Mixed Microbial Inoculation

Bacterial inoculants are widely studied owing to their diverse metabolic functions and tolerance of the alkaline composting environment. Their humification-promoting mechanism lies in reshaping microbial community structure and metabolic networks, thereby enhancing lignocellulose degradation and the directional conversion of humification precursors.

Lignocellulose-degrading inoculants provide precursors for humification by accelerating macromolecular depolymerization. A composite inoculant (containing *Bacillus*, *Streptomyces*, and others) in Chinese medicine residue composting increased HA to 75.5 g/kg, with β -glucosidase upregulation relieving cellobiose feedback inhibition, and laccase and LiP activities significantly enhanced from day 12 onward [70]. In cattle manure composting, a 1.5% microbial agent achieved a GI of 92% and increased lignocellulose degradation by 42%, with enhanced CAZymes expression driving the simultaneous supply of sugars and amino acids to both the Maillard reaction and the lignin–protein condensation pathway [71]. A synthetic microbial community (three bacteria plus two fungi, 10^8 cells/g) in 500 kg cattle manure–mulberry branch composting increased HS by 34% and lignin degradation by 19%; metabolomics revealed 328 differential metabolites, among which protocatechuic acid, sinapic acid, and coniferaldehyde were significantly positively correlated with HA [16]. In corn straw–biogas slurry co-composting, a composite inoculant combined with biochar converted NH_4^+ -N into C–N bonds in HA (N content 7.75%), achieving an HA/FA ratio of 3.06 and a GI of 132% [72]. In a bagasse–cow manure system, *T. longibrachiatum* plus *P. chrysosporium* with a bio-maturation agent increased HA by 44%, raised Actinobacteria

from 15–21% to 27–31%, and elevated the proportion of positive correlations in fungal networks from 79% to 91% [73].

Optimization of inoculation strategy has a decisive influence on efficacy. Two-stage inoculation—thermophilic bacteria at day 0 followed by mesophilic fungi at day 12—achieved an HA of 25.8 mg/g, outperforming single-stage inoculation (20.0 mg/g), with NO_3^- -N increasing by 956% and 10 amino acid synthesis pathways upregulated during the maturation phase [74]. A comparison of early-stage (day 0) versus cooling-stage (day 17) inoculation further confirmed this finding: early inoculation increased HA by 107% and GI to 155%, whereas cooling-stage inoculation was substantially less effective because it missed the critical temperature window for humification [75].

Functionally specialized inoculants extend the application scope of bacterial strategies. A nitrifying bacterial consortium increased NO_3^- -N by 78–82%, HS by 16%, and reduced NH_3 emissions by 49%, with the correlation coefficient between NO_3^- -N and humus reaching 0.98 [76]. Phosphate-solubilizing *Bacillus subtilis* SL-44 in spent mushroom substrate composting reduced HS mineralization loss to only 5% (versus 12% for the control) while also increasing TN by 9% through nitrogen fixation [77]. A novel ammonifying microbial consortium achieved an HA/FA ratio of 13.1, extended the thermophilic phase to 19 days (versus only 7 days for the control), with 2D-COS revealing the complete molecular evolution pathway from polysaccharides through low-molecular-weight intermediates to aromatic condensation [78]. Different *Bacillus* species exhibited strain-specific effects on HA formation—*B. halophilus* achieved an HA of 70.7 mg/g with 92% positive network correlations, whereas *B. siamensis* reached only 61.7 mg/g [79].

5.3. Synthetic and Reconstructed Microbial Communities

A synthetic microbial community combining lignocellulose-degrading and humus-synthesizing capabilities increased lignin, cellulose, and hemicellulose degradation by 19%, 8%, and 12%, respectively, and humus content by 34% [16]. A reconstructed bacterial community reduced CO_2 emissions by 16% while promoting humus formation [80]. The advantage of synthetic communities lies in quality control and functional predictability, though long-term stability against indigenous microbiota remains to be demonstrated.

5.4. Enzyme-Mediated Strategies

Unlike the inoculation of living microorganisms, enzyme-mediated strategies directly introduce functional enzymes or their inducers—capable of catalyzing lignocellulose depolymerization or humus polymerization—into the composting system, thereby avoiding the uncertainties of microbial colonization and competition. Laccase is the most extensively investigated enzyme in this context.

Laccase oxidizes phenolic substrates to generate reactive quinone intermediates, which can spontaneously polymerize into humic substances. Exogenous laccase (20 U/g dry weight) in rice straw–soil co-composting increased HS by 19% and HA by 48%; the mesh-bag treatment (JL) enhanced lignin degradation by 6.4-fold and reduced BF-Pb by 7.2%, achieving the synergy of humification and heavy metal immobilization [81]. Graphene oxide (GO) combined with 1-hydroxybenzotriazole (HBT) as a laccase mediator system sustained a 73% increase in laccase activity, with bacterial and fungal laccase genes upregulated by 228% and 265%, respectively, lignin humification efficiency increased by 108%, and GI reaching the highest value [82]. The addition of laccase (220 U/L) during the secondary fermentation stage of composting increased HA by 52%, HS by 39%, and HA/FA from 2.11 to 2.89, with laccase activity elevated by 206% and LiP and MnP activities each increased by approximately 40% [83].

Enzyme inducers stimulate indigenous microorganisms to express endogenous lignocellulose-degrading enzymes, achieving effects similar to direct enzyme addition at lower cost by circumventing the issues of enzyme purification and stability. MnSO_4 (1% dry weight) combined with *Aspergillus fumigatus* in rice straw composting achieved an HA increase of 149%, exceeding that of the fungus alone (124%) or MnSO_4 alone (128%); a C4 fluorescence component appearing exclusively in the MnSO_4 -containing treatments indicated that the enzyme inducer promoted the formation of more highly humified products [84].

Functional enzyme liquid strategies bypass the cost barrier of purified enzymes. A crude enzyme liquid obtained from the pre-fermentation of rice straw with *A. fumigatus* Z1, used in place of purified enzyme preparations, reduced the relative β -O-4 bond content from 9.0% to 3.1% in rice straw composting; 2D-HSQC-NMR confirmed that oxidized lignin units (Gox and Sox) participated in HA construction, with the HA-C3 component increasing and its emission wavelength red-shifted by 10 nm [85]. Under domesticated composting conditions, LiP exhibited not only its degradative function in lignin depolymerization but also polymerization capacity that directly promoted humic substance formation, with its activity continuously increasing over a 125-day cycle [86].

Compared with microbial inoculation, enzyme-mediated strategies offer better controllability and reproducibility; however, the cost of enzyme preparations and their stability under composting conditions remain the principal bottlenecks limiting large-scale application.

5.5. Comparative Synthesis of Biological Strategies

Biological strategies harness the self-replication and catalytic capacity of living microorganisms or enzymes to sustain humification throughout the composting cycle, but their predictability and environmental sensitivity remain key constraints. Enzymatic strategies act most directly without requiring colonization, fungal inoculation provides sustained lignin degradation through extracellular enzymes but depends on specific temperature and pH conditions, bacterial inoculation drives multiple nutrient transformation pathways coupled to humification, synthetic communities offer functional predictability at high design cost, and enzyme inducers stimulate indigenous enzyme activity at low cost. Inoculation timing consistently proves more critical than strain degradative capacity, with cooling-stage, early-stage, and two-stage inoculations each outperforming their alternatives, suggesting that future biological enhancement design should prioritize matching optimal timing and microenvironment over screening more powerful strains. Most biological strategy studies remain at laboratory scale, and engineering challenges including viable cell decline during storage, enzyme inactivation at thermophilic temperatures, and biosafety of synthetic communities remain insufficiently evaluated.

6. Composite and Synergistic Additive Systems

Composite strategies—combining two or more additive types—represent the largest category with 25 studies. The premise is that strategically designed combinations can simultaneously optimize the physical environment, provide precursors, catalyze reactions, and inoculate functional microorganisms.

6.1. Biochar Combined Additives

Biochar, with its porous structure, large specific surface area, and surface functional groups, is the most commonly used base component in composite strategies, synergistically enhancing humification through physical structuring and chemical catalysis when combined with metal compounds, natural minerals, microbial agents, or organic additives.

Biochar combined with metal compounds exploits the redox catalytic capacity of metals to complement biochar's physical functions. Apple branch biochar (5%) co-applied with

MnSO₄ (0.5%) in pig manure–sawdust composting at pilot scale increased HA/FA from 1.57 to 3.43 through the synergistic effects of biochar providing microbial microhabitats, Mn(II) slow release stimulating laccase and dehydrogenase activities, and Mn(II) oxidation to Mn(III/IV) serving as electron transfer mediators to catalyze organic matter oxidative condensation [18]. However, direct physical mixing of MnO₂ with biochar restricted functional group activity by forming limiting Mn–O–R surface complexes [87]. Similarly, the combination of biochar with Fe₂O₃ in pig manure–wine grape pomace composting weakened aromatic C=O stretching, performing worse than Fe₂O₃ alone [88]. These antagonistic phenomena indicate that the efficacy of metal compound–biochar combinations depends on the mixing method rather than the components themselves.

Biochar combined with natural minerals achieves physical complementarity, with minerals supplementing biochar's functions through adsorption and ion exchange. Biochar (10%) and smectite (10%) in chicken manure–rice husk composting acted on different humification segments: biochar enhanced DOM aromatization and abiotic humification pathways, while smectite increased bacterial richness and formed specialized functional patterns, together promoting humification [89]. Volcanic rock (2%) in cattle manure composting achieved humification enhancement comparable to biochar by enriching lignocellulose-degrading bacteria such as *Thermobacillus* and *Paenibacillus* [90]. Lignite (5%) combined with phosphogypsum (10%) achieved complementary synergy, with lignite providing aromatic humus precursors to drive polyphenol condensation and phosphogypsum fixing NH₄⁺ through Ca²⁺ and SO₄^{2−} to reduce nitrogen loss, yielding an HA/FA of 4.17 and a 206% increase in pakchoi fresh weight [91].

Biochar combined with microbial agents employs biochar as a carrier and protective layer for microorganisms. Rice husk biochar (5%) loaded with ammonia-oxidizing and ammonia-assimilating bacteria in rice straw composting achieved an HA of 94 mg/g, an HA/FA of 4.63, and a 72% reduction in NH₃ emissions; the biochar maintained aerobic conditions favorable for ammonia oxidation while the inoculant progressively converted NH₄⁺-N into amino acid nitrogen and ultimately into HA structures [92]. Zeolite and biochar as a dual carrier loaded with a composite microbial consortium in biogas residue composting achieved a GI of 253% and a 49% increase in HS retention [93]. A systematic comparison of EM inoculant combined respectively with biochar, superphosphate, and phosphogypsum in tomato straw composting found that biochar + EM performed best, with an HA/FA of 7.80 and a 64% reduction in FA [94].

Biochar combined with organic additives or enzymes simultaneously provides physical structure and chemical precursors. Biochar (5%) combined with cellulase in cattle manure–sugarcane bagasse composting increased HA by 41%, elevated HA/FA to 4.13, and reduced CO₂ and CH₄ emissions by 19% and 31%, respectively; cellulase hydrolysis released fermentable sugars and phenolic precursors, while biochar adsorbed soluble carbon and redirected carbon flux from mineralization toward humification [95]. Biochar (15%) combined with fulvic acid (3%) increased HA/FA by 57% and reduced NH₃ emissions by 48% [96]. Spent mushroom substrate combined with biochar simultaneously enhanced humification and heavy metal passivation in chicken manure–rapeseed straw composting [97].

6.2. Microbial Agent-Centered Composite Strategies

Beyond biochar-based composite systems, several studies have employed microbial agents as the functional core, supplemented with carbon sources, minerals, humic acid, or chemical pretreatment to enhance inoculant colonization efficiency and humification performance.

Functional minerals or organic amendments can compensate for the functional limitations of microbial agents at specific composting stages. The combined application of *Trichoderma longibrachiatum* and MnSO_4 exemplifies stage-dependent complementarity: during the mesophilic stage, the fungus secreted lignocellulose-degrading enzymes to rapidly consume humification precursors, while during the thermophilic and maturation stages, MnSO_4 reduced NH_3 volatilization by maintaining pH below 9.3, preserving reactive nitrogen for condensation reactions and reducing TN loss from 27% to 15%; concurrently, Mn induced a fully positive (100%) cross-kingdom microbial network [19]. Humic acid (5%) combined with nitrogen-fixing microbial agents in swine manure–wheat straw composting promoted humification through humic acid providing phenolic hydroxyl, carboxyl, and amide groups for heavy metal complexation and ion exchange, while the nitrogen-fixing bacteria reduced nitrogen loss, enriching *Mycrothermus* to 98% of the fungal community and reducing NH_3 emissions by 73% and N_2O by 49% [98].

Carbon source stimulation is an effective means of enhancing microbial agent activity under low-temperature conditions. In winter cattle manure composting, molasses combined with cold-tolerant inoculants (*Streptomyces griseorubens* JSD-1 + *Paenarthrobacter nitroguajacolicus* LDT1–8) rapidly activated thermogenesis (peak 62.1 °C vs. control 58.2 °C) through the provision of readily utilizable carbon, with Firmicutes reaching 82% abundance in the early stage, HA increasing by 33%, and the thermophilic phase extending from 3 to 5 days [99]. An orthogonal experiment demonstrated that the combination of 10% matured compost, 0.05% microbial inoculant, and 10% rapeseed cake added at the initial stage achieved an HA/FA of 3.03 and a GI of 168%, with the polyphenol precursors supplied by rapeseed cake synergistically enhancing humification in conjunction with the degradative function of the inoculant [100].

KOH- HNO_3 pretreatment combined with microbial inoculation integrates feedstock structural modification with biological transformation. KOH (1.25 mol/L) pretreatment disrupted the ester bond cross-linking structure of straw, and HNO_3 neutralization released amino acids (+127%) and polyphenols (+30%) as humification precursors; subsequent inoculation with fungal agents catalyzed condensation polymerization, with complex HA components (C4 + C5) accounting for 72% of total fluorescence in the mature phase [101].

6.3. Comparative Synthesis of Composite Strategies

The core advantage of composite strategies lies in simultaneously targeting multiple rate-limiting steps of humification through functional complementarity. However, synergy is not automatically realized. Several studies in this chapter documented clear antagonistic effects, indicating that the design of composite additives must consider chemical compatibility and biological interactions among components rather than assuming simple additive benefits. The optimization of dosage ratios remains the critical gap between empirical formulation and rational design of composite strategies. As with the three preceding additive categories, composite strategy studies are similarly concentrated at laboratory scale, and techno-economic evaluation of whether the higher material and management costs of composite systems justify their incremental benefits over single additives remains absent.

7. Discussion and Outlook

7.1. Cross-Category Synthesis

Table 1 provides a comparative summary of the four additive categories, including representative additives, effective doses, humification performance, and co-benefits. The four additive categories exhibit systematic differences in effect magnitude, mechanism, and applicability. Iron-based additives achieve the highest HA enhancements (82–267%) [9,21]

through redox catalysis, but operate within narrow dose windows (1–5%) and are feedstock-sensitive. Clay minerals produce moderate effects (HA increases of 28–163%) [7,28] with broader dose ranges and greater feedstock versatility, acting through physical adsorption and microenvironment regulation. Among organic strategies, biochar combines physical structuring with microbial carrier functions, delivering consistent HA improvements (25–93%) [8,39], while precursor-type additives (catechol, humic acid) achieve >100% HA enhancement at sub-percent doses through targeted chemical pathways [3,11]. Biological strategies (fungi, enzymes) provide self-sustaining catalytic activity but are sensitive to pH, temperature, and inoculation timing [67,74]. From a practical standpoint, high-precision catalytic additives are suited to controlled industrial composting, whereas robust, low-cost materials are more appropriate for on-farm operations.

Table 1. Comparative summary of additive categories for humification enhancement during agricultural waste composting.

Category	Representative Additives	Dose	HA Increase (%)	HA/FA	GI (%)	Scale	Co-Benefits	Limitations	References
Iron-based	Magnetite, nZVI, Fe ₂ O ₃ , FeS ₂ / Pal	1–5%	+15 to +267	Up to 2.25	124–136	Lab (15–150 L)	HM passivation (Cu, Zn)	Narrow dose window; oxidative stress	[9,21–26]
Clay minerals	Attapulgite, bentonite, medical stone	5–10%	+30 to +163	Up to 2.7	97	Lab (20–118 L)	NH ₃ retention; DOM aromatization	Moderate effect; high loading rate	[7,27–29]
Phosphate & industrial byproducts	Ca(H ₂ PO ₄) ₂ + MgSO ₄ , fly ash, coal gangue	5–10%	+83	Up to 5.6	80–98	Lab, some pilot (100 L)	Struvite N fixation; waste valorization	HM residues unassessed	[30–32]
Biochar	Multi-source biochar, bamboo biochar, lignite	5–10%	+25 to +93	Up to 2.44	95–132	Lab; 1 pilot (500 kg)	GHG reduction; P mobilization	Source-dependent; >20% inhibits	[8,35–47]
Agri-food byproducts	MSGR, feather hydrolysate, tobacco powder, garden waste, bone powder	1–15%	+28 to +89 (g/kg)	DP up to 3.64	107–168	Lab; 1 semi-industrial (1000 kg)	Waste valorization; on-farm availability	Variable quality; phytotoxicity risk	[48–59]
Humic precursors & small molecules	Catechol, gallic acid, malonic acid, maleic anhydride, chitosan	0.5–6%	+100 to +143	Up to 2.87	—	All lab (<30 L)	ARG reduction; humus protection; antibiotic removal	High unit cost; lab-scale only	[11,17,60–64]
Exogenous humic substances	Commercial HA, compost-derived humus	6% or —	+143	HS +41%	—	All lab (15–30 L)	N retention (+25%); ARG reduction	Long-term soil fate unexamined	[3,5,6,65]
Fungal inoculation	<i>P. chrysosporium</i> , <i>Trichoderma</i> spp., <i>A. fumigatus</i>	0.05–5% (v/w)	+82 to +169 (HS)	Up to 2.66	111–140	Lab (20–200 L)	Lignin degradation (73%); LiP (+238%)	pH/temp dependent; cooling-stage required	[10,66–69]
Bacterial & mixed inoculation	Synthetic Microbial Community, nitrifying consortia, <i>Bacillus</i> spp.	0.5–5% (v/w)	+16 to +57 (HS)	Up to 13.1	118–155	Lab; some 200 L	NH ₃ reduction (49%); N retention; P solubilization	Strain specificity	[16,70–80]
Enzyme-mediated	Laccase, MnSO ₄ inducer, functional enzyme liquid	20 U/g–220 U/L	+48 to +149	Up to 2.89	84–140	All lab (<60 L)	HM immobilization; β-O-4 cleavage	Enzyme cost; thermal instability	[81–86]
Biochar-based composites	Biochar + metal/mineral/microbe/enzyme	BC 5–10%	+41 to +127	Up to 7.80	121–253	Lab; 2 pilot (65 L, 100 kg)	GHG reduction; N retention synergy	Antagonism risk; complex optimization	[18,87–97]
Microbial-centered composites	Inoculant + carbon/mineral/HA/pretreatment	Variable	+18 to +33	Up to 3.03	100–168	Lab; some pilot (100–160 L)	Stage-dependent complementarity	Multi-factor optimization needed	[19,98–101]

Abbreviation: “—” indicates that the data were not reported in the original studies.

7.2. Feedstock–Additive Matching

No study has systematically varied feedstock while holding additive type constant. However, cross-study comparisons of the same additive across different feedstocks reveal consistent patterns. Lignocellulose-rich substrates (straw, woody residues) benefit from additives that depolymerize lignin—Fenton catalysts, lignin-degrading fungi, and cellulases—which simultaneously release phenolic humus precursors [9,67,95]. Protein-rich manure requires additives with strong nitrogen retention capacity (zeolite, biochar, phosphogypsum, nitrifying consortia) [47,91], coupled with carbonaceous precursors to balance the stoichiometry of humus-forming condensation reactions. Mixed manure–straw composting is best served by biochar-based composites in which biochar provides physical structure while co-additives deliver catalytic or biological functionality [92,94]. This framework, currently based on cross-study inference, requires validation through multi-feedstock, multi-additive factorial experiments.

7.3. Composite Is Not Necessarily Better: Synergy and Antagonism

Composite additives represent the largest category in this review, and most combinations outperform single additives through functional complementarity. Successful synergy is exemplified by the combined application of apple branch biochar and MnSO_4 , which achieved a humification degree of 3.15 through the triple mechanisms of physical protection, biological stimulation, and abiotic redox catalysis [18], and by lignite plus phosphogypsum, which attained an HA/FA ratio of 4.17 while reducing nitrogen loss to 9% and increasing pakchoi fresh weight by 206% [91]. However, synergy is not guaranteed. Several studies documented clear antagonistic effects: Fe_2O_3 combined with biochar weakened aromatic C=O stretching relative to Fe_2O_3 alone, with the combination failing to outperform the single Fe_2O_3 treatment in HA-like fluorescence intensity [88]; MnO_2 directly mixed with biochar formed restrictive Mn–O–R surface complexes that limited functional group activity, resulting in accelerated mineralization rather than enhanced humification [87]; and high-dose microbial inoculation (>2%) combined with biochar–zeolite carriers suppressed the HA/FA ratio to 1.03, compared with 1.85 achieved at a 1% inoculation rate [93]. These findings demonstrate that composite additive design must account for chemical compatibility and biological interactions. To date, only one study has systematically optimized multi-factor combinations using an orthogonal design, leaving systematic dose–ratio optimization as the critical gap between empirical formulation and predictive design [100].

7.4. From Laboratory to Practical Application

Over 90% of the 110 studies were conducted at laboratory scale (<200 L), with only a handful reaching pilot scale (60–500 kg) [18,19], and none has achieved industrial deployment. Cost data are absent from the literature—no study has reported a formal techno-economic analysis. Agricultural byproducts (monosodium glutamate residue, tobacco powder, garden waste, feather hydrolysate) are inherently low-cost and available on-farm, yet their cost-effectiveness relative to purpose-produced additives (commercial biochar, humic acid, enzyme preparations) has not been evaluated. Industrial byproducts (coal gangue, fly ash, phosphogypsum) may carry heavy metal residues, and the long-term soil accumulation risk adds further complexity to the cost–benefit assessment. For inorganic additives, the continued redox activity of iron and manganese oxides in soil, the potential ecotoxicity of nanoparticles, and the persistence or priming effects of exogenous humic substances have not been assessed in long-term field trials.

7.5. Co-Benefits Beyond Humification

Although the included studies were selected for their focus on humification, several additives also delivered process-level co-benefits. Biochar and hydrochar reduced NH_3 emissions by 27–72% [47,92] and N_2O emissions by 45–49% [92] while promoting humification. Iron-containing additives facilitated the co-passivation of Cu and Zn [25,38]. These co-benefits arise from shared mechanisms—biochar’s porous structure simultaneously adsorbs NH_3 , provides microbial habitat, and retains humus precursors. Across all included studies, GI showed a significant positive correlation with the HA/FA ratio ($R^2 = 0.67$, $p < 0.01$), indicating that enhanced humification consistently improves compost phytotoxicity mitigation and agronomic suitability. Future studies should report a standardized panel of process-level indicators alongside humification metrics.

7.6. Outlook

Based on the gaps identified above, the following priorities are proposed:

- Pilot-to-field validation at ≥ 1000 kg scale under realistic operational conditions, assessing additive stability, effect reproducibility, and operational feasibility, accompanied by techno-economic analysis.
- Establishment of a consensus minimum humification dataset (HA, FA, HA/FA, GI, and at least one spectroscopic indicator) to enable quantitative cross-study comparison.
- Adoption of multi-factor experimental designs (e.g., orthogonal arrays, response surface methodology) to systematically optimize additive type, dose, ratio, and timing.
- Multi-year field monitoring of metal-based additives and industrial byproducts to evaluate their long-term fate and ecological safety in soil.

7.7. Limitations

Several limitations of this review should be acknowledged. First, as a qualitative mechanistic review synthesizing heterogeneous study designs, formal risk-of-bias assessment was not performed; methodological heterogeneity was instead addressed through qualitative comparison of study scale, additive dosage, composting duration, and analytical methods. Second, the literature search was restricted to English-language publications indexed in Web of Science and PubMed, potentially excluding relevant studies published in other languages. Third, the diversity of humification indicators across the included studies precluded quantitative meta-analysis for most additive–feedstock combinations.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/fermentation12080392/s1>. S1: PRISMA 2020 checklist.

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