

Article

Municipal Sewage Sludge Composting in the Two-Stage System: The Role of Different Bulking Agents and Amendments

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Abstract: This study assessed the effect of different lignocellulosic amendments and bulking agents on compost stability (based on a 4 day respiration activity test, AT₄, and self-heating factor, SH_F) and maturity (based on the nitrification index I_{nitr} and the ratio of C in humic acids, HA, to total organic carbon, TOC, in compost, C_{HA}/TOC). With all feedstock compositions (FCs), the share of sewage sludge was 79% (wet mass). For FC₁, wood chips (13.5%) and wheat straw (7.5%) were used as bulking agents and amendments; for FC₂, instead of wood chips, energy willow was added; for FC₃, pine bark (13.5%) and conifer sawdust (7.5%) were used. All FCs produced stable and mature compost; however, with FC₂, the thermophilic phase last 3 days longer than with the other FCs. Moreover, an AT₄ value below 10 g O₂/kg dry mass (d.m.) was obtained the earliest with FC₂ (after 45 days, ca. 15–20 days earlier than with other FCs). With FC₂, I_{nitr} below 0.5 was obtained in ca. 60 days, 10 days earlier than with FC₃ and 30 days earlier than with FC₁. The highest net increases in HS (86.0 mg C/g organic matter (OM)) and HA (56.3 mg C/g OM) were also noted with FC₂; with other FCs, the concentrations of these compounds were from 1.3- to 1.5-fold (HS) and from 1.4- to 1.9-fold (HA) lower. With FC₂, the highest C_{HA}/TOC (15.5%) was also noted, indicating that this compost contained the largest share of the most stable form of organic carbon. The rates of OM removal in the bioreactor ranged from 7.8 to 10.1 g/(kg d.m.·day). The rates of SH and HA formation ranged from 1.63 to 4.83 mg C/(g OM·day) and from 1.23 to 1.80 mg C/(g OM·day), respectively. This means that, through the choice of the amendments and bulking agents, the length of the composting time needed to obtain a stable and mature product can be controlled.

Keywords: sewage sludge; compost stability; humic substances and humic acids; nitrification index; nutrients; heavy metals



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1. Introduction

Management of sludge generated in municipal wastewater treatment plants is one of the priorities in engineering and environmental protection. As the amount of municipal sewage sludge continues to grow, creating a global problem, it is necessary to develop technologies for its management/neutralization.

In Poland, according to the Update of the National Program for Municipal Sewage Treatment (UNMSTP 2022) [1], the following goals were adopted in the field of municipal sewage sludge management: (1) complete cessation of municipal sewage sludge storage; (2) increases in the amount of sewage sludge processed before being released to the environment and the amount of sludge subjected to thermal treatment; (3) an endeavor to maximize the use of nutrients contained in the sludges while meeting all sanitary, chemical, and environmental safety requirements. These goals can be achieved using the composting process, which is a form of organic recycling. Composting enables the stabilization and hygienization of sludge and the retention of nutrients in the final product.

Composting is a preferred method of sewage sludge treatment, mainly in small and medium-sized wastewater treatment plants. Due to its high moisture, low porosity, and low C/N ratio, sewage sludge cannot be composted alone and must be mixed with other materials that are rich in carbon, contain low nitrogen concentration, and are characterized by low moisture. These are mainly lignocellulosic materials, which serve as amendments and bulking agents. The addition of these materials decreases the moisture content, enables adequate gas exchange, and prevents excessive compaction of the feedstock by providing the structural support to create interparticle voids (increasing the porosity). Moreover, the amendments increase the C/N ratio, which also favors effective composting for both sewage sludge composting and other wastes with high nitrogen content [2–5].

Even though the composting process has been used for many years, currently mandated organic recycling rates have led to an increase in research focusing on how various additives influence the process and the quality of the final product. The physical and chemical properties of the composting feedstock and the resulting course of the composting process can be changed by adding to sewage sludge various materials that differ in composition but are rich in carbon. Several studies have looked into how the process and the quality of the final product are influenced by the introduction of amendments for co-composting [6–11]. Other authors used modified bulking agents. For example, Liu et al. [12] investigated the effect of a recoverable sulfuric acid- and sodium hydroxide-modified pinewood as a bulking agent during composting of sewage sludge with sawdust. They showed that the addition of recoverable pinewood promoted the degradation of organic matter, the extension of the high-temperature period by 4 days, and a reduction in the loss of nitrogen by 9.40%. Recently, easily biodegradable organic compounds (e.g., glucose and sucrose) have begun to be used as a carbon source [9]. According to the authors, these compounds accelerated organic matter degradation, increased dissolved organic carbon contents and CO₂ emissions, and reduced NH₃ emissions. All abovementioned papers indicate that the type of amendments/bulking agents that are added to composting feedstock affect transformations during the mineralization phase. However, it is very useful to check how different amendments/bulking agents influence compost stability and maturity, including the humification progress. This is important, as stable compost limits the emissions of carbon dioxide, and the HS content in compost plays a fundamental role in carbon sequestration in the soil in both natural and cultivated environments [13]. Moreover, the types of amendments/bulking agents that are used may determine the kinetic constants of mineralization and humification, and this information is important for process design, especially when estimating the time for feedstock retention during active composting in bioreactors and maturation in windrows. Therefore, these investigations were undertaken in this study, and the novelty follows from the multi-faceted analysis of the impact of feedstock composition on compost stability (based on AT4 and self-heating factor, SH_F) and maturity (based on the nitrification index I_{nitr} and the C_{HA}/TOC ratio), as well as mineralization and humification kinetics.

2. Materials and Methods

2.1. Experimental Setup

The process was carried out in a two-stage system, in which the first stage was an aerated bioreactor, and the second stage was a periodically turned windrow. The bioreactor with an active capacity of 100 dm³ was made of acid-resistant steel and was equipped with a fan forcing air into the aeration box, which was the floor platform of the bioreactor. The aeration intensity was regulated by an inverter coupled with the fan drive. In the upper part of the bioreactor, there was a cover that allowed batch loading and taking samples of the composted material. In the lower part of the reactor, there was a perforated plate enabling airflow and a chamber for collecting leachate. Temperature sensors were installed in the bioreactor. The second stage of composting was carried out in natural conditions, without forced aeration.

2.2. Sewage Sludge and Feedstock Characteristics

2.2.1. Sewage Sludge

The sewage sludge originated from a municipal wastewater treatment plant located in northeastern Poland (53°22'59" N, 19°55'58" E). Sewage sludge was collected three times (at intervals resulting from the implementation of subsequent research variants), and the sludge from each collection was used to prepare different feedstocks (abbreviated as FC₁ for feedstock of composition 1, FC₂ for feedstock of composition 2, and FC₃ for feedstock of composition 3). The characteristics of the sewage sludges (from each collection) are presented in Table 1.

Table 1. Municipal sewage sludge characteristics.

Characteristics	Unit	FC ₁	FC ₂	FC ₃
<i>Basic analysis of physicochemical characteristics and nutrient contents</i>				
Moisture	%	89.0	89.3	89.2
Dry mass (d.m.)	%	11.0	10.7	10.8
OM	% d.m.	84.6	85.3	84.9
Reaction	pH	8.16	8.06	8.12
N		70.1	70.3	71.3
N-NH ₄		6.49	7.82	6.78
P		11.2	11.5	12.3
P ₂ O ₅		25.8	26.4	28.3
K ₂ O	g/kg d.m.	10.10	9.11	9.58
K		8.38	7.56	7.95
Mg		2.91	2.87	2.68
MgO		4.82	4.76	4.44
Ca		5.02	5.18	5.71
CaO		7.02	7.24	7.98
<i>Heavy-metal contents</i>				
Cd (20 *)		1.15	1.43	3.80
Pb (750 *)		5.3	6.4	6.1
Ni (300 *)		7.1	7.4	7.6
Cr (500 *)	mg/kg d.m.	21.2	19.9	18.4
Cu (1000 *)		74.0	87.9	79.0
Zn (2500 *)		603.0	614.0	626.0
Hg (16 *)		0.0260	0.0841	0.0910

* Limit values in accordance with the Regulation of the Minister of the Environment of 6 February 2015 (item 257) on municipal sewage sludge (when using sludge in agriculture and for land reclamation for arable purposes) [14].

The sewage sludge had very high contents of moisture (89.0–89.3%) and OM (84.6–85.3%). The pH of the sewage sludge was 8.06–8.16. The sludge was rich in fertilizer components (NPK). In particular, the content of N was high (7.01–7.13%). The OM/N and C/N ratios in the sludge ranged from 11.9 to 12.1 and 6.7 to 6.8, respectively. The concentrations of heavy metals were much lower than the limit values given in the Regulation of the Minister of the Environment of 6 February 2015 (item 257) on municipal sewage sludge [14]. No live intestinal parasite eggs were found in any of the samples, but bacteria of the genus *Salmonella* were isolated from all samples (in 100 g).

2.2.2. Lignocellulosic Materials and Feedstock Composition

As sewage sludge cannot be composted alone, to prepare the proper feedstock, the sewage sludge was mixed with lignocellulosic materials with a low moisture content, low nitrogen content, and high organic carbon content. The characteristics of the lignocellulosic materials used to prepare the feedstock are given in Table 2.

Table 2. Characteristics of lignocellulosic materials.

Characteristic	Wood Chips	Wheat Straw	Energy Willow	Pine Bark	Conifer Sawdust
Moisture (%)	42.3	11.2	40.7	52.9	10.7
OM (%)	96.10	93.23	98.05	97.90	99.67
N (%)	0.84	0.66	0.49	0.47	0.39

All the lignocellulosic materials had low moisture content, high content of OM (over 90%), and low content of nitrogen (<1%), which indicates that they may be useful for use as bulking agents/amendments. The moisture of the lignocellulosic waste was much lower than that of the sewage sludge, which is a necessary condition for its use to prepare the feedstock.

Three FCs were tested for composting. In all FCs, the share of sewage sludge was 79% (wet mass; w.m.), and the FCs differed in the type of bulking agents and amendments: in FC₁, wood chips (13.5% w.m.) and wheat straw (7.5% w.m.); in FC₂, energy willow (13.5% w.m.) and wheat straw (7.5% w.m.); in FC₃, pine bark (13.5% w.m.) and conifer sawdust (7.5% w.m.). Because the sewage sludge was characterized by exceptionally high moisture, much higher than that of the bulking agents/amendments, the shares of individual components in the feedstock are also given below in terms of dry mass. Taking into account the moisture of all components, the shares of sewage sludge in the feedstock (on a dry weight basis) were 37.6%, 37.1%, and 40% in FC₁, FC₂, and FC₃, respectively (dry mass; d.m.); in FC₁, wood chips constituted 33.6% d.m. and wheat straw constituted 28.8% d.m.; in FC₂, energy willow constituted 34.4% d.m. and wheat straw constituted 28.5% d.m.; in FC₃, pine bark constituted 29.2% d.m. and conifer sawdust constituted 30.8% d.m.

The characteristics of the composting feedstocks are presented in Table 3. The moisture content was 73–76%, and the OM content was 88.9–89.8%. On the basis of the contents of OM, TOC, and N, the OM/N and C/N ratios were calculated, which were 27.8–29.8 and 15.6–16.7, respectively.

Table 3. Feedstocks characteristic.

Feedstock	Moisture (%)	OM (%)	TOC * (%)	N (%)	OM/N	C/N
FC ₁	73.1	89.7	50.23	3.14	28.6	16.0
FC ₂	74.2	88.9	49.78	3.19	27.8	15.6
FC ₃	75.8	89.8	50.30	3.01	29.8	16.7

* TOC = 0.56 × OM (based on Regulation (EU) 2019/1009 of the European Parliament and of the Council of 5 June 2019 laying down rules on making fertilizing products available on the EU market, amending Regulations (EC) No 1069/2009 and (EC) No 1107/2009, and repealing Regulation (EC) No 2003/2003) [15].

2.3. Kinetic Equations

2.3.1. Organic Matter Removal

The rate constant of organic matter removal (*k*) and the maximal organic matter removal (*A*) were determined from the first-order kinetics equation.

$$A_{\text{loss}} = A \cdot e^{-k \cdot t}, \quad (1)$$

where *A* (g/kg d.m.) is the maximal organic matter removal in the bioreactor (*A_b*) or the windrow (*A_w*), *k* (day^{−1}) is the rate constant of organic matter removal in the bioreactor (*k_b*) or in the windrow (*k_w*), and *t* (day) is the composting time. According to the *k* and *A* values, the initial rate of organic matter removal (*r*) was calculated.

2.3.2. Humic Substances and Humic Acid Formation

HS and HA formation proceeded according to a first-order kinetic model. The kinetics of humification (HS or HA formation) were calculated with the following equations:

$$C_{\text{HS}} = C_{\text{max,HS}} \cdot (1 - e^{-k_{\text{HS}} \cdot t}) + C_{\text{FC,HS}} \quad (\text{HS formation}), \quad (2)$$

$$C_{\text{HA}} = C_{\text{max,HA}} \cdot (1 - e^{-k_{\text{HA}} \cdot t}) + C_{\text{FC,HA}} \quad (\text{HA formation}), \quad (3)$$

where C_{HS} or C_{HA} (mg C/g OM) is the concentration of HS or HA over time, $C_{\text{max,HS}}$ or $C_{\text{max,HA}}$ (mg C/g OM) is the maximal increase in the concentration of HS or HA during composting, k_{HS} or k_{HA} (day^{-1}) is the rate constant of HS or HA formation, and t is the composting time.

2.4. Analytical Methods

Composting samples taken from the top, middle, and bottom of the reactor were collected and then mixed to prepare representative samples. After mixing, a portion of the sample was taken for AT4 and self-heating factor (SH_F) in a Dewar vessel. The values of AT4 and SH_F were used as indicators of compost stability. The AT4 was determined under aerobic conditions in an OxiTop Control system [16]. The SH_F compost was determined in a Dewar vessel with the methodology described by Brinton et al. [17] and Pecorini et al. [18].

With the rest of the sample, the pH, moisture content, and dry matter (d.m.) content were determined [19]. After drying the samples to a constant weight, the sample was ground (0.5 mm diameter, Retsch SM100 mill). In the ground sample, the organic matter (OM) content was determined by incineration at 550 °C [20], nutrient concentrations (N, P, K, Mg, and Ca) were assessed by a district accredited chemical–agricultural station laboratory, according to PN-Z-15011-3 [21], and heavy-metal (Cd, Pb, Ni, Cr, Cu, Zn, and Hg) content was measured in a specialized laboratory (with the atomic absorption spectrometry method, ASA). Additionally, the contents of HS and their fractions, i.e., the fulvic fraction (FF) and humic acids (HA), were determined with a procedure presented by Kuli-kowska and Klimiuk [22] and Kulikowska [2]. The content of FF was determined by taking the difference between the content of HS and that of HA. The content of TOC was determined using a Shimadzu Liquid TOC-VCSN analyzer.

In water extracts from samples, the ammonium nitrogen [23], nitrite nitrogen [24] and nitrate nitrogen [25] were determined.

The content of live eggs of intestinal parasites (*Ascaris* sp., *Trichuris* sp. and *Toxocara* sp.) was analyzed according to PB-102/LM, and the presence of *Salmonella* bacteria was analyzed according to PN-EN ISO 6579-1:2017-04 [26] (in an accredited laboratory).

During the experiment on sewage sludge composting, the temperatures in the middle of the reactor and at the bottom were measured automatically using a PC THERM REM-84 m dual-channel temperature sensor with an accuracy of ± 0.1 °C.

3. Results and Discussion

3.1. Temperature Profiles and Organics Removal

The temperature changes during composting in the bioreactors with different FCs are shown in Figure 1. During the first 3 days of the experiment with FC_1 , the temperature increased intensively, and the maximum temperature of 60 °C was reached. A temperature above 55 °C was maintained until the sixth day of the process, after which it started to decrease gradually. After about 2 weeks, the temperature was about 40 °C, and it remained at this level for another week (i.e., for the rest of the time the composting mass was kept in the bioreactor) (Figure 1a). With FC_2 , thermophilic conditions (temperature > 55 °C) were obtained after 29 h of the experiment. After this, the temperature continued to rise to 65 °C and then started to decrease, although it remained above 55 °C up to the ninth day. Then, during the next 3 days, the temperature dropped to approximately 43–45 °C, where it remained until the 21st day (Figure 1c). With FC_3 , thermophilic conditions (temperature above 55 °C) were achieved after 43 h, and the highest temperature (66.2 °C) was recorded

on the third day of the process. Similar to composting with FC₁, the temperature remained above 55 °C up to the sixth day with FC₃. Next, during the next 6 days, the temperature decreased to about 40 °C and then to ca. 35–36 °C, where it remained until the end of composting in the bioreactor (Figure 1e).

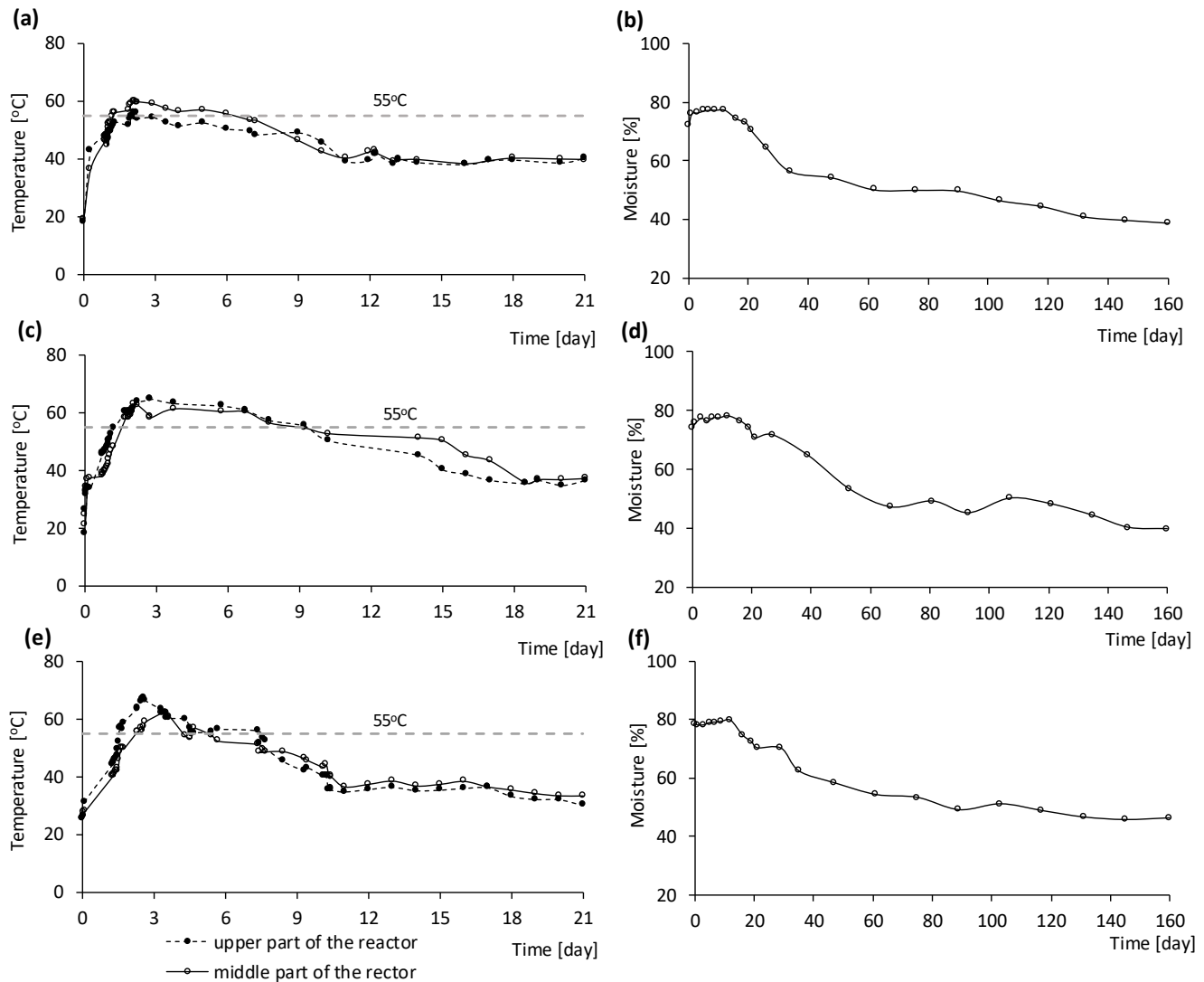


Figure 1. Temperature profiles (a,c,e) and changes in the moisture content (b,d,f) in the bioreactor during composting (until the 21st day in the bioreactor, and then in the windrow) of FC₁ (a,b), FC₂, (c,d), and FC₃ (e,f).

Although UE regulations state that a temperature of 55 °C or more should be maintained for at least 14 days, a temperature of 60 °C or more should be maintained for at least 7 days, a temperature of a 65 °C or more should be maintained for at least 5 days, and a temperature of 70 °C or more should be maintained for at least 3 days, in laboratory conditions, these temperatures last less time, regardless of the type of composted feedstock. This was the case in this study and others conducted in laboratory conditions. For example, Kulikowska and Bernat [5] found that, during composting of sewage sludge with willow-bark after salicylate extraction, thermophilic conditions lasted up to 5 days. Similarly, Bai et al. [4] observed that, although a higher share of cattle manure in a mixture with fermentative residue prolonged the duration of the thermophilic period during composting, the thermophilic phases lasted only 4–8 days. Kumar et al. [27] and Yuan et al. [28], on the basis of laboratory studies, also reported that the thermophilic phases were short: 5 days

when composting food waste with green waste and ca. 6–7 days when composting kitchen waste with dry cornstalks.

Since bacteria of the *Salmonella* genus were isolated from the sewage sludge, samples were taken to detect these bacteria after composting in the bioreactor was completed. No bacteria of the genus *Salmonella* were isolated from the samples taken from the bioreactor. This means that, in all FCs, the duration of the thermophilic phase was sufficient to hygienize the compost.

The changes in the moisture content during composting are shown in Figure 1. With FC₁, the initial moisture content in the feedstock was about 73% (Figure 1b). This relatively high humidity was a result of the high water content of the sewage sludge (89%). Although water is an important factor necessary for the growth of microorganisms, and its cooling ability prevents the compost from overheating, which would decrease the activity of microorganisms, the high moisture content and complete filling of the pores with water cause the composting material to have low structural strength and behave like a ductile material. Oxygen transfer is then limited, and practically no composting takes place. Transport of oxygen becomes possible, and composting can start if the pores are filled with air [29–32].

With FC₂, the initial moisture content, approximately 74% (Figure 1d), and the changes in moisture content during composting were similar to those observed with FC₁. With FC₃, the initial moisture content, approximately 76% (Figure 1f), was slightly higher than that of the other FCs. For the first 12 days of FC₃ composting, the moisture of the composted material increased slightly to 77–80%, and then it started to decrease. On the last day in the bioreactor, the moisture content with all the FCs was ca. 70%. After transfer to the windrow, the moisture content decreased intensively for the next 40 days (until the 60th day of the process), reaching approximately 50% with FC₁ and FC₂, and 54% with FC₃. With all the FCs, the moisture content was below 50% on the 90th day of the process. Throughout the process, the moisture content was monitored and regulated, so that it would not drop below 40%.

With all FCs and in both stages (bioreactor, windrow), the decrease in OM proceeded according to first-order kinetics. With FC₁, the concentration of OM in the feedstock was approximately 900 g/kg d.m. and, after 3 weeks of composting in the bioreactor, it decreased to ca. 800 g/kg d.m. (Figure 2). The maximum removal of organic matter in bioreactor A_b was 99 g/kg d.m. and the initial rate of organic matter removal, r_b, was 10.09 g/(kg d.m.·day) (Figure 2a), which caused an increase in the temperature of the composting material. The gradual depletion of organic compounds was accompanied by only a slight amount of OM removal in the windrow, which was 2.6-fold lower than in the bioreactor (A_w 37 g/kg d.m.) (Figure 2b). As a consequence, the rate of organic matter removal was almost sevenfold lower in the windrow than in the bioreactor. The final concentration of OM was 766 g/kg d.m. With FC₂, the course of organic matter removal in the bioreactor was similar to the course with FC₁; the maximum removal of organic matter in bioreactor A_b and the initial rate of organic matter removal, r_b, was equal to 93 g/kg d.m. and 9.67 g/(kg d.m.·day) (Figure 1c). In the windrow, A_w and r_w were 3.2-fold and 17-fold lower than in the bioreactor, respectively (Figure 1d). With FC₃, both the A_b and the r_b values in the bioreactor were 1.2–1.3-fold lower than those in FC₁ and FC₂ (Figure 1e,f).

The range of mineralization depends on the organic matter content in sewage sludge and the biodegradability of the organics used as amendments. For example, Meng et al. [9] showed that the OM contents in composted feedstock ranged from 65.3% to 67.3%, independently of the organics that were added (glucose, sucrose, starch, or cellulose). In all treatments, the OM degraded rapidly during the thermophilic phase of composting, and the OM contents dropped to 45.8% (control, sewage sludge without amendments), 44.8% (with glucose), 47.1% (with sucrose), 49.2% (with starch), and 51.3% (with cellulose). In other words, the respective losses of OM content with the control treatment and the cellulose treatment were 15.5% and 15.4%. However, the OM losses were higher with the glucose and sucrose treatments than with the other treatments: 21.9% and 20.2%, respec-

tively. Cardoso et al. [10] also reported similar decreases in OM content when composting sewage sludge with grasses and glucose or sucrose (22% less than in the initial sewage sludge). Although, in our study, the efficiency of OM removal was lower (10.7–14.6%) than in the above-cited study, it must be emphasized that we used lignocellulosic materials that improve not only the C/N ratio but also the porosity. Moreover, the decomposition products of the lignocellulose in substances such as these can serve as precursors for humic substances formation during cooling and maturation.

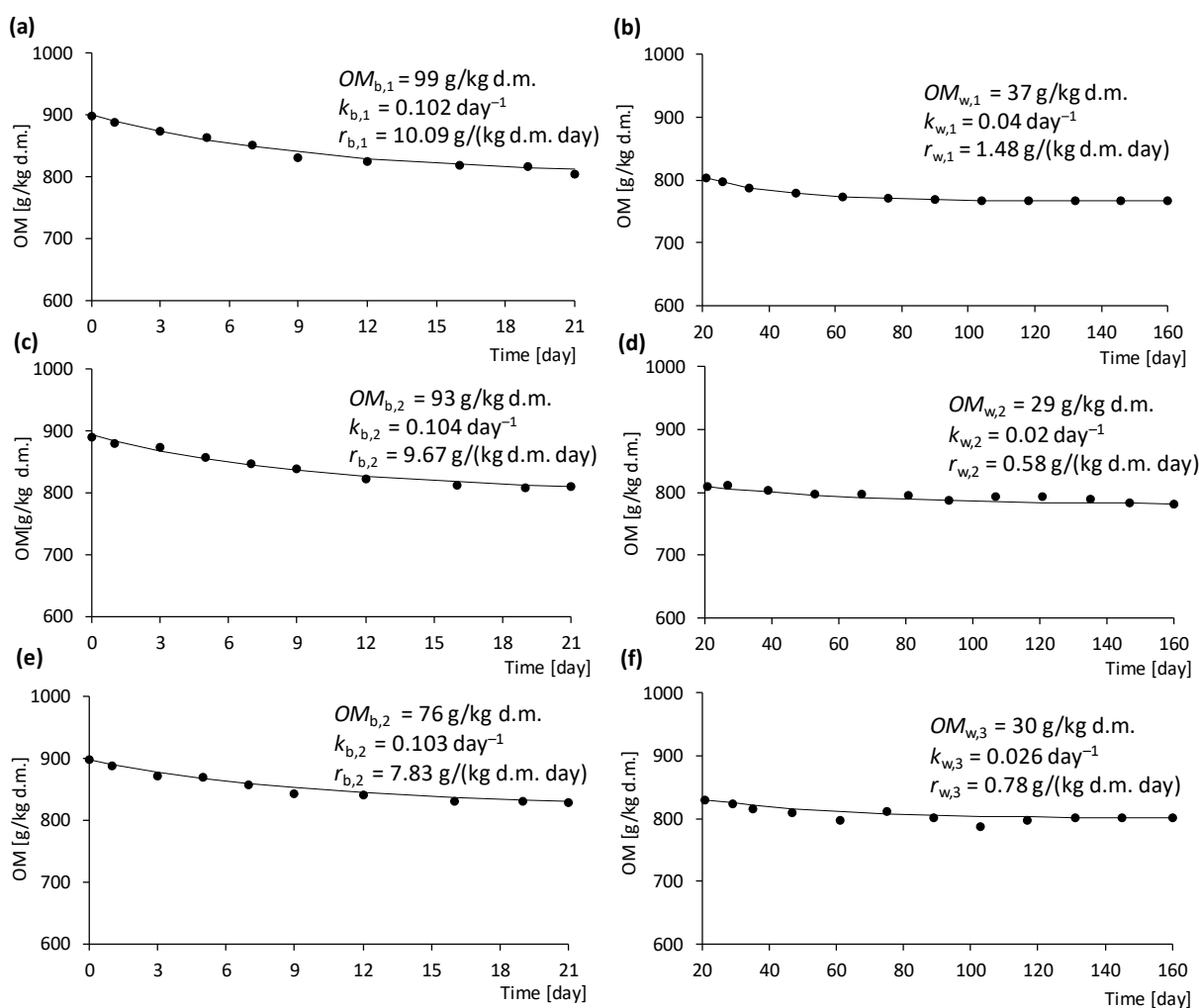


Figure 2. Changes in the organic matter (OM) content during composting (until the 21st day in the bioreactor, and then in the windrow) of FC₁ (a,b), FC₂, (c,d), and FC₃ (e,f) in the bioreactor (a,c,e) and the windrow (b,d,f); kinetic parameters are given, in terms of the maximal OM removal during the composting process (A_b —in the bioreactor, A_w —in the windrow), the rate constant of OM removal (k_b —in the bioreactor, k_w —in the windrow), and the rate of OM removal (r_b —in the bioreactor, r_w —in the windrow).

3.2. Nitrogen Profiles

During composting, the increase in the concentration of ammonia nitrogen was largest during the mineralization phase, when the decomposition of OM was the most intense. As the rate of organic metabolism decreases, the concentration of ammonia nitrogen also decreases, mainly due to the release of ammonia from the compost or its oxidation to nitrate nitrogen (V). Ammonia release is intensive in thermophilic conditions and with a high pH (>8.0). Nitrification leading to an increase in the concentration of oxidized forms of nitrogen begins after the temperature lowers to approximately 40 °C. In the

mature product, the concentration of nitrate nitrogen (V) is higher than the concentration of ammonium nitrogen.

During composting with FC₁, the concentration of ammonium nitrogen was highest on the fifth day of composting in the bioreactor (6.39 g N-NH₄/kg d.m.; Figure 3a). Then, its concentration gradually decreased to <0.1 g N-NH₄/kg d.m. in the mature product. As the concentration of ammonia nitrogen decreased, the concentration of nitrate nitrogen (III) and nitrate nitrogen (V) increased (Figure 3b,c), which meant that nitrification took place. Similar trends were noted with FC₂ and FC₃. With FC₂, the ammonia nitrogen concentration was highest on the third day, 6.1 g N-NH₄/kg d.m. and then it gradually decreased to 1.9 g N-NH₄/kg d.m. on the last day in the bioreactor and to ca. 0.1 g N-NH₄/kg d.m. in the mature compost. With FC₃, the ammonia concentration was highest on the 12th day at 5.09 g N-NH₄/kg d.m. On the last day in the bioreactor, it was equal to 2.07 g N-NH₄/kg d.m., whereas, in the mature compost, it was equal to 0.68 g N-NH₄/kg d.m., several times higher than the corresponding concentrations with FC₁ and FC₂. A similar increase in N-NH₄ concentration at the beginning of composting was noted by Liu et al. [12]. The initial N-NH₄ content in the two treatments was 3128.11 mg N-NH₄/kg d.m. On day 3, the N-NH₄ content in the two treatments increased to 9649.06 mg N-NH₄/kg d.m. and 7918.41 mg N-NH₄/kg d.m., respectively. With all FCs in the present study, nitrate nitrogen (V) dominated in the mature compost (concentrations of 1.21 g N-NO₃/kg d.m., 1.19 g N-NO₃/kg d.m. and 0.86 g N-NO₃/kg d.m. in FC₁, FC₂, and FC₃, respectively).

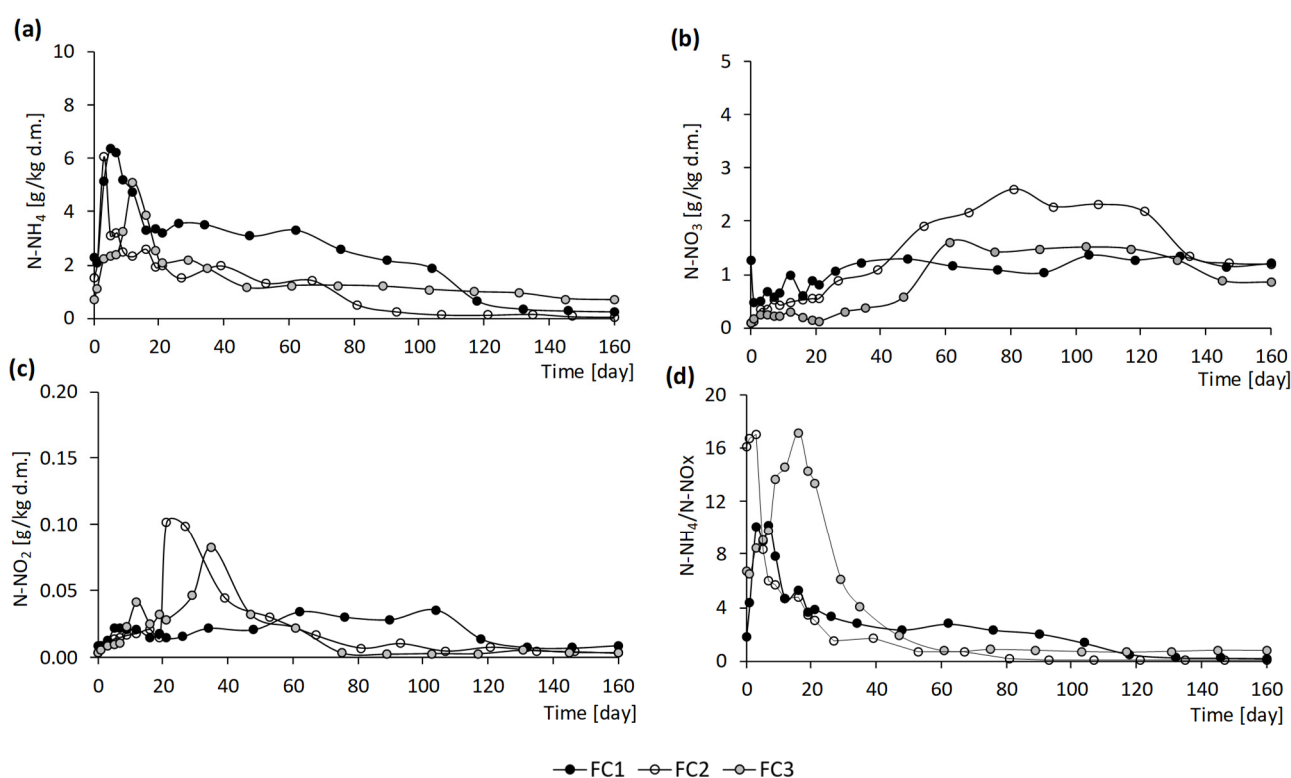


Figure 3. Changes in the nitrogen forms (a–c) and nitrification index ($N-NH_4/N-NO_x$) (d) during composting (until the 21st day in the bioreactor, then in the windrow).

The $N-NH_4/N-NO_x$ ratio, also known as the nitrification index (I_{nitr}), is an indicator of the maturation of organic compost. An I_{nitr} less than 0.5 indicates that the compost has reached full maturity; a value between 0.5 and 3 indicates that it has reached maturity; a value greater than 3 indicates that the compound is not mature [9]. In this study, the I_{nitr} was highest (up to 17) during composting in the bioreactor (Figure 3d), when intensive mineralization and ammonification took place. During maturation in the windrow, the

I_{nitr} gradually decreased, and a value below 0.5 was obtained after ca. 3 months with FC₁, after ca. 60 days with FC₂, and after ca. 70 days with FC₃. Cardoso et al. [10] showed that, during sewage sludge composting with grass, the I_{nitr} was 53.4 in raw sewage sludge, whereas, at the end of composting, it decreased to 0.101, indicating that the composted sewage sludge was highly mature.

3.3. Compost Stability

The stability of compost can be determined on the basis of the AT4 or SH_F values. This AT4 test is now commonly used to assess the degree of stability of a stabilizate from municipal solid waste. The condition for approval of stabilizate for landfilling is an AT4 value lower than 10 mg O₂/g d.m.; in this paper, this value was also applied as the limit when assessing the stability of the compost. A higher AT4 value reflects higher oxygen demand and indicates a higher microbiological activity, which results from the availability of readily biodegradable organics and the ongoing mineralization of OM during composting. With all the FCs used for composting, the AT4 value for the feedstock was approximately 70 g O₂/kg d.m. and it gradually decreased during the composting process. After FC₁ composting in the bioreactor, the AT4 value decreased more than twofold, reaching 32 g O₂/kg d.m. After FC₂ composting in the bioreactor, this value decreased 2.7-fold, to 26 g O₂/kg d.m. However, after this stage of FC₃ composting, the AT4 value only reached 44 g O₂/kg d.m., indicating that it was the most unstable compost at this time. After 60, 45, and 70 days of composting with FC₁, FC₂, and FC₃, respectively, AT4 values below 10 g O₂/kg d.m. were achieved. A similar trend was observed by Bożym and Siemiatkowski [33] during composting of sewage sludge, straw, wood chips, and sieved recirculated compost from a previous composting process. After 2 weeks of maturation the sample displayed high respiration activity, as shown by an AT4 value of 19.8 g O₂/kg d.m. During maturation, the AT4 value decreased; after 2 months, it was 6.5 g O₂/kg d.m. while, after 3 months, it was 4.6 g O₂/kg d.m.

SH_F, the maximum temperature reached by the sample (feedstock or compost sample taken from bioreactor/windrow) in a Dewar vessel, may also be used as an indicator of aerobic biological activity. According to the guidelines of the Regulation (EU) 2019/1009 of the European Parliament and of the Council of 5 June 2019 [15], the minimum required compost stability index is III (Rottegard III), which means an increase in temperature in a Dewar vessel of 10–20 °C [17]. With all FCs, the temperature of the samples taken on the last day of composting in the bioreactor increased in the Dewar vessel by 17–18 °C above the ambient temperature, which indicates class IV compost stability (moderately stable, curing compost, according to the classification given by Brinton et al.) [17]. In the samples collected in the initial stage of maturation in the windrow, only slight self-heating of the compost was noted; the temperature increase did not exceed 5–6 °C, which corresponds to class V compost stability (very stable, well-aged compost, according to Brinton et al. [17]. Finally, after approximately 30–35 days of maturation, no temperature increase was noted in the samples in the Dewar vessel.

3.4. Humification Progress

The HS content, the most stable form of organic carbon in compost, affects the quality of the compost and has a positive effect on soil fertility and carbon sequestration in soil. It is also known that the concentration of HS increases during the composting process; however, there is a period of composting time in which HS is formed most intensively. To find this time, an analysis of humification progress should be carried out. In the present study, the concentrations of HS and HA were determined during the time of composting, and the kinetic constants of HS and HA formation were determined.

With all feedstocks, the concentration of HS was relatively high (above 140 mg C/g OM), whereas the concentration of HA did not exceed 20 mg C/g OM (Figure 4). This means that, in the HS, the fulvic fraction (FF) predominated.

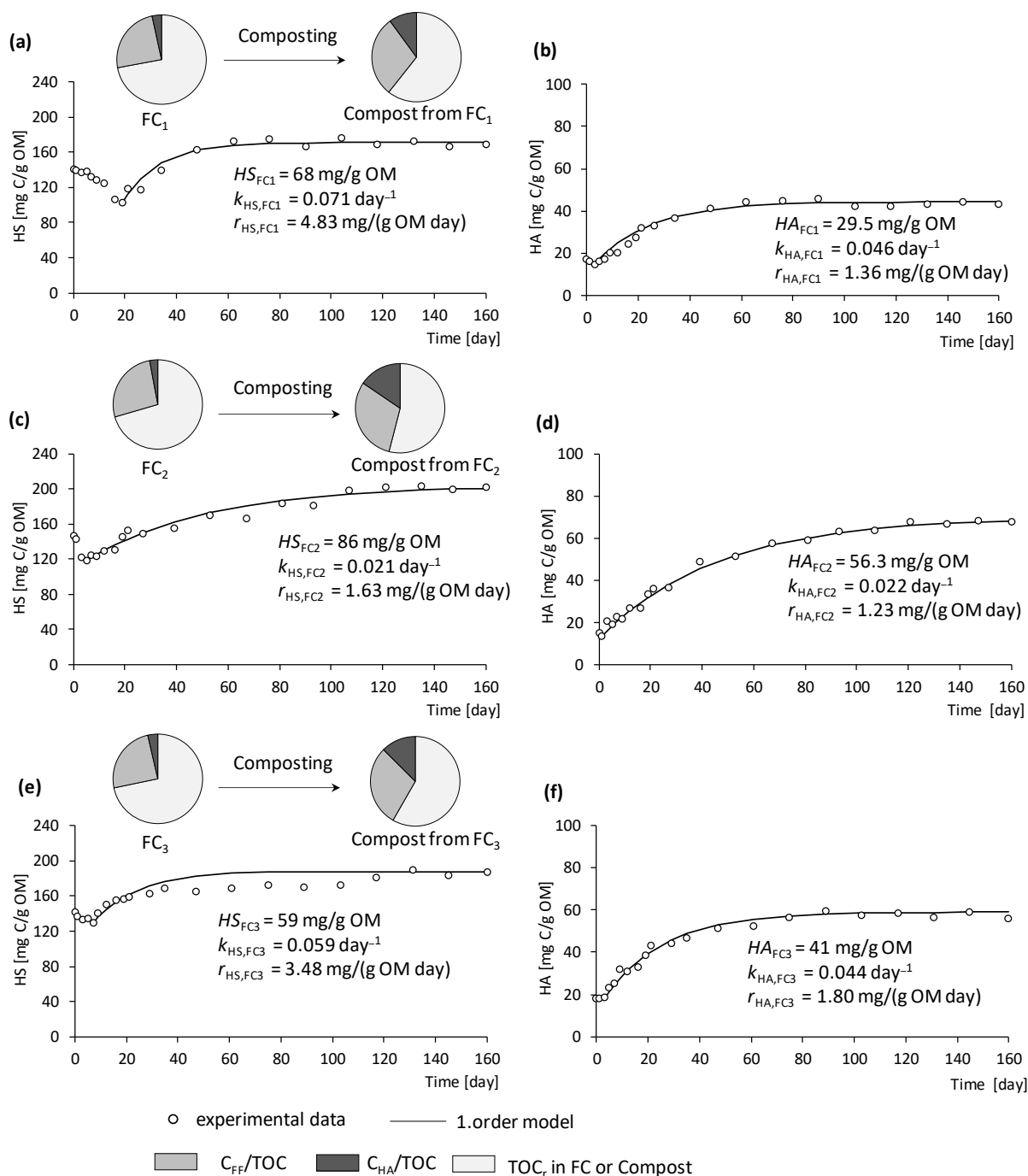


Figure 4. Changes in the concentration of HS (a,c,e), HA (b,d,f) during composting (until the 21st day in the bioreactor, and then in the windrow); kinetic parameters are given, in terms of the maximal increase in the concentration of HS ($C_{\max,HS}$), HA ($C_{\max,HA}$), the rate constant of HS (k_{HS}) and HA (k_{HA}) formation, and the rate of HS (r_{HS}) and HA (r_{HA}) formation; in the form of a circle, the share of carbon (C) in FF in the total organic carbon (TOC) of the feedstock or compost (C_{FF}/TOC , %), the share of C in HA in the TOC of the feedstock or compost (C_{HA}/TOC , %), and TOC_r (the remainder of the total organic carbon in the feedstock or compost) are presented.

During composting, after a short time at the beginning of the process when the HS content decreased (20, 5, and 10 days for FC₁, FC₂, and FC₃, respectively), the content of these substances started to increase. Similarly, the content of HA increased after a short-term decrease (1–2 days) at the beginning of the process. The respective contents of HS and HA increased to 168.7, 201.3, and 187.5 mg C/g OM, and to 43.3, 67.8, and

56.3 mg C/g OM with FC₁, FC₂, and FC₃, respectively. In all FC composts, however, FF predominated. The share of HA and FF in SH extracted from mature compost depends on feedstock composition, mainly the amendments added [34]. Becher et al. [35] stated, for example, that, in mature compost from municipal waste (collected selectively and organic fractions from waste sorting facilities), among HS, the HA prevailed, with a ratio of humic to fulvic carbon from 2.07 to 3.03.

In this study, HS and HA formation proceeded according to a first-order kinetic model. The kinetic constants of HS and HA formation had the same order of magnitude and ranged from 0.021 to 0.079 day⁻¹. The highest rate of HS formation, 4.83 mg C/(g OM·day), was found during composting of FC₁, whereas, with FC₂ and FC₃, the rates were lower, equal to 1.63 and 3.48 mg C/(g OM·day), respectively. As for the rates of HA formation, the values did not differ much, ranging from 1.23 to 1.80 mg C/(g OM·day). The highest net increase in HS (86.0 mg C/g OM) and HA (56.3 mg C/g OM) was noted in FC₂; in other FCs, the concentrations of these compounds were from 1.3-fold to 1.5-fold (HS) and 1.4-fold to 1.9-fold (HA) lower.

The shares of (i) carbon (C) in FF to the total organic carbon (TOC) in feedstock or compost (C_{FF}/TOC , %) and (ii) C in HA to the TOC in feedstock or compost (C_{HA}/TOC , %) were determined for each FC and the resulting composts. It was found that C_{FF}/TOC and C_{HA}/TOC were lower in the FCs (24.5–26.5% and 2.9–3.6%, respectively) than in the composts (29.2–30.5% and 10.1–15.5%, respectively). The highest proportion of C_{HA}/TOC was achieved in FC₂, which means that, in FC₂ compost, there was the largest share of the most stable form of organic carbon.

3.5. Compost Quality

The compost had a dry matter content of 62.7–63.6%, a relatively high content of organic matter (76.6–80.2%), and a high content of fertilizing elements (NPK). The composting product, in terms of its content of heavy metals, met the currently applicable requirements contained in the abovementioned regulation (Table 4).

Table 4. Compost characteristics.

Characteristics	Unit	FC ₁	FC ₂	FC ₃
<i>Basic analysis of physicochemical characteristics and nutrient contents</i>				
Moisture	%	37.3	36.8	36.4
Dry mass (d.m.)	%	62.7	63.2	63.6
OM	% d.m.	76.6	78.2	80.2
Reaction	pH	6.66	6.32	6.20
N		29.1	31.4	32.6
P		12.60	12.87	13.60
P ₂ O ₅		28.9	29.4	31.2
K ₂ O		14.56	15.67	16.28
K	g/kg d.m.	12.08	13.06	13.54
Mg		4.41	3.92	4.83
MgO		7.32	6.34	8.01
Ca		17.30	18.84	18.51
CaO		24.2	26.3	25.9
<i>Heavy-metal contents</i>				
Cd (5 *)		1.23	1.40	4.70
Pb (140 *)		6.1	6.2	7.4
Ni (60 *)		10.2	9.6	11.1
Cr (100 *)	mg/kg d.m.	22.4	20.0	21.1
Cu (n.r. *)		102.3	97.5	94.6
Zn (n.r. *)		734	806	761
Hg (2 *)		0.0940	0.1128	0.1230

* According to Regulation of the Minister of Agriculture and Rural Development of 18 June 2008 (item 765) on the implementation of certain provisions of the Act on fertilizers and fertilization (permissible content of pollutants in organic and organic–mineral fertilizers, as well as organic and organic–mineral agents supporting the cultivation of plants [36] (n.r.—not regulated).

4. Conclusions

All (FCs), i.e., sewage sludge with wood chips and wheat straw (FC₁), sewage sludge with energy willow and wheat straw (FC₂), and sewage sludge with pine bark and conifer sawdust (FC₃), enabled producing stable and mature compost. However, with FC₂, the thermophilic phase lasted longer, and some parameters of stability and maturity were obtained earlier (AT₄, I_{nitr}). Furthermore, the highest net increases in HS and HA in FC₂ were from 1.3- to 1.5-fold (HS) and from 1.4- to 1.9-fold (HA) higher than in other FCs. The highest C_{HA}/TOC (15.5%) in FC₂ means that, in FC₂, the compost had the largest share of the most stable form of organic carbon. The results indicate that, through the choice of the amendments and bulking agents, the length of the composting time needed to obtain a stable product with a high content of HS and HA can be controlled.

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