Research Article

Received: 18 October 2022 Revised: 28 February 2023 Accepted article published: 11 March 2023 Published online in Wiley Online Library: 29 March 2023

(wileyonlinelibrary.com) DOI 10.1002/jctb.7370

Differentiation of the composting stages of green waste using the CIELAB color model

Dimosthenis Tsivas,[a] Apostolos Vlyssides[a] and Anestis Vlysidis[a,b*]

Abstract

BACKGROUND: CIELAB color variables can be used to monitor a composting process and evaluate the maturity of the compost with a new, rapid, easy, and low-cost colorimetric analysis. However, to date there are no available data to support the ability of CIELAB color changes to depict the different stages of a composting procedure. This study aims to examine the correlation of CIELAB color variables with composting time to elucidate how color changes can be used to detect the different stages of a composting process. Two green waste industrial scale composting processes with different added materials were monitored using typical physicochemical and CIELAB color analyses.

RESULTS: During composting, color variables a*, b*, C*, and ΔE* exhibited fluctuations following a constant variation trend that correlated with each composting phase. This behavior depicts the transformation of the organic composition of compost, as described by Organic Carbon (OC), Carbon-to-Nitrogen ratio (C/N), Humic Acids (HA), and Fulvic Acids (FA). Moreover, color variables a*, b*, and C* showed strong relationships with OC, C/N, and HA/FA ($R^2 > 0.83$) and with HA ($R^2 > 0.74$). These results indicate that CIELAB color change follows the same general pattern for each composting procedure that utilizes the same main composting substrate, regardless of any differing additional materials.

CONCLUSION: Monitoring the CIELAB color variables made it possible to depict the different phases of composting, especially the transformation of the organic composition of the compost. Accurately monitoring CIELAB color variables distinguishes the different stages of a composting process through a rapid analysis at radically reduced costs compared to complex physicochemical analyses.

Supporting information may be found in the online version of this article.

Keywords: CIELAB color model; composting stages; green waste composting; maturity

INTRODUCTION

A major environmental issue that affects many developing and developed countries worldwide is the increase of Green Waste (GW) generated due to the growth of urban green areas.[1] Along with other organic waste (i.e., food waste), GW constitutes one of the highest fractions of municipal solid waste.[2] Specifically in the United States, GW (such as residential yard trimmings) accounts for 13.5% of the estimated annual total municipal solid waste being produced.[3] GW consists mainly of grass, leaves, tree trimmings, bark, pruning from young trees and shrubs, and garden litter, and it is mostly generated in municipal parks, gardens, reserves, and domestic yards and gardens.[4] GW management is often regarded as difficult due to the high principal collection and transportation costs to landfills or treatment facilities because of its low bulk density.[5] Recent data shows that only 40% of GW is effectively recycled into a high-quality compost and digestate, while the remaining is still disposed through landfilling or incineration.[6] Moreover, according to Al-Alawi *et al.*[4] inappropriate disposal of untreated GW, especially in landfills, leads to water and soil pollution that threatens environmental and human health.[4] Additionally, the European Union has set a goal to decrease municipal waste landfill disposal to 10% or less of the total amount of municipal waste generated by weight by 2035[7] with the immediate expansion of several environmentally friendly treatment methods.

A promising method applied for treating GW is composting, which is described as the biological decomposition of organic matter under controlled aerobic conditions.[8] Composting is a discontinuous bioprocess consisting of four phases. In the first phase, called the mesophilic phase, easily degradable compounds are decomposed at temperatures that usually remain under

* Correspondence to: A Vlysidis, School of Chemical Engineering, National Technical University of Athens, 15780, 9, Heroon Polytechniou St., 15780 Zografou, Greece. E-mail: *anestisvlysidis@chemeng.ntua.gr*

a *School of Chemical Engineering, National Technical University of Athens, Zografou, Greece*

b *School of Chemical and Environmental Engineering, Technical University of Crete, Chania, Greece*

45 °C.[9] During the second phase, called the thermophilic phase, decomposition continues quickly and accelerates until temperatures reach about 60–70 °C.[9] At the end of this phase, the thermophilic organisms decline due to exhaustion of substrates and the temperature starts to decrease. The two final phases of composting are the cooling and maturation phases, where the temperature remains lower than 45 °C. According to Diaz and Savage,[9] the coolik.ng phase is characterized by an increasing number of organisms that degrade starch or cellulose. During the maturation phase, the less easily degradable compounds are degraded at a low rate until compounds, such as lignin-humus complexes that do not degrade further, form and predominate.[9] Many studies have been conducted examining GW composting with and without added materials (i.e., bio-solids, seaweed, food waste, and zeolite).[1,4,10–15] Some of the aforementioned added materials include mature compost, compost tea, and zeolite. Mature compost is predominantly used in horticulture and agriculture to improve soil conditions and thereby enhance plant growth.[8] Compost tea is mainly used as an alternative to chemical fertilizers and pesticides.[16] Zeolite is applied to composting due to its notable cation exchange capacity, which improves soil quality and facilitates the growth of microbial populations with its enhanced surface area and porosity.[11] The compost obtained from GW treatment is an organic soil amendment and/or organic substrate that can be applied in almost all crops within the context of a circular economy[17] and can help solve the disposal problem of GW in landfills that has emerged in many countries.[2] Moreover, due to their usually low content of micro-pollutants, GW can produce a compost product rich in nutrients. This product can be used to maintain a suitable soil structure and to add organic material to soil whose organic matter content has been reduced by intensive agriculture,[9] in accordance with sustainable organic farming regulations and restrictions.[18]

GW composting has been reported to last between 21 to 220 days,[1,10,12,14,19] depending on handling conditions (i.e., oxygen supply, humidification, nutrient balance) and proper monitoring of the procedure.[2] GW are resistant to biodegradation or exhibit a slow degradation rate due to their high content in lignin, hemicellulose, and cellulose. Therefore, the production of a mature final product for commercial use can usually take several months. Compost maturity is essential for its use as a soil amendment since immature and poorly stabilized composts can lead to a series of problems with storage, use, and marketing.[20] According to many studies, compost maturity evaluation can be achieved by monitoring certain indices, such as C/N ratio, phytotoxicity, ammonia concentration, oxygen uptake, pH, and humic substances content.[8,9,12,20–24] Iglesias Jiménez and Pérez García[22] reported that a C/N ratio between 15 and 20 characterizes mature composts. Moreover, CCQC[20] has stated that a compost is considered mature if the C/N ratio is lower than 25. However these ranges are not absolute for all composting substrates and, therefore, the C/N decline should be monitored until a stabilization point is reached at the end of composting.[8,21] pH values have been observed to increase during composting and stabilize at alkaline values, usually between 7 and 9.[9] Moreover, compost maturity is achieved when the ammonia concentration and ammonia to nitrates ratio values are lower than 75 ppm[21] and 0.5,[25] respectively. However, if both levels of ammonia and nitrates are low in compost (i.e., less than 250 ppm), their ratio is not a reliable measure of maturity.[20] The humification progress has also been reported as a means to evaluate compost maturity. During composting, fulvic acids (FA) are formed as an intermediate step for the formation of humic acids (HA). According to recent studies,[9] the fraction of humic substances (HA/FA) should be greater than 1–3 in mature composts.[24] Another parameter is the oxygen uptake rate, whose monitoring is based on the respiratory activity of the microorganisms in the compost, as mentioned by Diaz and Savage.[9] As composting moves towards its maturation stage, the reduction and stabilization of oxygen uptake is an indicator of compost stability. Mature composts demonstrate oxygen uptake values lower than 0.4 mg O_2/g h.[20,25]

Most of the aforementioned indices require expensive and time consuming analyses of questionable reliability, making compost maturity evaluation impractical.[25] As mentioned before, GW composting has a wide duration range (21–220 days), therefore analyses need to be conducted regularly over an uncertain time period. This may lead to significant costs for compost analyses. To avoid operating expenses, composting evaluation is often compromised. This may result in the production of immature composts that may inhibit seed germination and reduce plant growth.[26]

Expensive and time-consuming analyses can be partially replaced with the use of a direct and rapid analysis that the CIELAB color model can offer.[23,27] CIELAB color variables are already being used to characterize the organic matter composition of soil. Results have revealed that these variables can be accurately correlated with organic carbon, humic acids, and minerals in soil.[23,28] Recent studies have measured compost color using the CIELAB color model and have correlated color change with certain composting parameters for different composting substrates (such as a mixture of green tea waste with rice barn[23] and three phase olive pomace[27]). As shown by these studies, even though most CIELAB color values appear to exhibit constant variation trends during composting, their absolute values seem to be greatly affected by the inherent characteristics of the raw materials. While CIELAB color variables were shown in previous studies to be correlated with the organic carbon content of mature composts,[29] no study has provided any data on the ability of certain CIELAB color variables to reflect the different stages of a typical composting procedure.

The current study is based on previous data, which reported the strong correlation of certain CIELAB color variables with the organic composition of soil and the fact that compost color shows a constant variation trend during composting.[27] In this study the relationship between CIELAB color variables and certain physicochemical variables that describe the transformation of organic composition, namely OC, C/N, FA, and HA, was investigated. To do so, the CIELAB color variables were monitored during GW composting with various added materials. The main goal was to examine the correlation of CIELAB color variables with composting time and elucidate if color change can depict the transformation of compost organic composition. This would allow for detecting the different stages of a GW composting process up to the point of maturity. This easy, fast, and low-cost analysis can vastly decrease the operational costs of a composting process. It is also noteworthy that this method can be immediately adopted in all composting facilities.

EXPERIMENTAL

Materials and Methods

All composting processes examined in this work contained GW as the main substrate, along with different added materials, such as zeolite, mature compost, and compost tea. GW consisting of fallen leaves and branch cuttings generated by urban landscape maintenance was sourced from three local municipalities in the province of Attica. Once received, GW was cut and passed through a woodchipper with a mesh diameter of 2.5 cm to increase the composting active surface. Zeolite, obtained from the market, consisted of 90% clinoptilolite and had a specific granulometry of less than 2 mm.

Compost tea preparation

The preparation of compost tea was based on an adaptation of the method described by Pant *et al.*[30] Specifically, 186 kg of the final GW composting product was added to a 1.2 m^3 reactor, along with 820 kg of tap water and 2 kg of molasses. The tap water was initially pretreated in an aerated vessel (1.2 m^3) with constant aeration for 24 h to reduce any possible chlorine concentration from the local water supply system. Molasses was used as substrate for microbial growth. The final mixture was constantly aerated and mixed at a constant temperature of 25 °C for 2 days. Maximum microbial growth was usually achieved after 24 h when the pH value reached 7.5. When maximum microbial population was achieved, the liquid fraction of the mixture was decanted and applied on the composting procedure.

Composting process

Compost pile preparation involved the initial mixing of GW with certain added materials, as listed in Table 1. Two different composting processes, both of which lasted for 112 days, were conducted using GW collected during autumn 2020. Once collected and transported to the composting facilities, GW was immediately composted. All composting procedures were conducted in an industrial scale open windrow system. Gore covers were applied to reduce significant heat losses and unwanted interference from weather conditions (i.e., heavy rains and snow). During composting, moisture levels were adjusted to approximately 50% by adding water when necessary. Turning of the piles was conducted every 5 days, depending on weather conditions, to maintain the oxygen concentration in the middle of the pile at around 5%,[24] to ensure uniformity of decomposition, and to avoid anaerobic conditions. Similar GW composting techniques have successfully been implemented by previous composting studies.[2,11,12]

Sample collection and physicochemical analysis of compost

Compost samples were collected every week according to the methods described by Thompson.[25] Sample collection and initial preparation were conducted following the procedures described by Tsivas *et al.*[27]

Temperature was measured at different points of the composting pile at approximately 30 cm depth using an electrical thermocouple. Microbial population, as general microbes, was measured using the plate count method described by Liu *et al.*[31] Oxygen uptake rate (OUR) was evaluated from CO_2 production using a modified method by Thompson[25] and described by Tsiodra *et al.*[32] Total Kjeldahl Nitrogen (TKN) was measured using TMECC Method 4.02-A,[25] as described by Tsivas *et al.*[27] Organic carbon (OC) was measured through combustion oxidation at high temperatures (900 °C). The detection of produced carbon dioxide was measured using TMECC Method 4.01-A.[25] pH was measured using a 1:2 (v/v) compost to water ration.[27] Ammonium (NH_4N) and nitrate (NO_3N) nitrogen content were measured with TMECC Methods 4.02-C and 4.02-B,[25] respectively, using a 1:5 (w/v) compost to KCl (2 M) solution slurry ratio. Humic acid (HA) and fulvic acid (FA) contents were determined according to the method described by Velasco et al.[33] Germination Index (GI) was measured using the method described by Zucconi *et al.*[34] with *Lepidium sativum* seeds. The average values of all physicochemical properties and their corresponding standard deviations were derived from three measurements.

Table 1. Characteristics of the composting procedures

Compost process	Compost 1	Compost 2
GW (m^3)	9	9
Mature compost (m^3)	0	1
Zeolite (kg)	0	20
Compost tea (kg)	6.5	0
GW collection period	Autumn 2020	Autumn 2020
GW moisture (%)	46.5	46.5

Compost color analysis

Dry compost samples were used to perform the color analysis using the CIELAB model. This color model describes color by three coordinates (L*, a*, b*). Color lightness is described by color variable L* with values ranging from 0 (black) to 100 (white). The green/red component of color is represented by color variable a*, whose values range from negative (green) to positive (red). The blue/yellow color component is described by variable b* with values ranging from negative (blue) to positive (yellow).[35] Other CIELAB color variables, such as L* difference ΔL, a* difference Δa, b* difference Δb, chromatic quotient a*/b*, Chroma C*, hue angle h^0, and total color difference ΔE*, can be derived as described by Tsivas *et al.*[27]

A portable colorimetric device was used to measure color. Specifically, a Dr. Lange spectrophotometer (Hach Lange LMG 183) with standard circular viewing geometry d/80, an illuminant D65/100, and two calibration standards (LZM 268) — one black ($X = 4.07$, $Y = 4.35$, $Z = 4.59$) and one white ($X = 83.78$, $Y = 88.60$, $Z = 90.39$), was employed. The compost color measurement procedure used in this study is the one described by Tsivas *et al.*[27]

Statistical analysis

Regression analysis was used to estimate the relationship of CIELAB color variables with composting time and with the studied physicochemical composting properties. Various equations were tested to determine the best fitting ones, with the strongest relationship as expressed by Pearson's correlation coefficient, R^2 (i.e., the highest value).

RESULTS AND DISCUSSION

Physicochemical parameter progression during the composting processes

During this study, two composting processes with different added materials were performed, as shown in Table 1. Zeolite was used due to its cation exchange capacity, which improves soil quality. Mature compost and compost tea were employed to assist biological transformation by providing the necessary microbial organisms at the beginning of composting. The initial physical, chemical, and microbiological characteristics of the raw materials are presented in Table 2. The progression of physicochemical properties for the two composting processes is presented in Figs 1–4.

Both compost experiments reached high temperature levels (above 45 °C) very quickly in only 6 days after the initiation of

Table 2. Physical, chemical, and microbiological characteristics of the raw materials

Raw materials	GW	Mature compost	Compost tea	Zeolite
Bulk density (g cm^{-3})	0.13	0.42	-	0.72
pH	6.22	8.82	7.51	12.2
GI (%)	71.23	124.50	79.46	-
General microbes (CFU/mL)	-	-	1×10^8	-
General microbes (CFU/g)	3×10^7	4×10^7	-	-
Moisture (%)	46.5	32.84	-	3.44

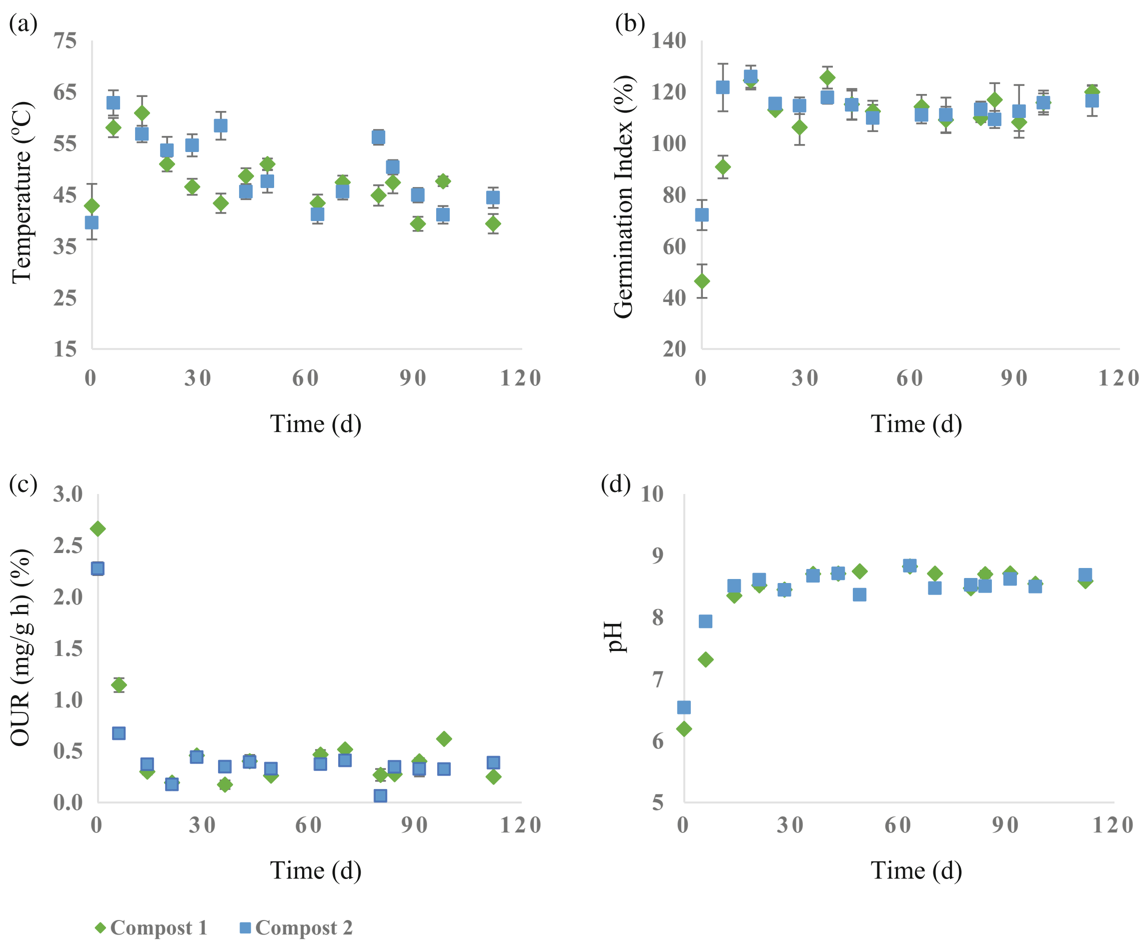

Figure 1. Temperature (a), Germination Index (b), oxygen uptake rate (c), and pH (d) progression during the two composting experiments (Compost 1, 2).

the experiments, indicating that the mesophilic phase of composting lasted from day 0 to day 6. The temperature of Compost 1 (Fig. 1(a)) remained high (>50 °C) for 36 days and then stabilized above 40 °C for the remaining composting period. This indicates that the thermophilic phase of composting lasted from day 6 until day 36. After that day, the cooling and maturation phases took over. Similarly, the temperature of Compost 2 remained high (>50 °C) until day 43, at which point the thermophilic phase ended and the two subsequent phases of cooling and maturation began (lasting until the end of the process). Typically, after the completion of the cooling phase and before the initiation of the maturation phase, compost temperature is below 30 °C.[9] As this was not the case in the compost trials of the current study (temperature values were higher than 35 °C after the thermophilic phase for both composting experiments), it can be assumed that the last two phases of composting intertwined.

Based on GI (Fig. 1(b)), NH_4N (Fig. 3(a)), and pH (Fig. 1(d)) values, the detoxification of Composts 1 and 2 can be identified at days 21 and 14, respectively. At these moments, the GI, NH_4N, and pH values stabilized. NH_4N decreased from 334.57 to 21.92 mg kg^{-1} (Compost 1) and from 256.79 to 17.05 mg kg^{-1} (Compost 2) primarily due to its volatilization to NH_4.[36] This also caused an increase in pH values that then stabilized at around 8.45–8.60 in both composting experiments. The reduction of NH_4 also lead to the disappearance of phytotoxicity,[37] which is expressed by the increasing GI values that reached a maximum of 125.63%

J Chem Technol Biotechnol 2024; **99**: 2198–2208 wileyonlinelibrary.com/jctb
Journal of Chemical Technology and Biotechnology published by John Wiley & Sons Ltd on behalf of Society of Chemical Industry (SCI).

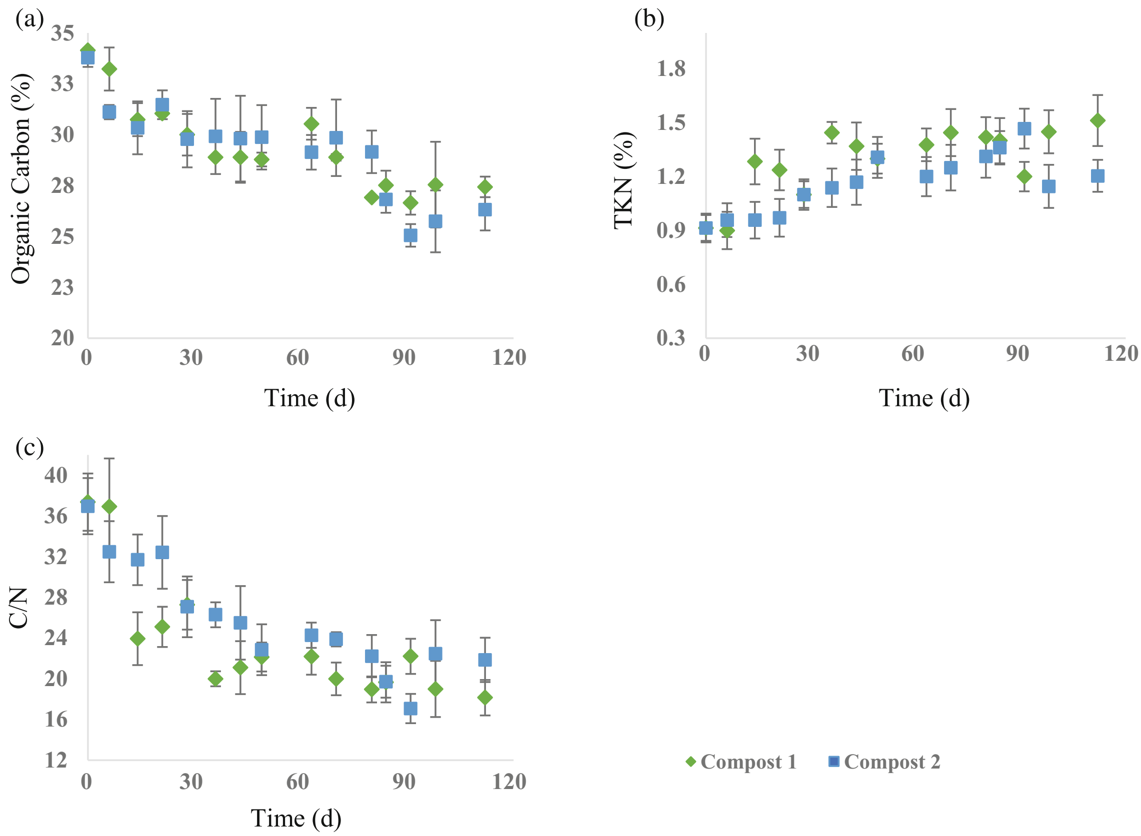

Figure 2. Organic carbon (a), total Kjeldhal nitrogen (b), and carbon to nitrogen ratio (c) progression during the two composting experiments (Compost 1, 2).

and 126.01%, for Composts 1 and 2, respectively. In both compost piles, NO_3N (Fig. 3(b)) values exhibited a constant decrease from 165.71 to 24.73 mg kg^{-1} until day 43 for Compost 1 and from 119.77 to 26.18 mg kg^{-1} until day 63 for Compost 2. Afterwards, these values stabilized.

Moreover, OUR (Fig. 1(c)) values indicate that high microbial activity for Composts 1 and 2 lasted for 21 and 14 days, respectively, and then remained stable at lower levels for the rest of the composting duration (with values at around 0.25–0.60 mg g^{-1} h^{-1} and 0.35–0.45 mg g^{-1} h^{-1} for Composts 1 and 2, respectively).

In addition, organic carbon degradation lasted 80 days for Compost 1 and 91 days for Compost 2. OC (Fig. 2(a)) values decreased from 34.14% to 26.92% and from 33.79% to 25.05% for Composts 1 and 2, respectively. During the two first phases of composting (mesophilic and thermophilic), OC values decreased rapidly and then reached a temporary stabilization point at the beginning of the cooling phase. After 63 days for Compost 1 and 70 days for Compost 2, OC values again showed a rapid decrease accompanied by a decrease in FA (Fig. 4(b)) values and an increase in HA (Fig. 4(a)) values. The TKN (Fig. 2(b)) values of Compost 1 increased and reached a stabilization point at day 36 at around 1.44%, while for Compost 2 the TKN continued to increase until day 91 and reached a maximum of 1.47%. Therefore, the C/N ratio (Fig. 2(c)) showed a more rapid decrease for Compost 2 until day 91 (C/N = 17.08), while for Compost 1 its decrease became less rapid after 36 days but continued until its stabilization (after 80 days, at around 18 to 20). The formation of humic substances began after 36 days (Compost 1) and 28 days (Compost 2), with the increase of FA (Fig. 4(b)) initiating the maturation phase, as described by Guo and Liu.[38] This indicates that, for Compost 1, the cooling and maturation phases occurred at the same time. For Compost 2, the production of FA after 28 days of composting indicates that the maturation phase began a few days before the completion of the thermophilic phase and that it continued after day 43, along with the cooling phase. The production of FA reached a maximum of 127.22 mg kg^{-1} and 153.19 mg kg^{-1} for Composts 1 and 2, respectively. Its subsequent decrease coincided with a simultaneous increase of HA (Fig. 4(a)) that continued until the end of both composting processes, with maximum values of 111.22 mg kg^{-1} (Compost 1) and 142.27 mg kg^{-1} (Compost 2). The initiation of the humification process is also indicated by the HA/FA (Fig. 4(c)) ratio, which reached its lowest value after 49 days (HA/FA = 0.42) for Compost 1 and 36 days (HA/FA = 0.59) for Compost 2. As the humification progressed, HA/FA values increased for both Composts throughout the remaining period.

In both composting experiments, the progression of each physicochemical parameter that was monitored agreed with data provided in the literature. The mesophilic stage ($T < 45$ °C) of composting was completed only a few days after the initiation of the process. After this stage, high temperature levels (>50 °C) occurred during the entire thermophilic phase, and after this point, it decreased gradually as the biodegradation of organic matter was stabilized.[4,8] The biodegradation of organic matter involves an OC decrease,[39] a C/N decrease, and a HA/FA increase.[13] As composting progresses, organic carbon is reduced due to the decomposition of the raw materials.[9] As a result, the C/N ratio decreases until it reaches the point of maturity.[12] NH_4N presents a decreasing trend partially due to the nitrification process and mostly due to its volatilization to NH_3, which also causes an increase in pH values.[37] Moreover, NO_3N losses that were monitored in this study can be attributed to the fact that denitrification occurs at a higher rate than

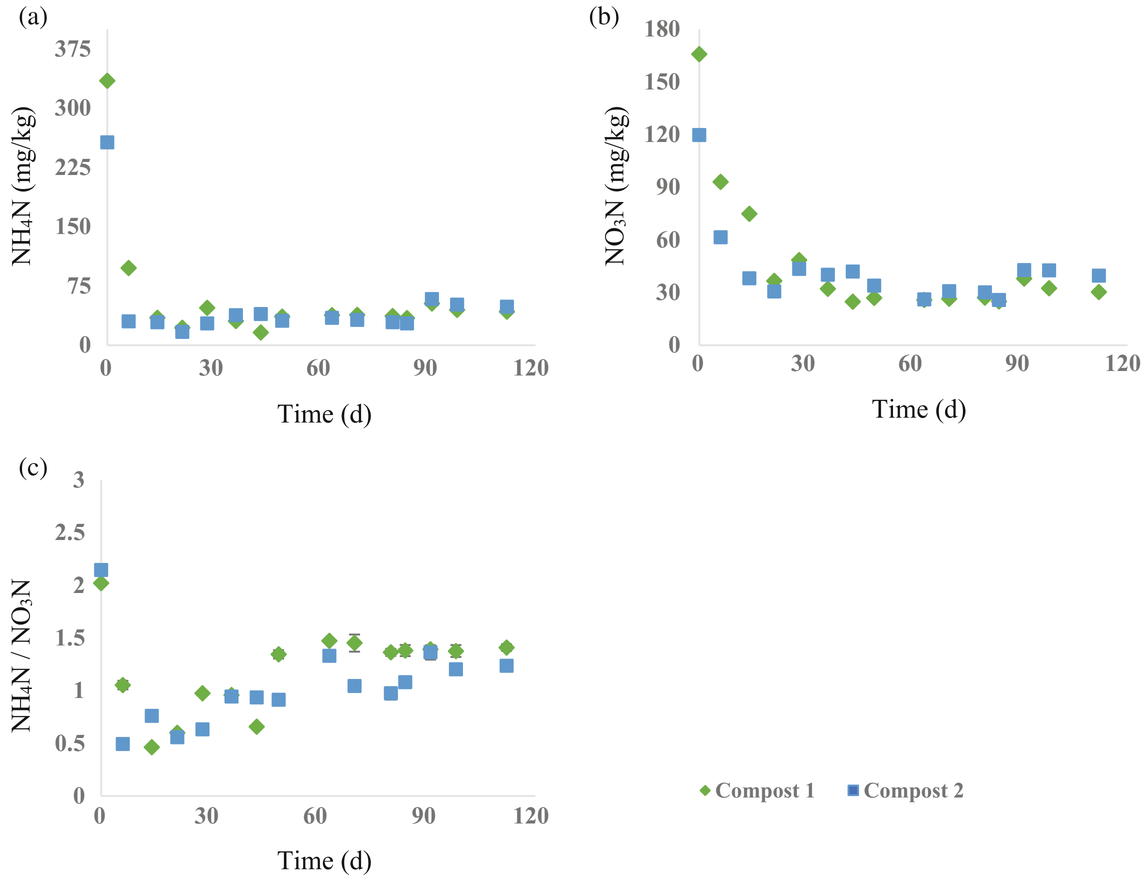

Figure 3. Ammonium nitrogen (a), nitrate nitrogen (b), and ammonium to nitrate nitrogen ratio (c) progression during the two composting experiments (Compost 1, 2).

nitrification, resulting in N_2O emissions.[36] The NH_4N/NO_3N ratio (Fig. 3(c)) reached its minimum during an early period due to the rapid decrease of ammonium and nitrate nitrogen. Furthermore, as both NH_4N and NO_3N values were very low, their simultaneous reduction did not affect TKN. TKN appeared to increase during composting because organic nitrogen losses were very limited for these composting experiments and because organic matter content was reduced through decomposition.[40] GI gradually increased during composting as the phytotoxicity of the pile was reduced, as described by Tiquia,[37] reaching values greater than 100% that indicate a final compost product rich in nutrients. High OUR values at the early stages of composting indicate an increased microbial activity followed by a constant reduction until the end of the process.[8] Humification is the process by which macromolecular carbohydrates are converted in humic substances by microorganisms.[13] During humification, FA values typically decrease after reaching a maximum point, while HA values increase as the composting process progresses.[41]

Evaluation of compost maturity

As mentioned previously, certain physicochemical parameters of composting can be used for the evaluation of compost maturity. Many of these parameters, specifically OUR, GI, NH_4N, NH_4N/NO_3N, and HA/FA, have specific absolute values that can be used as indicators for maturity. For other maturity indexes, such as pH and C/N, even though various studies have presented certain value ranges for compost maturity, these ranges are quite large and coupled to the composting materials. Therefore, pH and C/N values cannot be used directly for the evaluation of maturity without further examination and studies have proposed the monitoring of their behavior until certain requirements are achieved, such as stabilization at maximum and minimum values for pH and C/N, respectively.[9,21] In Supporting Information 1, the compost maturity values of C/N and pH derived from the literature and the current study are presented, along with all the other aforementioned compost maturity indexes (e.g., NH_4N, NH_4N/NO_3N, GI, OUR) from the literature.

To accurately evaluate compost maturity, all indices presented in Supporting Information 1 must reach their suggested maturity values.[20] The specific time periods when the composting procedures meet each of the aforementioned maturity requirements are presented in Supporting Information 2. Even though the NH_4N/NO_3N ratio can be regarded as a maturity index for composting procedures, it was not taken into further consideration due to low NH_4N and NO_3N concentrations (<250 ppm) for all composting experiments.[20] Based on the collected data, compost maturity was reached for Composts 1 and 2 during 80–84 and 84–91 days, respectively, as presented in Supporting Information 2. It should be noted that, even though compost added materials were different for these experiments, compost maturity was achieved in similar time periods, which agree with the typical GW maturity time presented in recent studies.[2]

Color determination during composting

The CIELAB color variables during the two composting processes are illustrated in Fig. 5. As the composting progressed, compost

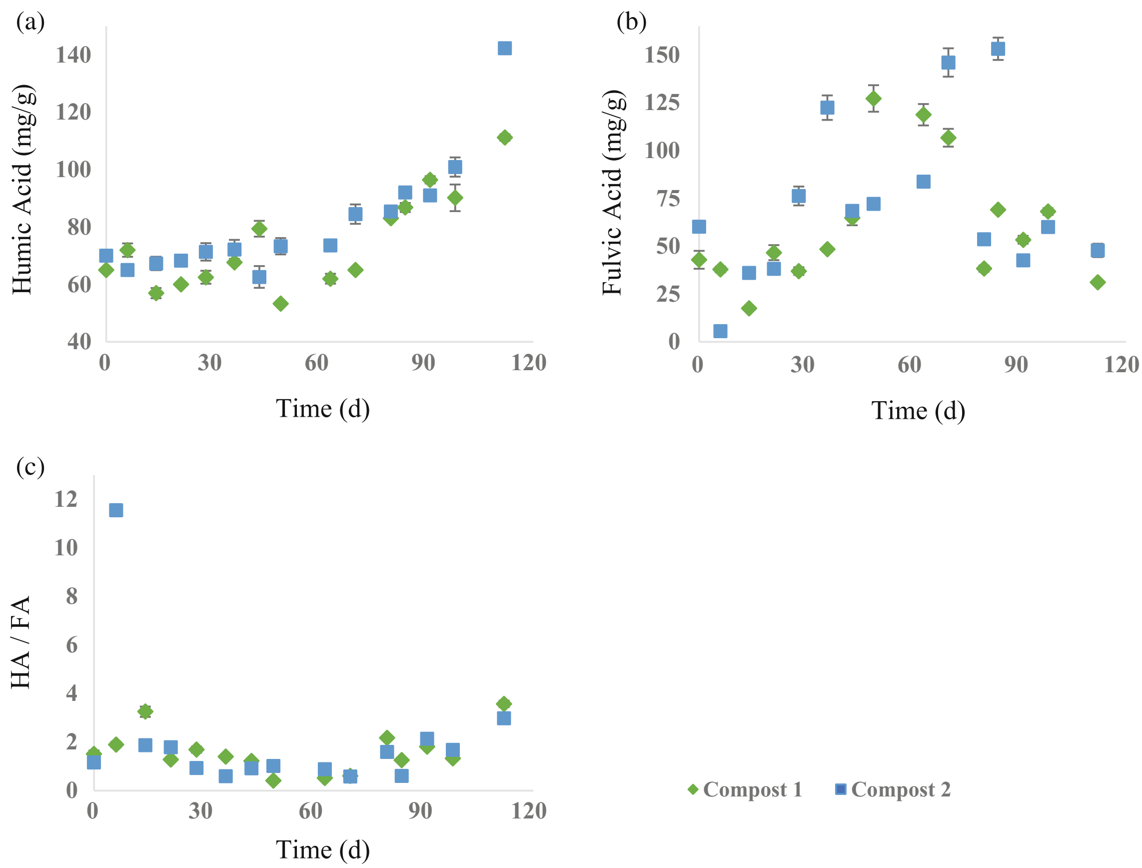

Figure 4. Humic acid (a), fulvic acid (b), and humic to fulvic acid ratio (c) progression during the two composting experiments (Compost 1, 2).

color alterations were recorded.[23,27] At the beginning of the process, the color of the material was usually light brown. As composting continued, the compost became darker and finally dark brown or black.[25] This color change is described by the variation trend of CIELAB color variables visually presented in Supporting Information 3. In both composting experiments, the decreasing trend of color variables a* (Fig. 5(b)) and b* (Fig. 5(c)) were observed throughout the whole composting period. This indicates that, as the compost darkened, its initial red and yellow color component turned into blue and green. Major changes in L* values occurred only during the first days of composting. After the fifth day, the L* values were reduced in a more gradual way until day 36, and then they remained stable for the rest of composting duration. Variables h^0 (Fig. 5(f)) and a*/b* (Fig. 5(d)) were very scattered during all composting experiments, exhibiting both increases and decreases in their values. Variable C* (Fig. 5(e)) showed a constant decreasing trend for both composting experiments throughout the whole composting period. Moreover, color variable ΔE* values (Fig. 5(g)) presented an increasing trend, with the difference that this increase was rapid at the beginning and then proceeded at a more moderate pace. The constant decreasing trend of color variables a*, b*, and C*, along with the constant increasing trend of color variable ΔE*, have also been reported in previous studies regarding different composting substrates.[23,27]

The best fitting equations that describe the relation between CIELAB color variables and time for the composting experiments, along with their respective R^2 values, are presented in Table 3. The two composting processes examined showed similarities in the fitted equations that describe color progression. However, differences were observed regarding the individual values of CIELAB color variables. These results can be attributed to the fact that the values of color variables appear to be affected by the inherent characteristics of the raw materials.

Relationship between color variables and composting phases

Color variables a*, b*, C*, and ΔE* were further evaluated for their ability to depict the various stages of a composting process. Based on the results showed in Fig. 5 and Table 3, the above mentioned color variables fluctuated for the entire composting period until the point of maturity. This is why this study primarily focused on the physicochemical properties that varied throughout the composting period (such as OC, HA, FA, and C/N) and not on the ones that remained constant after the first 21 days of composting (such as OUR, pH, GI, NH_4N, and NO_3N).

Color variable a* (Fig. 5(b)) presented a rapid decrease in its values for 36 and 28 days for Composts 1 and 2, respectively. This rapid decrease occurred during the first two phases of composting (mesophilic and thermophilic) when the declines of OC (Fig. 2(a)) and C/N (Fig. 2(c)) were also fast, and stopped at the initiation of the maturation phase, specifically with the increase of FA (Fig. 4(b)). As the cooling and maturation phases proceeded as a combined stage, a* values presented certain fluctuations. Specifically, during this period, color variable a* increased and decreased in accordance with the rise and fall of FA values. This

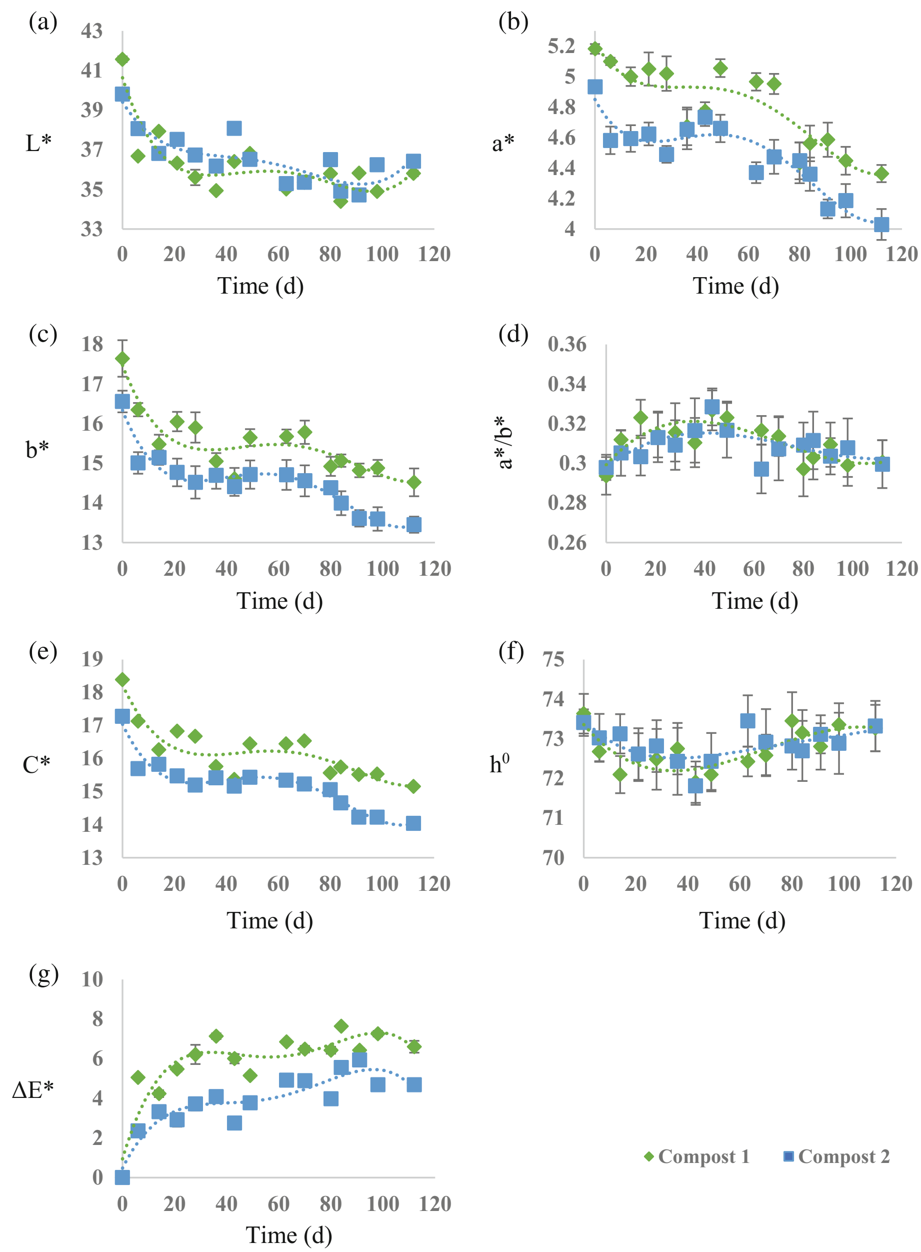

Figure 5. CIELAB color variables L* (a), a* (b), b* (c), a*/b* (d), C* (e), h^0 (f), ΔE* (g) during the two composting experiements (Compost 1, 2).

phenomenon can be attributed to the fact that FA has a yellow or light brown color[42] that influences the total color of compost. As the humification continued and FA reduction led to a gradual increase in HA (Fig. 4(a)), a* values continued to decrease due to the interference of the dark brown color of HA.[42] Specifically, after 70 (Compost 1) and 80 (Compost 2) days, a* values decreased rapidly until days 80 and 91, which signify the respective periods of maturity, as shown in Supporting Information 2, for each compost experiment. During this period, OC and C/N values decreased at a faster rate. After the point of maturity, variable a* remained fairly stable with a minor decrease until the end of the process.

Color variables b* (Fig. 5(c)) and C* (Fig. 5(e)) decreased rapidly for 43 (Compost 1) and 28 (Compost 2) days during the mesophilic and thermophilic stages of composting. At these stages, OC (Fig. 2(a)) and C/N (Fig. 2(c)) also decreased rapidly. When the rapid decrease in OC values reached a temporary halt, the maturation phase began after 36 (Compost 1) and 28 (Compost 2) days. Color variables b* and C* presented the same fluctuations as a*. However, b* and C* values in Compost 2 began to increase a few days after the rapid increase of FA (Fig. 4(b)) and especially when OC reduction reached a temporary halt at the end of the thermophilic phase. Additionally, b* and C* values in Compost 1 showed smaller range fluctuations due to FA production compared to the variations of the color variable a*. Therefore, it is safe to assume that b* and C* are more influenced by OC than a*. When FA values began to decrease (Fig. 4(a)) and HA increased along with the ultimate decrease of OC, b* and C* presented a rapid decrease until the point of maturity and then remained fairly stable with little decrease in their values.

ΔE* values (Fig. 5(e)) presented a very rapid increase during the first phase of composting and a slightly less increasing trend during the thermophilic phase for both composting experiments.

Table 3. Mathematical relationships of CIELAB color variables with time after regression analysis using various types of equations for the two composting experiments (Compost 1, 2)

Compost CIELAB color variables	Compost 1		Compost 2	
	Best fitting equation during composting period	Pearson's correlation coefficient, R^2	Best fitting equation during composting period	Pearson's correlation coefficient, R^2
L*	$L^* = 6E\text{-}07\ t^4 - 0.0001\ t^3 + 0.0117\ t^2 - 0.4047\ t + 40.638$	0.76	$L^* = 4E\text{-}07\ t^4 - 8E\text{-}05\ t^3 + 0.0061\ t^2 - 0.2079\ t + 39.411$	0.74
a*	$a^* = 5E\text{-}08\ t^4 - 1E\text{-}05\ t^3 + 0.0009\ t^2 - 0.0273\ t + 5.2196$	0.78	$a^* = 6E\text{-}08\ t^4 - 2E\text{-}05\ t^3 + 0.0011\ t^2 - 0.0308\ t + 4.8503$	0.87
b*	$b^* = 2E\text{-}07\ t^4 - 5E\text{-}05\ t^3 + 0.0048\ t^2 - 0.1715\ t + 17.475$	0.80	$b^* = 3E\text{-}07\ t^4 - 6E\text{-}05\ t^3 + 0.0052\ t^2 - 0.1654\ t + 16.34$	0.93
h^0	$h^0 = -7E\text{-}06\ t^3 + 0.0014\ t^2 - 0.073\ t + 73.342$	0.65	$h^0 = -4E\text{-}06\ t^3 + 0.0009\ t^2 - 0.0521\ t + 73.424$	0.44
C*	$C^* = 2E\text{-}07\ t^4 - 5E\text{-}05\ t^3 + 0.0048\ t^2 - 0.172\ t + 18.239$	0.80	$C^* = 3E\text{-}07\ t^4 - 7E\text{-}05\ t^3 + 0.0053\ t^2 - 0.1672\ t + 17.045$	0.94
ΔE*	$\Delta E^* = -6E\text{-}07\ t^4 + 0.0001\ t^3 - 0.0127t^2 + 0.4417\ t + 0.9419$	0.82	$\Delta E^* = -4E\text{-}07\ t^4 + 1E\text{-}04\ t^3 - 0.0078t^2 + 0.2634\ t + 0.4762$	0.84
a*/b*	$a^*/b^* = 1E\text{-}07\ t^3 - 3E\text{-}05\ t^2 + 0.0014\ t + 0.2992$	0.65	$a^*/b^* = 7E\text{-}08\ t^3 - 2E\text{-}05\ t^2 + 0.001\ t + 0.2976$	0.44

When the last two phases of composting (cooling and maturation) took place, ΔE* values continued to increase with fluctuations based on FA and HA values until the point of maturity. At this point, ΔE* achieved its greatest value. After the point of maturity, ΔE* values slightly decreased and remained stable until the end of humification process. However, it should be noted that the kinetics of ΔE* cannot depict the different stages of composting with the same accuracy as color variables a*, b*, and C* because ΔE* (apart from a* and b*) is also influenced by color variable L*.

To further evaluate the ability of color variables a*, b*, C*, and ΔE* to depict the changes of the organic composition of compost (OC, C/N, HA, FA, HA/FA) and, therefore, the different stages of composting, the statistical process of regression analysis was conducted. Various types of equations were tested and best fitting equations were determined by Pearson's correlation coefficient, R^2. To determine the relationship between CIELAB color variables and HA/FA, only data after the initiation of the maturation phase was used. The results of this statistical analysis are presented in Table 4. Specifically, the effects of OC, C/N, and HA/FA changes on compost color can also be mathematically described, as these physicochemical parameters showed a very strong relationship with color variables a*, b*, C* ($R^2 > 0.83$). HA also showed a strong relationship with color variables a*, b*, and C* ($R^2 > 0.74$). However, the effect of FA fluctuations on compost color change was not correlated efficiently with any CIELAB color variable presenting rather low R^2 values ($R^2 < 0.70$). This result can be attributed to the fact that FA does not present a constant variation trend but instead shows fluctuations during the cooling and maturation stages; thus, its influence on compost color change cannot be mathematically proven without the implementation of a more complex mathematical model. As expected, ΔE* did not present any strong relationship with any of the aforementioned physicochemical properties because it is primarily affected by L*.

In total, based on the findings of this study, CIELAB color variables a*, b*, C*, and ΔE* not only presented constant variation trends (i.e., decreasing or increasing) during composting but they also appeared to fluctuate differently for each phase of composting until the point of maturity based on the formation of the various organic compounds that take part in composting. This fact indicates that color change follows a general pattern for each composting procedure, especially regarding the transformation of organic matter as expressed by OC, C/N, FA, and HA. In previous composting studies with green tea waste[23] and olive pomace[27] as substrates, a*, b*, C*, and ΔE* presented a rather linear variation trend until the point of maturity. The absence of color variable fluctuations can be attributed to the fact that, during these composting experiments, the different stages of composting were indistinguishable from each other and progressed simultaneously, as observed by the formation of humic substances from

Table 4. Pearson's correlation coefficients (R^2) between physicochemical variables depicting compost organic composition and CIELAB color variables

	Compost 1				Compost 2			
	a*	b*	C*	ΔE*	a*	b*	C*	ΔE*
OC%	0.83	0.84	0.84	0.59	0.83	0.91	0.92	0.76
HA (mg g^{-1})	0.78	0.74	0.81	0.29	0.83	0.77	0.79	0.40
FA (mg g^{-1})	0.15	0.16	0.18	0.08	0.15	0.41	0.42	0.36
HA/FA	0.84	0.93	0.94	0.24	0.85	0.85	0.85	0.07
C/N	0.83	0.83	0.83	0.60	0.58	0.81	0.82	0.81

the first days of composting. As a result, in these studies, CIELAB color variables could not depict the different stages of composting and, thus, could only be used to monitor composting or evaluate the point of maturity.

CIELAB colorimetric application and future prospects

Based on the findings of this work, the CIELAB colorimetric method can be used to distinguish the different stages of a composting process and replace, to a significant extent, the typical physicochemical compost analyses that are usually expensive and time consuming.[25] In the work of Tsivas *et al*.,[27] a cost analysis between the CIELAB colorimetric monitoring method and typical physicochemical analyses for composting showed that this new proposed method dramatically reduces the operational costs and duration of analysis by up to 60.3% and 93.8%, respectively.

Additional research will reinforce the use of the CIELAB colorimetric method as a quick, simple, and sufficient indicator for compost evaluation. An open access data repository of composting data, including CIELAB values and critical physicochemical characteristics using various substrates, can serve as an ideal space for the analysis of possible similarities and differences between two sets of variables and for the inherent characteristics of the examined raw materials. This repository will provide all the necessary colorimetric data to distinguish the different composting stages and to rapidly and cheaply evaluate the level of compost maturity. Composting procedures with a wide range of composting duration periods, such as GW, can largely benefit from this new method of analysis that avoids to a great extent the tedious implementation of typical expensive and time-consuming physicochemical composting analyses.

CONCLUSIONS

To conclude, a more robust colorimetric method for the evaluation of compost stages would solve many operational problems. This method can be implemented directly, without further examination, in various laboratory and industrial composting applications to reduce the operational costs and duration of analysis. Further research is required to evaluate the effects of additional composting properties, such as microbiological analyses, on color change as they were not part of this study and were only investigated indirectly through their oxygen uptake rate.

SUPPORTING INFORMATION

Supporting information may be found in the online version of this article.

REFERENCES

1 Zhang L and Sun X, Addition of fish pond sediment and rock phosphate enhances the composting of green waste. *Bioresour Technol* **233**:116–126 (2017).
2 Reyes-Torres M, Oviedo Ocaña R, Dominguez I, Komilis D and Sánchez A, A systematic review on the composting of green waste: feedstock quality and optimization strategies. *Waste Manage* **77**: 486–499 (2018).
3 Zhu-Barker X, Bailey SK, Paw UKT, Burger M and Horwath WR, Greenhouse gas emissions from green waste composting windrow. *Waste Manage* **59**:70–79 (2017).
4 Al-Alawi M, Szegi T, El Fels L, Hafidi M, Simon B and Gulyás M, Green waste composting under GORE(R) cover membrane at industrial scale: physico-chemical properties and spectroscopic assessment. *Int J Recycl Org Waste Agric* **8**:385–397 (2019).
5 López M, Soliva M, Martínez-Farré FX, Bonmatí A and Huerta-Pujol O, An assessment of the characteristics of yard trimmings and recirculated yard trimmings used in biowaste composting. *Bioresour Technol* **101**:1399–1405 (2010).
6 Compostnetwork.info, Bio-Waste in Europe, https://www.compostnetwork.info/policy/biowaste-in-europe, 2020.
7 OJEU, Directive (EU) 2018/851 of the European Parliament and of the Council of 30 May 2018, https://eur-lex.europa.eu/legal-content/EN/TXT/HTML/?uri-CELEX:32018L0851&from=EN, 2018.
8 Epstein E, *The Science of Composting*. CRC Press, Boca Raton (1997).
9 Diaz LF and Savage GM, Chapter 4 factors that affect the process, in *Waste Management Series*, ed. by Diaz LF, de Bertoldi M, Bidlingmaier W and Stentiford E. Elsevier, Amsterdam, pp. 49–65 (2007).
10 Belyaeva ON and Haynes RJ, Chemical, microbial and physical properties of manufactured soils produced by co-composting municipal green waste with coal fly ash. *Bioresour Technol* **100**:5203–5209 (2009).
11 Kazamias G, Roulia M, Kapsimali I and Chassapis K, Innovative biocatalytic production of soil substrate from green waste compost as a sustainable peat substitute. *J Environ Manage* **203**:670–678 (2017).
12 Khalil A, Domeizel M and Prudent P, Monitoring of green waste composting process based on redox potential. *Bioresour Technol* **99**: 6037–6045 (2008).
13 Liu L, Wang S, Guo X, Zhao T and Zhang B, Succession and diversity of microorganisms and their association with physicochemical properties during green waste thermophilic composting. *Waste Manage* **73**:101–112 (2018).
14 Oviedo-Ocaña ER, Dominguez I, Komilis D and Sánchez A, Co-composting of green waste mixed with unprocessed and processed food waste: influence on the composting process and product quality. *Waste Biomass Valorization* **10**:63–74 (2019).
15 Montoneri E, Koutinas M, Padoan E, Negro V, Licignano C, Leone S *et al*., Integrated chemical and biochemical technology to produce biogas with a reduced ammonia content from municipal biowaste. Validating lab-scale research in a real operational environment. *Environ Sci: Adv* **1**:746–768 (2022).
16 Naidu Y, Meon S, Kadir J, Siddiqui Y and Malaysia S, Microbial starter for the enhancement of biological activity of compost tea. *Int J Agric Biol* **12**:1560–8530 (2010).
17 Negro V, Montoneri E, Negre M, Fabbri G, Boero V, Solaro S *et al*., Compost stream as a potential biomass for humic acid production: focus on compost seasonal and geographical variability. *Procedia Environ Sci Eng Manage* **5**:133–138 (2018).
18 Bustamante MA, Ceglie FG, Aly A, Mihreteab HT, Ciaccia C and Tittarelli F, Phosphorus availability from rock phosphate: combined effect of green waste composting and sulfur addition. *J Environ Manage* **182**:557–563 (2016).
19 Abbott JA, Quality measurement of fruits and vegetables. *Postharvest Biol Technol* **15**:207–225 (1999).
20 CCQC, California Compost Quality Council. Compost Maturity Index, Technical Report, Nevada (2001).
21 Azim K, Soudi B, Boukhari S, Perissol C, Roussos S and Thami Alami I, Composting parameters and compost quality: a literature review. *Org Agric* **8**:141–158 (2018).
22 Iglesias Jiménez E and Pérez García V, Composting of domestic refuse and sewage sludge. I. Evolution of temperature, pH, C/N ratio and cation-exchange capacity. *Resour Conserv Recycl* **6**:45–60 (1991).
23 Khan MAI, Ueno K, Horimoto S, Komai F, Someya T, Inoue K *et al*., CIELAB color variables as indicators of compost stability. *Waste Manage* **29**:2969–2975 (2009).
24 Vlyssides AG, Lamprou GK and Vlysidis A, Chapter 6-industrial case studies on the detoxificaton of OMWW using Fenton oxidation process followed by biological processes for energy and compost production, in *Olive Mill Waste*, ed. by Galanakis CM. Elsevier Academic Press, Amsterdam, pp. 119–138 (2017).
25 Thompson WH, *Test Methods for the Examination of Composting and Compost*. The US Composting Composting Council Research and Education Foundation and the United States Department of Agriculture, USA (2001).
26 Zorpas AA and Loizidou M, Sawdust and natural zeolite as a bulking agent for improving quality of a composting product from anaerobically stabilized sewage sludge. *Bioresour Technol* **99**:7545–7552 (2008).
27 Tsivas D, Vlyssides A and Vlysidis A, Monitoring of a III-phase olive pomace composting process using the CIELAB colorimetric method. *Waste Biomass Valorization* **12**:5029–5039 (2021).

28 Sánchez-Marañón M, Delgado G, Melgosa M, Hita E and Delgado R, Cielab color parameters and their relationship to soil characteristics in mediterranean red soils. *Soil Sci* **162**:833–842 (1997).
29 Palechor-Tróchez JJ, Ordóñez-Santos LE and Villada-Castillo HS, Relationship between color CIEL*a*b* and total organic carbon in compost. *Adv Mater Sci Eng* **2018**:1–6 (2018).
30 Pant A, Radovich T and Hue N, Compost tea production, 19–26 (2011).
31 Liu D, Zhang R, Wu H, Xu D, Tang Z, Yu G *et al*., Changes in biochemical and microbiological parameters during the period of rapid composting of dairy manure with rice chaff. *Bioresour Technol* **102**: 9040–9049 (2011).
32 Tsiodra C, Lambrou G, Seintis G and Vlysiddes AG, A novel respirometer for the determination of compost stability, in 4th International Conference on Sustainable Waste Management, Limassol (2016).
33 Velasco MI, Campitelli P, Ceppi SB and Havel J, Analysis of humic acid from compost of urban wastes and soil by fluorescence spectroscopy. *Agriscientia* **21**:31–38 (2004).
34 Zucconi F, Pera A, Forte M and de Bertoldi M, Evaluating toxicity of immature compost. *BioCycle* **22**:54–57 (1981).
35 CIE, Industrial colour-difference evaluation, in *Technical report 116/1995*, ed. by Bureu CIEC. CIE, Vienna, Austria (1995).
36 Hao X and Benke MBJDS, Dyn Plant, Nitrogen transformation and losses during composting and mitigation strategies. **2**: 10–18 (2008).
37 Tiquia SM, Reduction of compost phytotoxicity during the process of decomposition. *Chemosphere* **79**:506–512 (2010).
38 Guo X-x and Liu H, Humic substances developed during organic waste composting: formation mechanisms, structural properties, and agronomic functions. *Sci Total Environ* **662**:501–510 (2019).
39 Jiménez EI and García VLPJBW, Evaluation of city refuse compost maturity: a review. *Biol Wastes* **27**:115–142 (1989).
40 Michailides M, Christou G, Akratos CS, Tekerlekopoulou AG and Vayenas DV, Composting of olive leaves and pomace from a three-phase olive mill plant. *Int Biodeter Biodegr* **65**:560–564 (2011).
41 Tuomela M, Vikman M, Hatakka A and Itävaara M, Biodegradation of lignin in a compost environment: a review. *Bioresour Technol* **72**: 169–183 (2000).
42 Stevenson FJ, *Humus Chemistry: Genesis, Composition, Reactions*. John Wiley & Sons, New York, USA (1994).

Journal of Chemical Technology and Biotechnology published by John Wiley & Sons Ltd on behalf of Society of Chemical Industry (SCI).