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Effect of composting and vermicomposting on potentially toxic element contents and bioavailability in sewage sludge digestate

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ABSTRACT

Vermicomposts and composts prepared from sewage sludge digestate and additives (spent mushroom compost, straw, biochar) after 43 days pre-composting followed by 90 days vermicomposting with *Eisenia fetida* or by compost maturing were investigated regarding the potentially toxic element (PTE) As, Co, Cr, Cu, Mo, Ni, Pb and Zn contents. The average increment in the total PTE concentration for the entire process was ten times higher (104 %) compared to the increment solely in the composting or vermicomposting (9.3 and 9.5 %, respectively) after pretreatment. Compared to the untreated digestate the As and Co concentrations in the final mixtures were 26 and 51 % higher, respectively while for the other PTEs 26 ± 9 % average decrease was observed. Total PTE content was the same in composts and vermicomposts. Average PTE bioavailability (water soluble/total concentration) was statistically the same in vermicomposts (2.5) and composts (2.7), but lower in mixtures with biochar (2.5) than without it (2.8).

1. Introduction

Sewage sludge is used in agriculture in very high proportions worldwide, either directly or as processed products (Buta et al., 2021). These materials contain several valuable components, such as plant macro- and micronutrients, as well as organic compounds. Therefore, they can be used in the soil as both fertilizers and amendments (Aranyos et al., 2016). However, they also carry certain risks, one of which is the presence of inorganic contaminants, which are potentially toxic elements (PTE).

Sewage sludge processing may include certain stabilizing treatments like anaerobic digestion, composting or vermicomposting, which involve various additives aimed at producing a final product that has more favourable properties for soil application than the original material. These more favourable properties include stabilized organic matter, inactivated pathogens and reduced total PTE content and bioavailability (Amir et al., 2005; He et al., 2016; Swati and Hait, 2017; Thanh et al., 2016).

Unlike digestion and composting, where it is the C/N ratio that must be adjusted first and foremost, in the case of vermicomposting the raw or digested sewage sludge must first undergo at least aeration, a drying process or thermophilic pre-composting in order to create suitable living conditions for earthworms. During these pretreatments the material becomes stabilized and potentially toxic gases are vented out of the material (Malińska et al., 2017; Wang et al., 2013a). In the case of sewage sludge digestate (digestate) this pretreatment results in further stabilization of the already partly stabilized material, which means a further loss of organic matter by diminishing the ratio of high-energy organic compounds (Amir et al., 2005).

Most previous studies investigating the effects of vermicomposting on the PTE content and bioavailability in sewage sludge, however, have only addressed the effect of sludge pre-composting or pretreatment tangentially, if at all (He et al., 2016; Malińska et al., 2017; Wang et al., 2013a; Wu et al., 2018). Pre-composting may cause significant changes in the concentration of inorganic components, which is not negligible within the whole sludge treatment process (Zheng et al., 2004). Therefore, the effect and efficiency of vermicomposting on PTE content and availability must be evaluated together with the pretreatment process that precedes it (Khan et al., 2019; Ning et al., 2021). To the best of our knowledge municipal sewage sludge digestate has not yet been studied

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Table 1
Chemical properties and dry matter content of the raw materials. Values refer to D.M.

Parameter	Unit	Digestate	GW	WS	SMC	BC
pH_{H2O}		6,62	5.64	6.08	7.12	10.4
OC	m m^{-1} %	21,9	35.7	40.8	19.9	27.5
$CaCO_3$	m m^{-1} %	4,08	0.98	0	6.52	5.75
Total N	mg kg^{-1}	39,400	10,100	6900	19,700	9590
Total P	mg kg^{-1}	32,509	1559	1459	3708	6742
Total K	mg kg^{-1}	3370	8360	3388	19,806	15,380
C/N		6	35	59	10	29
Total As	mg kg^{-1}	3,38	0.56	9.03	0.83	<dl
Total Co	mg kg^{-1}	2,25	0.415	0.507	<dl	1.38
Total Cr	mg kg^{-1}	32,9	3.46	7.97	8.14	6.61
Total Cu	mg kg^{-1}	296	14.7	38.5	12.4	15.7
Total Mo	mg kg^{-1}	5,27	0.798	2.69	0.867	1.66
Total Ni	mg kg^{-1}	19,3	2.13	10	4.81	5.85
Total Pb	mg kg^{-1}	19,1	1.21	2.04	0.91	2.51
Total Zn	mg kg^{-1}	795	36.7	145	59.2	53.3
Dry matter	m m^{-1} %	18,4	73	65	40	–

Detection limit (dl) was 0.4 mg kg^{-1} for As and 0.04 mg kg^{-1} for Co.

from this aspect, despite the fact that there is an uptrend in digestion technology throughout the world, thus predicting an increase in the volume of digestate available (Hanum et al., 2019).

During pre-composting and composting, the change in the total concentration of PTEs depends on the balance of two processes: the decrease caused by leaching and the increase associated with the mineralization of organic matter, which leads to a reduction in the volume of the substrate. During vermicomposting, the same processes apply, but the absorption of elements by the worms may also contribute to a decrease in PTE concentration (He et al., 2016; Suthar, 2008; Swati and Hait, 2017). Hence, vermicomposting could be more effective in reducing the PTE concentration than composting (Mohee and Soobhany, 2014).

In order to increase the efficiency of the composting or vermicomposting process and to achieve a final product with better quality, additives can be applied at various stages in the sewage sludge treatment. The use of these materials has different purposes, like adjusting pH and C/N ratio, reducing the risk of PTEs or increasing aeration (Guo et al., 2022). Additives such as green waste (GW), wheat straw (WS), spent mushroom compost (SMC) and biochar (BC) may reduce the risk of heavy metal toxicity in sewage sludge-based composts and vermicomposts, and might also increase aeration, thus improving organic matter degradation (Meng et al., 2017; Wu et al., 2018).

The earlier work of Rékási et al. (2019) showed that, in terms of nitrogen, phosphorus and potassium availability and organic matter quality, the vermicomposting of a digestate has no advantage over composting. The results presented below provide information on how the total concentrations and bioavailability of PTEs in the digestate differ after composting and vermicomposting.

The main objectives of the present work were: i) To evaluate the extent to which the use of different additives modifies the enrichment of PTEs in the digestate during composting and vermicomposting. ii) To compare the total and water-soluble concentrations and bioavailability of the PTEs in compost and vermicompost made from different digestate mixtures. iii) To evaluate the effect of substrate properties on the elemental accumulation of worms from the digestate.

2. Materials and methods

2.1. Materials

The municipal sewage sludge digestate was collected from sewage treatment plant of Érd (Pest County, Hungary). The GW came from public parks and home gardens in Százhalombatta (Pest County, Hungary). The treatment is based on activated sludge process in a sequencing batch reactor technology. The waste activated sludge undergoes anaerobic digestion in order to produce biogas. In the experiment, the material left after the digestion was used. The WS was used unchopped. The SMC contained cow manure, wood chips and $CaCO_3$.

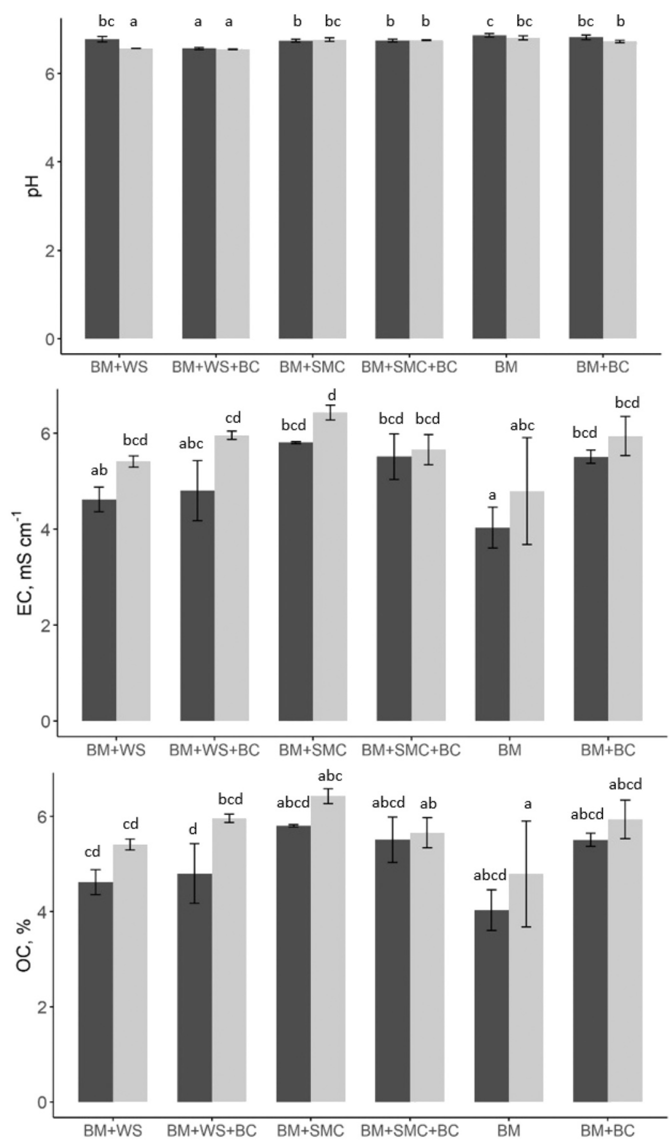

Fig. 1. The pH, electrical conductivity (EC) and organic carbon (OC) content in mature composts (COM) and vermicomposts (VC). Data are the means ±sd of the replicates (n = 3). Different letters indicate significant differences between columns (p < 0.05). Error bars represent the standard deviation (SD). Dark and light grey columns represent composts and vermicomposts, respectively.

The BC was produced by Sonnenerde GmbH (Austria) from grain husks and paper fibre sludge pyrolyzed at 450–500 °C for 20 min. Some properties of the materials are presented in Table 1.

2.2. Treatment process

For thermophilic pre-composting six compost piles weighing 1.4 t (1.1–1.4 m3 of volume) were made from the mixture of materials in different proportions. The piles were kept on an open field covered with geotextile. The pre-composting phase lasted for 43 days during which the heaps were mixed seven times and the core temperature was monitored daily at 50 cm depth. The main component of each pile was the basic mixture (BM) that consisted GW and digestate in 1:3 ratio based on their wet weight. The mixtures had the following wet mass ratios: 1) BM 80 % + WS 20 %; 2) BM 75 % + WS 20 % + BC 5%; 3) BM 80 % + SMC 20 %; 4) BM 75 % + SMC 20 % + BC 5%; 5) BM 100 %; 6) BM 95 % + BC 5%.

After pre-composting, the same amount of material from each prism was loaded into 6-l (25 × 25 × 10 cm) plastic containers without drainage for compost maturation and vermicomposting in three replications. The water content of the mixtures was maintained at 50 % relative saturation during the three months treatment period at room temperature. For vermicomposting *Eisenia fetida* previously bred in digestate and marc mixture was used in the density of 1 worm/35 g D.M. The average weight of the worms was 0.33 ± 0.11 g. The boxes for both composting and vermicomposting were covered with geotextile. After the end of the procedure the materials were dried and ground for analysis.

The worms were removed from the material and rinsed with distilled water. The cleaned worms were then kept at room temperature in Petri dishes with moist filter paper on the bottom for 24 h to void their gut contents. The worms were then killed by freezing at -80oC and dried to constant weight. The dried tissues were then ground for chemical analysis (Wang et al., 2013a).

2.3. Analysis

2.3.1. Material analysis

The pH of the materials was measured in a 1:2.5 solid:water suspension after 12 h (Mokolobate and Haynes, 2002; MSZ-08-0206/2, 1978). The organic carbon (OC) content was determined using the modified Walkley-Black method (MSZ-08-0452, 1980). The extraction and fractionation of the humus fractions were carried out by Stevenson (1994). The spectral characterization (E_4/E_6) of fulvic and humic acids was performed according to the method of Chen et al. (1977).

In this study the following elements were measured as PTEs: As, Co, Cr, Cu, Mo, Ni, Pb and Zn. The pseudo-total and water-soluble element concentrations were determined. Pseudo-total concentrations were determined after microwave teflon bomb digestion with aqua regia (MSZ-21470-50, 2006): 7 ml aqua regia was added to 1 g of soil. After digestion in microwave digester (Milestone Inc., Shelton, CT, USA), the filtered solution was filled up to 50 ml with ultrapure water for analysis. Water-soluble element contents were measured from 1:10 extract after 3 h of shaking with ultrapure water. The element concentrations in each extract were determined by ICP-AES technique (Jobin-Yvon Ultima 2 sequential instrument), using Merck calibration standards and following the manufacturer's instructions. In each measurement session the extract of a standard soil sample was also analyzed as a control. The calibration curves were determined after every 12th sample.

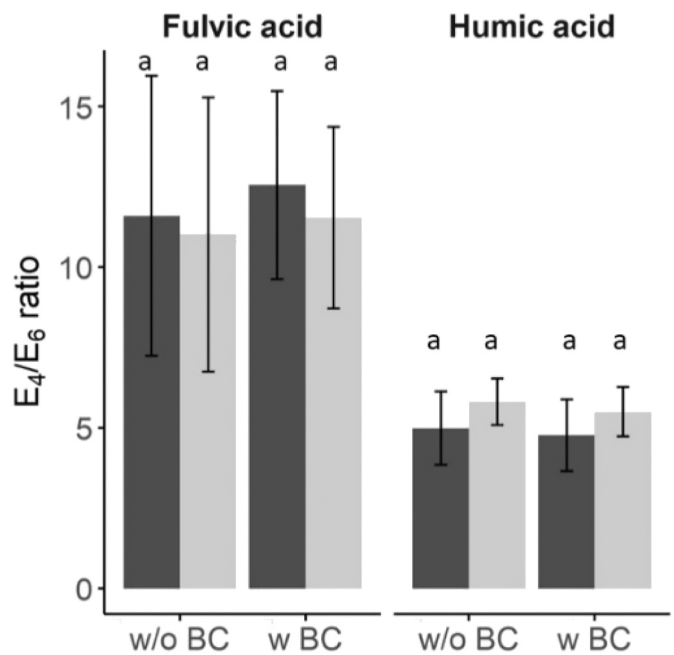

Fig. 2. Effect of BC, composting and vermicomposting on the E_4/E_6 ratio of fulvic and humic acids in the materials. Error bars represent the standard deviation (SD) of the mean with 9 replicates. Different letters indicate significant differences between treatments ($p < 0.05$). Dark and light grey columns represent composts and vermicomposts, respectively.

2.3.2. Data and Statistical analysis

Several factors were determined from the analytical data. Changes in PTE concentrations were examined both for the entire treatment process from the beginning of pre-composting to the finished mature compost or vermicompost, and from the beginning to the end of compost maturation or vermicomposting only. Modified versions of the equations used by He et al. (2016) were applied for the calculations:

$$A = (C_f - C_0)/C_0 \times 100 \text{ and } A' = (C_f - C_p)/C_p \times 100$$

where A stands for the concentration change during the entire process, A' for the concentration change for compost maturation or vermicomposting only, and C_f, C_0 and C_p for the concentration of the given element in the final products, the starting mixtures and the pre-composted materials, respectively.

The bioavailability factor (BF) was calculated as the ratio of the PTE concentration in the water soluble fraction to the total metal concentration and was expressed as a % (Wang et al., 2013a).

The accumulation of PTEs in worm tissue can be described by the bioconcentration factor (BCF), calculated using the following equation (Li et al., 2010):

$$BCF = \text{Worm element content}\left(\text{mg kg}^{-1}\right)/\text{Total element content in substrate}\left(\text{mg kg}^{-1}\right)$$

Treatment effects were analyzed using one or two-way analysis of variance (ANOVA) and Tukey's post-hoc test, where the factor was either the mixture (BM + WS, BM + WS + BC, BM + SMC, BM + SMC + BC, BM, BM + BC) or the treatment (composting, vermicomposting) or the presence or absence of biochar or a combination of two of these. Significant differences between the treatments were calculated at the $p <$ 0.05 level with Statistica v.13 (StatSoft Inc.) software, which was used for all the statistical evaluations. Data visualization was made with R

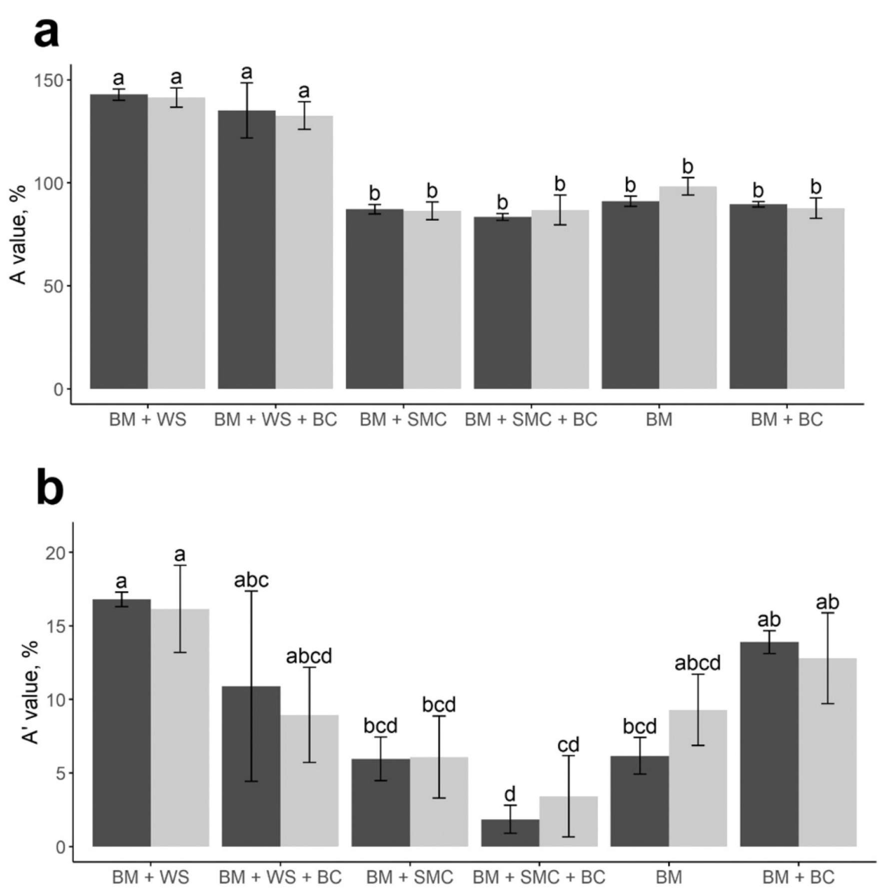

Fig. 3. Increments in the total PTE concentrations in the final mixtures (average values recorded in composts and vermicomposts) a) compared to the starting mixtures (A value), % and b) compared to the pre-composted mixtures (A' value), %. Data are the means ±sd of the replicates (n = 3). Different letters indicate significant differences between columns ($p < 0.05$). Dark and light grey columns represent composts and vermicomposts, respectively.

statistical software (R Core Team, 2022).

3. Results and discussion

3.1. Process parameters

Detailed results for the composting and vermicomposting processes can be found in (Rékási et al., 2019). During pre-composting the temperature reached 60 °C in each treatment. The thermophilic phase was significantly longer in mixtures containing WS. The average temperature was slightly higher in mixtures containing BC. During vermicomposting no worm mortality was detected. The worm population and biomass was the lowest in mixtures containing WS and SMC, though BC also significantly reduced biomass growth compared to BM.

Vermicomposting resulted in a greater decline in OC than compost maturation (12 and 7 %, respectively). BC also enhanced the degradation process during maturation and vermicomposting, but only to a limited extent (Rékási et al., 2019). However, the final OC content of the materials was nearly the same (Fig. 1). The EC values were higher in vermicomposts but the difference was not significant. This could have been the result of the increased concentration of soluble organic materials and salts due to the more intensive decomposition (Rékási et al., 2019). The pH values showed significant differences, but the range was narrow, between 6.54 and 6.86, which is around the pH value of the digestate (6.62), the main component of the mixtures (Table 1 and Fig. 1). Regarding the quality of the organic compounds in the materials, vermicomposts contained slightly more complex fulvic acid molecules than composts, while in the case of humic acids the opposite was observed. However, the differences were not significant (Fig. 2).

3.2. Changes in the total PTE concentrations

Leaching, element uptake by worms and the decreased volume of the material were all found to play a role in the changes in total PTE concentrations, when the whole process was considered. However, the PTE concentration was only influenced by volume reduction during compost maturing, while absorption by earthworms was also important in the case of vermicomposting.

The PTE content of the mixtures originated primarily from the digestate, though SMC also had a considerable concentration of As, Mo and Zn (Table 1). This meant that, as a consequence of additive application, the PTE concentration was lower in the mixtures than in the digestate.

When the whole process was considered, the PTE concentration was found to increase by an average of 105 % in the mixtures. No reduction was observed in any treatment. The concentration changes were the same in composts and vermicomposts made from the same mixture. The increments in the mean concentrations of the elements (A value) were as follows: Co (274 %), Pb (116 %), As (107 %), Cr (79 %), Zn (74 %), Cu (73 %), Ni (61 %), Mo (57 %). These values were comparable to those found by Cai et al. (2007). Similar increments were observed for a wide range of substrates during vermicomposting (Swati and Hait, 2017). However, according to Amir et al. (2005), in the course of sewage sludge composting, leaching resulted in a concentration decrease in the total concentrations of Zn, Cu, Pb and Ni within 30 days. The relatively high increases in Co and Pb could be explained by their being present in a high ratio in the residual fraction of sewage sludge (Malinowska and Jankowski, 2020; Zheng et al., 2004).

The total concentration of PTEs increased to the greatest extent in

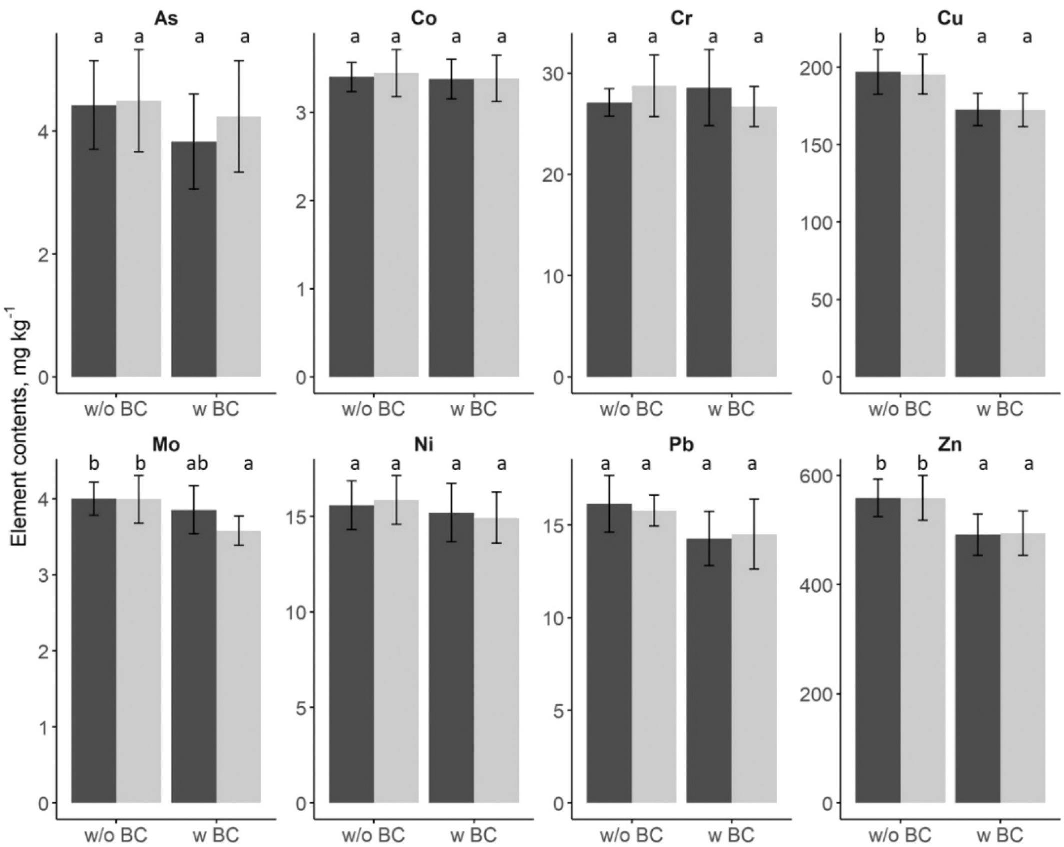

Fig. 4. Effect of BC, composting and vermicomposting on the total element contents of the materials. Error bars represent the standard deviation (SD) of the means with 9 replicates. Different letters indicate significant differences between treatments ($p < 0.05$). Dark and light grey columns represent composts and vermicomposts, respectively.

BM + WS, with an average of 142 %. This value was 134 % in the case of BM + WS + BC, 87 % for BM + SMC, 85 % for BM + SMC + BC, 95 % for BM and 89 % for BM + BC (Fig. 3), indicating that WS enhanced the enrichment of PTEs compared to BM, whereas SMC resulted in a moderate reduction. This was contrary to expectations, since SMC generally promotes decomposition processes and thus raises PTE concentrations (Meng et al., 2017; Zhang and Sun, 2014). However, the SMC used in the present experiment inhibited the decomposition of OC, and also had an adverse effect on worm reproduction probably due to its sawdust content that may be an unfavourable component for worms (Domínguez et al., 2000; Rékási et al., 2019).

When the whole process was considered, BC slightly reduced the PTE enrichment in the mixtures (mean A value without BC: 108 %; with BC: 103 %), but the difference was only significant for the Cu concentration in BM (58.6 %) and the BM + BC mixture (43.6 %). Due to its effect on aeration and microbial activity and to its adsorption capacity, BC is able to influence changes in the concentration PTEs in both positive and negative directions. On the positive side, at an incorporation rate of above 5 % BC enhances the mineralization of organic matter, thus increasing the concentration of PTEs (Sanchez-Monedero et al., 2018; Wang et al., 2018). BC can also increase PTE concentration due to its high CEC, which enables it to adsorb elements and prevent their being leached (Guo et al., 2020; Khan et al., 2020). However, in the present experiment, the following negative effects probably prevailed. At an incorporation rate of 5 % or less, although BC promotes the degradation of organic matter, it only moderately promotes or possibly moderates organic matter mineralization, so the PTE concentration remains unchanged (Sanchez-Monedero et al., 2018; Wang et al., 2018). The more intensive degradation induced by BC has a significant influence on PTE leaching by increasing their concentration in the solution. According to Hsu and Lo (2001) and Hanc et al. (2012), during the first three weeks of composting, the ratio of the water-soluble fraction of the elements might increase temporarily. This happens in parallel with a temporary increase in the concentration of water-soluble organic matter. In the present experiment, the temperature of prisms containing BC proved to be significantly higher (53 °C) during the first 20 days of pre-composting than that of those without BC (49 °C) (Rékási et al., 2019). It can be assumed that, as higher temperatures indicate a more intense degradation and transformation process, a higher ratio of PTEs was mobilised in mixtures containing BC. This could have led to greater leaching, resulting in more moderate accumulation. BC is also able to adsorb humic substances on its surface, thus protecting them from microbial decomposition, resulting in a smaller reduction in the volume of the material and decreasing the increment in PTE concentration. Moreover, BC itself is a recalcitrant substance and may remain intact during composting, thus reducing the volume loss of the material (Sanchez-Monedero et al., 2018). These processes may result in less organic matter loss in mixtures containing BC and thus a smaller increase in PTE concentration.

The A' values indicative of concentration changes during compost maturing and vermicomposting are shown in Fig. 3b. The average A' values calculated for each mixture followed the same order as those of the A values: 16 % for BM + WS, 10 % for BM + WS + BC, 6 % for BM + SMC, 3 % for BM + SMC + BC, 8 % for BM and 13 % for BM + BC (Fig. 3b). The A' values for the elements were the following: Zn (16.4 %), As (14.4 %), Cr (14.3 %), Co (7.7 %), Ni (6.4 %), Mo (6.4 %), Pb (4.9 %), Cu (3.4 %). The effect of BC was similar to that recorded for the whole process: the average PTE increment was 8.6 % for mixtures with BC and 10.1 % for those without BC, though the difference was not significant.

There was no considerable difference in the mean A' values of composts (9.3 %) and vermicomposts (9.5 %). Since there was an increase in the concentrations and the negative tendencies were not

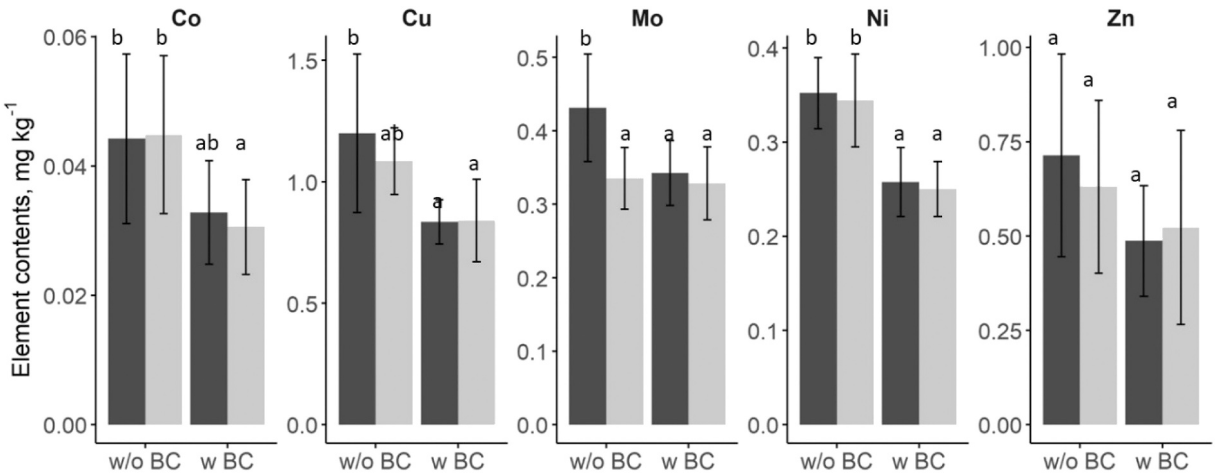

Fig. 5. Effect of BC, composting and vermicomposting on the water-soluble element contents of the materials. Error bars represent the standard deviation (SD) of the means with 9 replicates. Different letters indicate significant differences between treatments ($p < 0.05$). Dark and light grey columns represent composts and vermicomposts, respectively.

significant, the worms did not decrease the PTE content of the mixtures through element uptake. These results contradict those of other authors, who reported that worms decreased the concentration of PTEs in the course of sewage sludge vermicomposting (He et al., 2016; Wang et al., 2013a; Wang et al., 2013b; Wu et al., 2018). In these experiments, however, sewage sludge was applied as a substrate after pretreatment consisting of only drying or a short period (15 days) of pre-composting, presumably resulting in a less stabile substrate compared to the digestate employed in the present study, which was pre-composted for 43 days. The stabilization of the substrate results in PTEs with decreased bioavailability (Amir et al., 2005), so the ability of worms to reduce the PTE concentration may be limited in a more stabilized substrate.

It can therefore be concluded that both the entire process and the maturation/vermicomposting process result in the enrichment of PTEs. The data indicated that the total PTE content of the final materials (Fig. 2) was determined mostly by the processes that took place during pre-composting. On average, the whole process (average A value = 104 %) resulted in a concentration change ten times higher than that observed for the maturing and vermicomposting processes alone (average A' value = 9 %). This indicates that a remarkable amount of organic matter could have mineralized during the pretreatment process, which increased the ratio and thus the concentration of PTEs. The ability of vermicomposting to compensate for the concentration increment caused by pre-composting, as reported by Khan et al. (2019) in raw sewage sludge, was no longer manifested in the case of digestate.

3.3. PTE concentration and bioavailability in the final mixtures

3.3.1. Total PTE concentration

The total PTE concentrations of the final mixtures proved to be determined by the composition of the mixtures. Composts and vermicomposts made of the same mixtures did not differ from each other. The order of the elements according to their concentration was as follows: Zn (526 mg kg^{-1}) > Cu (184 mg kg^{-1}) > Cr (27.8 mg kg^{-1}) > Ni ≈ Pb (15.2 mg kg^{-1}) > As ≈ Mo (3.85 mg kg^{-1}) ≈ Co (3.40 mg kg^{-1}), which is in agreement with the results of (Cai et al., 2007).

Cu and Zn were present in the digestate with a concentration one order of magnitude higher than in the additives (Table 1). This means that the lower the additive ratio of the mixture, the higher the Cu and Zn concentrations. The highest Cu (213 mg kg^{-1}) and Zn (603 mg kg^{-1}) contents were found in the case of BM, and the lowest, averaging 166 and 467 mg kg^{-1}, respectively, in the BM + WS + BC and BM + SMC + BC mixtures. Due to the high As content of SMC (Table 1), mixtures containing this component had high As concentration (5.22 mg kg^{-1} on average). As for the other elements, these values were nearly the same in all the mixtures. It is important to note that the mean concentrations of As and Co in the final mixtures exceeded their initial concentration in the untreated digestate, while for the other elements the treatments resulted in a decrease in their total concentration.

No difference was found in the total concentrations of As, Co, Cr, Ni and Pb in mixtures with or without BC (Fig. 4), but the total concentrations of Cu and Zn proved to be 12 % lower and that of Mo 7 % lower in mixtures containing BC. As these differences were greater than the dilution resulting from the 5 % BC content of the mixtures, they were probably the consequence of the effects of BC on the composting and vermicomposting processes. A similar decrease was observed by Khan et al. (2019) in the total fractions of Cr, Cu, Mn, Pb and Zn after BC addition, in the course of sewage sludge vermicomposting. This was explained by the increased reproduction rate and thus the increased metal uptake by worms in substrates containing BC. In the present experiment, however, BC had a rather negative effect on the increment in worm biomass (Rékási et al., 2019). The decrease observed in the total concentrations of Cu, Zn and Mo was therefore probably caused by BC induced leaching during pre-composting.

Compared to composting, vermicomposting caused no difference in the total concentrations of any of the elements (Fig. 4). Contradictory results were found in the literature regarding in this respect. Hait and Tare (2012) observed that, due to the enhanced volume reduction caused by worms, the total concentrations of Co, Cr, Cu, Mn and Zn in the vermicompost produced from waste- activated sludge exceeded those measured in the compost. According to (Wu et al., 2018), on the other hand, the vermicomposting of sewage sludge resulted in significantly lower total Cd, Pb, Cu and Zn concentrations in the substrate compared to composting.

3.3.2. Water-soluble PTE concentration

Changes in the water-soluble PTE fraction during composting and vermicomposting are a sensitive indicator of the transformation of organic substances. This is also considered to be the most biologically active element fraction, and is thus a risk indicator for the contamination of the food chain (He et al., 2016; Paré et al., 1999). The water-soluble concentrations of As, Cr and Pb remained below the detection limit in the mixtures. The mean water-soluble concentration was 0.99 for Cu, 0.59 for Zn, 0.36 for Mo, 0.30 for Ni and 0.04 mg kg^{-1} for Co. These values are one or two orders of magnitude smaller than those measured by Hait and Tare (2012) in sewage sludge compost and vermicompost. The highest concentration was observed for Co (0.058 mg kg^{-1}) in BM and for Mo in the BM + WS vermicompost (0.505 mg kg^{-1}).

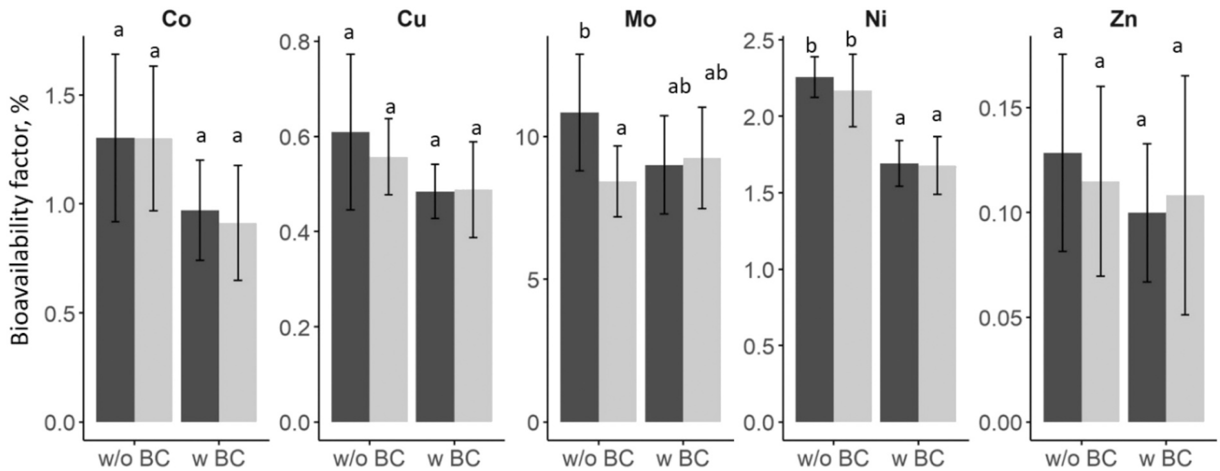

Fig. 6. Effect of BC, composting and vermicomposting on the bioavailability factor (water- soluble/total, %) of the elements in the materials. Error bars represent the standard deviation (SD) of the means with 9 replicates. Different letters indicate significant differences between treatments ($p < 0.05$). Dark and light grey columns represent composts and vermicomposts, respectively.

The highest value of water-soluble Ni was also detected in BM (0.402 mg kg^{-1}), with only half as much (0.216 mg kg^{-1}) in the BM + SMC + BC mixture (0.216 mg kg^{-1}). Irrespective of the components of the mixture and the treatment type, the water-soluble concentrations of Cu and Zn exhibited no significant differences. As regards the effect of BC, although the differences were not always significant, the water-soluble concentrations were 29, 27, 12, 27 and 25 % lower in mixtures with BC in the case of Co, Cu, Mo, Ni and Zn, respectively (Fig. 5). These results are in accordance with the findings of Hao et al. (2019), where adding BC to chicken manure compost reduced the extractable Cu and Zn fractions by 90 and 75 %. The lower water solubility of PTEs can be explained by increased adsorption on the BC surface (Huang et al., 2020).

When comparing composting and vermicomposting, no differences were found in the water-soluble concentrations of Co, Cu, Ni or Zn, while that of Mo was 22 % lower in vermicomposts than in composts (Fig. 5).

Organic matter properties have a significant effect on the PTE fractions. Both composting and vermicomposting stabilize organic matter, thus increasing the residual fraction of PTEs in the material, while the more available fractions exhibited a decrease. In the case of less stable substrates like dried waste-activated sludge the availability of PTEs at the beginning of the process may increase as the decomposition of the original organic matter begins. Later the PTEs released become bound to more polymerized organic fractions, leading to a decrease in their availability (He et al., 2016; Lv et al., 2016). Organic matter degradation during vermicomposting is more intensive than during composting, so the above process takes a shorter time to complete (Ndegwa and Thompson, 2001). This was demonstrated in a four-week experiment conducted by Hait and Tare (2012), where vermicomposts made from dried waste-activated sludge had lower water-soluble Co, Cr, Cu and Zn concentrations than composts. When the processing time was somewhat longer, e.g. 45 days, there was little or no difference in the proportion of bioavailable fractions of PTEs between sewage sludge-based composts and vermicomposts (Wu et al., 2018). The longer processing time of 90 days used in the present experiment may have allowed both composting and vermicomposting to reach the final stage of organic matter degradation and transformation, thus resulting in similar concentrations of the water-soluble fractions in both composts and vermicomposts. Moreover, in the present case, when the sludge forming the substrate was stabilized both by anaerobic digestion and by thermophilic pre-composting, the effect of composting and vermicomposting on the concentration of water-soluble PTE fractions may have become even less pronounced, explaining why the differences between the water-soluble PTE concentrations in composts and vermicomposts became negligible.

3.3.3. Bioavailability factor (BF)

The BF values could only be calculated for Co, Cu, Mo, Ni and Zn, as As, Cr and Pb were below the detection limit in water extracts. The BF of the PTEs showed significant differences depending on the composition of the mixtures, but these differences varied from element to element. The PTEs showed the following order according to their BF values: Mo (9.38 %) > Ni (1.95 %) > Co (1.12 %) > Cu (0.54 %) > Zn (0.11 %). As in the case of water-soluble concentrations, the BF values were several orders of magnitude lower than those measured in waste-activated sludge compost and vermicompost (Hait and Tare, 2012).

The average BF for each PTE in the vermicomposts (2.5) did not differ significantly from that in the composts (2.7). The lack of a significant difference can be attributed to the fact that due to the multiple stabilization and the 90-day compost maturation and vermicomposting time, the organic matter of the mixtures reached a similar maturity state, thus affecting the mobility of the PTEs in the same way. However, BF values for mixtures without BC (2.8) and with BC (2.5) were statistically different. BC probably exerted its BF-reducing effect during pre-composting by immobilizing PTEs (Khan et al., 2019). In mixtures without BC the BF of Cu, Mo, Ni and Zn was higher for composts than for vermicomposts, but this difference was only significant in the case of Mo (Fig. 6). Lv et al. (2016) also found higher element mobility in compost than in vermicompost.

Thus, both BC and vermicomposting slightly decreased the BF of the PTEs. In general lower BF values can be related to several factors, such as higher humic acid content, total organic carbon content, or better macroaggregate proportion in the substrate (Hait and Tare, 2012; Wang et al., 2013a; Wu et al., 2018). In the present mixtures the OC contents did not differ significantly (Fig. 1), they were slightly higher in mixtures with BC (21.4 vs. 22.0 %).

3.4. PTE concentration in worms and the bioconcentration factor

The advantage of vermicomposting over composting is that the earthworms reduce the PTE concentration in the substrate through element uptake (Swati and Hait, 2017). The average PTE concentration in worm tissues followed the order Zn (110 mg kg^{-1}) > Cu (32.2 mg kg^{-1}) > As (10.5 mg kg^{-1}) > Ni (2.08 mg kg^{-1}) > Co (1.29 mg kg^{-1}) > Mo (0.67 mg kg^{-1}) > Cr (0.31 mg kg^{-1}). These values are comparable with earlier results and fall into the range observed in worms feeding on different substrates (Li et al., 2010; Liu et al., 2012; Suthar et al., 2014; Swati and Hait, 2017; Wu et al., 2018).

As the element contents of worms living in different mixtures were nearly equal, it can be assumed that these materials had similar

Table 2
Parameters of stepwise regression equations between worm PTE concentrations and the properties of the vermicompost.

Worm element concentration	Vermicompost						Intercept	R^2	p
	Total element concentration	Water soluble element concentration	E_4/E_6 ratio of Fulvic acids	E_4/E_6 ratio of Humic acids	OC %	pH			
As	0.544	–	0.859	−0.431	–	−24.8	174	0.405	0.124
Co	–	–	–	–	–	–	–	–	–
Cr	0.021	–	–	–	−0.024	–	0.218	0.393	0.023
Cu	0.244	–	–	–	–	–	−12.70	0.323	0.013
Mo	–	1.81	0.015	–	–	–	−0.109	0.268	0.095
Ni	–	8.93	−0.219	–	–	−4.49	31.9	0.652	0.001
Zn	–	–	–	–	–	–	–	–	–

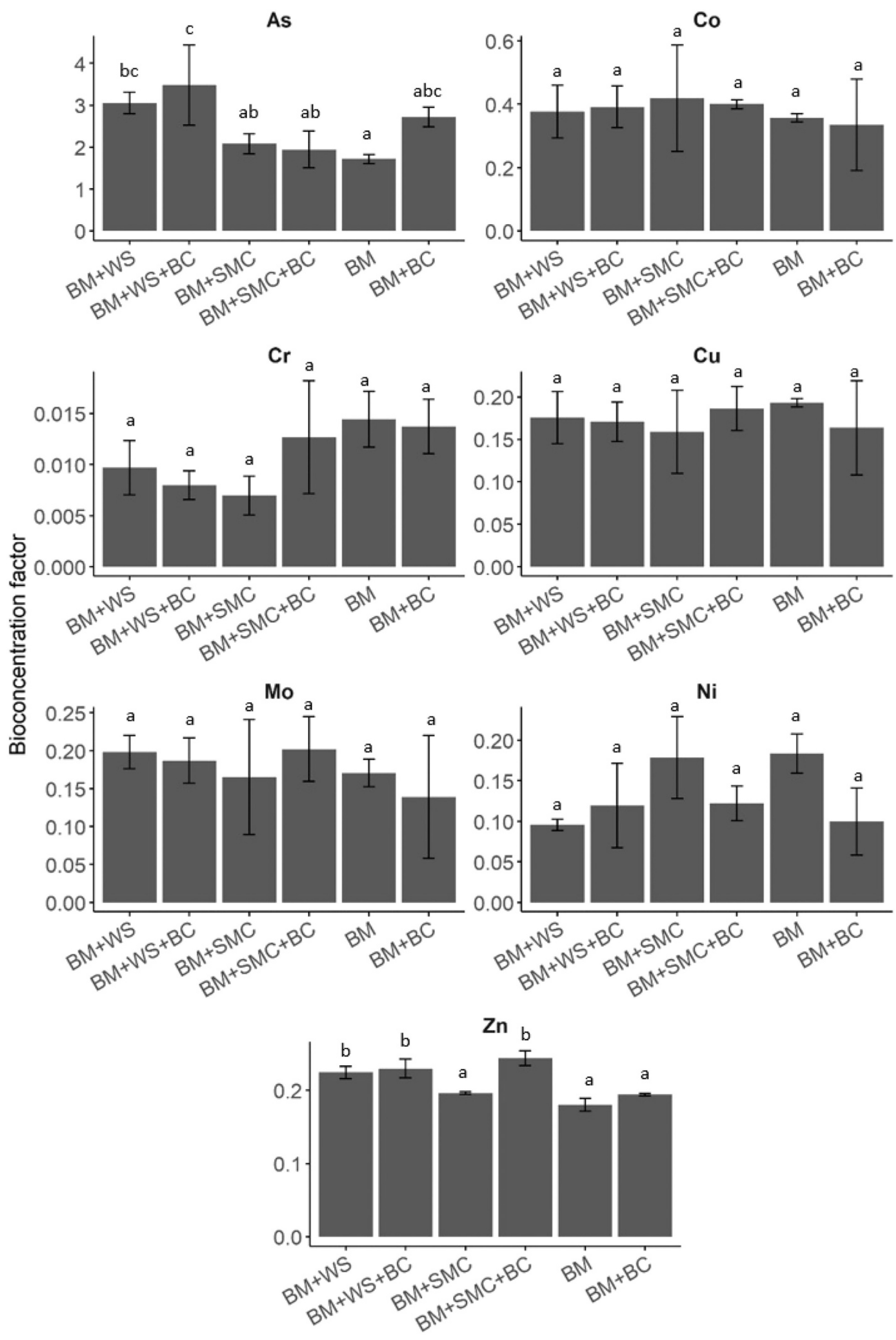

Fig. 7. Bioconcentration factor of the worms in different mixtures. Data are the means ±sd of the replicates ($n = 3$). Different letters indicate significant differences between columns ($p < 0.05$). Error bars represent the standard deviation (SD).

properties from the aspect of element uptake. The PTE concentration in worm tissues is related to the absorption of these elements through the skin or the gut of the animals. The worms can take up considerable amounts of contaminants, but this depends on several factors, including the concentration of the given element in the substrate, especially in the more bioavailable fractions (Li et al., 2010; Swati and Hait, 2017). However, other properties like organic matter content and quality, or the pH of the substrate have a considerable influence as well (Kang et al., 2011; Wang et al., 2013b). The substrate properties investigated (OC content, quality of humic and fulvic acids, total and water-soluble element content and substrate pH) only accounted for a minor part of the variance in worm PTE concentrations (Table 2). The R^2 value was only above 0.5 in the case of Ni. In the case of Co and Zn none of the examined properties were suitable for predicting the earthworm element content, which was also affected by the total or water-soluble element content in the case of As, Cr, Cu, Mo and Ni, and by the quality of the fulvic acids in the case of As, Mo and Ni. Low-molecular-weight organic compounds like fulvic acids can modify the solubility and facilitate the assimilation of PTEs by worms (Suthar et al., 2014; Zhu et al., 2014). Based on the regression parameters, the quality of fulvic acids influenced the uptake of elements in several ways. The value of the E_4/E_6 ratio is inversely proportional to the degree of complexity of the organic molecules. Thus, the uptake of As and Mo is promoted by lower molecular-weight fulvic acids, while in the case of Ni these molecules decreased the uptake. The pH value of the substrate only determined the uptake in the case of As and Ni, where an inverse relationship was observed in accordance with the inverse ratio of pH to element uptake (He et al., 2016). The OC % only affected Cr uptake.

The BCF value is a widely used tool to measure the bioaccumulation of a given pollutant in living organisms. Its value depends on the concentration of the pollutant and the characteristics of the medium and the organism (Wang et al., 2013b). The average BCF values decreased in the following order: As (2.50) > Co (0.38) > Zn (0.21) > Mo (0.18) ≈ Cu (0.17) > Ni (0.13) > Cr (0.01) (Fig. 7). The sequence of elements according to BCF indicates the bioavailability of the elements in the given material (Li et al., 2010). Comparing these values with data from the literature, the BCF values for Zn and Cu were close to those which can be calculated from the data of Wang et al. (2013a) in sewage sludge vermicompost, but the BCF for As was an order of magnitude higher in the present digestate-based mixtures, having a value >1, which means that As was not only accumulated but concentrated in worm tissues (Hartenstein et al., 1980). The only elements showing different BCF values as a function of substrate composition are As and Zn. For As the highest BCF values were detected in the BM + WS and BM + WS + BC mixtures, and for Zn in these mixtures and in BM + SMC + BC. On average, the BCF values were lower in mixtures without BC, but this difference was only significant for Zn (0.20 without BC and 0.22 with BC). This is in accordance with the findings of Khan et al. (2019) where BC increased the element uptake of worms.

4. Conclusions

The average increment in the total PTE concentration for the entire process including the pretreatment was ten times higher (104 %) compared to the increment solely in the composting or vermicomposting phase (9 %) after pretreatment. Worms were unable to reduce the total PTE content of pre-composted digestate. WS enhanced PTE enrichment whereas SMC and BC moderated the intensity of this process. BC also decreased the water-soluble element contents in the mixtures, though there was no difference between composting and vermicomposting in this respect. The results suggest that the 90-day composting or vermicomposting of a doubly stabilized (digested and pre-composted) sewage sludge caused no significant difference in total PTE content and bioavailability.

CRediT authorship contribution statement

Márk Rékási: Conceptualization, Data curation, Writing – original draft. **Péter Ragályi:** Writing – review & editing, Visualization. **Déniel Benjámin Sándor:** Project administration, Writing – review & editing. **Anita Szabó:** Funding acquisition, Project administration. **Pierre-Adrien Rivier:** Conceptualization. **Csilla Farkas:** Conceptualization, Writing – review & editing. **Orsolya Szécsy:** Writing – review & editing. **Nikolett Uzinger:** Conceptualization, Supervision.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

The raw data supporting the conclusions of this article will be made available by the authors, without undue reservation.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.biteb.2022.101307.

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