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Investigating the leaching properties of MBT wastes and composts from aerobic/anaerobic processes

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ABSTRACT

This work assessed and compared the leaching properties of two types of compost and stabilized waste from a mechanical-biological treatment (MBT) plant. The first type of compost and the MBT waste were produced by aerobic treatments, while the second type of compost was generated from a combination of anaerobic and aerobic biodegradation processes. Both static-batch leaching tests, carried out applying a single liquid to solid (L/S) ratio equal to 10 L kg^{-1}, and dynamic column percolation tests, investigating constituents release as a function of different L/S ratios, were performed. The three materials were also characterized in order to investigate their biological stability degree, organic matter content, and metals total content. As expected, due to the differences in feed input waste or treatment conditions, the three types of samples showed different characteristics and leaching concentrations. However, the leaching behavior of the three types of treated materials presented similar trends of dissolved organic carbon (DOC) and metals (Cu, Ni, Pb, Zn) leaching as a function of the L/S ratio. A screening model proposed and developed for MBT waste in a previous study, which allows the description DOC release as a function of the L/S ratio, was applied and discussed. The comparison of model predictions with the experimental leaching data highlighted that for all three samples the screening model describes quite well the release trends of metals.

Keywords: Compost, MBT waste, leaching properties, modelling.

Estudo da propriedade a lixiviação e de resíduos sólidos do tratamento mecânico biológico, da compostagem e dos processos conjuntos anaeróbico/aeróbico

RESUMO

O estudo atual aprofunda-se e compara o comportamento a lixiviação (leaching) de dois tipos de composto, o primeiro produto do tratamento aeróbio da fração orgânica selecionada (dos mercados ortofrutíferos), o segundo tratamento em conjunto com o anaeróbio/aeróbio de fração orgânica de resíduos sólidos da cidade, proveniente de seleção diferenciada, com o resultado da fração orgânica estabilizada produzida de planta do tratamento mecânico/biológico. As três matrizes evidenciaram propriedade e comportamentos diferentes

que dependem seja das várias características dos resíduos que foram tratados seja dos diferentes processos de tratamentos que foram feitos, com relação ao tratamento da lixiviação, foi determinante um desempenho igual para as três matrizes testadas em todos os três resíduos foram observados uma significativa semelhança entre as cinéticas, de lixiviação do carbônio orgânico solubilizado (BOC) e dos metais principais (Cu, Ni, Pb, Zn) em função do relacionamento. L/S. em base a essas evidências foi adotado o modelo interpretativo já proposto e desenvolvidos dos mesmos autores para uma fração orgânica estabilizada proveniente do tratamento mecânico biológico em outro estudo. O confronto entre os resultados obtidos com o modelo interpretativo adotado e com aquele obtidos dos testes de lixiviação realizados evidenciam como o modelo interpretativo replica bem o desenvolvimento real da liberação dos metais pesquisados liberados.

Palavras-chave: Composto, tratamento mecânico biológico (TMB), lixiviando proprietários, modelagem.

1. INTRODUCTION

In the last decades, since the issuing of the European Landfill Directive 1999/31/EC, Member States took action in order to divert biodegradable municipal waste from landfills. In fact, the anaerobic biodegradation reactions that ensue from this waste stream in the landfill body generate greenhouse gases and a liquid leachate that typically presents high concentrations of both organic and inorganic contaminants. Inadequate management of biogas and leachate flows over time may lead to significant impacts for the environment and human health. The source-segregated organic fraction of Municipal Solid Waste (MSW) is generally treated by aerobic and/or anaerobic biodegradation processes that produce compost and digestate, which can be used as soil amendments (Abdullahi et al., 2008). Residual MSW may be processed in mechanical–biological treatment (MBT) plants to produce biostabilised waste, besides waste-derived fuels aimed at energy recovery (Lornage et al., 2007; Di Lonardo et al., 2016; Di Gianfilippo et al., 2016).

As mentioned, compost is generally used in agriculture as a soil conditioner for improving soil physical and chemical properties and as a fertiliser, i.e., as a nutrient source for growing crops (Westerman and Bicudo, 2005; Hargreaves et al., 2008). On the other hand, the main fate of MBT waste is landfilling due to the higher content of non-compostable materials (e.g. plastic and glass pieces) and heavy metals compared to compost (Dimambro et al., 2007; Di Lonardo et al., 2015a; 2015b). However, interest in the possibility of recovering MBT waste is increasing (MacLeod et al., 2008), especially considering the large amounts that are being produced in efforts to divert waste from landfills (Farrell and Jones, 2010, Lombardi et al., 2017). Many studies investigated the recovery of MBT waste for land reclamation purposes obtaining positive performances, such as in landfills as cover layer material (Angermeier et al., 2011) or applied to degraded and/or contaminated soils for organic matter supply and for metals recovery, respectively (Farrell and Jones, 2010). In this light, assessing the environmental performance of different types treated biowastes is essential for increasing the material recovery by keeping the soil quality and preventing the risk to human health (Smith, 2009, Pantini et al. 2015a) and, based on the previous targets, defining end-of-waste criteria (EU, 2008). To date, the latter have not been issued at a European level, whereas some Member States (e.g. Italy and Austria) have their own regulations on treated biowaste requirements suitable for recovery. These characteristics are expressed in terms of the total composition (e.g. heavy metals total content), whereas no restrictions on leaching concentrations are prescribed (Vandecasteele et al., 2013). However, the leaching of soluble constituents from waste materials upon contact with water, regarded as the main mechanism of release, may result in a potential risk to the

environment during use or disposal (Van der Sloot et al., 2003; Singh and Kalamdhad, 2013). Therefore, understanding leachate composition is a key issue for the evaluation of long-term impacts either in the case of recovery or landfilling.

Many authors studied the release behavior of organic-rich wastes (generally by means of the static batch leaching test and the sequential extraction method) and various different results were found, as there are several variable factors affecting leaching, such as the origin of the biodegradable feedstock, the type of process employed to biostabilise it, the initial metals and organic content, pH conditions, etc. (Song and Greenway, 2004; Van der Sloot et al., 2003; Amir et al., 2005; Castaldi et al., 2006; Farrell and Jones, 2010; Smith, 2009; Van Praagh et al., 2009; Karak et al., 2013; Vandecasteele et al., 2013; Di Lonardo et al., 2015a; 2015b). An important and common finding raised by these studies was that, beside the pH, which is widely known to influence the solubility of heavy metals (Whittle and Dyson 2002; Van der Sloot et al., 2004; Vandecasteele et al., 2013), in the organic-rich matrices, the role of solid and dissolved organic matter may be considered a key factor in the transfer of the inorganic constituents to the water phase through the formation of soluble and insoluble complexes.

This work assessed and compared the leaching properties of two types of compost, one produced from an aerobic composting process (Compost A) and the other from a combination of anaerobic and aerobic biodegradation processes (Compost B), respectively (Lombardi et al., 2016). The experimental leaching data were then elaborated through a screening model proposed and developed in a previous study (Pantini et al., 2015b), which assesses organic carbon and metals release as a function of the L/S ratio. The experimental and modeled results obtained for the two composts were then compared with those found for an MBT waste (Pantini et al., 2015a), in order to point out analogies and differences in the release of constituents from the different types of organic wastes.

2. MATERIALS AND METHODS

2.1. Origin of the analysed samples

As previously mentioned, two compost samples of different origin were investigated. Compost A was collected, after completion of the maturation stage, at a composting plant located in Rome, (Lazio, Italy) fed by the source-segregated organic fractions of MSW (from local markets, restaurants and households) at maximum treatment capacity of 30000 MgOFMSW y^{-1}. The aerobic composting process is consists of 28 days active phase followed by a maturation stage of up to 180 days carried out on the undersize flow from the mechanical sieving unit at 10 mm. Both phases are carried out under covered conditions.

Compost B, after completion of the maturation stage, was collected at the biodigestor/composting plant located in Central Italy, which treats the source-segregated organic fractions of MSW (OFMSW) (mainly food and green waste) with a maximum treatment capacity of 43500 Mg OFMSW y^{-1}. Unlike the composting plant of Rome, the plant of Central Italy includes two different biodegradation process stages. First, an anaerobic digestion stage of 15 days is carried out in a horizontal biodigestor; then, the digestate output is mixed with "fresh" OFMSW and sent to the aerobic composting stage. The latter is performed in biocells for 20 days, followed by a 90-day maturation stage of the undersize flow from the mechanical sieving unit at 40 mm. Both phases are carried out under covered conditions. A secondary mechanical sieving at 10 mm is carried out at the end of the maturation phase. The oversize flows are recirculated at the beginning of the process in order to act as structuring materials during the two biodegradation stages.

The MBT waste, analyzed in the work of Pantini et al. (2015a), after completion of the stabilization process, was sampled in one of the four MBT plants located in Rome, having a maximum treatment capacity of 750 Mg MSW d^{-1}. Focusing on the biological process, after

metal removal by belt-type electromagnetic separators, the biodegradable fraction (i.e. the undersize flow coming from the primary mechanical sieving unit at 80 mm) is sent to a biostabilisation basin where biodegradation occurs for 28 days in forced aeration conditions. The biostabilised output is then mechanically sieved at 20 mm in order to separate an oversized fraction mainly composed of plastics and inert materials from the undersized fraction, namely the final biostabilised waste (Sample C) which is daily landfilled.

Random sampling from piles, mixing and quartering were performed according to the procedure laid down in the Italian standard UNI 10802 (2013), in order to obtain final samples of roughly 20 kg for lab analyses.

2.2. Analytical methods

Preliminary characterisation of the sampled materials was carried out in order to investigate the biological stability degree, the organic matter content, as well as metals' total content.

Biological reactivity was analysed by determining the dynamic respiration index (DRI), namely the maximum oxygen consumption rate due to microbial activity (Adani et al., 2004). DRI was measured and calculated according to the procedure reported in the Italian Standard UNI/TS 11184 (2006) by using a 30 L adiabatic respirometric reactor (Costech International Respirometer 3024). The organic matter content was determined by measuring the total organic carbon (TOC) in triplicate by means of a Shimadzu SSM-5000A instrument. In order to determine the heavy metals total content (Cu, Ni, Pb and Zn), air-dried and grinded samples were acid-digested at high temperature (USEPA, 1996; Kazi et al. 2009) in triplicate. The obtained solutions, after filtration at 0.45 μm, were analysed by inductively coupled plasma atomic emission spectrometry (Varian ICP-AES). The leaching behaviour of the analysed materials was evaluated by means of both static batch and dynamic column tests in order to identify the key factors affecting organic matter and metals release. The batch leaching test was performed according to the UNI EN 12457-2 (2002) method. A 0.001 M $CaCl_2$ solution was added to air-dried samples grinded to a size lower than 4 mm at a liquid to solid ratio equal to 10 ml/g and bottles containing the mixture were slowly shaken for 24 hours. The up-flow column percolation tests were carried out according to the prEN 14405 (2013) method. Each column was filled with roughly 1 kg of compost samples or 0.5 kg of MBT waste which were introduced in consecutive thin layers (2-3 cm) and compacted with a rammer. After saturation, columns were maintained disconnected from the pump for a few days before starting the test in order to ensure local equilibrium between the liquid solution and the solid matrix. During the experiment, columns were subjected to an upward flow (15±2 cm d^{-1}) of the 0.001M $CaCl_2$ solution. Different leachate samples were collected at fixed L/S ratios. More information about column test conditions are reported in Lombardi et al. (2016) for the compost samples and in Pantini et al. (2015a) for the MBT waste sample. Tests were conducted in duplicates for each sample. All the batch and column eluates as-collected were analysed for the pH (Eustech Instrument pH 700). Then, after filtration at 0.45 μm, the dissolved organic carbon (DOC) (Shimadzu TOC-V CPH/CPN analyser) and the metals concentrations (Varian ICP-AES) were measured.

2.3. Modelling experimental results

The constituent mass release measured in column experiments was modelled using the approach proposed by Pantini et al. (2015a). Considering an equilibrium condition between solid and liquid phases, in the early stages of the test (i.e. low L/S ratios) the mass release is governed by an advection-controlled transport and the cumulative mass release per unit mass of waste, $M_{cum,adv}$ (mg $kgTS^{-1}$), can be estimated as a function of the L/S ratio (Kosson et al., 2002) (Eq. 1):

$$M_{cum.adv} = L/S \cdot C_{sol} \quad (1)$$

Where C_{sol} is the concentration in the liquid phase (mg L^{-1}).

The liquid to solid ratio, L/S (L $kgTS^{-1}$), i.e. the amount of water percolated through the column after a certain time relative to the dry weight of the solids in the column (Grathwohl and Susset, 2009), is a function of the flow velocity attained in the column, as follows (Eq. 2):

$$L/S = \frac{v \cdot \theta \cdot t}{h_c \cdot \rho} \quad (2)$$

Where v is the average flow velocity (m s^{-1}), t the time (s), θ the porosity, h_c the column height (m) and ρ the dry bulk density of the waste in the column (kg L^{-1}).

For extended leaching times, non-equilibrium conditions, resulting from a switch to a mass transfer-controlled scenario, are usually observed (Grathwohl and Susset, 2009; Kosson et al., 2002) and Eq. (1) is not valid anymore. Under a mass transfer-controlled scenario, the cumulative mass release per unit waste mass, $M_{cum,dif}$ (mg $kgTS^{-1}$), can be estimated using the following simplified expression (Kosson et al., 2002) (Eq. 3):

$$M_{cum,dif} = 2\frac{C_0}{h_c}\left(\frac{D \cdot t}{\pi}\right)^{1/2} \quad (3)$$

Where D is the diffusivity of the constituent in the porous medium (cm^2 s^{-1}).

Hence, the cumulative mass release per unit mass of waste expected during the entire duration of the column test, M_{cum} (mg $kgTS^{-1}$), can be estimated as follows (Eq. 4):

$$\begin{cases} M_{cum} = L/S \cdot C_{sol} & L/S \leq L/S^* \\ M_{cum} = L/S^* \cdot C_{sol} + 2\frac{C_0}{h_c}\left(\frac{D \cdot (t - t^*)}{\pi}\right)^{1/2} & L/S > L/S^* \end{cases} \quad (4)$$

Where L/S* and t* are the critical liquid to solid ratio and time, respectively, above which the mass release switches to a mass transfer-controlled scenario.

L/S* can be estimated assuming the critical number of pore volumes, n_{pv}, delivered in the column required to achieve a mass transfer-controlled release condition (Eq. 5):

$$L/S^* = \frac{n_{pv} \cdot S_w}{V_c \cdot \rho} \quad (5)$$

Where S_w is the water required to saturate the packed material in the column (L) and V_c the column volume (L). As a result, the t* can be calculated from Eq. (2), as follows (Eq. 6):

$$t^* = L/S^* \cdot \frac{h_c \cdot \rho}{v \cdot \theta} \quad (6)$$

3. RESULTS AND DISCUSSION

3.1. Sample characteristics and leaching properties

The three considered organic waste samples showed different biological reactivity degrees in the following increasing order: Compost A < MBT waste < Compost B, with DRI values equal to 620, 1456 and 2857 mgO_2 $kgVS^{-1}$ h^{-1}, respectively. This variability was related to the different nature of the input to the biological processes (source-segregated organic fractions and mixed MSW), to the heterogeneity of the feedstock, which in turn is influenced by the location

and seasonal period of sampling, as well as to the type of treatment, i.e. the duration of the process and the type of treatment applied (aerobic or anaerobic). It can be noticed that only Compost A was biologically stable as the DRI was lower than the maximum limit value of 1000 mgO_2 $kgVS^{-1}$ h^{-1}, below which an organic waste presents a good biostability (Adani et al., 2004). This finding was associated to the longer treatment time which this sample underwent compared to the others (as above described). Also, the significant reactivity of Compost B was likely due to a non-efficient treatment process management.

Figure 1 shows the results of the organic carbon and the metals total content in the solid matrix (a), the characteristics of the eluates obtained from the batch leaching tests at L/S = 10 L kg^{-1} (b), as well as metals and organic carbon percentages of release (c), calculated as the ratio between the leached concentration and the total content. It can observed that MBT waste showed the highest metal total content (Figure 1a), especially for Cu and Pb, with a significant standard deviation. This is mainly because MBT waste derives from the mechanical separation of non-segregated MSW (residual from at-source separate collection) which might be composed of materials with high concentrations of toxic elements, whereas the compost samples derived exclusively from at-source separated biodegradable organic fractions. For the same reason, MBT waste presented the uppermost metal concentrations in the eluate (Figure 1b), as reflected by the release percentages (Figure1c), which were found to be particularly significant for Ni and Pb. The total and dissolved organic carbon were quite similar between the three samples, also in terms of release percentages.

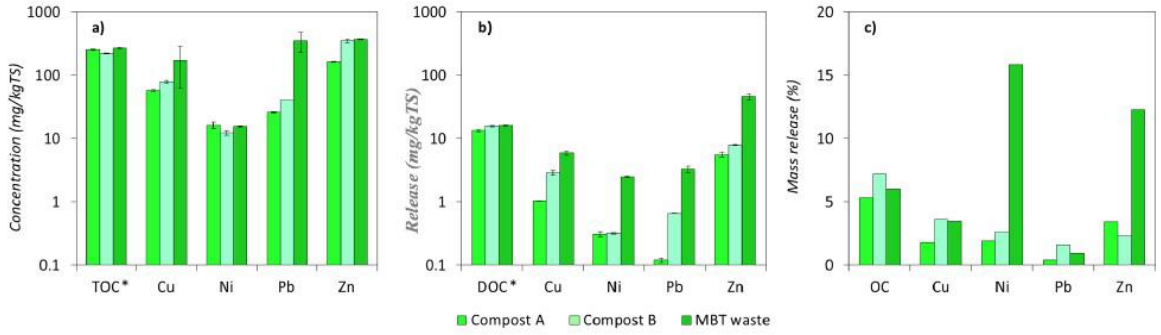

Figure 1. a) Organic carbon and metals total content (*TOC expressed in g $kgTS^{-1}$), b) characteristics of the eluates from batch leaching tests (*DOC expressed in g $kgTS^{-1}$), c) organic carbon (OC) and metals release percentages.

Figure 2 shows the pH trend and the cumulative mass release of DOC and metals measured during column leaching experiments as a function of the liquid to solid ratio (L/S). Data are reported as an average of the two column replicates per each sample.

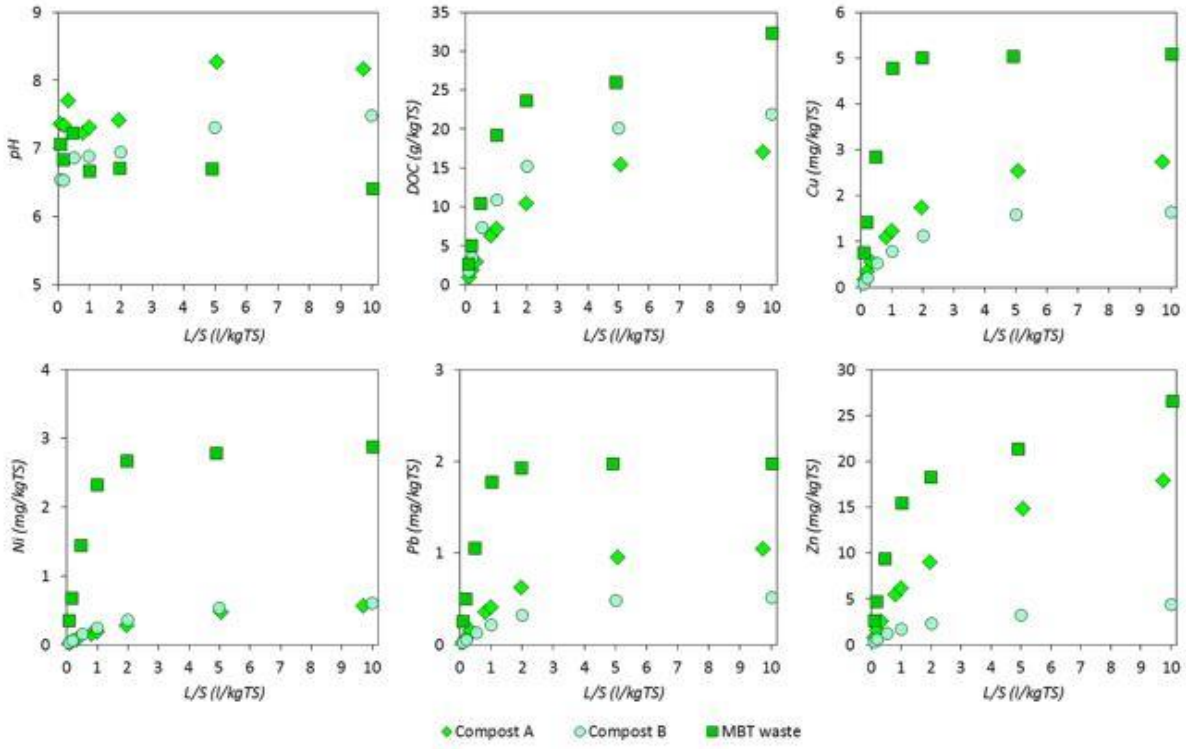

Figure 2. pH trend and cumulative mass release of DOC, Cu, Ni, Pb and Zn measured from column leaching tests as a function of the L/S ratio.

No significant variation of the pH trend as a function of increasing L/S ratios can be observed for all the three samples. However, the cumulative mass release of DOC and metals was found to be affected by the L/S value; in particular, a similar trend of the curves expressing metal/Doc release as a function of L/S was observed for all the tested samples and all the investigated contaminants. Specifically, the latter rapidly increased at low L/S ratios (L/S < 2) indicating a fast washout of the elements. Afterwards, at higher L/S values, the slope of the cumulative curve decreased, especially in the case of MBT waste. It can be further noticed that the DOC cumulative release was found to be higher for MBT waste compared to the compost samples, likely because of the higher biodegradability of the organic matter, that means a higher solubility (Said-Pullicino et al., 2007), due to the relatively short biological process carried out in the MBT plant. As for metals cumulative mass release, the two compost samples exhibited lower leaching of metals than the MBT waste. This can be again related to the longer duration of the decomposition process compared to that of the MBT waste, since metals tend to be bound to the more-mineralized solid organic matter (Castaldi et al., 2006; Smith, 2009; Di Lonardo et al., 2015a).

3.2. Modelling organic carbon and metals release

The results obtained from column and batch tests were interpolated through the model developed by Pantini et al. (2015a) in order to find the key mechanisms governing elements release and to compare the leaching behaviour of the three investigated samples.

Firstly, by applying the above-mentioned model, the DOC cumulative mass release trend was obtained by considering the highest DOC value measured (i.e., the one corresponding to the first eluate at L/S equal to 0.1 L kg^{-1}) and the set-up parameters of the column tests (from Pantini et al., 2015a and Lombardi et al., 2016) as input data. As shown in Figure 3, the modelled data were then compared with the experimental results measured during the column tests proving a very good fit for all the three samples, as confirmed by the significant correlation factor $R^2 > 0.95$.

Therefore, it can be deduced that at low L/S ratio organic matter release is mainly governed by a washing mechanism which switches to a mass-transfer controlled one after a certain period t*. It has to be pointed out that the latter is determined by choosing the diffusivity coefficient D and the critical number of pore volume n_{pv} as best-fitting parameters and, in this regard, it is interesting to note that the D values used resulted in the same order of magnitude for the three different samples (Figure 3).

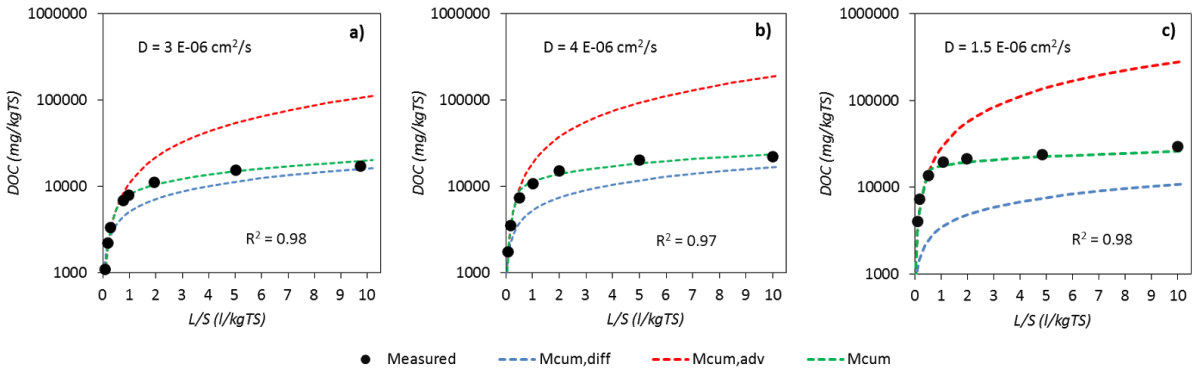

Figure 3. Comparison between measured and modelled data of DOC cumulative mass release as a function of the L/S ratio for a) Compost A, b) Compost B and c) MBT waste.

In Figure 2, a certain similarity between the elution profiles of DOC and those of the considered metals was observed, suggesting a possible interdependence. Hence, for all the three samples, the concentrations of Cu, Ni, Pb and Zn were plotted as a function of the DOC measured in each eluate of column and batch experiments, as shown in Figure 4.

A strong linear correlation ($R^2 \geq 0.9$, with the exception of Pb in the cases of Compost A and MBT waste) between the concentration of the metals in the eluates at different L/S ratios and the dissolved organic carbon can be observed. It must be further noted that, for the three investigated samples, the correlation coefficient $K_{DOC,Me}$ (i.e. the slope of the regression line) were in the same order of magnitude, with the exception of $K_{DOC,Cu}$, which was found to be lower for compost B compared to compost A and MBT waste, as well as $K_{DOC,Ni}$ which was higher for MBT waste than for the two composts.

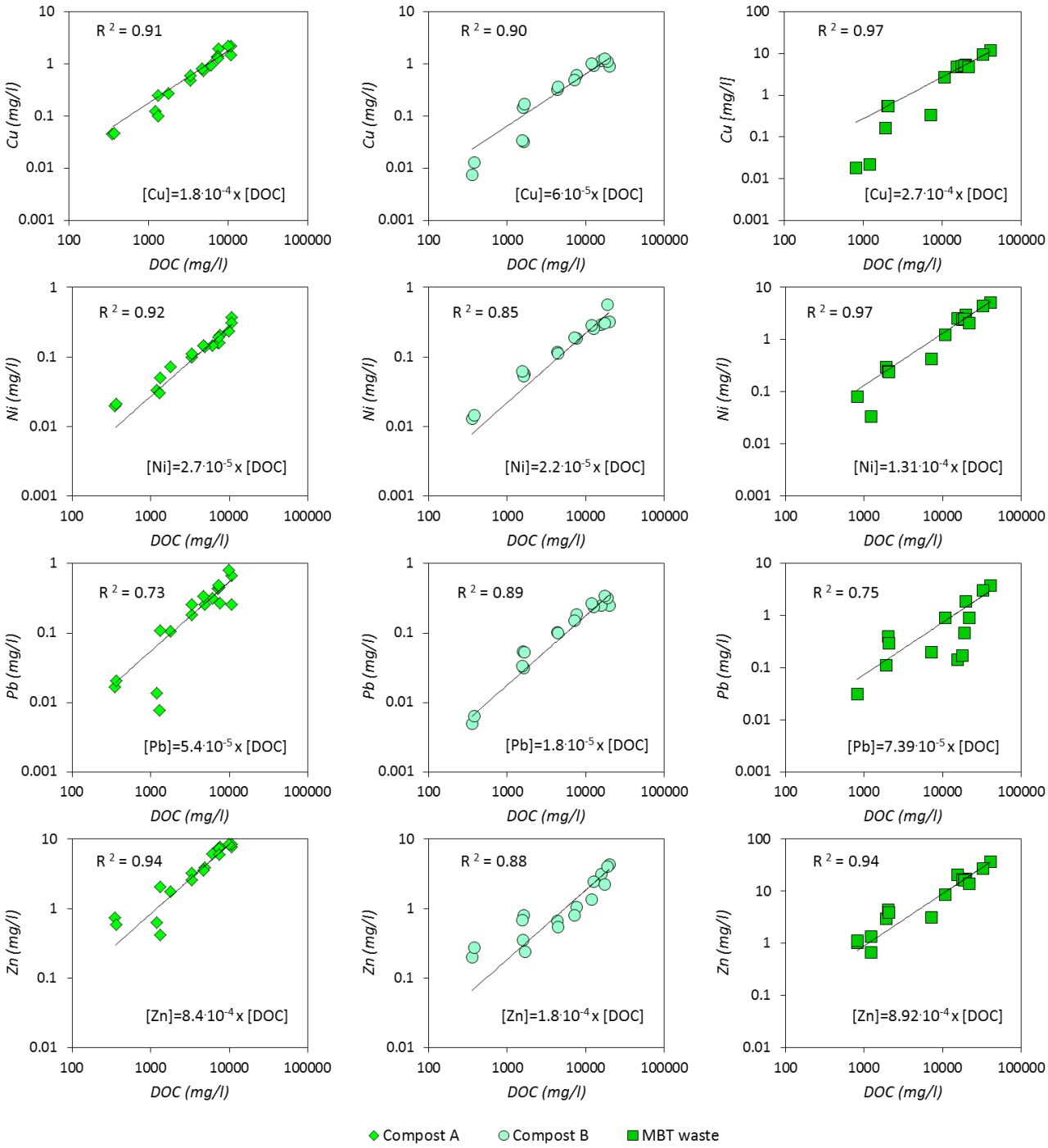

Figure 4. Metal concentrations (mg/l) plotted as a function of DOC concentrations (mg/l) per each investigated sample.

On the basis of the significant correlation found between metals [Me] and the dissolved organic carbon [DOC], the mass release of Cu, Ni, Pb and Zn may be derived using the correlation coefficients, $K_{DOC,Me}$ from the equation describing the regression lines shown in Figure 4, as follows (Pantini et al., 2015a) (Eq. 7):

$$[Me] = K_{DOC,Me}[DOC] \quad (7)$$

Hence, assuming that complexation to DOC is the main mechanism governing the release of the considered metals, results of column experiments were modelled combining Eqs. (4), (7) as follows (Eq. 8):

$$\begin{cases} M_{cum} = (L/S \cdot DOC) \cdot K_{DOC,Me} & L/S \leq L/S^* \\ M_{cum} = \left(L/S^* \cdot DOC + 2\frac{TOC}{h_c}\left(\frac{D\cdot(t-t^*)}{\pi}\right)^{1/2}\right) \cdot K_{DOC,Me} & L/S > L/S^* \end{cases} \quad (8)$$

Where TOC is the total organic content of the solid matrix, $K_{DOC,Me}$ the empirical correlation coefficient between metal (Me) and DOC, h_c the column height, D the diffusivity coefficient of DOC in the porous medium ($cm^2\ s^{-1}$), t* and L/S* the critical time and liquid to solid ratio that allows the switch from flux-controlled to mass transfer-controlled scenario, computed through the critical number of pore volume n_{pv} (Eqs. (5), (6)).

The results returned by Eq. (8) were then compared with the metals' cumulative mass release measured during the leaching experiments, as shown in Figure 5. It is interesting to note that this modelling approach may provide a good description of the cumulative mass release of Cu, Ni, Pb and Zn from organic waste material, as proved by the high correlation factors ($R^2 > 0.9$) found between the measured and the modelled data. This indicates that, in principle, from the total organic content (TOC) and dissolved organic content (DOC) of an organic-rich waste material, a reliable estimation of the metals release can be attained by employing a simplistic approach. It should be highlighted, however, that the model does not account for other processes (e.g. geochemical, biological) that may influence the long-term leaching behaviour of these materials.

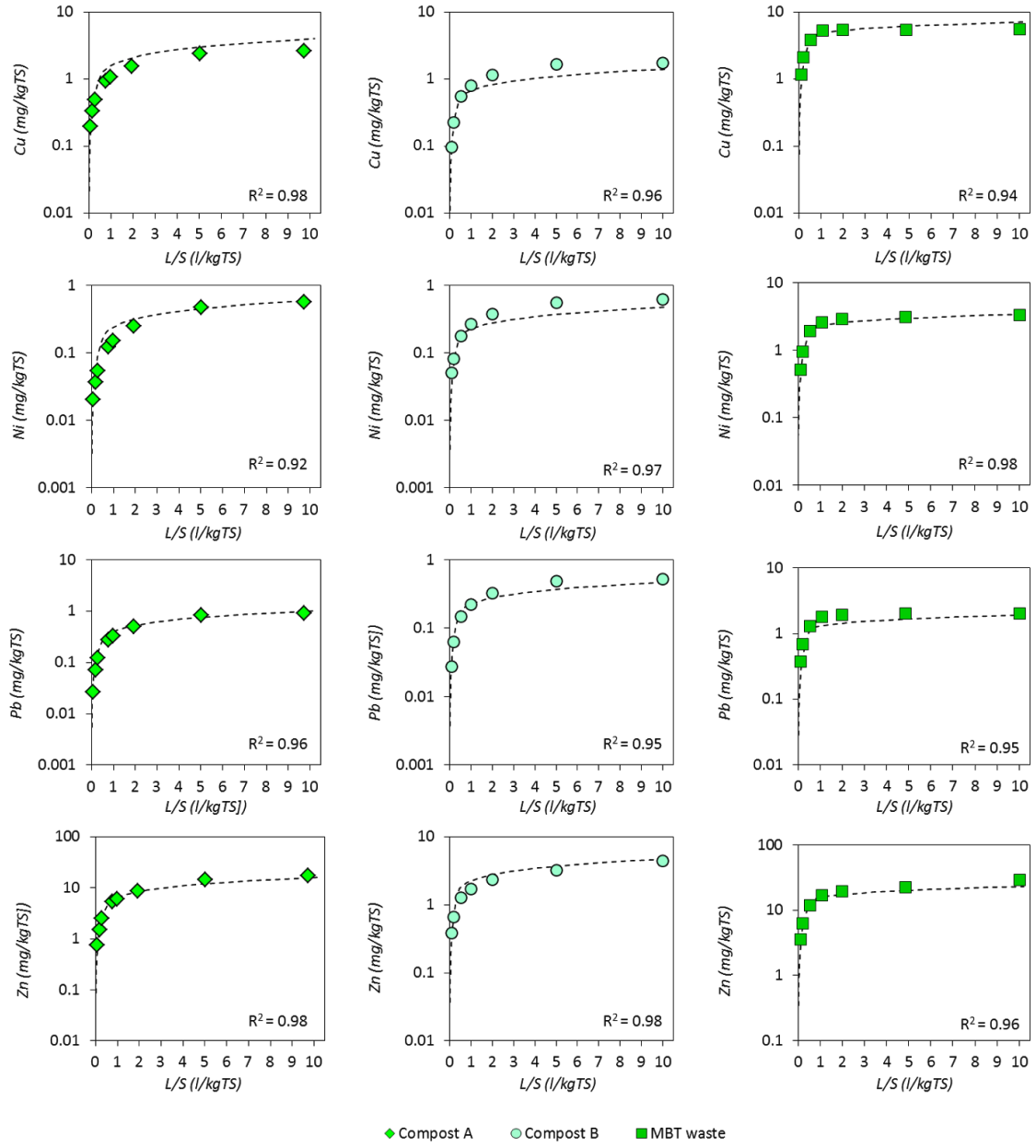

Figure 5. Comparison between measured data and modelled values (dotted line) of the metals cumulative mass release as a function of the L/S ratio.

4. CONCLUSIONS

The results obtained in this work showed that three different organic waste samples, coming from different feed-waste types and treatment processes, are characterized by similar leaching behaviors. Furthermore, for all the investigated materials, a significant similarity between the leaching pattern of the dissolved organic carbon and the one observed for the investigated metals (Cu, Ni, Pb and Zn) was found. Based on this evidence, by plotting the concentrations of the DOC and metals measured in each eluate of column and batch leaching experiments, specific correlation coefficients for each metal were derived. It is worth pointing out that these correlation coefficients were quite similar for the three samples. The correlation coefficients were then integrated in a simplistic model that simulates the cumulative release of DOC, allowing the description of the release of metals as a function of the L/S ratios applied. The comparison with the experimental data observed in the different column tests highlighted

that the screening model describes quite well the leaching trend of the metals. This result suggests that, from the total organic content (TOC) and dissolved organic content (DOC) of an organic rich waste material, a reliable estimation of the release of metals can be attained by employing a simplistic approach such as the one presented in this work. However, further investigation is needed to better understand release-controlling mechanisms and in which form metals are leached, e.g., by performing pH dependence leaching tests, sequential extraction for metals speciation, as well as DOC fractionation.

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