RELEASE OF POLLUTANTS IN MBT LANDFILLS: LABORATORY VERSUS FIELD

Ancella Molleda [a], Ana López *[a], Miguel Cuartas [b], Amaya Lobo [a]

[a] Environmental Engineering Group, Department of Water and Environmental Sciences and Techniques, University of Cantabria, Avd. Los Castros s/n, 39005 Santander, Cantabria, Spain

[b] Information Technologies Group, Department of Applied Mathematics and Computer Science, University of Cantabria, Avd. Los Castros s/n, 39005 Santander, Cantabria, Spain

* Corresponding author

ABSTRACT

The evolution of liquid pollutants over time remains one of the main issues in a landfill for decades. However, although the literature contains several works that attempt to model the release of pollutants from waste, there is still scant information on the matter, especially for MBT waste. In this study, contaminant washing laboratory tests and field trials have been carried out on different waste particle sizes and fractions to estimate to what extent the former provide adequate information about the evolution of the leachate in a real MBT waste landfill. The results show significant differences between the test results. The majority of inorganic contaminants (Ca, Cu, Mg, Na, Ni, Zn), however, complex with organic matter in all percolation column tests. The use of other materials, the early beginning of the methanogenic phase, and the increase in pH and temperature in the field alter the reactions and, importantly, the leaching of some of the components that precipitate, such as Ca, Fe and Mg, or NH_4-N, which increases its presence. It is therefore necessary to adapt the models and the current test standards to assess the fate of biodegradable waste such as MBT under field conditions.

KEYWORDS: MBT waste; field test; laboratory test; leaching behaviour; released contaminants

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1. Introduction

The emission of liquid contaminants is one of most relevant issues once waste has been disposed of in landfills. It may generate a major environmental impact and thus a significant part of the landfill operation and maintenance budget must be assigned to its treatment. Nevertheless, significant uncertainties exist about how the infiltrated water flows through waste and how the pollutants are released into the liquid over time. This uncertainty becomes greater for mechanical-biological treated (MBT) waste landfills, since most studies on this type of material focus on other physical-chemical properties (Münnich et al., 2007; Robinson et al., 2005) or on the gas emissions (Bockreis and Steinberg, 2005; De Gioannis et al., 2009; Pantini et al., 2015b) that they produce.

Five stages are usually distinguished in the degradation of municipal solid waste (MSW). When the waste is disposed of in the landfill, an initial adjustment is produced, followed by a transition phase in which all the oxygen present between the pores in the waste is consumed. Later the acidogenic phase appears, in which hydrolysis and acetogenic processes predominate until the generation of methane begins in the methanogenic phase. During the last phase of maturation, the slowly biodegradable organic matter can be consumed, and the generation of biogas gradually decreases (Cappai et al., 2005; Tchobanoglous et al., 1993) .

Some authors agree that the organic load of leachate decreases considerably after a mechanical biological treatment (MBT). This is due to the fact that the acidogenic phase in the landfill degradation process is considerably shortened or disappears and leachate characteristics similar to those of conventional MSW landfills in the methanogenic phase, which present much lower levels of contamination, are rapidly established (Robinson et al., 2005; Siddiqui et al., 2012; Sormunen et al., 2008). Nonetheless, the effect of MBT on other pollutants, which could be relevant, is still not well known. Some inorganic contaminants may gain relevance as a consequence of the treatment. The proportion of metals in the material increases with respect to untreated waste due to the loss of organic matter produced by the treatment (Tungtakanpoung, 2007). But the metal leaching potential is not always greater: many metals join the organic matter from the waste matrix after the biological stabilization process (Farrell and Jones, 2009; van Praagh et al., 2009). In addition, although some authors have reported that the biological treatment as a very efficient method for reducing the ammonia–nitrogen (NH_4-N) content (Cappai et al., 2005; Trois and Griffith, 2008), others have measured ammonium concentrations in the MBT leachate that remain high for long periods of time (Salati et al., 2013; Tran et al., 2014).

Different laboratory and field tests have been used in the last few decades to characterize the leaching potential of waste, depending on the objectives of the study and other factors (economic, time constraints, etc.). The easiest, fastest and most commonly used trial is the leaching test (Grathwohl and Susset, 2009; Krüger et al., 2012; Luo et al., 2019; Naka et al., 2016; Qiang et al., 2015). However, nowadays percolation tests are considered more suitable to simulate the flow of water through the material in the laboratory and the evolution of contaminants in the leachate over a long period of time (López Meza et al., 2008). This is mainly due to the different agitation and dilution conditions in the two tests, since the appearance of contaminants in the liquid depends not only on the total amount present in the washed solid, but also on the amount available and the leaching kinetics. Even though the available amount is usually estimated through the leaching test, the information gathered with this test is only partial, since information on the release kinetics is not obtained (López Meza et al., 2010).

The majority of studies published on percolation tests, performed in laboratory columns, do not follow any standard (Cossu and Lai, 2012; Fellner et al., 2009) and thus the size of the column, the particle size of the waste, as well as other test conditions, differ (Fang et al., 2017; Krüger et al., 2012; Naka et al., 2016; Qiang et al., 2015). This makes it difficult to compare different works. The European standard, CEN/TS 14405 (CEN/TS, 2004), which establishes specific laboratory conditions for a percolation test, is aimed at avoiding this difficulty. Yet, it does not reproduce a real scenario, since, for instance, it is run in saturated columns while in a landfill waste is only subjected to rainfall percolation. Nevertheless, the results obtained are taken as an estimate of the potential release of pollutants from waste in landfills (Butera et al., 2015; Geurts et al., 2016).

The CEN/TS test has been widely used to analyse the washing of pollutants in waste with low organic matter content (slag, ash, construction and demolition waste, etc.) (Di Gianfilippo et al., 2018; Geurts et al., 2016; Yin et al., 2018). Moreover, the influence of some parameters on it (column size, hydraulic regime, etc.) and the relation with other tests, such as the leaching test, have also been studied (Butera et al., 2015; Delay et al., 2007; Di Gianfilippo et al., 2018; López Meza et al., 2008; Qiang et al., 2015). On the other hand, the column and leaching tests require a reduction in the size of the residue that leads to an overestimation of the leachate contamination (Kosson et al., 2002) due to the increase in the contact surface (Galvín et al., 2014). In spite of this, both are used to establish environmental regulations, including, for example, the limits for waste admission in landfills (European Council, 2002).

In recent years, works have been published that use the CEN/TS 14405 test to characterize the washing of contaminants in MBT waste (Lombardi et al., 2018; Pantini et al., 2015a) or to evaluate the possibility of washing these materials prior to landfilling (Cossu and Lai, 2012). However, some authors have observed limitations of laboratory tests on biodegradable waste (Di Maria et al., 2013; Fellner et al., 2009), and they suggest that the best option to estimate the washing of contaminants in the landfill is in situ sampling. In an in situ test (Tatsi and Zouboulis, 2002; Tojo et al., 2011) measures must be taken frequently over a prolonged period of time to characterize the leachate generated under different meteorological conditions and degrees of decomposition, since the results must be representative of the entire landfill (Münnich et al., 2012). This increases the cost of the technique considerably and hinders the extrapolation of conclusions from some cases to others.

This study aims to investigate how useful laboratory tests are to estimate the evolution of leachate in MBT landfills and how test conditions influence the results. To this end, a type of MBT waste has been analysed and the results obtained through different laboratory tests have been compared with each other and with those obtained in the field, in an experimental cell.

2. Materials and methods

The material analysed was the rejected fraction from the refining stage of a Spanish MBT plant, where approximately 220,000 t of raw municipal mixed waste are treated every year. The first treatment stage in the facility includes the separation of recyclable and high calorific value materials, which are valorised in an annexed energy recovery plant. Then, the fraction under 90 mm, separated using a trommel, is biologically stabilized in a covered warehouse, where it is mechanically turned once a week for 8 weeks. There is no addition of water or other elements.

After aerobic stabilization, the raw compost-like material is subjected to a process carried out to recover the combustible fraction (> 42 mm) by means of a trommel. The fraction smaller than 42 mm undergoes a refining process that consists in screening the particles greater than 15 mm, separating the heavy materials (ceramics, glass and inert material) and removing the light fraction (plastics). The compost-like material is stored until its commercialization as a soil improver, while the rejects from refining make up the material destined for final deposition in the landfill. The composition of this latter fraction, which represents approximately 40% of the total amount of material deposited in the landfill, can be found in López et al. (2018).

In this study, four types of tests were carried out on the rejected fraction: three on a laboratory scale (one leaching test and two types of column test) and one on a field scale with a pilot cell.

2.1. Leaching tests

The leaching tests were performed on samples of the rejects taken from the facility described above and prepared including only the biodegradable materials (paper and cardboard, textile, wood, bones, shells and seeds, and mixed biodegradable material), in the same proportion as they appear in the studied waste (the rejects from the refining process of the compost-like material). The test followed the EN 12457/2 standard (ECS, 2002): approximately 100 g of sample was reduced in particle size to less than 4 mm and mixed with 1000 mL of deionized water in a sealed plastic container at a liquid to solid (L/S) ratio of 10.

Samples were introduced in a rotary shaker, where they were maintained at a speed of 10 rpm for 24 h at room temperature. The solid and liquid phases were then separated and the different parameters in the eluate were determined (see section 2.4.).

2.2. Percolation tests in columns

Two different column sizes were used in the percolation tests. “Small columns”, with a diameter of 80 mm and a height of 200 mm, and “Intermediate column”, 200 mm in diameter and 800 mm high. Both were tested under the procedure of the CEN/TS 14405 standard (CEN/TS, 2004), as upflow percolation tests in saturated regime. However, only the small columns fully complied with the standard, which establishes a diameter of between 50 and 100 mm. According to the standard, the waste size in the experiment must be smaller than a tenth of the column diameter. For the small columns this means a maximum particle size of 8 mm. In accordance with this requirement, two types of samples were prepared. On the one hand, the waste was screened using a sieve with a pitch size of 8 mm and the through material, the ‘fines’, was taken to columns A and B. On the other hand, the material smaller than 8 mm was mixed with the fraction greater than 8 mm, which was cut with scissors to reduce the size to below 8 mm. This mixed material was studied in columns C and D.

The intermediate column was filled directly with rejects without any special preparation, all the material being smaller than 30 mm. Initially two test columns were prepared, but problems with the watertightness of one of them prevented the test from being carried out in duplicate. Therefore, to ensure the quality of the results, special care was taken in the operation and monitoring of the single column that was tested.

Table 1 presents the details of the percolation experiments.

Table 1 Details of the percolation tests performed in the study.

COLUMN	**Small Φ = 80 mm**				**Intermediate Φ = 200 mm**
Material	Sieved waste < 8 mm		Sieved + reduced size waste < 8 mm		Raw waste (< 30 mm)
Test Identifier	A	B	C	D	
Dry weight (g)	447	448	561	586	12210
Flow (mL h^{-1})	32.29	33.27	38.21	32.03	202.20

In all cases, a layer of sand and a filter paper were included both at the base and at the top of the columns. Once filled, the columns were saturated with distilled water and left to rest for three days in order to establish equilibrium conditions throughout their entire height. The pump was then started again at a speed such that the flow rate in each case was equivalent to approximately 15 cm d^{-1} along the column, as required by the standard.

The collection times for the L/S = 0.1, 0.2, 0.3, 0.5, 1, 3, 5 and 10 ratios were calculated by means of Eq. 1.

$$t = \frac{\frac{L}{S} \cdot m_0 \cdot 1000}{24 \cdot C} \tag{1}$$

where t is the sampling time in days, m_0 is the initial dry weight of the waste in kg and C is the flow rate in mL h^{-1}.

Additional samples were taken in the small columns, some of them through an automatic sampler. The parameters specified in section 2.4 were determined in each of them.

2.3. Field test

The pilot cell measured 30 x 30 m^2 and 4 m in height and included 4,966 kg of the rejects coming directly from the refining stage of the MBT plant, with an average moisture content of 30%. It was located on the intermediate cover of one of the operation layers of the landfill, following the tipping front in an area delimited by a wall of clay soil. This perimeter wall, together with a plastic sheet protected with geotextile that was placed on the bottom of the cell, isolated the contour. Thus, the liquid accumulated on the bottom corresponded only to the leachate generated in the cell. The leachate collection system was placed on the diagonal of the cell, on the geotextile and following the slope of the landfill. It was formed by a coarse aggregate section with a perforated plastic tube in its final stretch. The leachate collection system ended in a pit placed at the base of the landfill, where the samples were collected for analysis. Once constructed, the cell was covered with the same clayey material that is used for the intermediate covers.

In addition, two flow meters were installed for continuous monitoring of the flow rate and surface runoff. The precipitation data and the data collected by the flowmeter allowed us to estimate an infiltration rate of approximately 20% through the clayey coverage. The L/S ratios in the field were determined with this rate. In the pilot cell, other measuring instruments were arranged that allowed the monitoring of other parameters such as the composition of the biogas or the temperature at several points inside it. Details of the cell and its monitoring results are presented in López et al., (2018).

In this case, the leachate samples were analysed monthly for 18 months. The analyses were performed in the laboratory, after previously preserving the samples according to ISO 5667-3 (2012).

2.4. Analysis of the samples

The parameters measured in all cases and compared in this study were pH, electrical conductivity (EC), total nitrogen (TN), total organic carbon (TOC) and different metals: Ca, Cd, Cr, Cu, Fe, Mg, Na, Ni and Zn. In addition, K was measured in the leaching and percolation tests. All the analyses were carried out according to standardized methods (APHA, 2005).

In order to calculate the load of pollutant released per unit mass from the measured concentrations, each measured concentration was assumed to be representative of a volume of water in accordance with Eq. 2:

$$L = \frac{C \cdot (v_1 - v_2)}{M_{dw}} \quad (2)$$

where L is the mass of contaminant released (mg kg^{-1} dw), C is the measured concentration of the contaminant (mg L^{-1}), M_{dw} is the mass (kg) of the tested waste in dry weight (dw) and v_1 and v_2 are the cumulative volumes (L) of leachate from which the measured sample was taken.

2.5. Correlations between variables

The relations between different characteristic variables, measured in the experiments, were studied using a customized Python program executed in Jupyter Notebook. The Pandas and Matplotlib Libraries were used for the analysis and graphic representation of the data. The standard correlation coefficient (Pearson's r) between series of pollutant concentrations (as a function of the L/S ratio) was calculated using the "corr" function of the dataframe object of the Pandas library.

A correlation matrix was represented with the results of each pair of variables, using colour codes to represent the coefficients of linear correlation between the variables, to allow the easy detection of those with a greater relationship between them. In addition, the information was completed with the coefficient of determination (R^2) for the linear model calculated for some variables.

3. Results

3.1. Evolution of pollutants in the percolation tests

The results of the evolution of some pollutants analysed in the different percolation tests can be seen in Figure 1. They show that the initial concentration of the inorganic compounds (Ca, Cr, Cu, Na, Ni and Zn) is significantly different in each type of test, and is generally higher in the small columns. Something similar happens with TOC: while the concentration at the beginning of the test is around 7,500 mg L^{-1} for the intermediate column, this value is between 12,000 and 14,000 mg L^{-1} in the small ones. There are three exceptions to this trend: Fe and Cd (not represented here) present a higher initial concentration in the intermediate column and TN is the only pollutant that shows similar initial concentrations in the three types of tests.

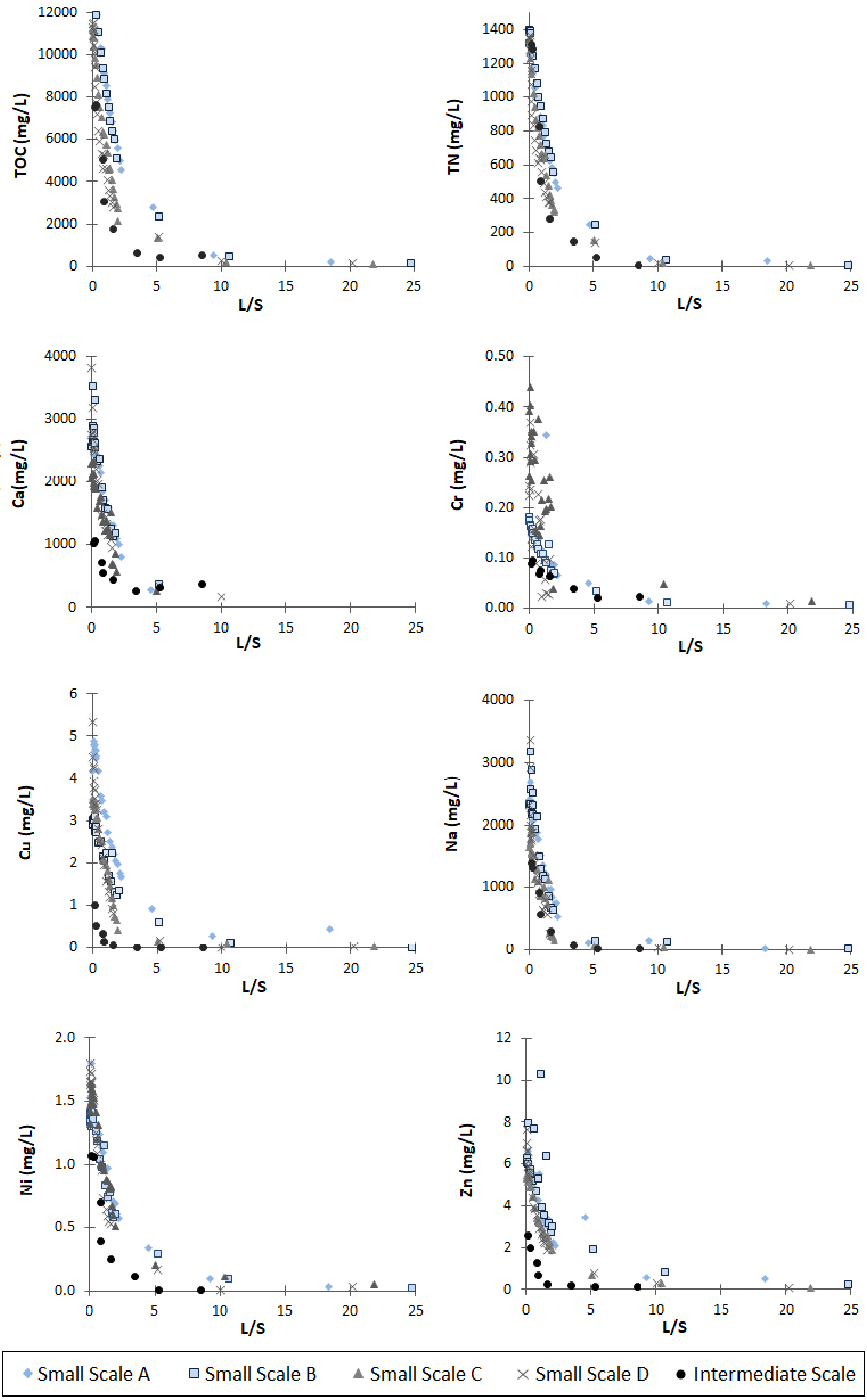

Fig. 1. Evolution of the concentration of some pollutants for the different columns

In most cases, there is a reduction greater than 75% of the initial concentration for L/S of 5 and more than 95% for L/S of 10. However, the evolution in each column depends on the contaminant. Figure 1 shows a slower trend in TN and TOC for the columns containing only fine material (columns A and B): while in these columns the concentrations of TOC and TN have decreased by 80% at an L/S ratio of 5, reductions of 90% occur in columns C and D at this same ratio. As occurs with a large part of the elements considered (Ca, Cr, K, Mg, Na, Ni), however, the concentration has a similar tendency for both scales containing the same material (columns C and D vs. intermediate column), even though the initial concentration changes. For the rest of the metals the minimum values are reached earlier than in the intermediate column in the cases of Ca, Cu and Zn, and in the small columns in the case of Fe.

Table 2 shows that, in general, the smoother tendency of the columns containing fines (A and B) leads to a greater washout of contaminants compared to the rest of the columns for similar L/S ratios at the end of the experiment. Columns A and B release on average almost twice as much TOC as columns C and D. The same is observed for the other organic component: columns A and B have a TN release of 3.62 mg kg^{-1}, while columns C and D release 2.50 mg kg^{-1}. The material smaller than 4 mm consists of the remains of non-identifiable organic waste, which accounts for 30% of the total amount of waste, and the fraction smaller than 8 mm is also mainly composed of organic remains. This composition, different from that of all the rejected material, justifies a greater proportion of pollutants in the leachate, especially organic, since the organic proportion in the solid is also greater.

There are also differences in the release of pollutants between tests with the same fraction of waste but a different column. Table 2 shows that for L/S close to 10, the release of TOC and TN in the intermediate column has been 40% and 45% of those in the small column, respectively. Moreover, the amount of metals released in the small columns (C and D) is greater than in the intermediate column, except in the case of Fe. This difference may be partly due to a greater presence of preferential paths for the flow of water in the larger columns, which favor the appearance of areas of waste isolated from washing that therefore do not contribute contaminants to the leachate.

Another factor that could affect the divergence between tests is the existence of different equilibrium conditions at the beginning (Kalbe et al., 2007). However, in all cases considered in this study equilibrium was achieved in the same contact time: three days. In addition, several authors (Kalbe et al., 2007; López Meza et al., 2010; Qiang et al., 2015) describe how other factors that change with the scale, such as the flow rate or the contact time of the eluate with the residue during the test, hardly modify the release of contaminants.

Table 2 Release of contaminants in the columns at L/S ratio 10.

Parameter	Small columns A-B	Small columns C-D	Intermediate column	Leaching test
L/S	9.97	10.22	8.45	10
pH	6.16-6.89	6.27-7.45	6.34-6.90	6.90
EC (mS cm^{-1})	0.41-18.96	0.42-16.92	1.41-18.12	
TOC (mg g^{-1} dw)	36.15±0.38	21.00±0.55	8.72	27.2
TN (mg g^{-1} dw)	3.65±0.24	2.50±0.12	1.12	2.52
Ca (mg kg^{-1} dw)	-	-	2697	2462
Cd (mg kg^{-1} dw)	0.08±0.003	-	0.09	0.06
Cr (mg kg^{-1} dw)	0.55±0.025	-	0.24	<0.26
Cu (mg kg^{-1} dw)	10.83±1.68	5.52±0.27	0.39	2.01
Fe (mg kg^{-1} dw)	44.38±6.90	37.39±4.18	47.37	-
K (mg kg^{-1} dw)	-	3868±61	934	-
Mg (mg kg^{-1} dw)	1161±299	563±26	239	-
Na (mg kg^{-1} dw)	4681±163	2652±291	1066	1805
Ni (mg kg^{-1} dw)	4.44±0.02	3.67±0.35	0.87	1.03
Zn (mg kg^{-1} dw)	25.48±0.46	13.27±0.08	1.95	3.00

Greater release of contaminants in the tests corresponds to higher initial concentrations. In general, the mobilization of contaminants is greater in the first period of the experiments (Delay et al., 2007), when the processes of dissolution and surface washing predominate, thus producing a depletion of the soluble or interchangeable components over time. The smaller size of the waste, the more available surface area per unit mass (Palmowski and Müller, 2000) and, therefore, the area that can be washed make contaminants easier to transport from the matrix solid to the liquid phase (Schmukat et al., 2012). The size of the waste therefore determines the available contact surface which causes a higher initial liberated concentration in the first stage of the tests, and this marks the released concentration for the rest of the test. The results in this study confirm that the particle size of the waste determines the release of contaminants, in both the short and the long term.

3.2. Leaching test vs. percolation tests

Table 2 compares the results of the leaching test and those of the washing in columns for an L/S ratio close to 10. The amount of TOC released in the leaching test is 22% greater than in the percolation test in columns C and D and 68% more than in the intermediate column. The amount of TN is similar to that in columns C and D and 56% more than in the intermediate column. However, the amount of inorganic contaminants released in the leaching test is generally lower than in the small columns but higher than in the intermediate column, except for Ca and Cd. These results indicate that there is no direct relation between the two types of test for the waste considered, despite the fact that other authors have observed a certain connection for non-biodegradable waste (López Meza et al., 2008).

The differences in the released organic load can be attributed to the particle size, which in the leaching test is 4 mm and therefore smaller than in all the column tests. However, the release of inorganic components is generally greater in the columns than in the leaching test. This may be due to the renewal of water and the longer duration of the experiments in the columns (Pantini et al., 2015a). In the leaching test the water in contact with the material soon reaches equilibrium conditions in which some parameters that affect the leachability remain practically stable. The balance cancels the formation of gradients of some parameters, such as pH or Redox potential, which can change the solubility of many inorganic substances (Kosson et al., 2002). In contrast, these gradients are favoured by the renewal of water in the columns and their effects will be greater due to the extended contact time between eluate and waste.

3.3. Laboratory tests vs. field experiment

The L/S ratio reached in the pilot cell after the 18-month test was 0.11. This value corresponds to the first points of the measurements in the laboratory tests but allows for the identification of some specific circumstances that occur in the field.

Figure 2 shows the evolution of the TOC concentration in the pilot cell compared to the evolution observed in the laboratory tests. The initial concentrations registered in the cell, which are higher than in the laboratory, are attributed to the saturated conditions of the columns and to the fact that the waste reaches the field at a high temperature (44ºC) and without having undergone the protocol for sample preservation that is followed in the laboratory (López et al., 2018). It quickly releases the simplest organic compounds, which appear in the first samples. In spite of this, it is clearly observed how the concentrations decrease drastically compared to the columns favoured by the biological activity in the landfill. The microbiological decomposition, helped by the high temperatures, easily reached the methanogenic phase in the first month. López et al. (2018) describe how the gas generated in the cell maintained a methane concentration over 50% after the first three weeks. Under these circumstances part of the organic matter dissolved in the liquid is degraded to gas and therefore disappears from the leachate. In contrast, the laboratory tests were conducted in a significantly shorter time (maximum 30 days compared to 18 months) and at a lower temperature (approximately 20ºC), which reduces the relevance of the biological activity in the degradation process against the continuous passage of water.

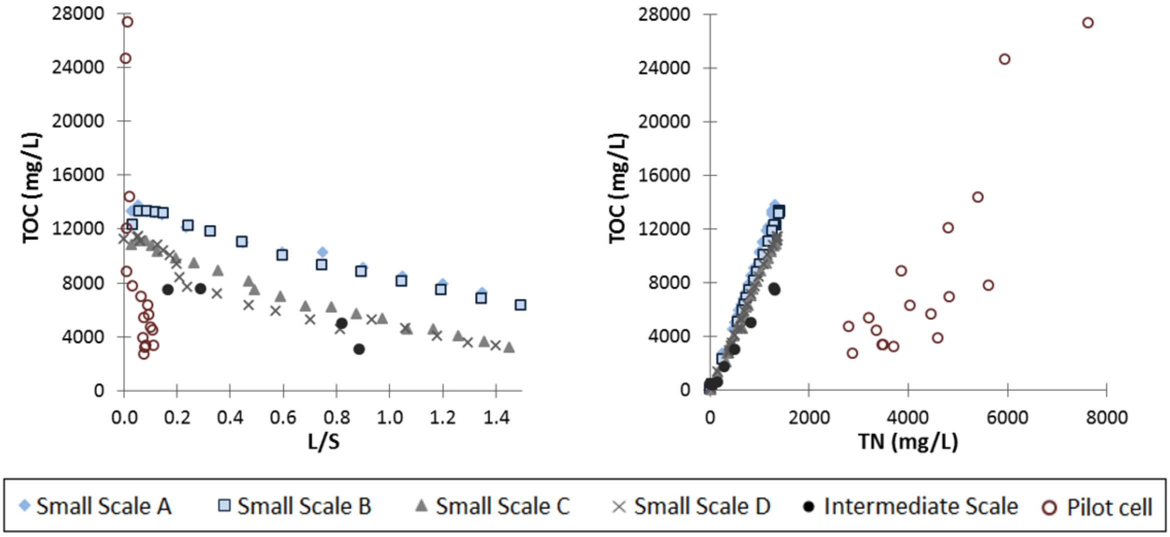

Fig. 2. TOC evolution and correlation of TOC and TN in the cell and in the small and intermediate column

Table 3 shows the total load released in the cell at the final L/S ratio of 0.11 for some pollutants compared to the laboratory results of the tests in small columns C and D and in the intermediate column for a similar L/S ratio, as well as in the leaching test (L/S = 10).

One important point that stands out is the difference in pH: while in the cell it is basic and increases over time (López et al., 2018), it is below 7 in the columns. The lack of activity of microorganisms in the laboratory prevents the biological processes through which the pH rises in the cell.

The amount of inorganic contaminants washed at similar L/S ratios is higher in the cell than in the columns, with the exception of Ca, Cd, Fe and Mg in the intermediate column and Cu and Zn in the small column. The magnitude of the difference between tests varies significantly for each pollutant and, in some cases, is strongly influenced by the pH (Quina et al., 2009): the high pH of the cell leachate favours the precipitation and adsorption of some cations (Ca, Fe or Mg), as in Kjeldsen et al. (2002). Others, such as Na, whose solubility is more influenced by its availability than by pH (Quina et al., 2009), leach more quickly in the cell, reaching higher loads released for the same L/S ratio.

pH is usually basic both in landfills for untreated waste in the methanogenic phase (Christensen et al., 2001) and in MBT landfills with different degrees of decomposition (Robinson et al., 2005). Yet, it rarely exceeds values of 9, as it did in the pilot cell, probably due to the alkalinity released from some of the construction materials used, especially from the calcareous gravel used for the leachate collection network. Over time, as these materials are washed, the pH would probably drop, thereby increasing the solubility and availability of cations that have precipitated as a result (Di Gianfilippo et al., 2016).

Table 3 Release of contaminants in the pilot cell at the end of the field test and in the intermediate and the small columns at L/S ratios similar to those of the cell and in the leaching test

	Pilot Cell	Intermediate column	Small Columns (C-D)	Leaching test
L/S	0.11	0.16	0.13	10
pH	9.6-7.9 (8.9)	6.9	6.3	6.9
EC (mS cm^{-1})	61.8-31.5 (46.8)	17.0		
TOC (mg kg^{-1} dw)	857	618	1234	35320
TN (mg kg^{-1} dw)	496	108	147	2520
NH_4-N (mg kg^{-1} dw)	418			290
Ca (mg kg^{-1} dw)	13	84		2462
Cd (mg kg^{-1} dw)	0.005	0.005		0.056
Cr (mg kg^{-1} dw)	0.091	0.007	0.035	<0.258
Cu (mg kg^{-1} dw)	0.203	0.083	0.442	2.193
Fe (mg kg^{-1} dw)	1.5	2.2	1.3	
Mg (mg kg^{-1} dw)	15	22	39	
Na (mg kg^{-1} dw)	522	114	236	1805
Ni (mg kg^{-1} dw)	0.177	0.088	0.174	1.204
Zn (mg kg^{-1} dw)	0.427	0.214	0.681	3.048

Table 3 shows that 20% of the nitrogen in the leaching test has already been released in the cell, almost 70% more than in the columns at a similar L/S. This high release corresponds mostly to NH_4-N, since more NH_4-N has been released into the cell than in the leaching test (418 mg kg^{-1}vs. 290

mg kg^{-1}). Part of the released NH_4-N is formed during the biological pretreatment in the MBT plant, whereas the rest is generated in the cell as a consequence of the high temperatures that produce the decrease in the nitrification and the emission of nitrogen in the form of ammonium (Beck-Friis et al., 2001; Pagans et al., 2006).

Furthermore, the concentration of NH_4-N can decrease principally through its release in the leachate and secondarily through its volatilization, since there is no mechanism for its degradation under methanogenic conditions (Kjeldsen et al., 2002). This results in the fact that, for the same TOC values, the released TN is greater in the cell with respect to the columns (Figure 2). As a consequence, the linear correlation of the two pollutants is lost.

3.4. Correlation between the different pollutants

Figure 3 left shows graphically the values of the Pearson coefficient as a measure of the correlation between the different pollutants in the leachate considering the whole experimental period (a series of 8 data for each parameter) in all the percolation tests: small (A, B, C, D) and intermediate column. Some parameters, such as TOC, TN, Ca, K, Cu, Mg, Na, Ni and Zn, show a strong direct relationship (Pearson coefficient close to 1). Conversely, values of the coefficient close to -1 show an inverse relationship between pH and organic (TOC and TN) and most inorganic parameters (Ca, Cu, K, Mg, Na and Zn).

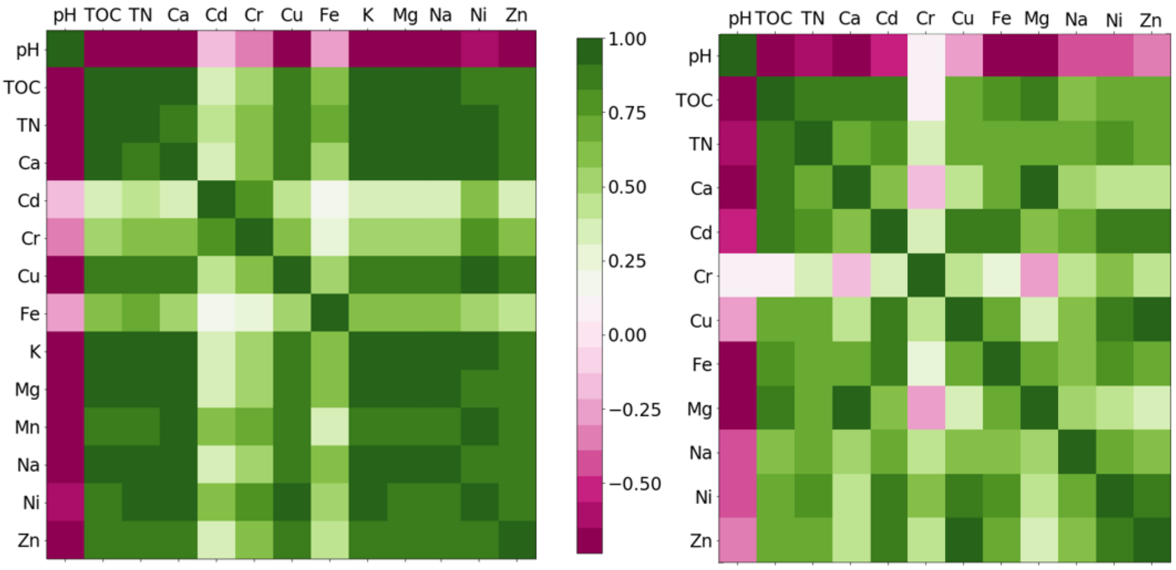

Fig. 3. Pearson correlation coefficient between leached pollutants observed in the percolation tests (left) and in the cell (right)

Figure 3 right shows the values of the Pearson coefficient as a measure of the correlation between the different pollutants measured in the cell leachate throughout the 18-month monitoring period (a series of 17 data for each parameter). It can be seen that the correlation between inorganic contaminants and TOC decreases in the cell, while pH keeps its, strong influence in some cases, such as Ca, Fe and Mg (Pearson coefficients above 0.8).

Figure 4 shows the good linear fit between TOC and a large part of the inorganic components in the column percolation tests. For Mg, Na and Zn the R^2 is higher than 0.90. Thus, despite including particles of different sizes and even different compositions (in the case of columns A and B), the released compounds maintain the organic matter/inorganic elements ratio in all the tests. This linear relation also appears, albeit less strongly ($R^2 > 0.8$), for Ca, Cu and Ni.

This result reinforces those obtained in other works (Lombardi et al., 2018; Pantini et al., 2015a), in which metals such as Mg, Ni or Zn showed a linear correspondence with the concentration of organic matter (represented by the COD in those cases) for different samples of the same MBT material. In this study it is also observed how this correspondence persists even when the sizes or fractions of the studied waste are different. This seems to be due to the effect of complexation of these inorganic pollutants with organic matter. Multiple works demonstrate this complexation in metals, mainly in other media such as soil (Antoniadis and Alloway, 2002; Weng et al., 2002). Moreover, organic matter forms soluble and stable organic complexes, aided by its high solubility, with metals such as Cr, Cu, Ni, Pb or Zn (Pantini et al., 2015a). It has also been reported that other cations such as Ca, Fe, Mg and Mn are released from the waste matrix to form compounds with dissolved organic matter in the leachate (Kjeldsen et al., 2002). In addition, the formation of organic complexes is favoured by the acidic pH values during the tests, which help the extraction of some metals (Antoniadis and Alloway, 2002). Some works indicate that complexation of Zn is not as strong as for other elements, such as Cu (Zhao et al., 2007). However, the results obtained show a linear relation with TOC, even though its concentration evolves differently in some of the columns with regard to the rest (Figure 4).

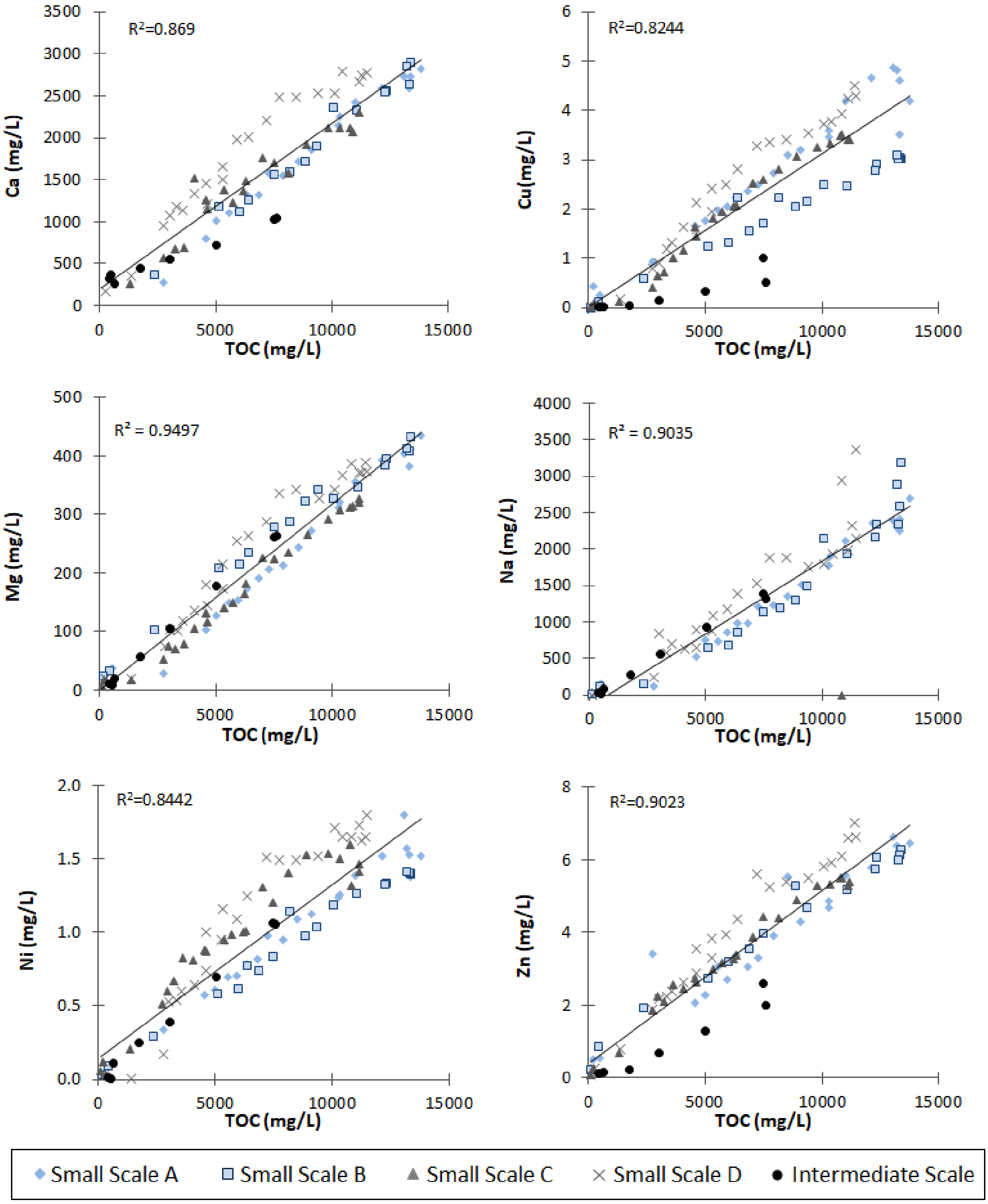

Fig. 4. Correlation of some inorganic components with TOC in the column percolation tests

3.4. Implications regarding waste characterization tests

The CEN/TS 14405 (CEN/TS, 2004) test standard used in the column tests has been incorporated (as a European standard) into the European waste management legislation (European Council, 2002) to assess whether the residual materials meet the leaching limit values associated with their use as substitutes for raw materials or their acceptance in landfills (Hjelmar et al., 2013).

However, with these tests it is not possible to take into consideration the interaction with other materials and the biodegradation that occurs in landfill or the different pH and temperature conditions, which have effects on the washing of contaminants. The results of this study reveal the need to consider the waste biodegradability, that is, to make distinctions between waste with a significant presence of biodegradable organic matter and mainly non-biodegradable waste when establishing criteria, as other experts have already pointed out (European Commission, 2009).

The curves obtained for the pollutants in the columns, which have been shown to follow an advection-dispersion model, do not correlate with those of the cell (López et al., 2018), which is never saturated and only subjected to rainfall percolation. Thus, while this model could sometimes be useful to predict the release of contaminants prior to discharge (Cossu and Lai, 2012), this is not the case. In waste with a significant presence of degradable organic matter such as the one studied, the model should include a term that takes into account waste biodegradation. Moreover, biodegradation in MBT waste becomes significant as of the moment it is deposited, due to the shortening of the non-methanogenic phase (Bockreis and Steinberg, 2005; De Gioannis et al., 2009). A combination of microbial activity, high temperature and different pH levels may affect the release and transport of components into the leachate within the landfill.

Some authors (Di Gianfilippo et al., 2018; López Meza et al., 2008) have studied the influence of pH on the leaching properties of inorganic waste through the CEN/TS 14997 standard (CEN/TS, 2006). Similarly, adjusting the pH values in the laboratory tests could help to analyse the release of inorganic contaminants in MBT waste. The interactions with the organic matter should be carefully considered in those cases.

4. Conclusions

The results presented in this work show that both the particle size and the tested fraction condition the results of the laboratory percolation and leaching tests on MBT waste.

It has been proved how the MBT tested in the laboratory release organic compounds that complex some inorganic elements (Ca, Cu, Mg, Na, Ni and Zn), giving rise to a constant TOC/inorganic elements ratio which does not vary with the size or fraction of the material under consideration.

Yet, the release of pollutants changes under landfill conditions because of the relevance that biodegradation gains, since the washing by the flow of water through the waste is much slower, the temperatures (which may be higher), and the possible changes in pH due to the presence of auxiliary materials. These differences in the pilot cell that was studied significantly affect the release of Ca, Fe and Mg, which decrease, and that of NH_4-N, which increases. This should be taken into account when considering the laboratory results of MBT waste.

In addition, column tests on MBT waste should be carried out, at least, with a basic pH and with a particle size that is similar to real values. This would allow results to be obtained that are as close as possible to those found in a real landfill, using a very simple method.

Likewise, both the regulations used in the admission of landfill waste and the models used in predicting the transport of pollutants in waste with an important presence of pre-treated organic matter should consider the biodegradation of waste. In the case of MBT waste, which rapidly becomes methanogenic as a consequence of pretreatment, this consideration gains importance with respect to fresh waste.

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