



Variations in the properties of leachate according to landfill age

Marta Gómez^{a,*}, Francisco Corona^{a,b}, María Dolores Hidalgo^{a,b}

^aCARTIF Centro Tecnológico, Boecillo, Valladolid, 47151, Spain, Tel. +34983546504; emails: margom@cartif.es (M. Gómez), fraenc@cartif.es (F. Corona), dolhid@cartif.es (M.D. Hidalgo)

^bITAP Institute, University of Valladolid, Valladolid, 47010, Spain

Received 17 October 2018; Accepted 7 March 2019

ABSTRACT

A comparative study of the leachate composition between different types of landfills in Spain and Europe from the point of view of one of the factors that most influence in the leachate composition, such as the landfill age, among others, was carried out. For this purpose, 13 leachate samples from different Spanish landfills were analysed to determine concentrations of organic compounds, for instance COD and BOD₅, inorganic compounds, as calcium, magnesium, potassium, sodium, ammonia, chlorides, sulphates, etc., heavy metals and other physical parameters including pH and conductivity. The leachate composition varied widely because it is affected by many factors such as age and quality of waste, climatic conditions, among others. The results show that there are significant differences between the leachate samples. As the landfill age is higher, the concentration of the leachate constituents decreases due to the processes of stabilization of the waste that occurs within the landfill. On the other hand, comparing the results obtained between the samples of young leachates from different Spanish landfills and young leachates from different European landfills, large variations are observed due to not only the seasonal variations, but also the type of waste that is mostly treated in landfills.

Keywords: Landfill; Leachate; Waste treatment; Municipal waste

1. Introduction

In the last years, the amount of municipal waste generated per person has been decreasing. According to the latest data published by the Municipal Waste Statistics from Eurostat database in July 2018, in EU a total of 483 kg of municipal waste were generated per person (equivalent to 246,377 thousand tonnes) in 2016. This amount of municipal waste varies significantly across the EU Member States. Denmark (777 kg per person), Norway and Switzerland are the countries that generate the highest amount of waste per person and, Romania (261 kg/person) and Poland the countries with the lowest amount of waste generate. In Spain, 443 kg of municipal waste were generated per person in 2016 [1].

Each European country deposits waste in landfills in a greater or lesser amount, that is, it uses the landfilling as

waste treatment. By 2016, in EU, 29% of all treated waste was recycled and 24% was sent to landfill, according to the latest data collected by the Municipal Waste Statistics (Eurostat2018). In the case of Spain, the number of landfills has decreased last years, mainly due to the need to promote other waste management methods, e.g. recycling. However, despite this decrease, Spain continues to deposit its municipal waste mainly in landfills, about 55% in 2016, and this percentage remains in the last years (Fig. 1). Therefore, Spain is the second country that more wastes are landfilled.

One of the biggest problems in landfills is the discharge of leachate. These are generated mainly by the percolation of the water through the waste as a result of incoming rainfall in the landfill, causing its pollution [2].

Directive 1999/31/EC establishes the requirements, which must be complied by the landfill in terms of design and operation, as well as the collection of leachates generated and

* Corresponding author.

their appropriate treatment before being discharged into the environment.

The main component of leachate is organic matters but also it contains ammonia-nitrogen, heavy metals, inorganic salts and chlorinated organics pigments. These pollutants are categorized into four groups as: organic matters, for instance COD (chemical oxygen demand); specific organic compounds, inorganic compounds and heavy metals [3].

The general characteristics of leachates as composition, quantity and potential pollutants are affected by many factors such as quality of solid waste, degree of compaction in landfill, age of waste, climatic condition, pH, chemical and biological process which occurs during degradation [4].

Mainly, the composition of landfill leachate depends on the landfill age and, more specifically of the landfill phases, which are categorized into: aerobic phase, anaerobic acid phase, methane fermentation phase and maturation phase. Each of these phases has different concentrations of the main pollutants.

According to the landfill age, there are leachates called young and old. Young leachates correspond to a landfill age less than 5 years and old leachates correspond to a landfill age of more than 10 years. There may also be leachates called intermediates corresponding to a landfill age between 5 and 10 years.

Characteristics of leachate at different ages of landfill are shown in Table 1.

Another factor that affects the leachates composition is the type of waste present in landfill (hazardous and

non-hazardous waste and inert waste). In Spain, the most generated waste by households are the mixed ordinary wastes followed by the recyclable wastes in smaller amounts in 2016 (Fig. 2). So, depending on the type of waste that is treated primarily in the landfills, the leachate composition is different.

Taking into account all the factors that affect the leachate composition, the aim of this study was to compare the leachate composition from different types of landfills in Spain with respect to the landfills in Europe and to study the effects of one of the factors that most influence in the leachate composition: the landfill age.

2. Material and methods

2.1. Sampling and analytical method

This study was carried out between March and May 2017. Leachate samples from 13 different landfills located in Spain were collected and analysed. After the sample collection, the leachate samples were transported to the laboratory. The collected leachate was stored at 4°C during the period required to complete all experimental analysis. Physicochemical parameters such as pH, conductivity, solids, biochemical oxygen demand (BOD₅), chemical oxygen demand (COD), ammonia nitrogen, nitrate nitrogen, nitrite nitrogen, total nitrogen, total phosphorus, phosphate, sulphate, chloride, sodium, potassium, magnesium, calcium and heavy metals were measured in the leachate samples according to Standard Methods [7].

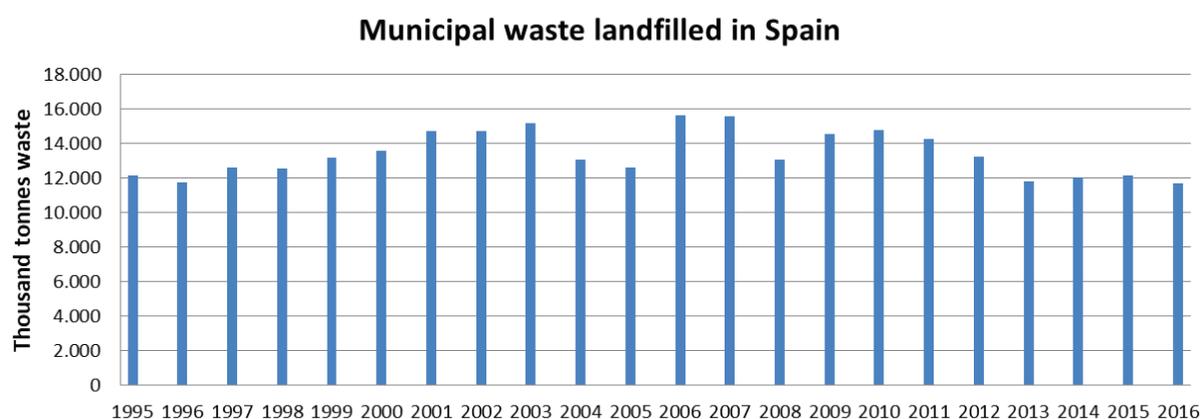


Fig. 1. Municipal waste landfilled in thousand tonnes in Spain [1].

Table 1
Characteristics of leachate at different ages of landfill

Parameters	Ages		
	Young (<5 years)	Intermediate (5–10 years)	Old (>10 years)
pH	6.5	6.5–7.5	>7.5
COD (mg/l)	>10,000	4,000–10,000	<4,000
BOD (mg/l)	>2,000	150–2,000	<150
BOD/COD	>0.3	0.1–0.3	<0.1
Organic compound	80% VFA	5–30% VFA+humic & fomic	Humic & fomic
Heavy metals	Medium	Low	Low

VFA – Volatile fatty acid. Source: Renou et al. [5].

3. Results and discussion

3.1. Overview of the leachates composition from several European landfills of different ages

As already mentioned, there are several factors that affect to the leachate composition. These factors vary considerably from landfill to landfill because of the wastes generated from European Union countries are basically different in terms of composition. It is difficult to find two leachates with the same characteristics, either in different countries or within the same country. Even in the same landfill, the leachate composition changes, because the waste is buried for many years in layers, and therefore, is very common that different parts of landfill are in different phases of decomposition [8].

A comparison of the leachate composition of different ages from different European landfills is shown in Table 2.

All the leachate results shown in Table 2 are from municipal solid waste landfills. According to the results shown in Table 2, in general, as the landfill age increases, the parameters

that indicate the organic matter, such as COD and BOD₅, decrease due to the leachate decomposition. Values of COD vary from 115,000 mg/l of the leachate sample obtained from a landfill in Thessaloniki (Greece) to 500 mg/l of the leachate sample from old landfills in Sweden and Germany. The pH also varies with the leachate age, being higher in the older landfills.

In some parameters, the differences or variations between leachates of the same age and different countries may be due to the different waste composition of each of the landfills or other factors already mentioned.

3.2. Leachate characterisation from different Spanish landfill

The results of the leachate characterisation study in Spain are shown in Tables 3–5. The composition of young leachates from Spanish landfills is shown in Table 3; the composition of intermediate leachates is shown in Table 4 and the composition of old leachates are shown in Table 5.

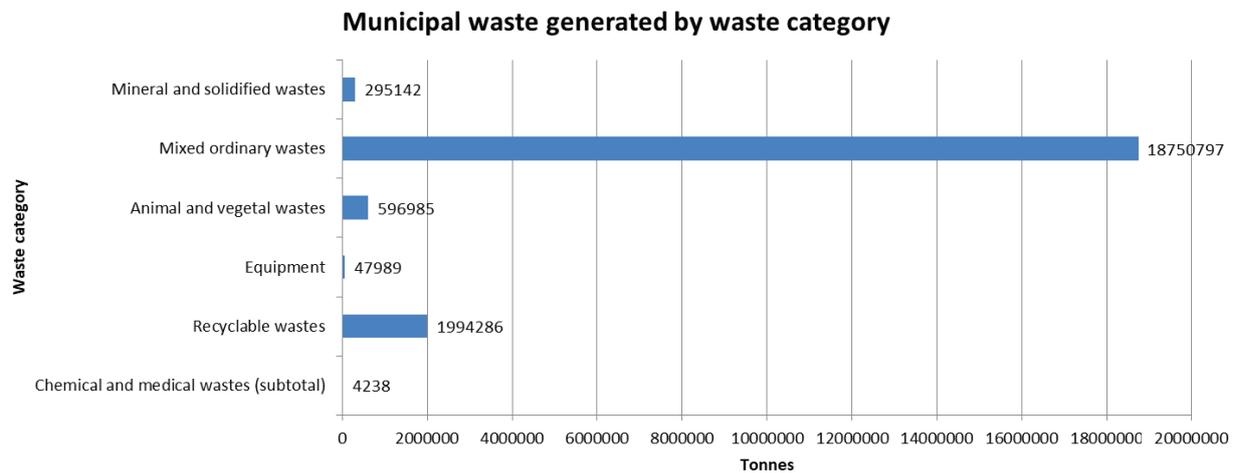


Fig. 2. Municipal waste generated in 2016 classified according to the waste category [6].

Table 2
Leachate composition from different European landfills

European landfills	Age	Parameter							
		COD (mg O ₂ /l)	BOD ₅ (mg O ₂ /l)	pH	Conductivity (mS/cm)	N-NH ₄ ⁺ (mg/l)	SO ₄ ²⁻ (mg/l)	Cl ⁻ (mg/l)	
Ireland [9]	Young	411–7,160	36–984	7.6–8.5	3.09–28.43	130–4,000	7.2–1,950	160–2,620	
Germany [10]		6,000–60,000	4,000–40,000	–	–	–	70–1,750	–	
Greece [11]		44,000–115,000	9,500–80,800	4.9–6.7	23–35.5	10–840	–	–	
Sweden [12]		1,000–30,000	–	5–6	0.5–14	150–560	–	(5)–1,300	
Italy [13]	Inter	2,525–3,879	500–1,800	7.2–8.7	–	512–2,229	23–61	–	
Poland [14]		1,873–3,600	150–273	–	–	971–1,250	–	–	
Ireland [9]		190–748	6–33	6.8–8.4	2.60–10.44	63–378	21–445	130–669	
Germany [10]		Old	500–4,500	20–550	–	–	–	10–420	–
Italy [14]			966–1,784	110–600	7.8–8.5	–	715–1,259	9–25	–
Romania [15]			6,200	–	7.75	–	–	1,670.75	5.03
Sweden [13]			500–4,000	–	8–9	0.5–14	80–370	–	1,000–6,000
Greece [16]	685–15,000	50–4,200	7.3–8.8	6.2–34.0	–	55–500	1,162–9,209		
Poland [17]	1,183	331	8.0	–	743	–	–		

Table 3
Composition of young leachates from different Spanish landfills

Parameter	Young leachate				
	Landfill 4	Landfill 5	Landfill 8	Landfill 9	Landfill 11
Ammonia nitrogen, mg N/l	4,238.7 ± 386.9	5,245.5 ± 465.8	2,302.1 ± 227.4	3,990.8 ± 367.1	2,702.2 ± 261.4
Chloride, mg/l	7,870 ± 1,180	6,310 ± 946	10,300 ± 1,545	7,640 ± 1,146	9,360 ± 1,404
Conductivity, mS/cm	42.8 ± 2.4	44.7 ± 2.5	44.8 ± 2.5	43.2 ± 2.4	40.5 ± 2.2
BOD ₅ , mg O ₂ /l	12,250 ± 2,205	12,690 ± 2,284	11,912 ± 2,144	4,874 ± 877	2,310 ± 416
COD, mg O ₂ /l	29,219 ± 4,010	13,533 ± 1,818	26,592 ± 3,640	14,194 ± 1,909	16,849 ± 2,277
BOD ₅ /COD	0.42	0.94	0.45	0.34	0.14
Total phosphorus, mg P _T /l	148.4 ± 17.4	74.0 ± 9.3	75.4 ± 9.4	97.1 ± 11.8	148.7 ± 17.4
Nitrate, mg/l	<3	<3	<30	<3	<3
Nitrite, mg/l	<4	<4	<40	<4	<4
Total nitrogen, mg/l	5,323.8 ± 1,064.7	5,709.0 ± 1,141.8	3,608.0 ± 721.6	4,438.0 ± 887.6	3,264.0 ± 652.8
Phosphate, mg/l	23.1 ± 4.6	24.1 ± 4.8	56.7 ± 11.3	29.2 ± 5.8	58.7 ± 11.7
pH	8.24 ± 0.39	8.17 ± 0.38	7.63 ± 0.36	8.03 ± 0.38	8.69 ± 0.41
Sulphate, mg/l	9.6 ± 1.4	213.0 ± 31.9	<60	2,250.0 ± 337.5	511.0 ± 76.6
Total suspended solids (TSS), mg/l	3,104.6 ± 392.2	159.0 ± 24.7	639.0 ± 90.0	398.6 ± 58.0	185.5 ± 28.5
Volatile suspended solids (VSS), mg/l	1,815.0 ± 237.9	126.0 ± 19.9	430.0 ± 62.3	229.3 ± 34.7	116.0 ± 18.4
Total dissolved solids (TDS)	26,619 ± 2,899	25,235 ± 2,759	36,222 ± 3,863	28,217 ± 3,061	33,324 ± 3,574
Total solids (TS), mg/l	29,724 ± 2,926	25,394 ± 2,759	36,861 ± 3,864	28,616 ± 3,062	33,509 ± 3,574
Calcium, mg/l	542.59 ± 108.52	47.75 ± 9.55	184.08 ± 36.81	78.49 ± 15.70	9.12 ± 1.82
Magnesium, mg/l	161.35 ± 32.27	194.66 ± 38.93	113.33 ± 22.66	161.70 ± 32.34	13.18 ± 2.64
Sodium, mg/l	5,617.84 ± 1,123.57	4,832.05 ± 966.41	6,869.45 ± 1,373.89	4,694.29 ± 938.86	5,511.10 ± 1,102.22
Potassium, mg/l, mg/l	3,172.96 ± 634.59	2,513.91 ± 502.78	2,959.91 ± 591.98	3,129.35 ± 625.87	4,853.65 ± 970.73
Zinc	0.99 ± 0.20	1.27 ± 0.25	2.68 ± 0.54	1.16 ± 0.23	1.58 ± 0.32
Copper, mg/l	0.51 ± 0.10	4.13 ± 0.83	3.18 ± 0.64	0.65 ± 0.13	1.35 ± 0.27
Chromium, mg/l	7.21 ± 1.44	1.55 ± 0.31	0.85 ± 0.17	2.05 ± 0.41	4.04 ± 0.81
Manganese, mg/l	0.24 ± 0.05	0.15 ± 0.03	0.21 ± 0.04	0.36 ± 0.07	0.15 ± 0.03
Lead, mg/l	0.11 ± 0.02	0.10 ± 0.02	0.07 ± 0.01	0.15 ± 0.03	0.13 ± 0.03
Iron, mg/l	6.42 ± 1.28	5.61 ± 1.12	6.76 ± 1.35	17.25 ± 3.45	29.91 ± 5.98
Nickel, mg/l	0.37 ± 0.07	0.67 ± 0.13	0.25 ± 0.05	0.89 ± 0.18	0.55 ± 0.11
Cadmium, mg/l	0.004 ± 0.001	0.003 ± 0.001	0.029 ± 0.006	0.076 ± 0.015	0.075 ± 0.015

According to the results obtained in the leachate samples, there are significant differences between the different ages of the leachate. It is already known, because it is one of the factors that most affect the leachate composition. As the landfill age is higher, the concentration of the leachate constituents decreases due to the processes of stabilization of the waste that occurs within the landfill. These differences are found in some parameters such as COD, BOD₅ and ammonia nitrogen, mainly.

The trend of the organic compounds concentrations, as COD and BOD₅, with respect to the time (leachate age), is decreasing. It is mainly due to the leachate decomposition because of biodegradable nature and washout [4]. According to the results obtained, the parameter of COD varies from 29,219 mg/l for young leachates samples, to 2,725 mg/l for old leachates, while BOD₅ values vary between 12,690 and 164 mg/l, respectively. These variations of the COD parameter with the leachate age are shown in Fig. 3.

In Fig. 3, it is observed that old leachate samples show low COD values while intermediate and young leachates show higher COD values with a wider range.

The BOD₅/COD ratio is a factor of biodegradability of organic matter and according to Table 1; this ratio tends to decrease as the landfill age increases. Therefore, the higher the ratio is the most biodegradable the organic matter is. This tendency was observed in general in the leachate samples analysed, with a minimum value of 0.05 for old leachates and a maximum value of 0.94 for young leachates.

Apart from organic matter, ammonia is the principal pollutant in leachate. According to Kulikowska and Klimiuk [18], the landfill age has a significant effect on its composition, especially on organics and ammonia concentrations.

In general, the ammonia nitrogen concentration is high, especially in young leachates. In these cases, the ammonia nitrogen values vary between 5,245.5 and 2,302.1 mgN/l, decreasing this concentration when the leachate age increases. It is due to the deamination of amino acids and destruction of

Table 4
Composition of intermediate leachates from different Spanish landfills

Parameter	Intermediate leachate				
	Landfill 1	Landfill 3	Landfill 6	Landfill 12	Landfill 14
Ammonia nitrogen, mg N/l	1,385.8 ± 146.1	3,084.1 ± 293.3	3,568.1 ± 333.0	1,999.9 ± 201.1	2,216.9 ± 220.0
Chloride, mg/l	3,140 ± 471	7,240 ± 1,086	4,940 ± 741	5,780 ± 867	8,970 ± 1,345
Conductivity, mS/cm	16.2 ± 0.9	36.9 ± 2.0	32.2 ± 1.8	29.4 ± 29.4	39.3 ± 2.2
BOD ₅ , mg O ₂ /l	5,550 ± 999	4,100 ± 738	1,184 ± 438	2,307 ± 415	2,450 ± 441
COD, mg O ₂ /l	12,554 ± 1,683	13,701 ± 1,841	10,026 ± 1,336	8,938 ± 1,187	13,646 ± 1,834
BOD ₅ /COD	0.44	0.30	0.12	0.26	0.18
Total phosphorus, mg P _T /l	45.4 ± 5.9	94.4 ± 11.5	64.2 ± 8.1	56.9 ± 7.3	90.2 ± 11.1
Nitrate, mg/l	<3	<3	<30	<3	<3
Nitrite, mg/l	<4	<4	<40	<4	<4
Total nitrogen, mg/l	2,101.5 ± 420.3	3,642.6 ± 728.5	3,386 ± 677.2	2,550.0 ± 510.0	2,928.0 ± 585.6
Phosphate, mg/l	10.7 ± 2.1	48.6 ± 9.7	25.1 ± 5.0	29.3 ± 5.9	20.2 ± 4.0
pH	7.73 ± 0.36	7.81 ± 0.37	8.25 ± 0.39	7.96 ± 0.37	8.43 ± 0.40
Sulphate, mg/l	65.8 ± 9.9	81.0 ± 12.1	<60	7.6 ± 1.1	824.0 ± 123.6
Total suspended solids (TSS), mg/l	595.0 ± 84.2	117.5 ± 18.6	197.5 ± 30.2	153.5 ± 23.9	257.0 ± 38.6
Volatile suspended solids (VSS), mg/l	446.7 ± 64.5	79.0 ± 12.9	142.5 ± 22.3	120.5 ± 19.0	175.0 ± 26.9
Total dissolved solids (TDS), mg/l	14,166 ± 1,612	27,372 ± 2,976	20,361 ± 2,259	19,076 ± 2,126	31,923 ± 3,434
Total solids (TS), mg/l	14,761 ± 1,614	27,489 ± 2,976	20,558 ± 2,260	19,229 ± 2,126	32,180 ± 3,434
Calcium, mg/l	809.97 ± 161.99	70.23 ± 14.04	96.88 ± 19.38	174.85 ± 34.97	–
Magnesium, mg/l	186.56 ± 37.31	65.81 ± 13.16	90.46 ± 18.09	98.88 ± 19.78	222.71 ± 44.54
Sodium, mg/l	1,435.18 ± 287.04	4,802.39 ± 960.48	3,451.17 ± 690.23	2,541.10 ± 508.22	4,846.64 ± 969.33
Potassium, mg/l	1,430.39 ± 286.08	3,196.61 ± 639.32	1,932.08 ± 386.42	2,763.94 ± 552.79	4,891.99 ± 978.40
Zinc, mg/l	10.69 ± 2.14	1.74 ± 0.35	1.61 ± 0.32	0.86 ± 0.17	0.38 ± 0.08
Copper, mg/l	3.28 ± 0.66	1.93 ± 0.39	2.57 ± 0.51	1.07 ± 0.21	0.53 ± 0.11
Chromium, mg/l	1.05 ± 0.21	2.68 ± 0.54	0.47 ± 0.09	2.05 ± 0.41	13.75 ± 2.75
Manganese, mg/l	0.75 ± 0.15	0.13 ± 0.03	0.31 ± 0.06	0.75 ± 0.15	0.36 ± 0.07
Lead, mg/l	0.09 ± 0.02	0.13 ± 0.03	0.04 ± 0.01	0.09 ± 0.02	0.08 ± 0.02
Iron, mg/l	14.87 ± 2.97	9.45 ± 1.89	9.24 ± 1.85	9.62 ± 1.92	4.35 ± 0.87
Nickel	0.46 ± 0.09	0.55 ± 0.11	0.31 ± 0.06	0.39 ± 0.08	0.58 ± 0.12
Cadmium, mg/l	0.003 ± 0.001	0.002 ± 0.000	0.028 ± 0.006	0.075 ± 0.015	0.072 ± 0.014

–not detected.

organic compounds that occurs in young landfills [18]. It can be observed in Fig. 4.

The results for the pH value are all greater than 7.5, which according to the characteristics of leachates (Table 1) all landfills would be old; however it is known that this is not the case. It may be because some leachates that are exposed to the atmosphere could cause some removal of carbon dioxide from the leachate which increases the pH value [19].

In terms of anions, it is worth mentioning the high concentrations of chlorides. Like organic matter, the chlorides concentration decreases as the landfill age increases, due to a washing phenomenon [20] but to get low chloride concentrations can last for years. High chloride content in the leachate samples reflects the presence of significant amount of soluble salts in the municipal solid waste materials. It will affect the COD analysis but it has been treated by dilution of the samples and adding mercury to the sample when the COD has

been analysed. According to the results obtained, the lowest chlorides concentration in old leachates is 2,440 mg/l.

Sulphates concentration also decreases with the landfill age due to the reduction to sulphides in anaerobic conditions. However, in the results obtained for this parameter, several variations in its concentration can be observed. It can be due to seasonal variations.

The cations concentrations such as calcium, magnesium, iron and manganese depend on the stabilization of the landfill. The concentrations of these cations are lower in methanogenic phase due to higher pH and lower organic matter content [21]

Conductivity of the leachate samples is mainly due to the presence of the cations such as sodium or potassium. High conductivity values are observed in young leachates, due to a high presence of these cations, mainly sodium. The average conductivity value obtained in old leachates is 16.6 mS/cm

Table 5
Composition of old leachates from different Spanish landfills

Parameter	Old leachate		
	Landfill 7	Landfill 10	Landfill 13
Ammonia nitrogen, mg N/l	757.3 ± 86.3	486.0 ± 58.7	377.5 ± 47.1
Chloride, mg/l	2,440 ± 366	2,480 ± 372	4,740 ± 711
Conductivity, mS/cm	15.0 ± 0.8	17.1 ± 0.9	17.7 ± 1.0
BOD ₅ , mg O ₂ /l	632 ± 234	164 ± 29	443 ± 163.9
COD, mg O ₂ /l	2,725 ± 350	3,460 ± 448	4,777 ± 623
BOD ₅ /COD	0.23	0.05	0.09
Total phosphorus, mg P _T /l	38.7 ± 5.1	37.8 ± 5.0	50.4 ± 6.5
Nitrate, mg/l	<30	<30	<3
Nitrite, mg/l	<40	<40	<4
Total nitrogen, mg/l	1,541.0 ± 308.2	1,401.0 ± 280.2	981.0 ± 196.2
Phosphate, mg/l	19.6 ± 3.9	55.8 ± 11.1	18.1 ± 3.6
pH	8.05 ± 0.38	8.22 ± 0.39	8.36 ± 0.39
Sulphate, mg/l	99.2 ± 14.9	<60	627.0 ± 94.1
Total suspended solids (TSS), mg/l	96.5 ± 15.5	106.0 ± 16.9	110.5 ± 17.6
Volatile suspended solids (VSS), mg/l	68.5 ± 11.3	64.5 ± 10.6	69.5 ± 11.4
Total dissolved solids (TDS), mg/l	9,004 ± 1,057	11,089 ± 1,283	15,333 ± 1,735
Total solids (TS), mg/l	9,100 ± 1,057	11,195 ± 1,283	15,443 ± 1,735
Calcium, mg/l	120.61 ± 24.12	49.14 ± 9.83	95.33 ± 19.06
Magnesium, mg/l	65.95 ± 13.19	–	39.30 ± 7.86
Sodium, mg/l	1,624.57 ± 324.91	1,900.12 ± 380.02	2,327.95 ± 465.4
Potassium, mg/l	714.54 ± 142.91	1,601.03 ± 320.21	2,262.17 ± 452.43
Zinc, mg/l	1.13 ± 0.23	2.17 ± 0.43	1.77 ± 0.35
Copper, mg/l	5.36 ± 1.07	3.32 ± 0.66	1.36 ± 0.27
Chromium, mg/l	0.44 ± 0.09	1.03 ± 0.21	1.45 ± 0.29
Manganese, mg/l	0.41 ± 0.08	0.27 ± 0.05	2.70 ± 0.54
Lead, mg/l	0.05 ± 0.01	0.11 ± 0.02	0.15 ± 0.03
Iron, mg/l	4.09 ± 0.82	15.80 ± 3.16	34.73 ± 6.95
Nickel, mg/l	0.21 ± 0.04	0.34 ± 0.07	0.51 ± 0.10
Cadmium, mg/l	0.029 ± 0.006	0.079 ± 0.016	0.076 ± 0.015

–not detected

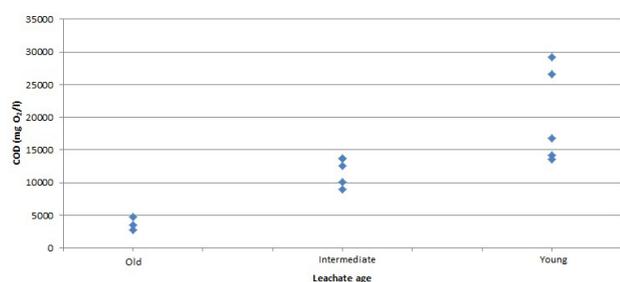


Fig. 3. COD parameter variation with the leachate age.

with low sodium concentrations that vary between 1,624.57 and 2,327.95 mg/l, while in young leachates the average conductivity is 35.2 mS/cm with high sodium concentrations.

With respect to the heavy metal concentrations, when the landfill age increases, the solubility of the metal decreases due to the increase in pH values, so decreasing the metal concentrations [16] but in general, these concentrations do not appear to follow patterns such as COD or BOD [20].

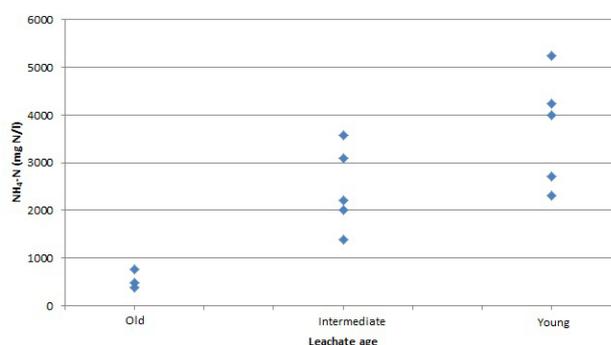


Fig. 4. Ammonia nitrogen parameter variation with the leachate age.

In general, fluctuation of parameters such as phosphorus, chlorides, calcium, magnesium, sulphates and heavy metals depends on seasonal variations rather than on landfill age.

Table 6
Characteristics of young leachate (range of values) from different European landfills

Parameter	Young leachate						
	Ireland landfill	Germany landfill	Greece landfill	Sweden landfill	Italy landfill	Poland landfill	Spain landfill
Ammonia nitrogen, mg N/l	130–4,000	–	10–840	150–560	512–2,229	971–1,250	2,300–5,300
Chloride, mg/l	160–2,620	–	–	(5)–1,300	–	–	6,300–10,300
Conductivity, mS/cm	3.09–28.43	–	23–35.5	0.5–14	–	–	40.5–44.8
BOD ₅ , mg O ₂ /l	36–984	4,000–40,000	9,500–80,800	–	500–1,800	150–273	2,300–12,700
COD, mg O ₂ /l	411–7,160	6,000–60,000	44,000–115,000	1,000–30,000	2,525–3,879	1,873–3,600	13,500–29,219
pH	7.6–8.5	–	4.9–6.7	5–6	7.2–8.7	–	7.63–8.69
Sulphate, mg/l	7.2–1,950	70–1,750	–	–	23–61	–	9.6–2,250

3.3. Comparison of European landfills with Spanish landfills

A comparison of some data obtained in the different landfills of Spain with the characteristics of young leachates from European landfills is presented in Table 6. In general, these data indicate large variations, even though all the leachates are from municipal solid waste landfills but it is necessary to consider another factor that also influences the leachates composition, in addition to the landfill age, such as the nature or types of waste that the landfill contains [20].

Therefore, these large variations between the different landfills will be due, in addition to seasonal variations, among others, to the type of waste that is mostly treated in landfills.

Ammonia nitrogen is contained in plant and animal waste and in industrial waste such as fertilizers, gums, plastic, etc. [22]. On the other hand, sulphate is contained in soluble waste, such as construction wastes or ash, synthetic detergents and inert waste [23].

In general, the range of data obtained in each one of the analysed parameters of the young leachates of Spain, is higher than for the rest of the young leachates from European landfills. High concentrations of ammonia and chlorides show a typical mineral profile from household waste and industrial waste, mentioned earlier. The organic load is very variable because depends on the degradation of the organic products present in the landfill. Most of these organic products are soluble waste derived from decomposition products of the biodegradable fraction of the waste [24].

4. Conclusions

The present study contains results of the main physico-chemical parameters of leachate samples from different Spanish landfills. The presented data indicate that the leachate age has a significant effect on its characteristics and composition but other factors, such as type of waste that is mostly treated in landfills, also have an important effect on its composition.

As the landfill age increases, the concentration of the leachate constituents decreases due to the processes of stabilization of the waste that occurs within the landfill.

The leachate samples from different Spanish landfills show the general tendencies of the effect of the leachate age

on its composition and this effect is shown in several parameters for instance COD or ammonia nitrogen, being the lowest concentration as the leachate age increases.

Comparing the results obtained in Spanish landfills with other European landfills, specifically the young leachates, there are large variations between the same parameters, although in both cases they are municipal solid waste landfills. These variations will be due to seasonal variations, type of waste that is mostly treated, among others.

Wastes generated from European Union countries with regard to composition are basically different. The leachate composition changes because waste is piled for many years in layers that will be in different phases of decomposition.

Acknowledgements

The authors gratefully acknowledge support of this work by the LIFE+ Program under the responsibility of the Directorate General for the Environment of the European Commission through the agreement LIFE15 ENV/ES/000530)-LIFE LEACHLESS project.

References

- [1] Eurostat Database for Waste Statistics, last update: July 2018. Available at: <http://ec.europa.eu/eurostat/data/database>.
- [2] M.S. Raghad, A.M. Abd, H.A. Hegazi, Treatment of leachate from municipal solid waste landfill, *Hous. Build. Nat. Res. Center J.*, 9 (2013) 187–192.
- [3] N. Ragle, J. Kissel, J.E. Ongerth, F.B. Dewalle, Composition and variability of leachate from recent and aged areas within a municipal landfill, *Water Environ. Res.*, 67 (1995) 238–243.
- [4] B. Adhikari, K.R. Dahal, S.N. Khanal, A review of factors affecting the composition of municipal solid waste landfill leachate, *Int. J. Eng. Sci. Innov. Technol.*, 3 (2014) 273–281.
- [5] S. Renou, J.G. Givaudan, S. Poulain, F. Dirassouyan, P. Moulin, Landfill leachate treatment: review and opportunity, *J. Hazard. Mater.*, 150 (2008) 468–493.
- [6] Eurostat Database for Waste Statistics, Waste Generated by Households by Year and Waste Category, last update: August 2018, Available at: <https://ec.europa.eu/eurostat/data/database>.
- [7] APHA/AWWA/WEF, Standard Methods for the Examination of Water and Wastewater, 21st ed., United Book Press, Baltimore, MD, 2005.
- [8] P. Kjeldsen, M.A. Barlaz, A.P. Rooker, A. Baun, A. Ledin, T.H. Christensen, Present and long-term composition of MSW landfill leachate: a review, *Crit. Rev. Environ. Sci. Technol.*, 32 (2002) 297–336.

- [9] R.B. Brennan, M.G. Healy, L. Morrison, S. Hynes, D. Norton, E. Clifford, Management of landfill leachate: the legacy of European Union Directives, *Waste Manage.*, 55 (2016) 355–363.
- [10] B. Ślomożyńska, T. Ślomożyński, Physico-Chemical and toxicological characteristics of leachates from MSW landfills, *Pol. J. Environ. Stud.*, 13 (2004) 627–637.
- [11] A. Tatsi, A. Zoubolis, K. Matis, P. Samaras, Coagulation-flocculation pre-treatment of sanitary landfill leachates, *Chemosphere*, 53 (2003) 737–744.
- [12] S. Morling, Landfill Leachate, Generation, Composition, and Some Findings From Leachate Treatment at Swedish Plants. KTH, School of Architecture and Built Environment (ABE), Land and Water Resources Engineering, Vann, vol. 2, pp. 172–184, 2007.
- [13] A. Del Borghi, L. Binaghi, A. Converti, M. Del Borghi, Combined treatment of leachate from sanitary landfill and municipal wastewater by activated sludge, *Chem. Biochem. Eng. Q.*, 17 (2003) 277–283.
- [14] J. Kalka, Landfill leachate toxicity removal in combined treatment with municipal wastewater, *Sci. World J.*, 2012 (2012) 202897.
- [15] A. Schiopu, B.M. Sluser, I. Apostol, M. Gavriescu, Impact of landfill leachate on soil quality in Iasi County, *Environ. Eng. Manage. J.*, 8 (2009) 1155–1164.
- [16] A. Tatsi, A. Zouboulis, A field investigation of the quantity and quality of leachate from a municipal solid waste landfill in a Mediterranean climate (Thessaloniki, Greece), *Adv. Environ. Res.*, 6 (2002) 207–219.
- [17] J. Bohdziewicz, Treatment of landfill leachate by means of pressure driven membrane operations. *Architecture civil engineering environment, Silesian Univ. Technol.*, 4 (2008) 123–132
- [18] D. Kulikowska, E. Klimiuk, The effect of landfill age on municipal leachate composition, *Bioresour. Technol.*, 99 (2008) 5981–5985.
- [19] J.B. Christensen, D.L. Jensen, C. Gron, Z. Filip, T.H. Christensen, Characterization of the dissolved organic carbon in landfill leachate-polluted groundwater, *Water Res.*, 32 (1998) 125–135.
- [20] T.H. Christensen, R. Cossu, R. Stegmann, *Landfilling of waste: leachate*. Elsevier Applied Science, London, 1994.
- [21] A. Bikash, N.K. Sanjay, Qualitative study of landfill leachate from different ages of landfill sites of various countries including Nepal, *J. Environ. Sci. Toxicol. Food Technol.*, 9 (2015) 23–36.
- [22] A. Pivato, L. Gaspari, Acute toxicity test of leachates from traditional landfills using luminescent bacteria, *Chemosphere*, 64 (2006) 1777–1784.
- [23] T.H. Christensen, P. Kjeldsen, P.L. Bjerg, D.L. Jensen, J.B. Christensen, A. Baun, H.J. Albrechtse, G. Heron, Biogeochemistry of landfill leachate plumes, *Appl. Geochem.*, 16 (2001) 659–718.
- [24] D. Reinhart, A review of recent studies on the sources of hazardous compounds emitted from solid wastes landfills: a U.S. experience, *Waste Manage. Res.*, 11 (1993) 257–268.