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EFFECT OF THE RECIRCULATION OF A REVERSE OSMOSIS CONCENTRATE ON LEACHATE GENERATION: A CASE STUDY IN AN ITALIAN LANDFILL

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ABSTRACT: "Fossetto" landfill has been operating in the municipality of Monsummano Terme (Pistoia Province, Italy) since 1988; the authorized volume for landfilling is about 1,000,000 m^3; at the moment the plant is being mainly used to dispose of mechanically and biologically treated residual municipal solid waste. Since September 2006, an in-situ reverse osmosis leachate treatment plant has been operating to treat leachate. The treated water is being discharged into a small nearby stream while the concentrated leachate is being recirculated back into the landfill body following Italian Regulations and an authorization from the local authority (Pistoia Province). This paper presents monitoring results on leachate generation rates and composition for the past fifteen years. A moderate increase of the concentration of some of the monitored parameters occurred (e.g. ammonium, chlorides) and a decrease for most heavy metals. The increase of concentrations for Cl^- and NH_4^+ was more evident in the leachate coming from the wells closer to reinjection area. However, the change in leachate composition did not affect the quality of the effluent from the leachate treatment plant. The annual volume of the generated leachate increased significantly right after the recirculation started.

Keywords: concentrate, landfill, leachate, reverse osmosis, recirculation

1. INTRODUCTION

Landfilling is still the most widely used waste management system in the world. Landfilling is still relatively cheap, simple and is not linked to uses of complicated and patented technologies. Unfortunately, the environmental impacts associated to landfills are not negligible. Even after fifty years of research focusing on the complex physical, chemical and biological processes occurring within landfills to design technologies to minimize environmental impacts, much work is still needed.

Modern landfills are equipped with multi-barrier systems (Cossu, 1995) designed to minimize the environmental impact (leachate impact among others) both in the active and in the post-closure periods. One of the barrier systems is the leachate drainage and collection systems that allow the treatment of this potentially hazardous liquid discharge.

One of the most widely used leachate treatment technology in many countries and in Italy in particular Among various options is the co-treatment of landfill leachate with municipal sewage after its transportation by trucks in off-site authorized plants. Other techniques exist, such as the co-treatment with municipal sewage and the treatment in dedicated plants using advanced oxidation or adsorption processes, on or off-site (Renou et al., 2008; Wiszniowski et al., 2006). Another option increasingly considered is the on-site treatment using reverse osmosis facilities. A comparison between co-treatment with sewage and reverse osmosis is outlined in Table 1.

It is therefore clear that the economic sustainability of the adoption of a leachate treatment based on reverse osmosis is directly connected to the management of the resulting concentrated leachate. The most economically convenient option is the recirculation of concentrated leachate into the same landfill (Calabrò et al., 2010; Liu et al., 2008; Qu et al., 2008; Renou et al., 2008; Sluiter et al., 2012; Wiszniowski et al., 2006). The specific studies present in scientific literature on this practice are not numerous and opinions are often conflicting. Some researchers support that the impact of the recirculation of concentrated leachate is negligible or at least limited in time (Heinigin, 1995; Peters, 1998); others declare that its application is not sustainable in the long term (Heyer and Stegmann, 2002).

Table 1. Advantages and disadvantages of landfill leachate treatment options most commonly adopted in Italy.

Co-treatment with sewage off-site		**Reverse osmosis treatment on-site**	
Advantages	***Disadvantages***	***Advantages***	***Disadvantages***
Simplicity	Cost (about 50 - 100 €/m^3 in Italy including transportation)	Highly efficient pollutants removal from purified water	Concentrated leachate generation (about 30% of incoming leachate)
	Excess sludge often non-usable for agriculture due to the presence of heavy metals and other pollutants		Non-competitive if concentrated leachate must be treated in an external plant
	Some of the pollutants are simply diluted	Cost (about 15 - 40 €/m^3 in Italy)	
	(Off-site treatment)	(On-site treatment)	

This paper aims to advance the knowledge published previously on the same topic (Calabrò et al., 2011, 2010; Calabrò and Mancini, 2012). Specifically, we analysed the long-term effect of concentrated leachate recirculation in an Italian landfill (Fossetto) where the reverse osmosis technology to treat leachate is applied since September 2006. In particular, we aimed to analyse the effect of recirculation on the qualitative and quantitative leachate characteristics.

2. MATERIALS AND METHODS

The landfill under study (including all its ancillary plants such as mechanical-biological treatment (MBT), leachate treatment and biogas extraction and utilisation plants) is considered a complex, partially controlled, reactor where physical, chemical and biological processes occur. The study uses leachate data from a database of 15 years (2002-2016). Between years 2002 to 2006, no leachate recirculation was practised since the reverse osmosis system had not been installed yet. Therefore, data from years 2002-2006 provide a baseline of leachate quantity and composition when no concentrate recirculation existed. Concentrate recirculation was applied beyond year 2006, and its impact on leachate characteristics is investigated here.

2.1. The landfill “Il Fossetto” in Tuscany (Italy)

“Il Fossetto” landfill has been operating since 1988. It is located in the province of Pistoia (Northern Tuscany, Italy) in a flat area and has a total authorized volume of about 1,000,000 m^3; it is used to dispose of municipal waste after mechanical and biological treatment (MBT) and small amounts of street-cleaning residues and some bulky waste. Until 2011, also non-hazardous bottom ash and slag coming from a municipal incinerator were landfilled there, while until June 2003 (when the on-site MBT plant entered in operation) mixed municipal waste were directly landfilled. In addition to the MBT plant, a biogas recovery and energy production and a leachate treatment plant are operating in the landfill. For more information on “Il Fossetto” landfill see available literature (Calabrò et al., 2010).

In “Il Fossetto” landfill, leachate collected by the drainage system is extracted by 13 wells; until 2006, all the leachate produced by the landfill was sent to external plants for treatment. Since September 2006, most of the extracted leachate is treated on site in a reverse osmosis plant. This plant includes mixing and pre-aeration, sieving, pre-filtration by cartridge filters, membrane ultrafiltration, chemical conditioning to reach a pH of about 5 by adding sulphuric acid, membrane reverse osmosis (two modules), chlorination, activated carbon filtration. Purified water obtained by leachate treatment is discharged into a small nearby channel while the generated concentrated leachate is recirculated back into the landfill by a vertical reinjection well located in the 2nd cell of Landfill 4. The generated concentrated leachate that is reinjected into the landfill represents about 30% of the total incoming leachate.

2.2 Monitoring activities

According to the requirements of the Control Authority (Pistoia Province), an extensive monitoring program is being regularly carried out in “Il Fossetto” landfill.

Data available are related to the meteorological parameters (e.g. temperature and rainfall), to the amount of waste landfilled (detailed for each single type), to leachate produced and biogas extracted.

Once a year, the leachate from each recovery well is sampled and analysed according to Standard Methods (Eaton and Franson, 2005) to measure the pH and to determine the concentration of COD, ammonia nitrogen, chloride and of several metals and metalloids (As, total Cr, Cu, Hg, Ni, Zn). Moreover, since 2005, samples are being collected four times per year from the homogenisation tank to measure pH, conductivity, suspended solids, COD, BOD_5, ammonia nitrogen, chloride, sulphides, total Cr, Ni, Zn, As, Hg, Cu. Similar analyses are being carried out on the concentrated leachate too.

2.3 Statistical analysis

Statistical analysis was performed to check differences between the mean annual leachate generation (i.e. amounts) prior to and after leachate recirculation (i.e. before and beyond 2006). The normality criterion for the data was checked using the Shapiro-Wilk test. A parametric independent t-test was then employed to check the statistical differences between leachate quantities and leachate quality for the two aforementioned periods. Only the statistically significant regression equations are presented. Statistics were done with Minitab® v17.

3. RESULTS AND DISCUSSION

3.1. Leachate generation

Due to an increasingly efficient separate collection in the area served by “Il Fossetto” landfill, incoming MSW decreased from about 50,000 t/y in 2000 to about 28,000 t/y in 2015 and 2016. From year 2000 to early 2011, an average of about 7200 t/year of non-hazardous bottom ash and slag were also landfilled there.

Figure 1 depicts the cumulative leachate recovery as a function of the cumulative waste amounts entering the landfill. It is clear that a sharp increase in the leachate recovery exists beyond 2006, as witnessed by the increase of the slope of the line fitting the data right after year 2006, that signifies the initiation of the concentrated leachate recirculation project. It is noted that leachate recovery (i.e. amount withdrawn via pumping) is similar to leachate generation as long as the leachate level remains constant at the landfill bottom. This is true for the leachate recovery wells, since according to the permits leachate level must be kept almost constant. In that sense, the terms “leachate recovery” and “leachate generation” are used interchangeably here.

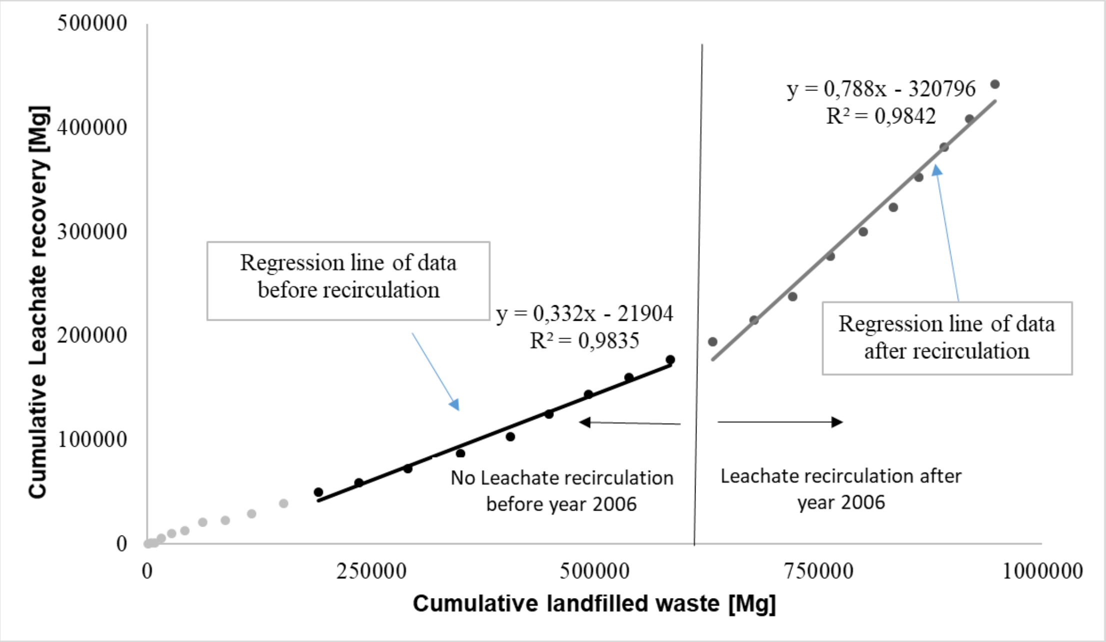
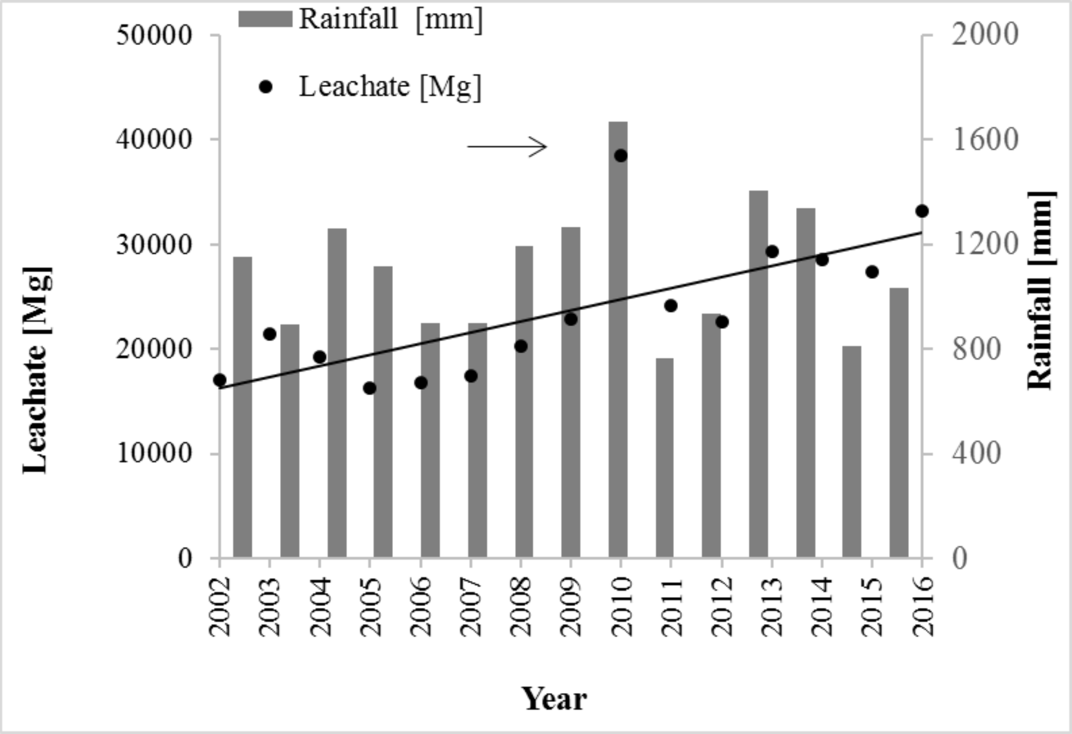
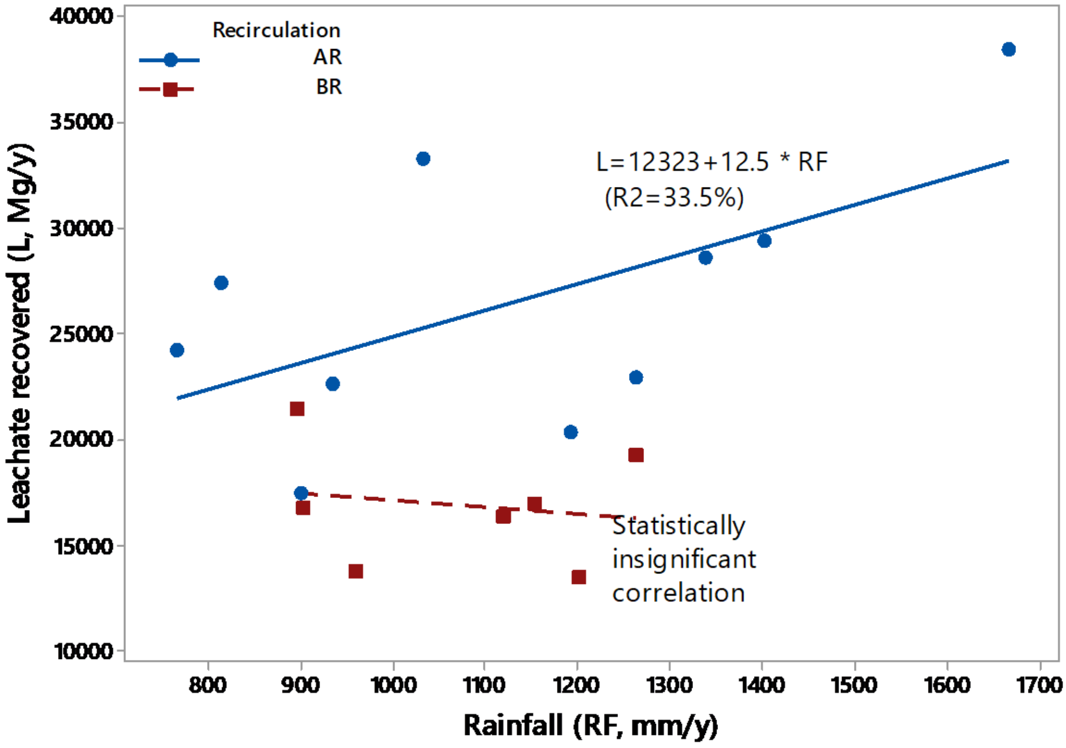


Figure 1. Cumulative leachate recovery versus cumulative landfilled waste.

Leachate recovery increased from about 45 t/d in years 2005/2006 (before the start of recirculation) up to about 90 t/d in 2016, while the maximum value was recorded in 2010 (105 t/d). The increase of the recovery in the period 2007-2016 (after the beginning of recirculation) with respect to the period 2002-2006 (the configuration of the landfill and the procedure of leachate recovery are comparable) is 45%, while in the same period rainfall increased only 6% (see Figure 2a).



(a)



(b)

Figure 2. Annual rainfall and leachate recovery in the period 2002-2016 (a: mean annual leachate generation and mean annual precipitation for 15 years, b: a statistically significant linear correlation was calculated only during the years after 2006 in which leachate recirculation was initiated – BR: Before recirculation; AR: After Recirculation).

Interestingly, a correlation between rainfall and the leachate recovered was feasible in the years following the recirculation (AR), as shown in Figure 2b. On the other hand, in the years before recirculation (prior to 2006), the correlation was statistically insignificant. This fact could be attributed to operational changes that occurred in the period 2001-2005, namely the: (i) reduction of the maximum level of leachate on the landfill bottom as requested by the control authority, and, therefore, the consequent increase of leachate recovery; (ii) initiation of pretreatment of MSW via MBT (Calabrò and Mancini, 2012)

Table 2 reveals that although the rainfall was statistically similar in both periods (prior to and after recirculation), the annual leachate generation was statistically different. In particular, leachate recovery was statistically higher (at $p<0.01$) after the initiation of recirculation

(September 2006) compared to before recirculation.

Table 2. Descriptive statistics of rainfall and leachate quantities before and after concentrate recirculation

	Period before recirculation (years 2000-2006, n=7)*	Period after recirculation (years 2007-2016, n=10)
Rainfall (mm/year)	1070 ± 150^A	1131 ± 291^A
Leachate recovery (Mg/year)	16867 ± 2826 ^{A}	26441 ± 6273 ^{B}

Means ± standard deviation; different letters indicate statistically different means per row at $p < 0.05$; *: Recirculation started on September 2006. However, for calculation purposes, year 2006 was considered a year without recirculation.

This is an indication that it was not the rainfall, but rather the recirculation process that led to the higher amounts of generated leachate beyond year 2006. The increase in leachate generation after recirculation was about 40% (the difference between the total leachate generation increase and the corresponding rainfall increase) over a 10-year period and can be attributed to the fact that around 30% of the generated leachate was reinjected back into the landfill in a concentrated form.

The volume of leachate concentrate recirculated each year in the landfill is about 30% of the raw leachate generated and treated; therefore the fact that the total leachate generation increase has been gradual and never exceeded 40% indicates that the landfill has the capability of retaining the recirculated leachate by increasing the average water content of the landfilled waste. The stabilized organic fraction of MSW (SOFMSW) produced in the MBT plant and used as daily cover can play a role in the above phenomenon thanks to its high water retention capacity. This hypothesis had been reported in Calabrò and Mancini (2012).

Recent measurements that were carried out according to the IPLA F4 1998 method revealed an average retention capacity equal to about 55-60% wb (wet weight basis).

3.2 Leachate composition

Table 3 and Figure 3 summarize raw leachate and recirculated leachate characteristics in the period 2000-2016 for the wells closer to reinjection area, for the homogenization tank (data refer to years beyond 2005 when the tank was constructed) and for the reinjected leachate (years beyond 2006). The leachate wells considered are those draining of cell 4 (well 44.1), cell 2 (well 42.1), cell 3 (well 43.1 until 2012 and beyond that, due to malfunction, of the adjacent newly built well 43.2).

In the homogenization tank, after an increase in the first years of recirculation, COD concentration had a tendency to reduce. A similar behaviour was observed for some heavy metals (i.e. Cu and Ni) while for others (i.e. Pb and Cr) a reduction trend was observed right after recirculation started. Zn and As concentrations showed a steep increase in the first years of recirculation followed by a decrease after 2010. In general, considering the already mentioned increase in the overall leachate generation, only the lead (Pb) and total chromium (Cr) concentrations decreased.

The concentration of some metals (i.e. Pb, Ni, As) in the homogenization tank during the period 2011-2016 was already below discharge limits. Given the existing trend in the concentration of hazardous metals, it is possible that in the next years other metals (i.e. Zn, Cu and total Cr) will also decrease below discharge limits.

As shown in Table 3, the characteristics of concentrated leachate are stable and the effluent quality was always below standard values (data not shown) that demonstrates the efficiency of the reverse osmosis plant. The plant is apparently not influenced by the changes in raw leachate quality.

The data of raw leachate composition from the wells confirm a strong decrease of Pb and Cu concentrations (rarely found in recent samples) and an increasing trend in COD, ammonium and chlorides. This increase is extremely high for well 44.1, in which, also, total chromium (Cr), nickel (Ni) and arsenic (As) had more than double concentrations in the period 2010-2016 compared to before recirculation.

Ammonium concentration increased over time since it is the product of decomposition of organic N present in the incoming waste and in the concentrated recirculated leachate. The ammonium remains in the ionic form at the pH of the Fossetto landfill (i.e. 7.8), which prevents N loss as NH_3 (which would occur in pHs above around 9.2). Also, NH_4^+ accumulation is explained by the presence of anaerobic conditions that prevent the oxidation of ammonium to nitrites and nitrates. The chloride (Cl^-) concentration increase is even sharper than that of ammonium. This is attributed to the chloride's high solubility and its typical conservative nature, regardless of pH. Since recirculation prevents the escape of pollutants outside the landfill, eventually both Cl^- and NH_4^+ will accumulate within the landfill, as was actually observed from the data.

However, only for the samples coming from wells 43.2 and 44.1, the ammonium and chloride concentrations were higher than those in the samples from the homogenisation tank. These wells were probably the ones most influenced by leachate recirculation.

The evaluation of the influence of concentrated leachate recirculation on the quality of leachate in the homogenization tank is not easy. Most probably, due to the increasingly high efficiency of the source separation of wastes in the wider area, that led to a consequent 50% reduction of landfilled waste, the leaching of pollutants has been constantly decreasing since the initiation of those source separation efforts. However, considering that recirculated leachate has a pollutant load of the same order of magnitude of that of raw leachate and that the average retention time of leachate in the landfill is less than a year, a dramatic increase in pollutants concentrations would have been expected. In fact, assuming no retention capacity in the landfill, the addition of the pollutants' mass normally leached in one year from the landfill with the one contributed by the recycled leachate would lead to a 100% increase of the pollutants' loading (compared to when no recirculation occurred). Average leachate composition in the period 2010-2016 (after 4-10 years from the start of recirculation), however, does not show such a doubling trend. Only the mass of chloride in the leachate increased in one year by a factor of around 2.4 (this value has been assessed combining the increase of about 60% in concentration with the increase of about 50% in leachate generation). For ammonium, that increase was slightly lower (2.1 times). The increase of other heavy metals (i.e. Ni and Zn and As) is less relevant since a clear reducing tendency had been observed.

These changes in the concentrations of various compounds (ions, metals) after recirculation can be explained by several concurring phenomena that can be both operating as well as chemistry based. Operating parameters can be the reduction of heavy metals in the incoming MSW stream as a result of the efficient source separation in the nearby area as well as the effective operation of the preceding MBT plant. However, those observations also support the hypothesis that the landfill has an inherent attenuation capacity with respect to some of the pollutants present in the recirculated leachate (Calabrò et al., 2010). This attenuation capacity is most probably linked to chemistry-based processes and other factors, such as:

i) the dilution of recirculated leachate with the less polluted leachate generated by the raw MSW,

ii) adsorption/complexation phenomena favoured by the presence in the landfill of SOFMSW (Calabrò and Mancini, 2012; Xie et al., 2015). and,

iii) the so called "sulphide barrier" effect that affects the concentration of heavy metals. In fact, due to the use of sulphuric acid during leachate treatment, noticeable quantities of sulphates (about 15000 mg/l on average) are present in the reinjected leachate. This occurs since sulphuric acid fully dissociates to sulfates at pHs higher than around 2. This explains the presence of sulfates in the landfill leachate that has a pH of 7.8. Those sulphates are expected to be further biologically transformed into sulphides in the landfill anaerobic environment. The formation of the scarcely soluble metal sulphides can, then, effectively reduce the presence of metals in the landfill leachate, particularly during the methanogenic phase that has relatively high pH values (Calabrò and Mancini, 2012; Möller et al., 2004; Qu et al., 2008). For example, zinc (Zn) and copper (Cu) concentrations in the wells are lower than in the homogenization (equalization) tank. It would have been expected that those metals would have been higher due to the additional amounts provided by the recirculated leachate. These lower concentrations can be attributed to the presence of sulfides that aided in the precipitation of those metals. On the other hand, Cr (that cannot precipitate at this pH), and partially Ni, tended to increase after recirculation, since they do not precipitate as sulfides.

Some other mechanisms that may explain the attenuation of metals are related to the increase of the superficial reactivity of the wastes after MBT (i.e. since the waste can be able to form superficial complexes with the metals) and to the presence of organic substances (i.e. humic and fulvic acids). The latter organic formations can create complexes with metals.

The release, on the other hand, of some other metals could be also related to the MSW inicineration bottom ash and slag landfilled in Fossetto up to 2011. The ageing and weathering phenomena occurring at the landfill site may lead to a significant metal release over time from both bottom ash and slag (Sabbas et al., 2003).

In general, the increase of water content in the landfill, also favoured by the presence of the SOFMSW, can increase the capacity of the landfill to retain soluble contaminants such as ammonium and chloride.

However, it must be pointed out that the increase of the ammonium and chloride concentrations can lead to a slow-down of the overall landfill stabilisation period and, therefore, to an extension of the period during which leachate is an environmental threat.

Table 3. Characteristics of the leachate recovered and of the recycled concentrated leachate

	pH	**COD [mg/L]**	**NH_4^+ [mg/ L]**	**Cl^- [mg/ L]**	**Pb [mg/ L]**	**Cr_{tot} [mg/ L]**	**Cu [mg/ L]**	**Ni [mg/ L]**	**Zn [mg/ L]**	**As [mg/ L]**
Treated leachate discharge limits	***5.5 – 9.5***	***160***	***15***	***1200***	***0.2***	***2***	***0.1***	***2.0***	***0.5***	***0.5***
					Homogenization Tank					
Average Before Recirculation (2005-2006)	7.69	3366	1832	2179	0.61	6.55	0.25	0.62	0.87	0.06
Average in the first 5 years of Recirculation (2006-2011)	8.15	4329	1652	2432	0.38	6.48	0.37	1.20	2.05	0.39
Difference with respect to the period before Recirculation	6.0%	28.6%	-9.8%	11.6%	-37.6%	-1.1%	50.7%	91.7%	134.9%	591.1%
Average in the period 2011-2016	7.70	3427	2514	3524	N.A.	2.46	0.18	0.58	0.86	0.07
Difference with respect to the period before Recirculation	0.2%	1.8%	37.2%	61.7%	N.A.	-62.5%	-28.9%	-7.6%	-1.4%	21.1%
					Well 42.1					
Average Before Recirculation (2000-2006)	7.74	2219	1956	2385	0.32	1.21	0.10	0.42	0.46	0.03
Average in the first 5 years of Recirculation (2006-2011)	7.68	2095	2182	2599	0.31	0.98	0.17	0.77	0.58	0.07
Difference with respect to the period before Recirculation	-0.8%	-5.6%	11.5%	9.0%	-5.1%	-19.2%	68.7%	86.2%	25.3%	98.3%
Average in the period 2011-2016	8.28	3032	2760	3499	N.A.	2.34	N.A.	0.49	0.31	0.14
Difference with respect to the period before Recirculation	7.0%	36.6%	41.1%	46.7%	N.A.	92.9%	N.A.	17.9%	-32.0%	336.2%

	Well 43.1-2*									
Average Before Recirculation (2000-2006)	7.78	2566	2392	2670	0.27	1.42	0.09	0.39	0.50	0.03
Average in the first 5 years of Recirculation (2006-2011)	7.70	3071	3087	4058	0.34	1.71	0.11	0.65	0.65	0.17
Difference with respect to the period before Recirculation	-1.0%	19.7%	29.1%	52.0%	25.5%	19.8%	22.3%	64.4%	29.2%	397.7%
Average in the period 2011-2016	7.78	3665	3585	4603	N.A.	2.48	N.A.	0.60	0.37	0.05
Difference with respect to the period before Recirculation	0,0%	42,8%	49,9%	72,4%	N.A.	74,3%	N.A.	53,2%	-25,4%	52,3%
	Well 44.1									
Average Before Recirculation (2001-2006)	7,61	1753	1770	1987	0,23	0,87	0,11	0,30	0,68	0,02
Average in the first 5 years of Recirculation (2006-2011)	7,62	3202	3116	3961	0,40	1,96	0,33	1,06	0,71	0,11
Difference respect to the period before Recirculation	0,2%	82,7%	76,0%	99,4%	71,0%	124,9%	206,9%	254,6%	5,5%	360,7%
Average in the period 2011-2016	7,93	3576	3654	4513	N.A.	2.43	N.A.	0.69	0.24	0.08
Difference respect to the period before Recirculation	4.2%	104.0%	106.4%	127.2%	N.A.	179.2%	N.A.	131.3%	-64.0%	203.1%
	Concentrated leachate									
Average in the period 2006-2011	6.17	4754	4059	5975	0.28	2.88	1.40	1.59	2.17	0.12
Average in the period 2011-2016	6.50	4512	4387	5955	0.10	2.88	1.49	0.88	1.95	0.09
Average in the period 2006-2016	6.34	4633	4227	5965	0.19	2.88	1.45	1.23	2.06	0.10

During 2012 Well 43.1 was substituted by the adjacent new well 43.2; N.A.: generrally below method detection limit

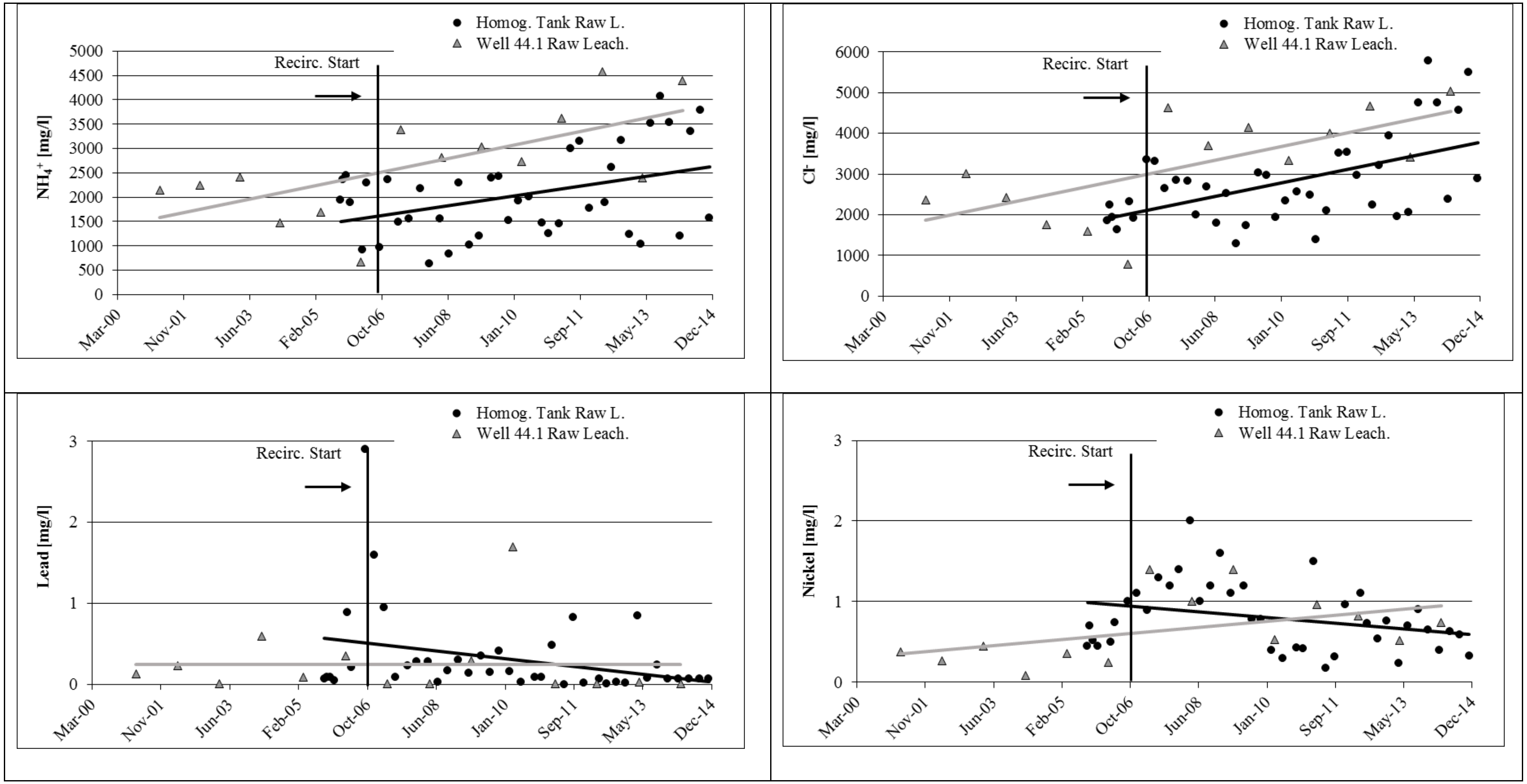


Figure 4. Raw leachate composition trend in the homogenization tank and in well 44.1.

3.2.1 Quality of raw leachate in the equalization tank

Table 4 shows that, in general, the quality of the leachate in the equalization tank was not statistically affected by the recirculation of the concentrate that was initiated at the end of 2006. However, it is noted that the number of data for the pre-recirculation period were relatively limited (n=8) compared to the post-recirculation period (n=40). Thus, although a one-way ANOVA can be theoretically applied on all data, it would have been desirable to have both databases at approximately similar size. In addition, the fact that the concentrations of all parameters was found to be statistically similar in the pre and post recirculation period may be attributed to the homogenization that the equalization tank achieved. The only exception was Pb, in which statistically higher values were measured during the pre-recirculation period compared to the post-recirculation one. Chromium, although statistically similar in both cases, had higher values in the pre-recirculation period compared to the post-recirculation period.

Table 4. Comparison of mean values of certain parameters in the leachate collected in the equalization tank before and after the recirculation was initiated

Parameter	Before Recirculation (n=8)	After Recirculation (n=40)
pH	7.80 ± 0.68 ^{A}	7.82 ± 0.34 ^{A}
COD	3490 ± 1540 ^{A}	3700 ± 1000 ^{A}
NH_4^+	1900 ± 621 ^{A}	2240 ± 1040 ^{A}
Pb	0.74 ± 1.0 ^{A}	0.18 ± 0.23 ^{B}
Cr_{tot}	6.3 ± 2.4 ^{A}	3.7 ± 4.9 ^{A}
Cu	0.26 ± 0.15 ^{A}	0.24 ± 0.29 ^{A}
Ni	0.68 ± 0.25 ^{A}	0.77 ± 0.42 ^{A}
Zn	0.99 ± 1.15 ^{A}	1.23 ± 1.35 ^{A}
As	0.043 ± 0.05 ^{A}	0.17 ± 0.23 ^{A}
Hg	0.0162 ± 0.042 ^{A}	0.033 ± 0.073 ^{A}
Cl	2320 ± 658 ^{A}	3170 ± 1328 ^{A}

Means ± SD; Means on the same row that share different letters are significantly different based on pairwise comparisons with the Tukey test at $\alpha=5\%$.

3.2.2 Correlation among parameters for the raw and concentrated recycled leachate

Table 5 presents all linear Pearson correlation coefficients among 11 parameters of the raw leachate in the equalization basin. The correlations were based on 48 measurements that were performed between 29/9/2005 and 2/12/2016. Figure 5 graphically presents those correlations. The first 8 measurements correspond to the period before the initiation of recirculation, whilst the following 40 measurements correspond to the period in which the leachate concentrate was recycled back to the landfill. Table 5 reveals that certain correlations exist among some parameters. For example, Hg and Pb observed a high correlation indicating that likely the same source is responsible for the presence of those two metals in the leachate. Similarly, there was a strong correlation between Zn and Cu. Table 5 also reveals a strong positive correlation between NH_4^+ and Cl^-, Other strong correlations were found between COD and certain hazardous metals (e.g. Zn, Ni, Pb, Cu, Cr). This is likely explained by the fact that those metals are sorbed onto the solid organics that were measured in the total COD.

Table 6 and Figure 6 show all pair-wise correlations among 15 parameters measured in the recycled leachate concentrate beyond year 2006. Figure 6 reveals a strong correlation between Cl^-, NH_4^+, $SO_4^=$ concentrations and conductivity. This is expected since those 3 ionic compounds are the dominant components of conductivity in leachate. A relatively weak correlation between the BOD_5 and COD was also calculated, which is also expected. Still, as in the case of raw leachate, the strongest positive correlation was between NH_4^+ and Cl^-.

Table 5. Statistically significant Pearson correlation coefficients among parameters of the raw leachate present in the equalization basin

	Cl^-	NH_4^+	COD	Cr_{tot}	Hg	Pb	Zn	Cu	Ni	As
pH						0.392**			0.376**	
Cl^-	-	0.879**								
NH_4^+		-			-0.323*		-0.406**			
COD			-	0.432**		0.423**	0.676**	0.402**	0.662**	
Cr_{tot}				-	0.345*		0.455**		0.623**	
Hg					-				0.309*	0.433**
Pb						-	0.335*			
Zn							-	0.400**	0.689**	0.291*
Cu								-		
Ni									-	0.501**
As										-

*: significant at $\alpha < 0.05$; **: significant at $\alpha<0.005$; non-shaded blank cells indicate that no statistically significant correlation was calculated

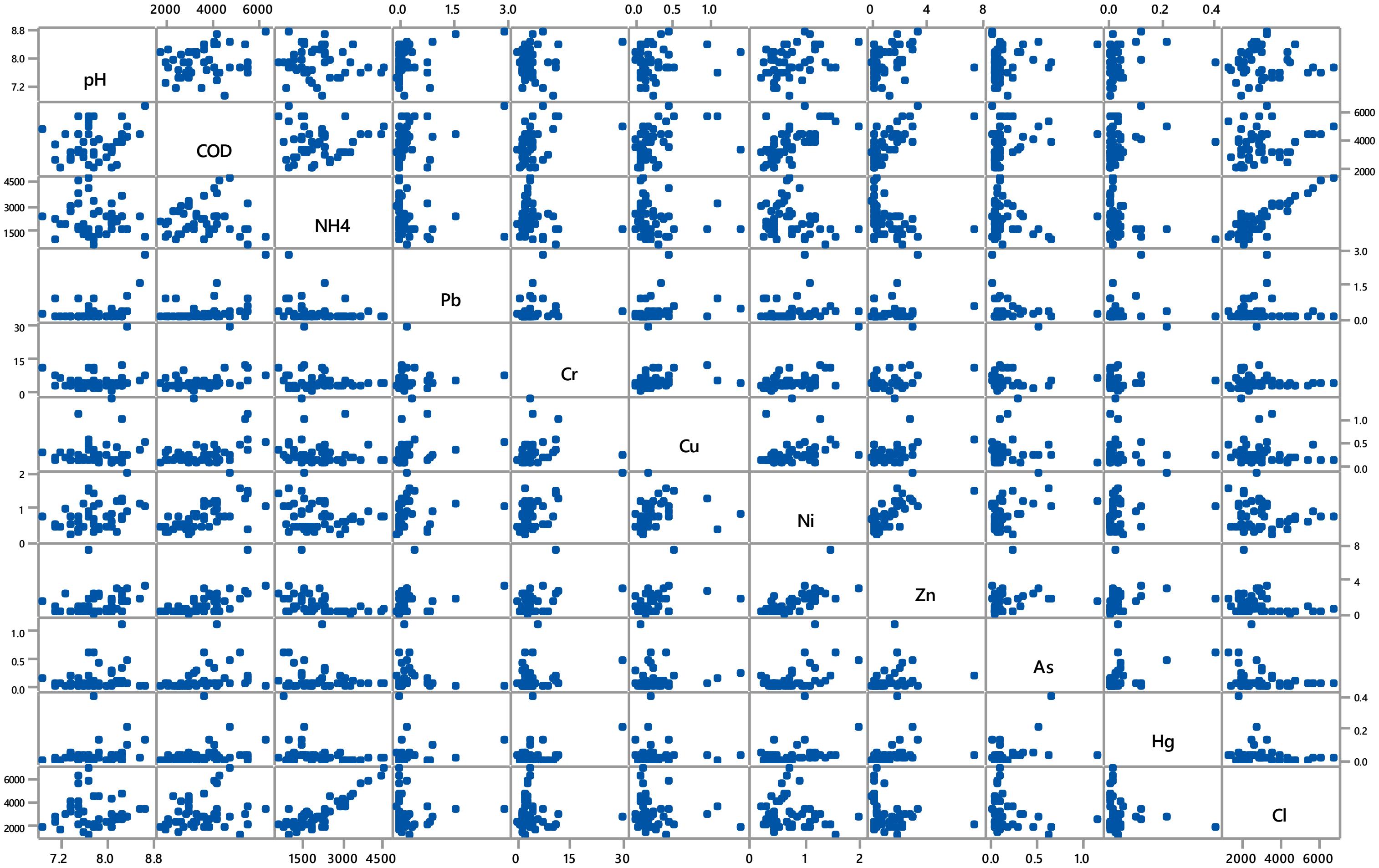


Figure 5. Scatter-plot matrix among concentrations of raw leachate present in the equalization basin based on 48 samples collected during 2006 to 2016 (pH in pH units and all other units in mg/L)

Table 6. Statistically significant Pearson correlation coefficients among parameters of the recycled leachate concentrate

	TSS	$SO_4^=$	Cl^-	NH_4^+	Conductivity	BOD_5	COD	Cr_{tot}	Hg	Pb	Zn	Cu	Ni
pH	-0.419**									0.315*		-0.464**	
TSS	-		-0.344*			0.320*						0.336*	
$SO_4^=$		-	0.673**	0.686**	0.701**								
Cl^-			-	0.600**	0.780**	-0.454**		0.371*					
NH_4^+				-	0.854**								
Conductivity					-		0.368*	0.425**					
BOD_5						-	0.396*						
COD							-				-0.326*	-0.378*	
Cr_{tot}								-					0.359*
Hg									-	0.473**			
Pb										-			
Zn											-	0.728**	
Cu												-	
Ni													-

*: significant at α < 0.05; **: significant at α < 0.005, non-shaded blank cells indicate that no statistically significant correlation was calculated.

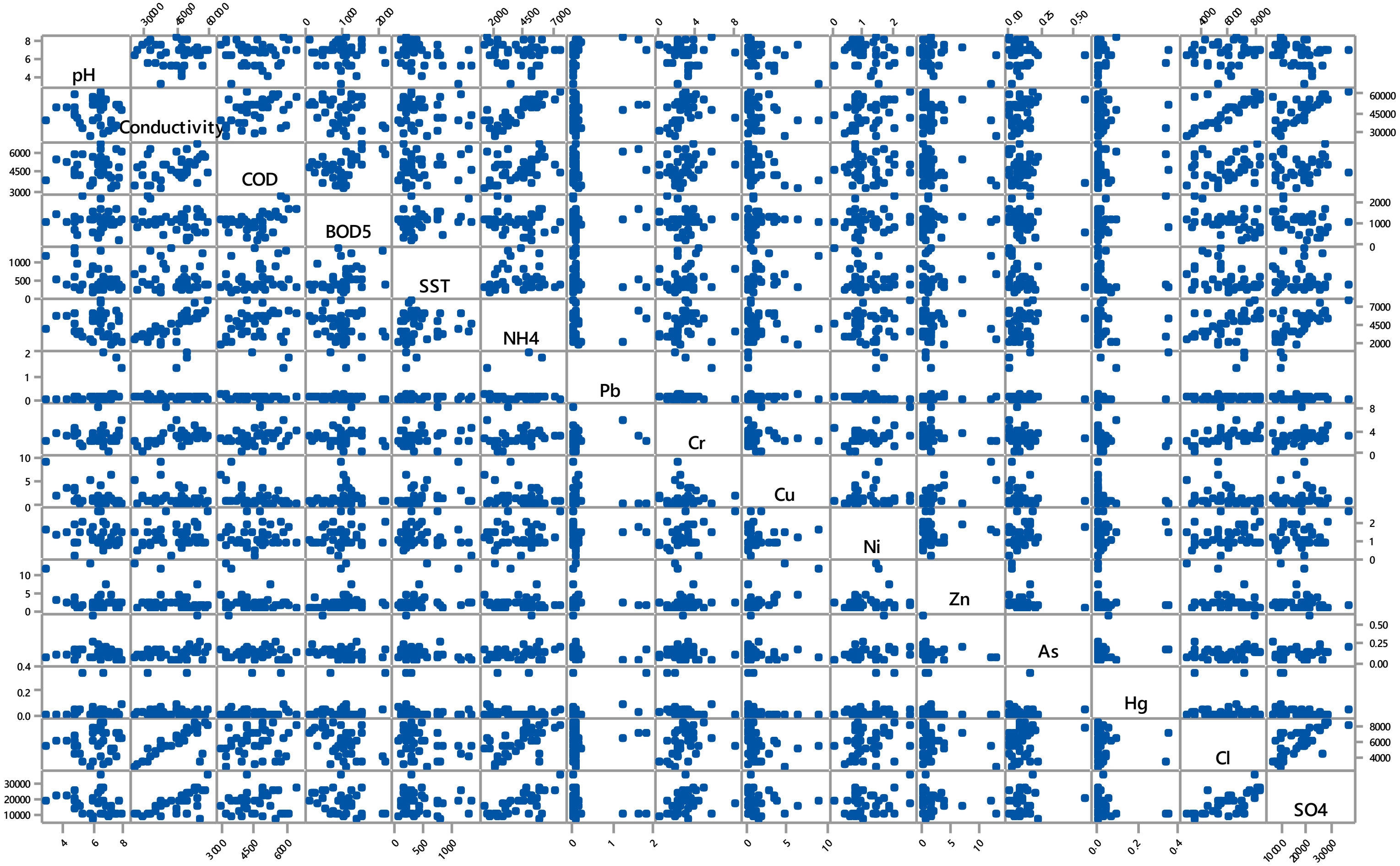


Figure 6. Scatter-plot matrix among parameters of the recycled leachate concentrate based 39 samples collected during 2006 to 2016 (pH in pH units, conductivity in mS/cm and all other units in mg/L)

4. CONCLUSIONS

The conclusions from this work are:

- An increase of the annual leachate volumetric rates occurred over a 10-year period of recirculating concentrated leachate back into the landfill. This increase was found to be irrelevant to the annual rainfall (which was statistically similar in the periods before and after recirculation).
- The concentration of certain parameters (NH_4^+, Cl^- and $SO_4^=$) was found to be higher in the leachate recovered after recirculation compared to the period before recirculation. However, this increase was small and therefore recirculation of concentrated leachate can be still considered a sustainable leachate treatment approach. Nevertheless, the overall leachate management would benefit from an optimized reinjection system (e.g. more reinjection points, sub-horizontal wells).
- Insignificant statistical differences were calculated between the concentrations of the raw leachate collected in the equalisation tank before and after recirculation. Only the mean concentration of Pb during the pre-recirculation period was statistically higher compared to the post-recirculation period.

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