Co-landfilling of pretreated waste: Disposal and management strategies at lab-scale

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Abstract

The present paper deals with the possible advantages that can be obtained by co-landfilling of municipal solid waste organic fraction (MSWOF) and bottom ash (BA) from the incineration of municipal solid waste. In particular, the aim of the research hereby presented is to check the effect exerted by different disposal (mixed or layered) and management strategies (anaerobic or semiaerobic conditions) for landfills in which MSWOF and BA are co-disposed. Three lab-scale reactors were set-up: the reactor A with mixed BA and MSWOF in anaerobic conditions, the reactor B with mixed BA and MSWOF in semiaerobic conditions, the reactor C with layered BA and MSWOF in anaerobic conditions. The results obtained showed that the aeration at the beginning of the experimental period for about 60 days led to a more rapid biodegradation of the organic matter and to an improved leachate quality in terms of both organic load and nitrogen content. Also a significant increase in the settling rate was observed at the end of the aeration phase. Therefore, the aerobic management can be advised as the most available strategy providing a more rapid biological and mechanical stabilization of the bulk waste. Otherwise, the disposal strategy did not exert any significant effect on the leachate characteristics; however, the layered configuration may be adopted in order to accelerate the main settlements.
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Keywords: Pretreated waste; Co-disposal; Landfill; Aerated landfill

1. Introduction

In Italy, as well as in many other countries, landfilling still represents the final fate for most of the municipal solid waste (MSW) produced. However, the law in force requires landfilling to be limited to only inert, or at least pretreated waste, in order to reduce the related environmental impacts and to allow a faster site recovery.

At present, technical indications on the characteristics of waste to be disposed in landfills have not been formulated yet by the current legislation; particularly, the setting of a standardized duration of each pretreatment process prior to the final disposal of waste is still lacking.

In order to reduce the environmental as well as the social impact caused by a landfill, some management strategies can be adopted:

- waste mechanical-biological and/or thermal pretreatment, leading to a significant reduction in the COD, BOD_5, and ammonium nitrogen release via the leachate [1–6] as well as to an acceleration of the landfill gas production [2,3,5,7,8];
- in situ aeration, providing a rapid and significant oxidation of the organic fraction as well as ammonium nitrogen consumption through nitrification [4,9–13];
- in situ water supply (*flushing bioreactor*), allowing a faster reduction of the leachate organic load; however a higher leachate quantity to be treated is produced [4,14].

A management including one or more of these strategies ensures several advantages in terms of both emissions quality and environmental impact.

Despite the influence exerted by the waste pretreatment on the emission intensity, the residual COD and ammonium nitrogen in the leachate can still be too high and their environmental impact is still not negligible. Besides, the length of the post-closure phase of the landfill management is strictly linked to the mechanical stability of the waste, which can require several

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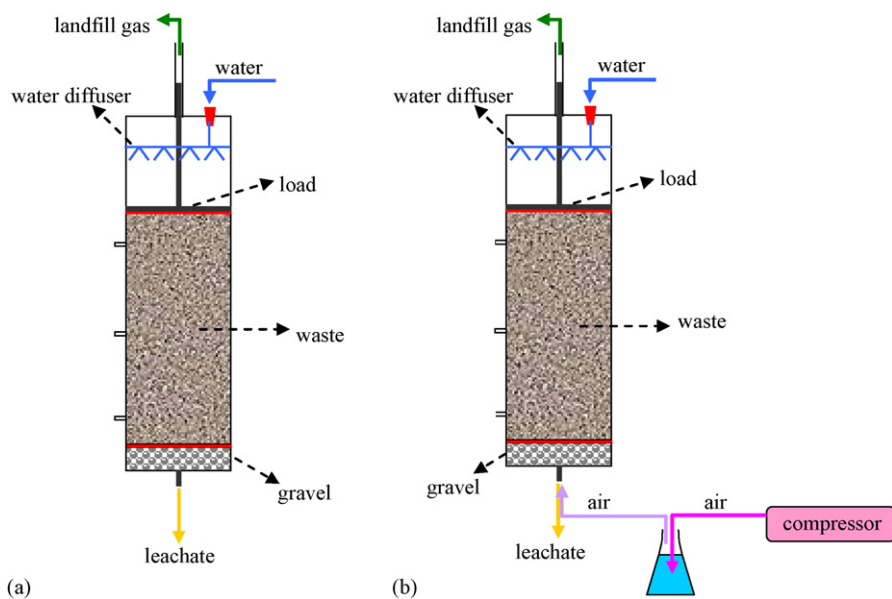*E-mail address:* silvia.sbaffoni@uniroma1.it (S. Sbaffoni).

Fig. 1. Scheme of the anaerobic reactors (a) and of the semiaerobic reactor (b).

decades to be achieved, even for mechanically-biologically pretreated (MBP) waste.

In recent years, several experimental studies involved the suitability of co-landfilling of different pretreated waste, in particular the residuals of both the mechanical-biological and thermal pretreatment of the municipal solid waste [4,14–16]. A significant reduction of the organic load of the leachate produced was observed [4,14,16–18], along with the dilution effect exerted by the incineration bottom ash (BA), being inert to the biological degradation process. The waste obtained by mixing the MBP waste and BA presented high buffering capacity, thus allowing a faster transition into the methanogenic phase [4,14–16,19]. In addition, a more rapid reduction of the BOD_5/COD ratio [4] and the acceleration of the landfill mechanical stability [16] can be achieved. On the other hand, some authors [14,20] found an increased heavy metal release, due to the complexing effect exerted by the organic and inorganic soluble ligands present in the waste; this behavior was not observed in the short term by Heyer and Stegmann [21], Lechner et al. [22], Boni et al. [15,16], and Lo [19].

The aerobic conditions in landfill reactors in which MBP waste and BA were co-disposed led to a significant improvement in the leachate quality in terms of organic and nitrogen load [4,17], even if the heavy metal concentration in the liquid phase can result in higher discharge [4] due to the metallic sulphides solubilization. Additional processes, such as carbonates and hydroxides precipitation, can occur, resulting in the heavy metals to be entrapped in the solid phase.

Based on these considerations, the aim of the experimental research presented here was the evaluation of the influence exerted by different managing and disposal strategies for co-disposal of the pretreated municipal solid waste organic fraction and BA in terms of both the leachate characteristics and the mechanical stability of the waste.

2. Materials and methods

2.1. Experimental setup and operating methods

The experimental phase was carried out in three lab-scale sanitary landfill reactors made of plexiglas cylinders, each one having the following dimension: 29 cm diameter and 100 cm height, for a net volume of about 66 l (Fig. 1). On the bottom of each reactor, a 6.5 cm deep drain made of gravel was located in order to simulate the in situ leachate drainage, whereas a channel allowed periodical leachate removal. A water diffuser with spray nozzles was added to the cylinder cover plates in order to simulate rainfall on top of the waste pile: care was taken in the water addition procedure, in order to ensure the distribution to be as uniform as possible over the entire waste pile, so as to minimize the channel effect.

During the entire experimental pilot study, a constant vertical loading was applied in order to simulate the weight of the upper layers of waste in a real landfill. For instance, a 5 kg steel plate was placed upon the waste pile in each reactor. On the plate several uniformly and closely disposed holes of about 0.8 cm diameter were present, through which a homogeneous distribution of the water on the waste could be guaranteed. The plate diameter was slightly smaller than the reactor's one, thus avoiding friction between steel and plexiglas, which could have slowed or even prevented the settlement. A disk of 29 cm diameter made by nonwoven fabric was placed between the steel plate and the bulk waste surface.

For landfill reactors set up, the MSWOF collected after the separation and composting process was used, along with BA from incineration of MSW. The separation and composting plant treats 500 t/d of unsorted MSW and consists firstly of a physical/mechanical sorting phase for the separation of the biodegradable organic fraction, which undergoes a composting

Table 1
Refuse filling and operating conditions adopted in the test columns

Column	Materials	Disposal	Operating conditions
A	30% BA + 70% MSWOF	Mixed	Anaerobic
B	30% BA + 70% MSWOF	Mixed	Semiaerobic
C	30% BA + 70% MSWOF	Layered	Anaerobic

process (active phase in windrows and curing phase) and then is sieved at 10 mm. The dry fraction (paper, plastic, textile residues) is converted into Refuse Derived Fuel (RDF), while the metals and glass are recovered. The incineration plant treats 120 t/d of unsorted MSW and a maximum of 10 t/d of pretreated hospital waste.

The three reactors namely A, B, C were all filled with 30% BA and 70% (by weight) 15 days aerobically biostabilized MSWOF and were realized and managed as shown in Table 1. The waste, after being sieved at 30 mm, was inserted into the reactors by adding 10 subsequent layers, each one compacted by applying about 61 kJ/m^3 energy, to reach a final unit weight of 8.9, 8.0, and 7.9 kN/m^3 for A, B, and C, respectively.

Some engineering properties of the waste, such as compressibility and permeability, are affected by the state of packing; thus the relative states of compaction were evaluated by means of the density index (or relative density), obtained by comparing the in situ void ratio of the waste into the reactors, with the minimum and maximum practical values. The minimum and maximum void ratios were determined with the waste compacted densely and poured loosely, respectively, in a metal mould. The density index (DI) was equal to 61.4, 46.9, and 47.8% for the reactors A, B, and C, respectively. Due to the higher density index, retarded or even minor settlements can be expected in the reactor A.

The rainfall was simulated by addition of 1.4 l of demineralized water twice a week until field capacity (FC); this amount was halved after FC was reached. Table 2 lists the FC and leachate production for the three reactors. The reactors A and B can be compared in order to evaluate the effect of the compaction. In fact, even if these reactors were both set up using the mixture of MSWOF and BA, they present different initial density index, as previously mentioned. The higher DI and unit weight in the reactor A resulted in a minor porosity as well as minor permeability, compared to reactors B and C. This led to a lower flow rate in the bulk waste and to a longer time needed to achieve the FC. Moreover, due to the similar DI and unit weight values, the reactors B and C can be compared in order to evaluate the influence of the waste disposal on their behavior in terms of hydraulic conductivity. BA and MSWOF monolayers in the reactor C seemed to increase the FC and the time needed for its

Table 2
Water addition and leachate production throughout the experimental period

Reactor	Field capacity (FC) (l)	Time for reaching (FC) (d)	Cumulative water addition (l)	Cumulative leachate production (l)
A	7.6	25	38.5	25.0
B	4.5	14	40.3	26.9
C	6.4	28	43.9	32.2

achievement. It was probably due either to the low permeability of the BA layers or to the very high water absorption capacity presented by the MSWOF layers: more water and a higher hydraulic head were needed to obtain the leachate outflow.

A compressor was used for the upward air supply in the reactor B. The air was supplied at a flow rate equal to 60 l/d for 14 h/d for five days per week, performed since day 35 for 57 days [4].

Settlements of the bulk waste in the reactors were followed by daily monitoring vertical drops of three comparators located on the top of each reactor.

2.2. Waste composition and characteristics

Data in Table 3 represent the initial characterization of the waste, prior to be disposed within the landfill reactors, in terms of pH, water content with respect to the wet weight (U), total volatile solids (TVS), total organic carbon (TOC), Total Kjeldhal Nitrogen (TKN), chlorides (Cl^-), heavy metals (Cd, Cr, Cu, Ni, Pb, Zn) concentration, and biological stability expressed by means of the dynamic respirometric index (DRI) [23,24]. The data reported in Table 3 for the mixtures are the weighted average of the corresponding BA and MSWOF values, using the BA and MSWOF percentages in each mixture as weighting factor. As it can be noted, the waste used in the three reactors had very similar values for several parameters such as U, TVS, TOC, and heavy metals. Due to the different characteristics of the MSWOF used, pH and DRI were found different in reactor A compared to reactors B and C. DRI, TVS, and TOC were quite high in the MSWOF. This was due to the short residence time in the biostabilization plant, which did not allow for complete degradation and consumption of the organic matter.

The dilution effect on the overall organic load, due to the presence of BA in the mixture was lower than expected. This result seems to be related to the low percentage of BA used, together with a nonnegligible content of organic matter in the BA [25], as indicated by TOC and TVS values. Contrarily, a strong dilution on the heavy metals content of BA can be observed. Such an effect was due to the very small amount of heavy metal in the MSWOF (with respect to BA) as well as to the high percentage of MSWOF used for the reactor set up. This resulted in significantly lower heavy metals content in all the reactors, thus avoiding a possible inhibitory effect on the microorganisms metabolism [19].

2.3. Acid neutralization capacity

Probably the most relevant waste property affecting the pH of the leachate is represented by the acid or base neutralization capacity (ANC/BNC), which are a measure of the ability of a system to neutralize the influence of acids or bases and assess the sensitivity of the material itself to external influences and/ or internal stresses (e.g. mineralization, organic matter degradation).

ANC (or BNC) represents the amount of acid (or base) needed to reach a final end pH. The amount of acid (or base) was evaluated [26] through acid (or base) titration with HNO_3 1N (or NaOH 1N). The test was carried out in polyethylene bottles by

Table 3
Initial characterization of the waste

Parameter	MSWOF in A	BA in A	Waste in A	MSWOF in B	BA in B and C	Waste in B	MSWOF in C	Waste in C
pH	6.60	11.8	8.30	8.70	11.35	9.49	8.70	9.50
U (%)	42.6	33.1	35.5	37.5	33.6	36.3	33.6	33.6
TVS (%)	69.0	14.8	52.7	69.1	10.2	51.5	69.6	51.8
TOC (%)	33.5	8.0	26.9	36.5	11.7	29.1	34.4	27.6
TKN (%)	0.60	n.d.	0.40	2.2	1.8	2.07	2.2	2.1
Cl^- (mg/kg)	6.80×10^3	11.4×10^3	7.80×10^3	5.67×10^3	3.90×10^3	5.14×10^3	5.52×10^3	5.03×10^3
DRI ($mgO_2\,kg\,VS^{-1}\,h^{-1}$)	5.04×10^3	–	3.53×10^3	4.19×10^3	–	2.94×10^3	4.19×10^3	2.94×10^3
Cd (mg/kg)	1.5	7.5	3.3	1.5	4.20	2.3	1.5	2.3
Cr (mg/kg)	0.045×10^3	1.08×10^3	0.356×10^3	0.0567×10^3	0.894×10^3	0.308×10^3	0.0567×10^3	0.308×10^3
Cu (mg/kg)	0.148×10^3	8.73×10^3	2.72×10^3	0.164×10^3	0.431×10^3	1.41×10^3	0.164×10^3	1.41×10^3
Ni (mg/kg)	29.0	170	71.3	38.2	182	81.3	38.2	81.3
Pb (mg/kg)	0.278×10^3	4.36×10^3	1.50×10^3	0.213×10^3	4.43×10^3	1.48×10^3	0.213×10^3	1.48×10^3
Zn (mg/kg)	0.321×10^3	3.96×10^3	1.41×10^3	0.479×10^3	5.66×10^3	2.03×10^3	0.479×10^3	2.03×10^3

adding demineralized distilled water to the waste samples with a liquid to solid ratio (L/S) equal to 10 l/kg. The bottles, one for each pH value (at least eight in the range 4–12), were agitated through an end-over-end tumbler at 10 rpm for 48 h. The pH values were measured after 4, 44 and 48 h. Equilibrium condition is defined when the pH deviation between 44 and 48 h does not exceed 0.3 pH units. For the pH measurement the agitation was stopped and the solid phase was allowed to settle for about 5 min.

ANC and BNC were evaluated for the waste used and their mixtures, allowing forecasting the waste behavior corresponding to pH variation under different environmental conditions established in landfills (e.g. acidogenic and methanogenic phases). The buffering capacity of the material affects the evolution of the pH of the leachate over time, thus allowing the expected pH range for the application site to be estimated.

In the case of MSW incineration residues, which are most often basic in their nature, alkalinity of the material is the relevant parameter, so that ANC is the appropriate measure of neutralization capacity.

2.4. Leachate characterization

Leachate was collected twice a week from each reactor. On leachate samples biochemical oxygen demand (BOD_5), chemical oxygen demand (COD), chlorides, TKN, NH_4^+-N, NO_3^-–N and NO_2^-–N concentrations were measured according to the Standard Methods for the Examination of Water and Wastewater [27]. Heavy metal concentrations—the same as measured on the solid waste samples (i.e. Cd, Cr, Cu, Ni, Pb, Zn)—were determined by means of atomic absorption (Perkin-Elmer 3030B) along with pH by a standard probe (Corning: Checkmate II).

3. Results and discussion

The most relevant results obtained during the experimental period are presented and discussed below.

3.1. Leachate

3.1.1. pH

Leachate from reactor A showed initial pH values around neutrality (7–7.5), as shown in Fig. 2. After 80 days pH decreased

to a minimum value equal to 6.2, due to the acidogenic phase of the anaerobic degradation of MSWOF; pH increased since day 140 up to 7.8 after 170 days evidencing the beginning of the methanogenic conditions and maintaining values over 7.5 until the end of the test. Leachate from reactors B and C was initially characterized by a moderately acid pH (6.3–6.4). The pH profile for C showed a similar behavior with respect to reactor A, due to the evolution of the anaerobic degradation process. Nevertheless, both acidogenic and methanogenic phase began earlier in reactor C, probably due to the initial slightly acid pH values and to the lower content of biodegradable organic matter, as shown by DRI values.

In reactor B, instead, a strong relation between the aeration and pH was observed; indeed, pH values were always above 7.5 when the aeration was on and dropped down to neutrality when air was no longer supplied.

The results show the occurrence of the two following phenomena:

- the presence of BA in the anaerobic reactors limited the length of the acid phase [16,19], due to the reduction of acid compounds content, as well as to the high BA buffer capacity, able to neutralize the volatile acids produced by the anaerobic metabolism, thus promoting a more rapid transition into the methanogenic phase;
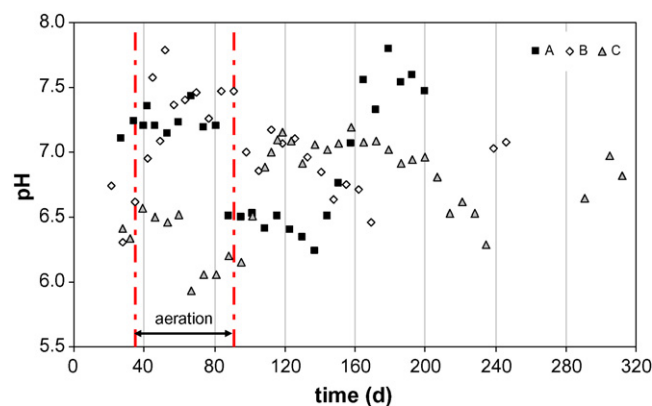

Fig. 2. pH time profile in the leachate collected from the reactors A, B, and C.
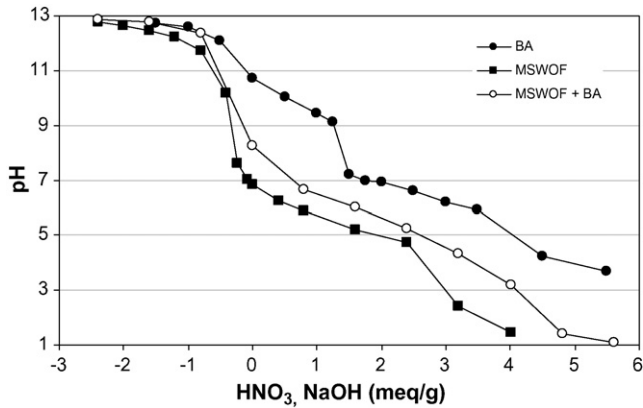
Fig. 3. ANC/BNC curves for BA, MSWOF and the mixture (negative values correspond to NaOH addition, while positive ones to HNO_3 addition).

- the forced aeration in the reactor B allowed to maintain pH values above the neutrality: the aerobic environment avoided the establishment of the acid phase and the air flow led to the stripping of the CO_2 produced by the oxidation of organic matter.

The high pH values, especially in the anaerobic reactors, can be explained by taking into account the ANC curves for the materials used (Fig. 3). As expected, BA presented the highest ANC, due to the presence of calcium minerals (calcium hydroxides, silicates and carbonates), which determine chemical conditions of the aqueous solutions [19,28]. An increase in the MSWOF ANC can be observed after BA addition. This parameter can be of great importance in the case of landfilling: the presence of materials with high ANC implies a short acidogenic phase; consequently some advantages can be obtained in terms of:

- lower metal release in leachate (especially for those metals with high solubility at low pH);
- less landfill gas production;
- faster biodegradation of the waste organic content, due to a more rapid establishment of the methanogenic phase.
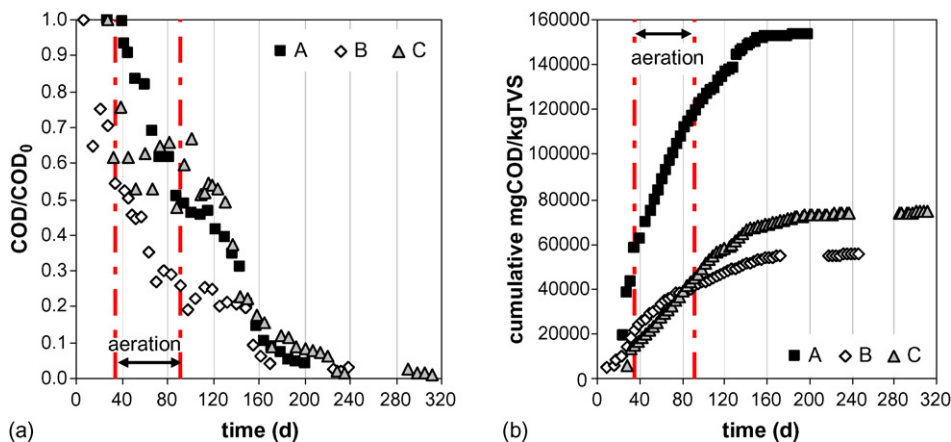
At the time of disposal, BA display their maximum alkalinity level, decreasing as the material comes into contact with the leachate and dissolved alkalinity is removed from the system by the leachate. As a consequence, the residual alkalinity at any time depends on the initial alkalinity of the material, on the dissolution of alkalinity at various pH values in the leaching scenarios and on the extent of the water infiltration through the waste.

3.1.2. COD and nitrogen

As expected, initial COD values in the leachate were significantly high, due to the almost negligible dilution effect exerted by BA. In fact, the TOC of the mixtures in the reactors were near 30% and quite similar to the MSWOF ones, because of both the high percentage of MSWOF used and the nonnegligible TOC of BA (Table 3).

At the beginning of the test, COD values were up to about 1,30,000 mg/l for A and 60,000 mg/l for both B and C; these initial differences were probably due to the different initial content of biodegradable organic matter and to not yet stable hydraulic conditions within the bulk waste, thus producing a different and unsettled initial release. In fact, as shown in Table 3, the waste used in the reactor A presented the highest DRI value (3526 mgO_2 kg VS^{-1} h^{-1}); moreover, for reactor A more water was needed to reach the FC (Table 2) and this probably caused the very high initial release in the leachate of the soluble organic matter present in the bulk waste. The time evolution of COD/COD_0, where COD_0 represents the initial COD value, showed a similar behavior for the two anaerobic reactors; the concentration values (Fig. 4(a)) and the cumulative release curves (Fig. 4(b)) indicate the presence of two mechanisms in the organic matter removal: washing out and biological degradation, the latter occurring at the highest rate after the establishment of the methanogenic environment (about 140 and 100 days for reactors A and C, respectively). Instead, the aerobic conditions in the reactor B allowed the highest biodegradation rate of organic substance, with respect to the other reactors, thus reducing also the cumulative organic release [4,13,29]: COD values for reactor B went down to 10,000 mg/l after only 90 days, whereas the same values were reached only after 160 and 180 days for reactors
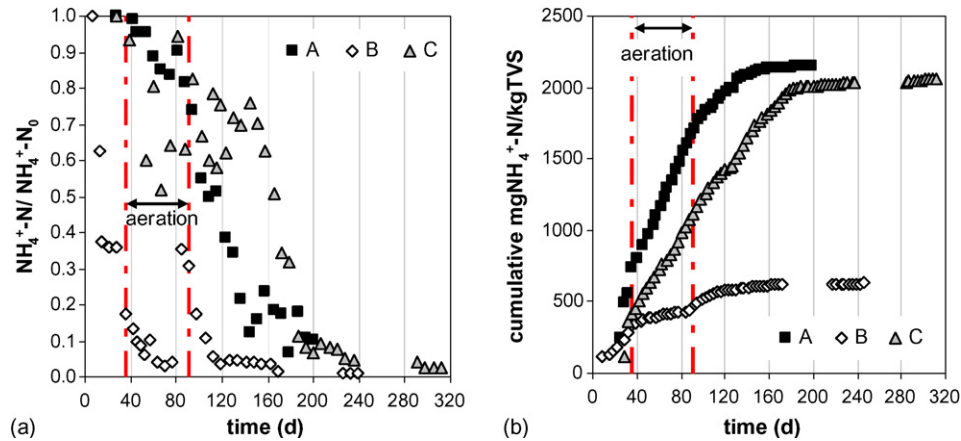

Fig. 4. COD (a) and cumulative COD (b) time profiles in the leachate collected from the reactors A, B, and C.

Fig. 5. NH_4^+–N (a) and cumulative NH_4^+–N (b) time profiles in the leachate collected from the reactors A, B, and C.

C and A, respectively, when the methanogenic condition was established. At the end of the stabilization process, organic matter consists mainly of hardly or nonbiodegradable compounds.

As mentioned above, the cumulative release proved to be higher in reactor A, especially at the beginning of the experimental period; the plateau was reached after about 160 days for both the anaerobic reactors, while the aerobic one presented a lower stable cumulative release (about 50,000 mgCOD/kgTVS) in a shorter period, due to the further-reaching degradation of the bioavailable waste components: the higher the degradation rate, the lower the cumulative release.

The ANC value of the mixture, higher than the MSWOF one, enabled a quick establishment of the methanogenic phase [15] and a rapid biostabilization of the organic matter in the anaerobic reactors. In the semiaerobic reactor the acceleration of the biodegradation was enhanced by both the high ANC value and by the air supply, which avoided the establishment of the acid phase.

The initial ammonium nitrogen concentration was equal to about 1200 mg/l for both the reactors B and C and 1600 mg/l for A (Fig. 5(a)). For the anaerobic reactors the main removal mechanism was the washout: more than 200 days were needed to obtain the same ammonium nitrogen levels as those observed at the day 120 for reactor B. As shown in Fig. 5a, where NH_4^+-N_0 represents the initial ammonium nitrogen concentration in the leachate, the curves for the anaerobic reactors A and C were quite similar, while the time profile curve for the aerobic reactor rapidly decreased in correspondence to the aeration phase. Also the cumulative release (Fig. 5b) for A and C reactors was quite similar, with a plateau reached at a value of about 2000 mgNH$_4^+$-N/kgTVS after about 140 and 180 days, respectively.

Instead, for the aerobic reactor, as already observed for COD curves and shown by the ammonium nitrogen cumulative release, the total release was far lower (about 600 mgNH$_4^+$–N/kgTVS) and the stability was achieved in a shorter time lag, after about 100 days. In fact, two mechanisms occurred in the ammonium nitrogen removal: nitrification and washout, even if the former was typical only of the aerobic reactor. As the forced aeration was initiated in the reactor B, the ammonium nitrogen concentration dropped from 500 to 70 mg/l after 25 days,

reaching a stable value of about 50 mg/l until the end of aeration. Once the aeration was stopped, the ammonium nitrogen concentration rapidly increased up to 400 mg/l, indicating that during the aerobic phase a third removal mechanism occurred: ammonia stripping through the forced upward airflow. The nitrate curve confirmed the effective presence of the nitrification process within the bulk waste, as shown in Fig. 6, where the nitric and nitrous nitrogen time profiles in the leachate collected from reactor B are presented; in the first anaerobic phase the decrease observed was due to nitrate solubilization and leaching. During the aeration of the bulk waste, both nitrate and nitrite concentration rapidly increased, as a consequence of the nitrification process established inside the reactor; such an increase corresponded to the above mentioned ammonium nitrogen decrease.

The nitric and nitrous nitrogen in the leachate collected from both the anaerobic reactors, were always below 40 and 0.01 mg/l, respectively, and significantly lower than the concentrations detected in the leachate from the aerobic reactor. The oxidized forms of nitrogen present in the bulk waste used for the experimental activity were produced during the aerobic biostabilization pretreatment in the MBP plant.

Similar results, in terms of ammonium release via the leachate were obtained by Cossu et al. [4] and Heyer et al. [13] for both aerated and anaerobic landfill reactors.
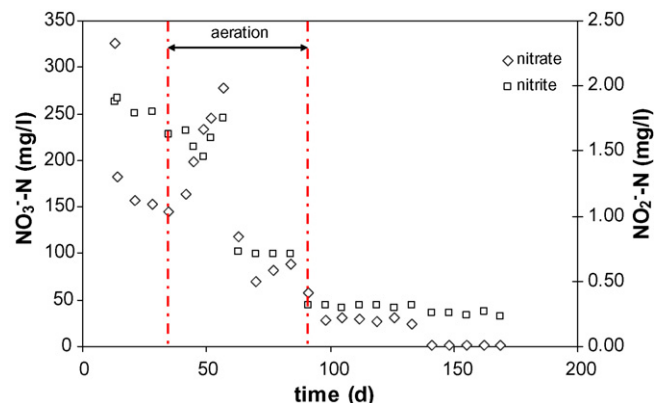

Fig. 6. NO_3^-–N and NO_2^-–N time profiles in the leachate collected from reactor B.
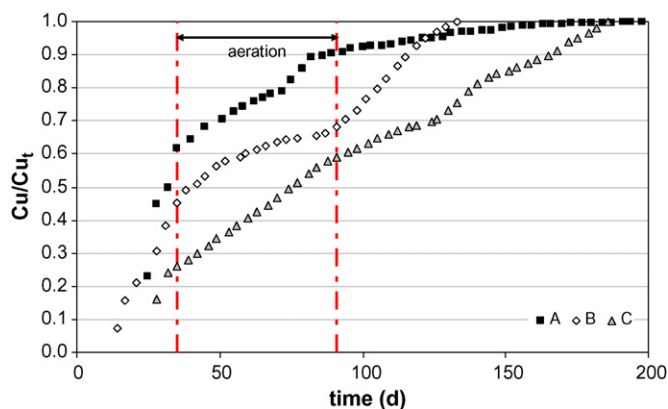
Fig. 7. Cumulative Cu time profiles in the leachate collected from the reactors A, B, and C; Cu_t represents the total Cu release.
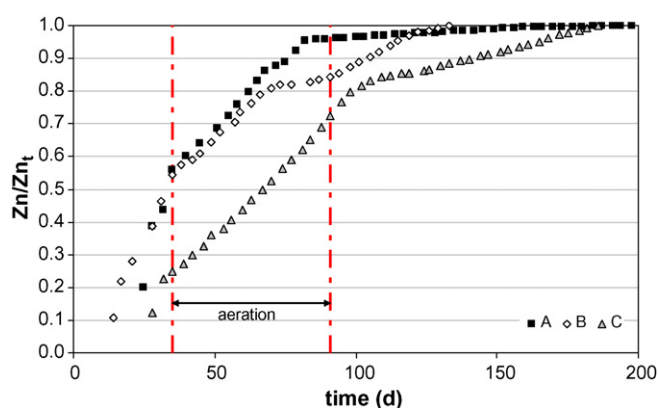

Fig. 8. Cumulative Zn time profiles in the leachate collected from the reactors A, B, and C; Zn_t represents the total Zn release.

3.1.3. Heavy metals

With respect to the heavy metals concentration measured in the leachate, Figs. 7, 8 and 9 show the cumulative release of Cu, Zn, and Cr, respectively, from the three reactors. The data presented are dimensionless and were calculated by dividing the milligrams of heavy metal released in time by the total release at the end of the experimental period.
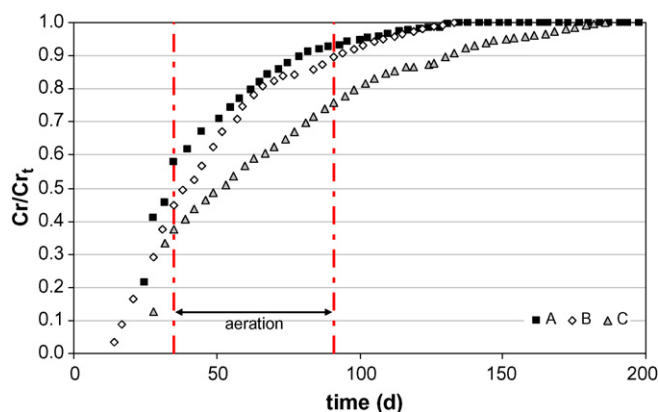

Fig. 9. Cumulative Cr time profiles in the leachate collected from the reactors A, B, and C; Cr_t represents the total Cr release.

Cu reached a stable value (about 130 mg) in the leachate from the reactor A, after about 100 days; the cumulative release from C was still occurring at the end of the experimental period, even if the values were significantly lower than the ones from the reactor A (about 30 mg). For reactor B, during the aeration phase, the Cu release was quite stable, increasing only at the end of the period to values similar to those reached in reactor A, due to the pH decrease; therefore, it can be inferred that the Cu solubility is mostly influenced by pH rather than by the redox potential. The same results were obtained for the Pb cumulative release curves not presented.

The cumulative Zn time profiles reached a stable value, equal to about 480 mg, after about 100 days in the leachate from the reactor A. The release can be considered quite stable also for the other two reactors, even if the final values reached were lower (about 80 and 60 mg for reactors B and C, respectively). The shape of the curves obtained for the anaerobic reactors were quite similar, even if the absolute release values were different, probably due to the different biological stabilization of the MSWOF used. Besides, no effect seemed to be exerted by the forced aeration on the cumulative release: the release occurred at almost the same rate observed in the reactors A and C during the aeration period, while an increase in the Zn release occurred in correspondence of the air disruption, probably due to the lower pH values reached which increased the Zn solubility and thus its concentration in the liquid phase. It can be asserted that Zn solubility, in the environmental conditions established during the present experimental activity, was affected by the pH more than by the redox potential, which was positive during the aeration turning into negative values after the aeration suspension. Ni, not presented, showed the same behavior as the Zn.

Cr cumulative release was not influenced neither by the aeration nor by the different disposal of MSWOF and BA: in fact, the curves obtained for the three reactors presented very similar shape and slope.

Cd was not detected in the leachate from any reactor, due to the very small content present in the bulk waste.

In general, it was observed that the heavy metals release from the anaerobic reactors always started during the hydrolytic and acidogenic phase, and it slowed down as soon as the methanogenic conditions were established. The higher complexity and biological stability of the organic matter being initially present in the reactor C, as proved by the lower DRI value, decreased the total amount of heavy metal finally released. The metals leached from BA were likely to bind onto the organic ligands in MSWOF, leading to equilibrium between solution and waste [19]. However, the heavy metal release pattern was very similar to that observed in reactor A.

As mentioned above, in the experimental conditions simulated in the present research, the redox potential, affected by the aeration condition, seemed not to exert any significant effect on the solubility of the heavy metals investigated. On the contrary, pH can be considered to be the parameter controlling the release [28], as shown in Fig. 10 for Cu released by the waste in the aerated reactor. The Cu concentration in the leachate increased once the aeration was stopped, probably due to the combined effect of the lower pH values, leading to a higher solubility of
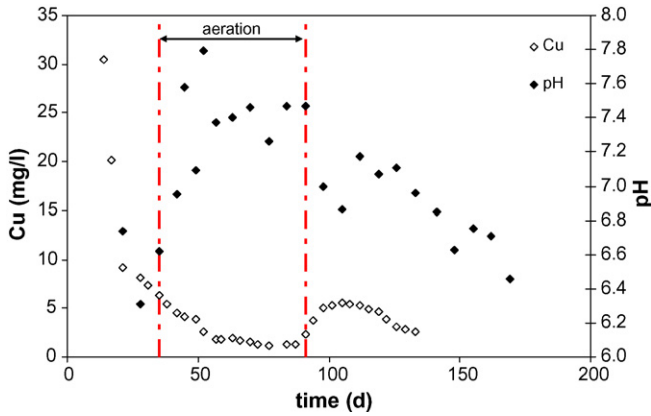
Fig. 10. pH and Cu time profiles in the leachate collected from reactor B.

Fig. 12. Heavy metals bound in the solid matrices within the reactors A, B, and C at the end of the experimental period.

the metal [30], and to the presence of high sulphate concentrations in the waste. Under aerobic conditions, sulphate is not reduced into sulphides, which would have provided heavy metal precipitation. The Cu time profile in the leachate produced by the anaerobic reactor A, is shown in Fig. 11: the decrease of the Cu concentration was mainly due to the reducing environment established in the reactor leading to a rapid sulphide generation and to a consequent metal precipitation; Cu release did not increase, even during the acidogenic phase in correspondence to pH values lower than 7. It can be concluded that metals leaching and binding were dynamic behaviors, involving kinetic and thermodynamic mechanisms of release, adsorption/desorption, chelation, precipitation. The variability of available concentration was strictly dependent on pH changes and on the dynamic binding of metals with functional groups in the total solids.

It is noteworthy that for all the reactors the total amount of heavy metals released during the experimental activity represented only a very limited fraction of the initial content measured in the bulk waste; in fact, the total bound percentage of heavy metals was always higher than 94%, as shown in Fig. 12; the values presented in Fig. 12 were calculated, for each metal, as the ratio between the total amount (expressed as grams) of metal entrapped in the waste at the end of the experimental period (not released via leachate) and the total amount of metal initially present in the waste used (expressed as grams).
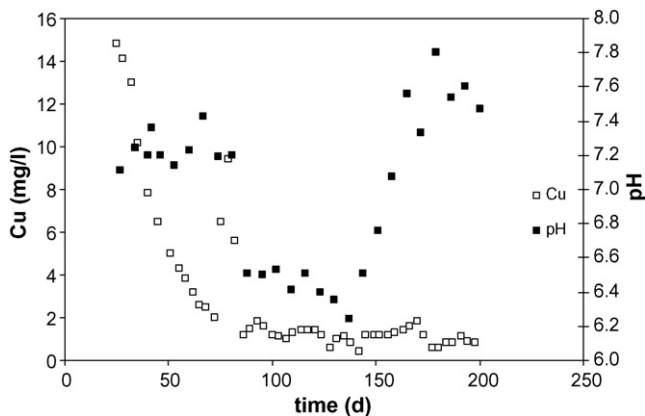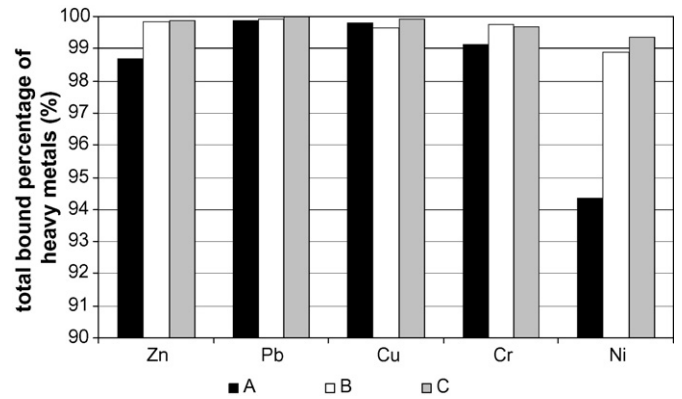
This phenomenon, also observed by other authors [31–33], was probably due to the high content of the organic matter (especially as humic acids), increasing the ligand effect, which retained the heavy metals within the solid matrix. In addition, the reducing environment in the landfill (no access for oxygen, sufficient nutritional matter and moisture for bacteria) promoted the growth of sulphur-reducing bacteria and the consequent precipitation and retention of metals in the reactor; moreover, the anoxic environment promoted metal hydroxide precipitation and retention.

The total release was quite negligible in the short-term; therefore, the potential heavy metals leaching could represent a risk source in the long-term, in case of a change in the environmental conditions within the bulk waste, especially in terms of pH as well as ORP.

In all the reactors, the heavy metals concentrations were low enough to avoid a potential inhibitory effect on the microorganisms metabolism [19]. Besides, the total heavy metals mass detected in the leachate was quite lower than the one released by BA mono-landfills [18], due both to the dilution effect exerted by MSWOF and probably to the adsorption of the heavy metal on the organic matter [15].

3.2. Settlements

Fig. 13 shows the cumulative settlement time-profile for the three reactors; the settlements can be attributed to both the compaction of the bulk waste because of its own weight and to the waste mass reduction, as a consequence of the biological decomposition. Therefore, it can be assumed that the settlement rate slowed down appreciably as soon as the biological stability is achieved.

The settlements started earlier (after two days) in reactor C, mainly due to the lower saturation degree (34.5%); moreover, the MSWOF monolayer probably presented a higher compressibility, if compared to the mix. Conversely, reactors A and B had greater initial saturation levels (42.6 and 38.2%, respectively), which caused stronger interstitial forces and higher material's resistance, which retarded the bulk wastes to settle. Due to the higher values of both saturation degree and initial compaction (as shown by the higher DI and unit weight
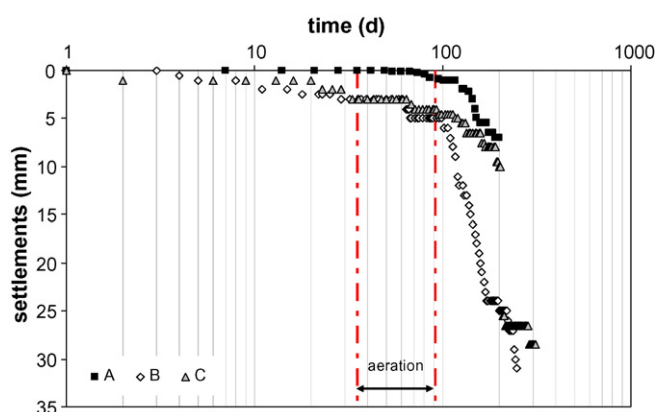

Fig. 11. pH and Cu time profiles in the leachate collected from reactor A.

Fig. 13. Cumulative settlements time profiles for the reactors A, B, and C.

Fig. 14. Linear fitting of the settlement experimental data for the reactor A.

values), the settlement occurred later in reactor A, compared to B. The settlement was first registered after 14 days for reactor A, while it was registered after only four days for B.

In all the reactors, the initial settlements were quite low or even absent, because of the lag phase in the biodegradation activity: the biomass, coming from the aerobic conditions of the composting process, once transferred into an anaerobic environment, needed some time to adapt its metabolic pathway to the new conditions. Afterwards, the settling process started as a consequence of the biological activity in the acid phase of the anaerobic biodegradation; then, it significantly increased because of the acceleration of the organic fraction consumption during the methanogenic activity. Therefore, in both the reactors A and C, the settling rate increased appreciably after about 100 days, due to the establishment of methanogenic conditions within the bulk waste which corresponded to the higher rate of biodegradable organic matter degradation in the anaerobic environment.

The behavior of reactor B was quite different: no significant differences could be observed during the aerobic phase if compared to the anaerobic reactors. In fact, despite the higher biodegradation rate achieved, only 5 mm of settlement were observed in 100 days, probably due to the strong interstitial forces generated by the air present in the bulk waste and to the expansion effect caused by the upward air flow. In correspondence of the anaerobic phase, the bulk waste settlements reached 31 mm, due to the weakening of the structure produced by the remained waste matrix [13]: the waste replaced the void spaces left in the wake of the organic matter consumed during the biodegradation occurred in the aerated phase.

At the end of the experimental period, the bulk waste of the three reactors presented a DRI value lower than 500 mgO$_2$/kgTVS h, which, according to the Italian guidelines, the European draft on the biowaste and some experimental evidences [24,34], corresponds to a biological stability state of the organic fraction. This assumption was confirmed by the COD time profile: at the same time COD concentration measured in the leachate showed constant pattern with values below 2000 mg/l and a plateau was reached. The corresponding value of the cumulative settlement in reactor A was 7 mm (1% of the total initial waste height); such a value must therefore be com-
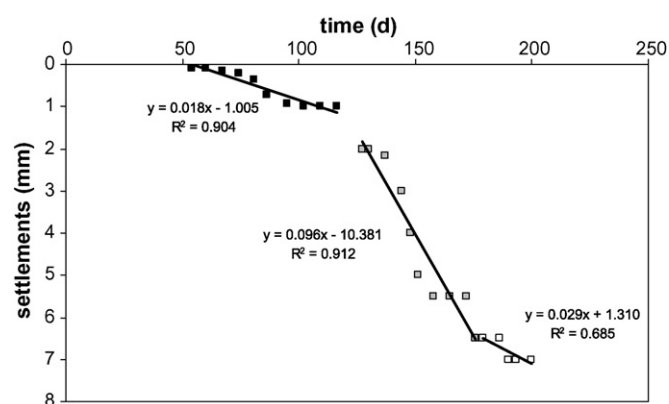
pared with the cumulative settlement measured at the same time in reactor C, which is equal to 9.5 mm (1.5% of the total initial waste height). The two anaerobic reactors showed essentially the same behavior in the first 200 days; the negligible differences in this period and the significant shift in the last 100 monitored days can be explained by the following considerations.

The presence of BA mixed to the MSWOF in the reactor A probably worked as a rigid skeleton which firstly retarded the starting of settling and secondly hindered the settlement rate, thus leading to a lower value reached at the biological stability occurrence. Despite the increase in space made available by the biodegradable fraction consumption, the bulk waste did not settle appreciably because of the BA. Due to aggregation, the residual waste particles created a cement-like form, which made it necessary to use mechanical tools to dismantle reactor A. Probably BA supplied aggregating and stabilizing properties to the materials which they were added to and functioned as a structural and support material [16]. Instead, the BA layers in the reactor C probably represented a significant weight on the MSWOF layers below, thus increasing the total settlements to about 28 mm after more than 300 days. However, the settlement of the reactor A can be expected to continue after 200 days, but the settling rate is likely to be much smaller, due to the exhaustion of the biodegradable fraction.

Figs. 14 and 15 show a linear fitting of the settlement data measured in the A and B reactors, respectively. It can be observed
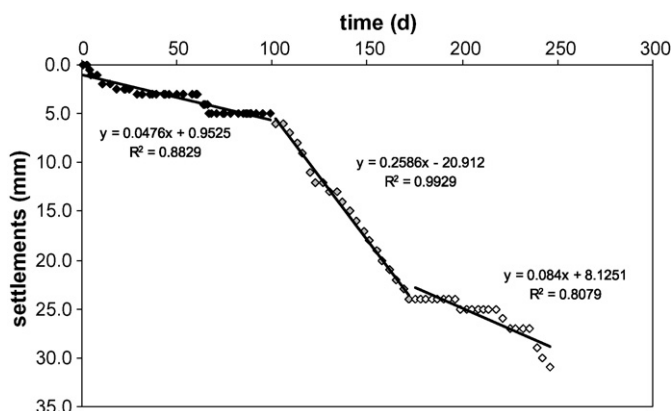

Fig. 15. Linear fitting of the settlement experimental data for the reactor B.

that a good correlation was always present in the different periods of the biological activity:

- for the anaerobic reactor: adaptation and acid phase, methanogenic phase and stability phase;
- for the aerobic reactor: initial period of very low settlements due to the interstitial pressure caused by the presence of air, significant settlements after the end of the aeration period, stability phase.

In the same figures, the corresponding settling rates are also shown, as measured by the slope of the best fitting straight line; the maximum settling rates were reached:

- for reactor A as the methanogenic phase was established; the first settling phase corresponded to the acid phase in the anaerobic metabolism and to lower pH values. When the pH rose, indicating the beginning of the methanogenic phase, a more rapid consumption of the organic matter occurred along with a clear change in the slope of the settlement time-profiles due to the settling rate increase;
- for reactor B as the aeration was stopped, as a consequence of the above mentioned reasons.

The additional settlements that occurred in correspondence of the stable organic load release in the leachate and after the achievement of the biological stability, cannot be ascribed to the biodegradation of the organic matter, but to either crushing of the solid frame or creep.

4. Conclusions

A comparison of the emission from the three lab-scale reactors and the evolution of the settling process indicate the following:

- the semi-aerobic conditions concurred to an increase in the biodegradation rate of the organic matter and promoted the nitrification process, thus providing the lowest emission level; besides, the waste settlements occurred earlier. Compared to the strictly anaerobic conditions, the aftercare for the leachate emission path could be reduced by at least several decades under in situ aeration;
- the different disposal strategies (mixed or layered) of MSWOF and BA did not exert any significant effect on the leachate quality in the anaerobic reactors; however, the disposal of MSWOF and BA in layers seemed to influence the settlements of the bulk waste only in the long-term, probably due to the intense weight of the BA on the MSWOF layers;
- the heavy metal release from the bulk waste via the leachate seemed not to be influenced neither by aeration (i.e. ORP values) nor by waste disposal; instead the release was controlled essentially by the pH, as well as by a variation in both the stability degree and the complexing capacity exerted by the organic matter on Cu, Pb, Zn, and Ni release.

On the basis of these considerations, the aerobic management can be advised as the most suitable strategy providing a more rapid biological and mechanical stabilization of the bulk waste; nevertheless, it implies significant economical as well as operational difficulties, related to the optimization of the quantity and the distribution of water and air in the landfill. Thus a proper costs-benefits analysis must be carried out in the project phase.

With respect to the disposal strategy, it can be suggested that subsequent layers of BA and MSWOF should be realized in a co-landfill, just in order to accelerate the main settlements.

Moreover, despite the presence of BA, the solubilization of heavy metals was quite negligible and their concentration in the liquid phase did not seem to exert any significant inhibitory effect on the biomass metabolism.

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