

Management of wastewater from landfill of inorganic fiberglass

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Abstract. Sustainability is one of the key factors in smart environment management and include the reduction of environment footprint. The waste and wastewater management plans are aimed on actions to reduce the amount of waste and environmental pollution. This includes collection of waste, logistics, storage, processing or valorisation and also treatment. The aim of this research was to evaluate environmental pollution risk and to demonstrate one of the wastewater management schemes to reduce the pollution level.

Evaluation of the better management scheme was performed in one of the landfills in Latvia, where fiberglass waste and other inorganic waste is stored. Onsite evaluation results demonstrated the need to develop a better wastewater management scheme of inorganic fiberglass landfill. After that, laboratory–scale experiments for conventional coagulation and biodegradation tests have been performed for efficient management.

Key words: landfill leachate, fiberglass, wastewater management, wastewater treatment, environmental pollution.

INTRODUCTION

Over the recent years, people have become aware that environment is a comprehensive resource that requires smart management. This has led to an increased need for sustainable energy management, steady energy supply, reduced energy consumption and search of new renewable energy sources (Coaffee, 2008). In the meantime the security policy has been focused on terrorist hazards and natural disasters (Collier et al., 2013), resource recovery and reduction of wastes.

Efficient waste management is one of the priorities in EU use of resources and is described in Waste Avoidance and Resource Recovery Act from 2001, Waste Avoidance and Resource Recovery Amendment (Container Deposit Scheme) Act 2016 No. 57 and Waste Avoidance and Resource Recovery (WARR) Strategy updated every five years. In 1992 the Convention on the Protection of the Marine Environment of the Baltic Sea Area (Helsinki Convention) was signed to reduce the inflows of nutrients and hazardous substances, as well as to promote a cleaner future of the Baltic Sea. The document was adopted by HELCOM in 1993 (Fuller, 2013). The international convention encompasses various measures for prevention and elimination of pollution of the Baltic Sea. Due to

high anthropogenic activities, increase in the level of the nutrients (nitrogen and phosphorus) in the Baltic Sea catchment area, eutrophication process occurs faster than expected (Ærtebjerg et al., 2003). The waste and wastewater management descriptions are aimed on actions to reduce the amount of waste and environmental pollution generated by households and industry, resource recovery including re-use, recycling, reprocessing and also better waste and wastewater management procedure. This includes collection of waste, logistics, storage, processing or valorisation and also treatment.

Although glass can be repeatedly recycled with no loss of quality, there are still large volumes of glass ending up in the landfill. Glass recycling rates in European Union nations are the world's highest, averaging 73% (FEVE News Corner, 2017), but in other countries glass recycling rate is less significant, for example in the US (34%) and Singapore 20%. One major barrier to conventional recycling is the need to separate glasses into its various types (Heiriyanto et al., 2018), e.g., fiberglass, cullet, container glass, mirrors and other metal-coated products. Thus, the formation of highly pollutant leachate by water infiltration through waste deposits is inevitable. The control of a landfill site and effective appropriate treatment has a high-priority importance for the present and future environmental conditions.

The most popular method to reduce the pollution from landfill leachates is reverse osmosis (RO). RO systems were developed in 1950 in USA for water desalination. These systems use a semipermeable membrane to remove ions, molecules and larger particles from solution (Busch et al 2010; Šir et al., 2012). These are effective, but have several disadvantages, for example, become clogged often and produce a limited amount of water per day. The RO filter system management is quite expensive and it is predicted that the amount of global landfill wastewaters will increase in the future; therefore, it is necessary to apply cost effective and alternative wastewater treatment methods such as coagulation and biofiltration. Certainly, advantages and disadvantages depend on the untreated water quality and requirements for treated water quality before it can be released into the environment.

The aim of this research was to evaluate environmental pollution risk and to demonstrate one of the wastewater management schemes to reduce pollution level with cost effective approaches like coagulation and biofiltration. The proposed technological approach was tested on wastes from a landfill where fiberglass and inorganic material is traditionally deposited.

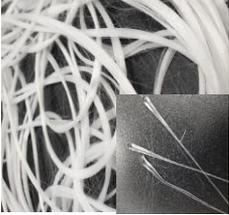
MATERIALS AND METHODS

Sample collection

Wastewater samples (storm water after the direct contact with fiberglass waste) were collected from a landfill in Latvia in plastic carboys (50 mL⁻² L) stored in a refrigerator (2 °C to 5 °C) after transport and analysed within 24 h for their chemical characteristics.

After field inspection there were six types (Table 1) of fiberglass solid wastes, based on the difference on its structure, hardness and thickness. All types were collected in plastic bags and stored at room temperature (RT) before experiments.

Table 1. Description of fiberglass samples

Sample	Picture of fiberglass	Description
1.		White, hard, dense, long threads with glass capsules
2.		Yellow, hard, dense, long threads
3.		Light yellow, soft fibre, short threads
4.		Yellow, hard, dense, long threads, similar to glass with fluffy surface
5.		Natural white, soft fibre, short threads
6.		Light white, hard, dense, very long threads, similar to fabric yarn

Experimental setup

In this study, the management scheme for wastewater from landfill of inorganic fiberglass was developed via: field inspection, sampling on the field, contaminant migration tests, coagulation tests, biodegradation tests, development of recommendations for waste management on the landfill.

Contaminants migration tests

To determine the contact time required to extract the maximum amount of contamination from the fiberglass samples and its migration into the water phase 200 g of fiberglass samples were soaked in 2 L of water (10% w v⁻¹) and kept for 24 hours at dark (RT = 21 ± 2 °C) in reservoirs with headspace. Six type of collected fiberglass were used to determine the amount of contamination from different type of fiberglass samples. Subsamples were taken for analysis after 48 h (Table 2).

To prepare average artificial wastewater fiberglass sample were homogenised through the mixing in equal amount fiberglass, an average sample were soaked in tap water (10% w v⁻¹) and kept for 24 hours.

Coagulation or Jar tests

Standard Jar tests (Phipps & Bird PB-900 Six Paddle) were used to investigate the removal of the organic matter. Aliquots (2 L) of sample were dosed with 3 stock solutions of coagulant (aluminium sulphate, iron chloride, bentonite). Samples were refrigerated prior to coagulation Jar testing, which was undertaken at room temperature (21 °C) and involved a 1 min rapid mix at 313 rpm, a 30 min slow mix at 41 rpm and 1 h sedimentation period. The pH was adjusted before the coagulation using concentrated hydrochloric acid and 10 M sodium hydroxide. At the end of the sedimentation period subsamples were taken for analysis (Table 2).

Biodegradation tests

The biodegradation kinetic tests were performed by the manometric methods (Jouanneau et al., 2014) for BOD determination using the OxiTop® IS 12 system (WTW, Germany). The samples were incubated for 5 days at 20 ± 1 °C (incubator) in 510 mL nominal volume brown bottles closed tightly with the OxiTop® measuring head and placed at the stirring platform. The results were expressed as mg O₂ L⁻¹ and this allowed to investigate the BOD removal efficiency.

Analysis of wastewater samples

All analyses were conducted according to the standard methods (Table 2).

Total organic carbon (TOC) measurements were performed with Primacs MCS TOC analyser (Scalar, Netherland) based on high temperature and acidification of sample and by the difference of the total carbon and inorganic carbon measurement. Each sample was tested in duplicate and the mean values were calculated (CV ≤ 2%).

Table 2. Water quality analytical methods

Parameter	Reference
Total Organic Carbon	LVS EN 1484:2000
Chemical Oxygen Demand	LVS ISO 6060:1989
Biological Oxygen Demand	LVS EN 1899-2:1998
Total Nitrogen	LVS EN ISO 11905-1:1998
Total Phosphorus	LVS 6878:2005 (part 7)
Metals	LVS ISO 17294-2:2016
pH	LVS EN ISO 10523:2012
Dissolved oxygen	LVS EN ISO 5814:2013
Turbidity	LVS EN ISO 7027-2002
Conductivity	LVS EN 27888:1993
Elements	LVS EN ISO 17294-2:2016

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sample and by the difference of the total carbon and inorganic carbon measurement. Each sample was tested in duplicate and the mean values were calculated ($CV \leq 2\%$).

Determination of concentration of Total Nitrogen (TN) and Total Phosphorus (TP) were performed with UV–Vis spectrophotometer M501 (Camspec, UK) after sample mineralisation.

Electrical conductivity (EC), pH, RedOx potential and dissolved oxygen were determined using Multi 340i SET B (WTW, Germany).

Turbidity was measured using the turbidimeter HACH 2100 P (HACH, USA).

Elements (boron (B), sodium (Na) and zinc (Zn)) concentrations were determined by Agilent 7700x ICP–MS (Inductively Coupled Plasma Mass Spectrometer, USA) using Nitric acid (69.0%, Trace Select) to preserve the samples.

Z–potential was determined using Zetasizer Nano ZS90 (Malvern, UK). The samples were taken for measurements of Z–potential at different pH level. Each sample was analysed at least 15 times and the arithmetic mean and standard deviation was calculated. Measurements were made at 20 °C. Analysis of control solutions was performed with each series of samples to evaluate the accuracy of the results.

Several repetitions of each sample ($n = 3$) were analysed to check the reproducibility of each method.

RESULTS AND DISCUSSION

In this study, the management scheme for wastewater from landfill of inorganic fiberglass was developed through various laboratory scale tests.

Field inspection

Landfill under study can be divided into old organic and intermediate inorganic wastes sectors. If the organic waste sector is fully managed, thus inorganic wastes are located on a field without drainage and treatment system.

About 80% of wastes in inorganic sector are fiberglass, and other 20% form rubber tires, plastic, metal, textile and organic wastes, including wood.

Sampling on the field was performed in 3 sampling points. Sample points can be characterised as water ditches or pits on different landfill sites, where storm water after contact with wastes on landfill was collected. Results of the analysis of wastewater samples collected on the field showed that pollution risk is high (Table 3).

Table 3. Results of chemical characterization of samples from the field

	1.	2.	3.
pH	10.2 ± 0.1	9.2 ± 0.1	8.84 ± 0.1
EC, $\mu\text{S cm}^{-1}$	2,100 ± 21	2,030 ± 40	1,783 ± 35
COD, mg L^{-1}	2,580 ± 258	153 ± 13	200 ± 19
BOD ₅ , mg L^{-1}	1,491 ± 218	40 ± 3	12 ± 1
TN, mg L^{-1}	149 ± 12	5.41 ± 0.43	17.7 ± 1.8
TP, mg L^{-1}	10.6 ± 0.5	0.24 ± 0.02	9.62 ± 0.98
B, mg L^{-1}	102 ± 10	46 ± 3	199 ± 16
Na, mg L^{-1}	13,452 ± 1,345	397 ± 27	2,694 ± 215

COD maximum was $2,580 \pm 258 \text{ mg L}^{-1}$, BOD₅ $1,491 \pm 218 \text{ mg L}^{-1}$, total nitrogen $149 \pm 12 \text{ mg L}^{-1}$ and total phosphorus $10.6 \pm 0.5 \text{ mg L}^{-1}$. Due to the fact that main industries using the landfill are linked with glass and fiberglass production, boron and

sodium concentrations were evaluated primarily (EPA Report 815–R–08–012, 2008). Presence of high amount of boron was determined $102 \pm 10 \text{ mg L}^{-1}$. With respect to the industry specifics (fiberglass processing), presence of elevated concentrations of other metals (especially Ni and Cr) might be regarded as an environmental risk a problem (Modin et al., 2011). According to the results, less than half of the organic substances are biodegradable (Table 3), indicating the need to combine various treatment methods, e.g. biological treatment with coagulation or other physicochemical methods.

The wastewater formed at the landfill can be compared with industrial wastewater that has strict management requirements. Difference between the results can be explained with the following remarks after field inspection: (I) the waste is located on an area without drainage system; (II) therefore, the collection of a representative sample was difficult. According to Brennan et al. (2017) inorganic waste on landfills are more biodegradable during the first 10 years.

After that, laboratory–scale experiments with collected fiberglass solid wastes for conventional coagulation and biodegradation tests have been performed.

Migration tests

The goal of the migration tests was to evaluate the pollution level from various fiberglass waste types. The results showed that the migration of contamination occurred already after 15 minutes, the peak was reached after 3 h and did not change significantly during the next 48 h. A tap water was used as a blank sample and a correction was made to calculate the contamination level for analysis (Table 4).

Table 4. Results of chemical characterization of samples after 24 h of contact time of fiberglass and water ($n = 1$)

Sample*	pH	EC, $\mu\text{S cm}^{-1}$	NTU	TOC, mg L^{-1}	B, mg L^{-1}	Na, mg L^{-1}	Zn, mg L^{-1}
Blank	7.39	451	0.88	7.4	0.003	9.60	0.025
1.	10.98	649	143	34.5	0.006	152.00	5.320
2.	6.94	514	61.2	25.8	0.243	10.30	0.077
3.	7.55	535	6.60	23.6	0.373	10.50	0.050
4.	7.55	504	16.9	26.7	0.101	9.76	0.033
5.	11.97	1,620	299	79.5	0.032	730.00	7.430
6.	7.56	544	16.7	25.6	0.584	20.40	0.214

*Sample identification according to Table 1.

Results of samples after 24 h contact of fiberglass and water showed that pH increased in the water and reached 10.98 and 11.97 in samples No. 1 and 5 after 24 h contact time. Turbidity level increased up to 143 and 299 NTU, respectively. The conductivity level increased to $1,620 \mu\text{S cm}^{-1}$ only in the solutions with sample No. 5. As a result of the migration process, a solution containing several elements is obtained (Table 4). Higher concentrations of boron were found in samples No. 6 (0.584 mg L^{-1}), 3 (0.373 mg L^{-1}) and 2 (0.243 mg L^{-1}), sodium – in sample No. 5 (20.40 mg L^{-1}), and zinc – in sample No. 5 (7.430 mg L^{-1}) and 1 (5.320 mg L^{-1}). According to results, there are two samples that have significant influence on the environment (No. 1 and 5). If it is not possible to sort the waste, homogenisation (e.g. mixing in equal amount) of all fiberglass waste is recommended for the treatment plant to obtain average leached wastewater from the field.

To determine the stability of the fiberglass solutions, measurements of Z-potential at different pH level were performed for average artificial wastewater solutions (Fig. 1).

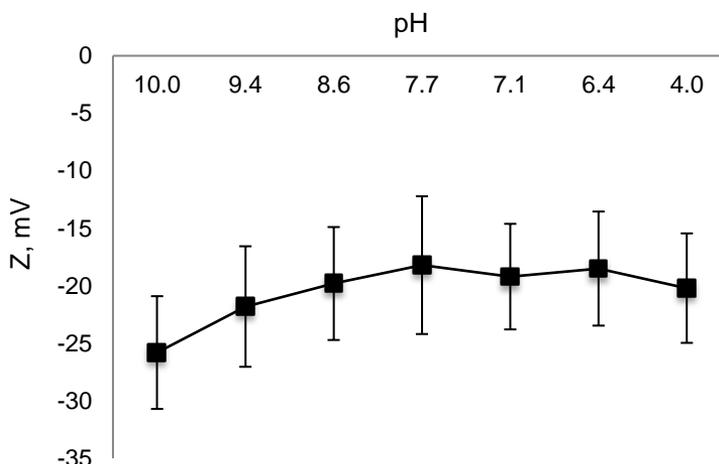


Figure 1. Changes of Z-potential in solution at different pH level.

Usually pH has a significant effect on Z-potential and indicates that coagulation process is active and agglomeration of contaminants and coagulant is performed. The results showed that the Z-potential ranged from -20.2 to -25.8 mV with pH of 3.99 to 10.02. This means that the solution contains a suspension whose balance does not change with changing environmental conditions, thus the optimisation of coagulation to increase efficiency by using the pH control is impossible.

Coagulation tests

To evaluate the potential of coagulation for removal of the fiberglass pollution, three coagulants were tested on average artificial wastewater solutions: aluminium sulphate; iron chloride and bentonite.

The results with aluminium sulphate showed that pH of the solution decreased from 5.17 to 4.45, but the EC of the solution increased from 686 to 828 $\mu\text{S cm}^{-1}$ with an increase in coagulant dose from 35 mg Al L⁻¹ to 60 mg Al L⁻¹, respectively. TOC removal was not effective (24%). The boron concentration in the initial solution was 3.50 ± 0.35 mg L⁻¹ and ranged from 2.90 ± 0.29 to 2.83 ± 0.28 mg L⁻¹ in samples after coagulation. Z-potential in the initial solution was -17.50 ± 5.08 mV and increased to 1.27 ± 4.33 mV with alum dose of 35 mg Al L⁻¹ and reached a maximum of 5.81 ± 3.03 mV at alum dose of 60 mg Al L⁻¹. The maximum removal of TOC concentration was only 30% after coagulation with alum dose 35 mg Al L⁻¹ and pH 5. After coagulation with 55 mg Al L⁻¹ coagulant doses, the maximum turbidity reduction (2.59 NTU) was observed (Fig. 2). No changes in element concentrations at different coagulant doses and pH level were observed.

Similar tendency was observed with iron coagulant. The pH decreased from 9.55 to 6.48 and the EC of the solutions increased from 685 to 1,030 $\mu\text{S cm}^{-1}$ with increase in coagulant dose from 35 mg Fe L⁻¹ to 60 mg Fe L⁻¹. TOC removal was not effective (only 20%). Removal of elements was not observed. Z-potential ranged from -7.72 ± 5.72 mV

in the initial solution to 2.75 ± 5.28 mV and 2.28 ± 3.92 mV at various Fe doses. The lowest turbidity was obtained at a coagulant dose of 60 mg Fe L^{-1} (4.68 NTU) and the same dose at pH 5 (8.91 NTU) (Fig. 2).

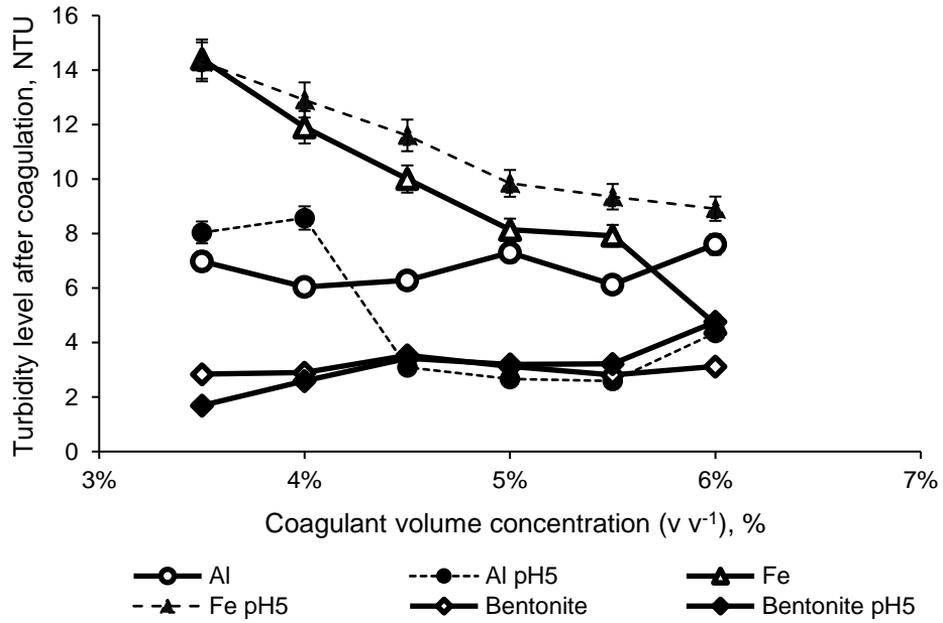


Figure 2. Changes of turbidity level during the coagulation process with alum, iron and bentonite coagulants (to compare process efficiency the volume concentration of coagulants was used in this graph).

Bentonite was chosen as natural and low cost coagulant. It demonstrated the maximum reduction of turbidity (97%) and TOC (71%) at the dose of $1,050 \text{ mg L}^{-1}$ and pH 5 (Fig. 2). Unfortunately, element concentrations in solutions before and after coagulation did not change (including coagulation with aluminium and iron) or slightly increased boron by 0.4% and sodium by 20% after coagulation with bentonite. This observation can be explained by the fact that bentonite coagulants are clays containing natural hydrated aluminium silicates, in which some magnesium and iron atoms replace aluminium and silicon atoms. Due to the inefficient coagulation process, overall concentration of elements in wastewater sample and added coagulant solutions can increase. At the same time bentonite coagulant showed significantly higher efficiency on the removal of organic and colloidal particles.

Approximately 90% of hydrophobic contaminants can be removed by coagulation (Hu et al., 2016; Bu et al., 2019) due to the solubility (hydrophobic is less soluble) and stronger interaction with metal coagulant (hydrophobic fractions form stable metal – organic complex) (Sharp et al., 2006; Ghernaout, 2014). Wastewater formed after fiberglass contact with water contains high concentrations of organic matter, metals and elements. Main component of fiberglass is the silicon that form complex compounds with metals, resulting in low efficiency of surface discharge (e.g. coagulation).

Biodegradation tests

The aim of the experiments was to estimate the degradation rate of BOD in pre-treated wastewater samples. BOD kinetic tests were performed with an untreated and 6 pre-treated samples after coagulation: (1) IN (pH 10.63); (2) 35 mg Al L⁻¹ (pH 6.75); (3) 35 mg Al L⁻¹ (pH 5.40); (4) 35 mg Fe L⁻¹ (pH 9.37); (5) 35 mg Fe L⁻¹ (pH 5.20); (6) 1,050 mg of Bentonite L⁻¹ (pH 10.35); (7) 1,050 mg of Bentonite L⁻¹ (pH 5.42).

Fig. 3 shows the removal efficiencies after pre-treatment with three different coagulant types at the best pH level.

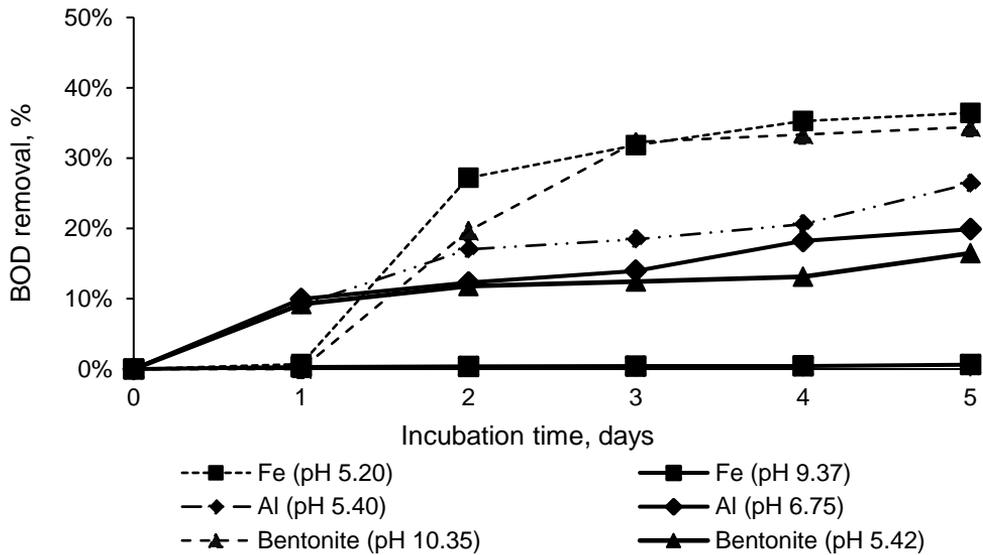


Figure 3. BOD removal through 5 incubation days for pre-treated samples after coagulation.

After the pre-treatment, among three coagulants, iron chloride at pH 5.20 showed the highest removal of BOD – 36%, the concentration reduced from 131 mg L⁻¹ to 83 mg L⁻¹. Similar result was observed with the bentonite coagulant at pH 10.35, 34% BOD removal was achieved. The aluminium sulphate coagulant showed the lowest BOD removal efficiency – 26%. The average BOD removal after coagulation process can be estimated at about 30% (not effective).

The results showed that the COD value in the initial (IN) fiberglass solution was 635 ± 64 mg L⁻¹ and the biodegradable fraction was only 20%. In the pre-treated samples after coagulation, the COD concentration decreased to 346 mg L⁻¹ with 35 mg Al L⁻¹, 279 mg L⁻¹ – with 35 mg Fe L⁻¹ and 299 mg L⁻¹ – with 1,050 mg Bentonite L⁻¹. The biodegradable fraction was 31%, 30% and 38% respectively.

The efficiency of biological treatment depends on the type of substrate (hydrophobicity/hydrophilicity, concentration of contaminants), the age of landfill and continuous biological activity (Costa et al., 2019). BOD can be calculated from the decrease of the dissolved oxygen concentration that is proportional to the initial biodegradable substrate concentration. (Pitman et al., 2015). Biological treatment

processes usually remove about 70–90% of BOD (Peng, 2017), thus, increasing water stability. To avoid bacterial regrowth after wastewater treatment, the threshold value for the BOD₅ is set to $\leq 25 \text{ mg L}^{-1}$, however, the effluent limit values for WWTPs with pollution load less than 2,000 population equivalent (PE) are not regulated by these requirements (Council Directive 91/271/EEC, 1991).

Taking into account the maximum process efficiencies, obtained during the laboratory tests, it can be concluded that by coagulation about 40% of organic compounds and 90% of colloidal particles can be removed. Element concentrations were not significantly affected.

Approximately 30% of organic contaminants can be removed by biodegradation (Table 5).

Existing rules (Council Directive 91/271/EEC, 1991) set that the BOD and COD should be reduced by 50–70% and 50–75%, respectively (for pollution load 200–2,000 PE). Laboratory experiments have shown that combination of chemical coagulation and biological purification can reduce organic matter and turbidity according to the rule requirements (Table 5). However, these methods were not effective to reduce metals concentration and boron. Therefore, further purification with activated carbon or biosorption technologies can be recommended.

Table 5. Efficiency of used methods

Coagulant	Coagulation stage	Biological treatment	Total efficiency of contaminant removal, %
Alum	46%	31%	77%
Iron	56%	30%	86%
Bentonite	53%	38%	91%

CONCLUSIONS

Management scheme, developed during this study is based on the field survey and laboratory scale tests and include two main steps:

1) Provide a waste sorting process and suitable drainage system for landfills that provide collection of one type wastewater in the receiving chamber (avoiding different consistency of wastewater mixing). The volume of the receiving chamber should provide well homogenisation of wastewater contaminants in this volume.

2) Plan for two-stage treatment: coagulation/sedimentation with bentonite and biological treatment (aerobic). The dose of coagulant (bentonite) is about $1,000 \text{ mg L}^{-1}$ without changing the pH of the water. Both active sludge reactors and biofilter can be used for biological purification, but effective separation of phosphorus and nitrogen must be ensured.

To provide efficient separation of metals and other specific chemicals, e.g., boron, post-treatment with filtration system (ex. activated carbon or biosorbent) are necessary.

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