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Alice Limoli

Sustainable landfilling of municipal solid waste

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Landfill of Guiri-Gui, Dominican Republic, picture by Cecilia Pradella

University of Trento
Doctoral School in Civil, Environmental and Mechanical Engineering
http://web.unitn.it/en/dricam
Via Mesiano 77, I-38123 Trento
Tel. +39 0461 282670 / 2611 - *dicamphd@unitn.it*

Abstract

The deposition of waste in a landfill can be a threat to the environment and human health; in spite of their potential pollution, landfill are still of grate use for the residual municipal solid waste, thus efficient and cost effective technologies need to be studied in order to minimize aqueous and gaseous emissions.

The present work focuses on the evaluation of the remediation of old landfill sites that pollutes groundwater and on the determination of a new pre-treatment of fresh waste upstream of landfilling. First the biosparging technology has been applied to remediate an aquifer polluted by leachate. The biosparging stimulates the growth of indigenous bacteria able to convert pollutants, such as ammonium nitrogen, in harmless compounds. The technology shows high efficiency in ammonium nitrogen removal via nitrification processes. The biosparging remediation technology prevents the mobilization of metals and removes the nitrates produced in the nitrification process when the organic carbon source is conveniently dosed. The application of the biosparging on site has proven to be feasible.

The Solidification/Stabilization (S/S) technology is a pre-landfill waste treatment process, which has been used for different types of hazardous wastes since it has a proved efficiency on heavy metal immobilization. The S/S process uses chemically reactive formulations that, together with the water, form stable solids; it also insolubilizes, immobilizes, encapsulates, destroys, sorbs, or otherwise interacts with selected waste components. The S/S process improves the physical characteristics of the waste and reduces the mobility of the hazardous compounds, thus the waste leaches less contaminants into the environment. The result of this process is a less hazardous solid. The experimental evidences proved that this technology reduces volumes used for landfilling and inhibits the methanogenesis blocking greenhouse gases emissions. The reduced permeability and the leaching test results show that the leachate produced is of a smaller amount and less polluted. The enhanced mechanical properties and the reduced emissions both in bodies of water and atmosphere have proven the worth of this technology. Therefore an alternative waste treatment plant involving S/S pre-treatment is proposed.

Keywords: sustainable landfill, biosparging, waste, solidification, stabilization

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Chapter 1

Introduction

1.1 Thesis outline and objectives

The present work aims at analyzing the current situation of municipal solid waste landfilling and at proposing new sustainable techniques able to reduce emissions into the environment.
The landfills have multiple harmful impacts and they affect mostly the environment and human health; these effects are stronger if there is a low level of containment or waste stabilization. A multidisciplinary study on illegal landfill has been performed with the support of specialists in medicine, law and sociology. This study proved that environmental and health impact can be seriously harmful; however, in case of problems of illegal landfilling the social mobilization lead the people to reach the awareness of the problem. The illegal landfills are forbidden and there is a need of sustainable technologies able to prevent harmful emissions. The illegal landfills show the highest potential for environmental pollution, however they are not the only sources of waste pollution. Regular landfills can show similar pollution levels depending on the design, the collection systems for leachate and biogas, and the isolation structures. Mostly in the past, but also in developing countries open dumps are built for waste disposal. This type of landfills pollutes the environment and causes the same health problems of illegal landfills, because of the lack of prevention systems.
The pollutants of the waste in aged MSW landfill leaches into groundwater and contaminate the aquifer. Among the liquid pollutant ammonia increases over time [1] while metals concentrations does not vary

significantly over time. Therefore a sustainable technology able to reduce these pollutants is needed. The technologies mostly used to contain the aquifer pollution are usually expensive and hard to perform. On-site cost effective techniques able to remove the pollutants from the aquifer are needed. Therefore the study went on to analyze the biosparging technology. The biosparging is a treatment performed by a series of injection wells that blow air or oxygen under the plume of contamination. Biosparging stimulate the growth of indigenous microorganisms that decompose the ammonia into nitrites and nitrates, which are subsequently converted into gaseous nitrogen via denitrification. Moreover, the process stimulates the immobilization of metals by oxidation and precipitation.
In order to prevent problems of pollution new models of landfills are necessary. Sustainable landfilling of municipal solid waste is aimed at reducing the long-term impacts by waste stabilization, namely green house gases emissions and leachate contamination. Mechanical biological treatment (MBT) plants are used as a pre-landfill treatment, as they enhance waste stabilization and reduce the volume occupied into the landfill. MBT reduces as well methane production but does not block it completelly. Therefore, a further step of solidification and stabilization, able to lower substantially the methanogenesis, is needed. The S/S of aerobically stabilized waste (fraction lower than 20mm) has been implemented and tested. This model has been called Trentino Sustainable Landfill (TSL) as it has been firstly tested in the autonomous province of Trento. The model can be described as a mechanical biological treatment coupled with a solidification stabilization (S/S) process, that uses slow quantities of cement and lime to obtain good physical properties, reduction of pollutant leaching and inhibition of biogas formation. The TSL model is also cost-effective and is able to reduce the volume occupied into the landfill, a crucial point as the landfill sites are gradually closing. This work will provide a whole understanding of the landfill problems, possible solutions for aged landfill pollution into groundwater and it proposes a new and sustainable model of waste treatment prior to landfilling able to prevent both liquid and gaseous emissions.

1.2 The problems of landfilling

The landfills can generate environmental pollution and problems to human health linked to the type of waste in the landfill body and the type of landfill in use. Hazardous waste such as radioactive and electronic waste should be treated separately and accurately because of their high content of contaminants such as radiation, pathogens and metals that are a significant danger to human health. Municipal solid waste can be rich in organic materials and metals that can leach into groundwater; moreover, organic matter causes greenhouse gases (GHGs) emissions into the environment. Systems of biogas and leachate collection and proper isolation of the landfill can help reducing the amount of pollution emitted. However, aged landfills can pollute the aquifer and spread GHGs causing harm to the environment and human health for a long time. The landfills of inert waste are less dangerous because of the waste characteristic: construction and demolition waste have a relatively low impact degree [2].

The municipal solid waste landfills are the object of the present study, the next paragraph will disclose properly the different types of landfills. Some models are nowadays obsolete, however aged landfill can present an obsolete structure and they keep on polluting the environment. The systems of pollution control (leachate and biogas capitation) do not prevent completely the pollution potential of landfills, as they are not always effective. New types of sustainable landfills prefer to treat the waste before the landfill in order to produce a more stable waste that will pollute less the environment.

As already stated, the pollution caused by landfill depends on the waste stored in the landfill and the technology used to prevent or reduce pollution. The main environmental impacts due to waste disposal can be summarized as:

- Groundwater quality degradation
- Eutrophication
- Acidification
- Phytotoxicity and aquatic toxicity
- Oxygen depletion in the root zone

- Animal toxicity and bioaccumulation
- Greenhouse effect and global warming

It appears clear that the pollution generated by waste landfilling affects the air, the bodies of water, the groundwater the soil the flora and the fauna.
Landfill pollutants have also harmful effects on human beings raising the risk of illness such as acute intoxication, cancer, infectious diseases, and respiratory and cardiovascular problems. Some pollutants show, as well, toxic effects on endocrine, reproductive and nervous system.
The local communities are also affected by: discomfort and change in habits, economical issues and aesthetic degradation of the landscape depending on the effectiveness of the landfill design.
The impacts of landfill pollution are widely disclosed in chapter 2, where they have been analyzed in the case of illegal landfills. Illegal landfills can be compared to open-dump sites as defined in the next paragraph. Other types of landfills show a lower pollution degree; however, the main impacts in case of dangerous uncontrolled emissions can be severe as well. Therefore, the impacts described for illegal landfills are a good indicator of potential impacts of regular landfills. Illegal dumps have a higher and more acute effect than legal landfill, thus the acute problems of regular sites are generally dangerous only for landfill workers that are provided with personal protective equipment, while in case of illegal landfills the whole local population can be affected.
The rising concern about the impacts of landfilling led to specific law regulations that prescribe duties and sanctions.

1.3 The landfill

The landfill has been defined by the International Waste Association as an “engineered deposit of waste onto and into land in such a way that pollution and harm to the environment is prevented and through restoration, land provided which may be used for another purpose” [3].
The waste management hierarchy is: prevention, reuse, recycling, waste-to-energy, and disposal [4]. Therefore, the landfill disposal is the least preferable option. The rising concern on GHGs emissions and global

warming, since the Kyoto protocol agreement, has led to find new chances of carbon sinks. The natural carbon sinks, such as oceans and forests, are starting closing up [5], thus the emissions of GHGs should be lowered. Approximately one third of the GHGs generated by the human beings in the European union comes from municipal solid waste landfills [6]. In case of waste stabilization and/or for some fraction of waste the landfill disposal could be identify as an artificial carbon sink able to store part of the potential GHGs emissions [7–10].
The landfill Directive (1999/31/EC) defines the types of landfill disposal according to the waste stored at the site; the options are:

- Hazardous waste landfills
- Municipal solid waste landfills
- Inert waste landfills.

This work focuses municipal solid waste (MSW) landfills. Both the treatment for aquifer pollution removal and the TSL model are studied in the case of MSW landfills.

1.3.1 Municipal solid waste

Municipal solid wastes are defined by Eurostat as the waste fraction collected by municipality, concerning waste from offices, houses, institutions and small businesses; MSW produced per capita in all countries of Europe are shown in figure 1.1.

In Italy MSW produced per capita in 2017 is about 489 kg; this value is slightly lower than the same parameter in 2005 and aligned to the one for Europe (487 kg per capita in 2017). This means that the production of waste is decreasing over time. However the countries show different trends (at least 19 out of 31 states increased MSW production); thus, a regulation aimed at decreasing waste production and practices for end-of-waste strategies should be introduced.

The differences between per capita waste productions in the European countries are due to variations in consumption patterns and economic wealth, but also it depend on municipal waste management and collection strategies.
Figure 1.2 shows the economic activities and household that produce total

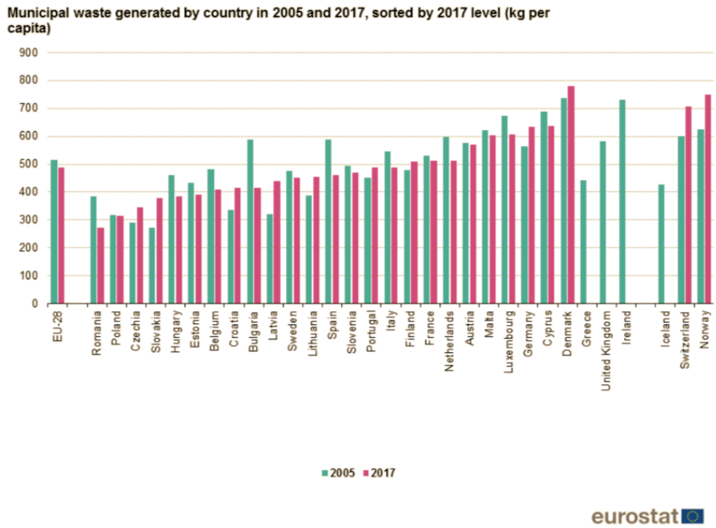

Figure 1.1: Waste generation in Europe 2016, data source Eurostat [11]

waste in Europe. The major part is given by construction/demolition processes and mining activities. Manufacturing produce more than the 10% of the waste and households about 8.5%. The municipal solid waste fraction is all the non-hazardous non-inert solid waste. The majority of the waste produced is disposed in inert waste landfills that need a lower degree of stabilization and lower costs due to pollution prevention. This type of waste will not be analyzed in this study because of their low pollution potential. The focus will be on municipal solid waste, thus the following data are linked to MSW collection and fate.

To date the fate of the municipal solid waste in Europe is shown in figure 1.3 approximately one fourth of the waste produced in Europe is disposed into landfills. The category of "other treatments" has been estimated as the difference between the sum of the amounts treated and the amounts of waste generated. The difference should be equal to zero; however, in countries that need to estimate waste generation in areas not covered by a municipal waste collection system, it is possible to have pockets of untreated waste. Therefore, in these states the generation of waste can be higher than the waste treatment. Moreover, part of the total mass of the waste can be lost during the transportation process and through other processes such as mechanical biological treatment. MBT reduces the total mass mostly because of the evaporation of water

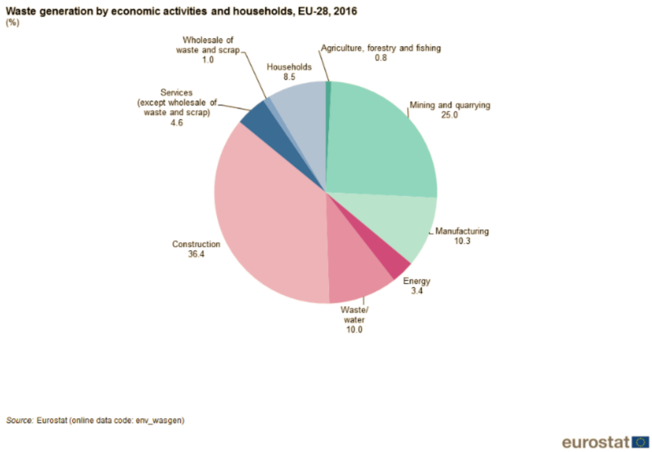

Figure 1.2: MSW per capita in Europe, data source Eurostat [11]

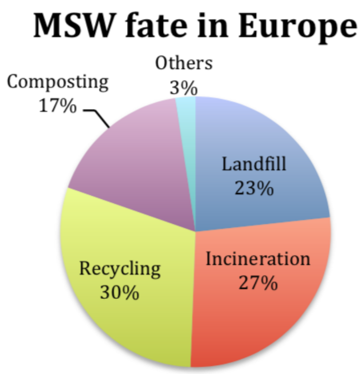

Figure 1.3: MSW fate in Europe 2017, data source Eurostat [11]

during the biostabilization process.
The trend of waste treatment over the last twenty years (Figure 1.4) shows a decreasing use of landfills and an increase in waste recycling and incineration. The MSW landfilling has decreased from 64% in 1995 to 23% in 2017.
This significant reduction of landfilled waste can be attributed mainly to two European regulations.
The Directive 62/1994, that introduced a threshold for packaging recycling; the minimum waste to recover was 50% until 2007 and in the following years this threshold has been raised to 60%. Moreover, the Directive 31/1999 set a threshold for biodegradable waste to be disposes into the landfill. This directive led to a more intensive recycling of

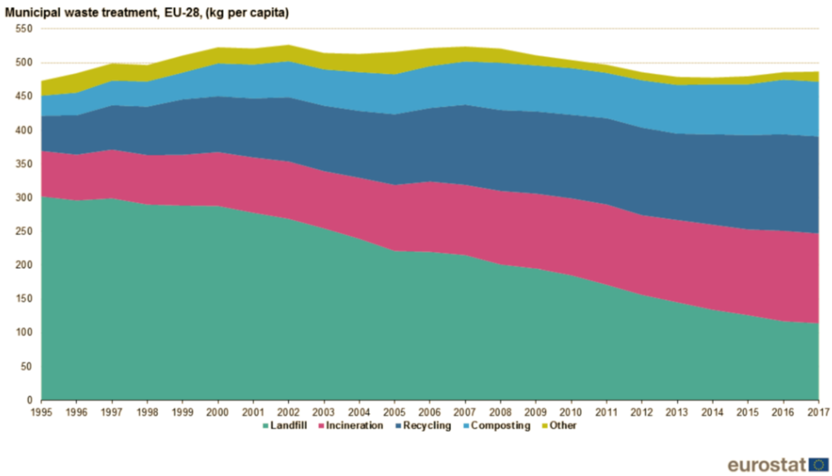

Figure 1.4: Trend of MSW fate in Europe, data source Eurostat [11]

organic waste with subsequent composting and to the introduction of pre-landfill treatments.
Moreover, individual countries and regions prepared strategies for progressive landfill closure. However, landfilling is a useful option needed as well for disposal of incineration fly ashes and wastewater plant solid residues, thus it is hard to plan the complete closure of landfills.
Eurostat collected and published data from the member countries and some of the candidate countries from 1995. The main source of data and information is the OECD/Eurostat-Joint Questionnaire [11].

1.3.2 History of landfilling

Since the human started to produce waste the main path for disposal were waste burning and waste landfilling [12]. In the past waste landfilling has been the easiest and cheapest waste treatment option. In recent years the landfilling of waste changed according to new disposal concepts and environmental targets [13]. Landfills have developed from open dumps to modern highly engineered facilities involving monitoring systems and enhanced control measures for GHGs and leachate. The first landfills built were open dumps, a design that can cause severe pollution problems. Therefore since the 70s the research aimed at finding pre-treatment and/or containment technologies [14].

1.3.2.1 Open dumps

Open dumps are unconfined sites without any means of leachate or GHGs collection systems and covers. The principle driving this disposal technology is the dilution and attenuation of pollutants. The open dumps are common in rural undeveloped areas because of the simple construction characteristics, which do not need trained manpower, and the low costs [15,16]. The dilution and attenuation process is achieved by allowing the water (rain, surface and groundwater) to pass through the landfill and dilute the leachate, thus the open dumps are environmentally risky [17]. The harmful effects of open dumps are severe and well known [16,18–20], the impacts are comparable to the ones of illegal landfills described in chapter 2.

1.3.2.2 Entombment/containment landfills

The containment landfill is a more sophisticated and engineered method than the open dump that allow an extremely low amount of pollutants to be released into the environment [15]. The principle of containment landfill is that the leachate and the biogas should not leave the landfill area; physical barriers and collection systems are installed for this purpose. Pollutants are retained into the landfill body and are slowly degraded [17]. Entombment is a similar method of landfill design, however the driving concept is that no liquids are allowed to infiltrate the waste in order to block biological processes. This technique is intended to isolate the waste from the environment by means of plastic sheets and compacted soil maintaining the waste unchanged over time.
Both the technologies of entombment and containment require effective management and are expensive to build and manage [17]. The entombment inhibits waste degradation and leaves the waste in potentially polluting conditions for several decades. This has resulted in low rates of waste degradation, thus the complete stabilization time reaches hundreds of years [21]. Moreover the performance of both containment and entombment systems decrease over time as the plastic sheet can brake and clay liners can interact with leachate pollutants

leading to a reduction in swelling capacity and an increase in hydraulic conductivity [22]. These types of landfills can show a high pollution rate after closure, freeing leachate and biogas because of the deterioration of isolation methods.

1.3.2.3 Sustainable landfills

The target of sustainability is an environmental friendly and equal development. Allen in 2001 pointed out that a sustainable landfill "should encompass the following basic principles:

- Reduction in the generation of waste
- Waste streaming at source
- Recycling and reuse
- Pre-treatment of waste to minimize quantity and volume." [22]

The collection and recycling systems play a fundamental role in sustainable waste landfilling. The project of a new sustainable model (TSL) has been applied at a situation that involves a proper waste management able to minimize the influent fluxes.
To date, sustainable landfill technologies involve pre-treatment such as mechanical biological treatment or the design of landfills working as bioreactors [15,21,23,24]. Studies have been conducted on the definition of proper sites for sustainable landfills in order to minimize environmental and health impacts [25,26]. However, the pre- treatment and evaluation of proper sites are not effective in complete stop of methanogenesis and the leachate produced is still strongly polluted [6]. The solidification/stabilization pre-treatment fulfill the objective disclosed by Allen [22]. Therefore, this work focuses on the comparison of different pre-landfill treatment scenarios to enhance waste stability and volume reduction. The best performing pre-treatment is the Mechanical Biological Treatment (MBT) coupled with solidification and stabilization (S/S) technology, this treatment scheme is defined as Trentino sustainable landfill (TSL) model.

1.3.3 Regulations

The rising concern about the impacts of landfilling led to specific law regulations that prescribe duties and sanctions. The General Framework Directive 2008/99/EC has been adopted in 2008 by the Council and the European Parliament [27]. This directive aims at:

- Reducing waste production
- Recycling and using the waste as a resource
- Minimizing environmental and health impacts.

The General Framework Directive imposes, on member states, to establish local waste prevention programs. It defines the principle that who pollute must pay and extends the responsibilities of waste prevention to the producers [28].
The Landfill Directive (1999/31/EC) aims at reducing and preventing harmful effects of landfilling on the environment end human health. Both the Landfill Directive and the Water Framework Directive (2000/60/EC) push for the protection of bodies of water and groundwater preventing leachate contamination [29].
The Waste Shipment Regulation (1013/2006) aims at supervising and controlling shipments of waste in order to avoid illegal trafficking. The regulation applies to all international waste shipments into or out of the European Union.
Italy has implemented the European Directive 2000/60/EC by a Legislative Decree 152/2006. The 6th section of the Italian decree defines the crimes against the environment and subjects the polluters to sanctions. The 4th section, namely norms concerning waste management and protection of contaminated sites, sets the proper parameters and virtuous behavior that should be respected in solid waste management and pollution prevention [30].
Waste management is a public interest activity, as the environment is a constitutional value set by articles 9 and 32 of the Italian Constitution.
Waste management is established by:

- Regulations that govern proper waste management
- Regulations that punish irregular waste management by means of sanctions.

The Italian law defines specific administrative authorizations to conduct a proper waste management, which are granted if certain requirements and procedures are respected.
Waste management activities involve: collection, transport, treatments, disposal, aftercare of treatment sites and intermediary activities.
Waste management activities should be traceable; they must not be a threat to the human beings and the environment; and they must respect the principles of: precautionarity, sustainability, proportionality, accountability and cooperation between the stakeholders. Therefore the waste management should be efficient, effective, transparent and cost-effective.
The Italian law imposes sanctions to punish irregular waste management, namely:

- Uncontrolled disposal of solid wastes on lands and bodies of water (articles 192-255 Legislative Decree 152/2006)
- Illegal landfill (article 256 Legislative Decree 152/2006)
- Illegal incineration of waste (article 256 bis Legislative Decree152/2006 introduced by Legislative Decree 10.12.2013 and modified by Law 6/2014)
- Unauthorized waste management such as refuse collection, transport, trade, intermediation and reuse (article 256 Legislative Decree 152/2006)
- Illegal trafficking of waste (article 260 Legislative Decree 152/2006)
- Behaviors and actions that lead to loss of waste traceability (article 258 Legislative Decree 152/2006)

The sanctions and duties prescribed by law against the crime of illegal landfilling and uncontrolled waste disposal are:

- Criminal or administrative sanctions to the responsible for these illegal actions (depending on the seriousness of the action)
- Duty of environmental remediation of the contaminated site
- Compensations to the aggrieved parties (the environment and/or the people affected by the illicit actions)

- Confiscation of the illegal landfill site (if owned by the responsible of the illicit actions) and of the means of transportation (in case of illegal waste trade and/or transport of abusively incinerated waste)

- Administrative sanctions prescribed by the Legislative Decree 231/2001 if the crime is committed in favor of a legal person by a person covering a senior position (administrative process against the guilty company).

Supplemental penalties are prescribed as the landfill can damage the environment affecting the soil, the bodies of water, the groundwater and the public health. The offences, in this case, can be more severe; the Law n. 86 22/05/2015 define and condemn the environmental pollution (article 452 bis), the death or injuries due to the environmental pollution (article 452 ter), and the environmental disaster punishable even without express malice (article 452 quater).
The Legislative Decree 152/2006 extends the duty of environmental remediation to the owner of the land even in case of unawareness.
Regulations settle as well good practices for landfill management. The Landfill Directive (1999/31/EC) settles its field of application to solid waste disposal and defines the procedures for waste acceptance. Moreover guidance on landfill gas control defines the best biogas collection practices.
The United States Environmental Protection Agency (EPA) in 2000 approved and promulgated the implementation alternative liner performance, leachate recirculation, and bioreactor landfills in order to encourage the use of new technologies in landfill building.
The European "landfill directive" defines the procedure and the permit for landfills building. The permit application should include an environmental risks assessment as defined by Council Directive 85/337/EEC.
The regions have the power to define guidelines for landfill construction. The guidelines are helpful and disclose all the necessary steps to follow during the construction of a landfill.
Moreover, the Council Decision of 19 December 2002 established criteria and procedures for the waste acceptance at landfills sites, stating that the limits values of leaching test "apply to granular hazardous waste acceptable at landfills for non-hazardous waste, calculated at L/S = 2

and 10 l/kg for total release [...]. Granular wastes include all wastes that are not monolithic. Member States shall determine which of the test methods and corresponding limit values should be used." The law limits are reported in the landfill decision 2003/33/EC Annex "Criteria and procedures for the acceptance of waste at landfills."

1.4 Landfill gas

Landfilled organic waste, degraded in an anaerobic environment, such as the inner body of the landfill, generates GHGs. Without pre- landfill treatments the released biogas can highly enhance global worming [31].
In Europe, landfill owners have to report accurate data on annual methane emissions; direct measurements of biogas emissions are important for the evaluation and improvement of the collection systems eventually implemented. Moreover the measurements can be useful in the quantification of biogas migration [32]. The estimation of the surface emission rate of GHGs from landfills is very hard to perform, because of the high number of factors that affect the biogas formation and migration into the landfill body: i.e. meteorological factors can enhance lateral migration [33,34]. Fugitive GHGs emissions from landfill sites are significant sources of global warming and pockets of methane gas can lead to explosions [33].
Landfill gas emissions should be trapped and extracted from the landfill body even during the landfill operation. The gas extraction pipes must be a separate system, not combined with leachate extraction pipes to prevent clogging problems [35]. The landfill gas can be used as a source of energy, producing power and/or heat; however, direct utilization is not possible as the gas shows various impurities and thus it must follow a process of cleaning and amelioration [36]. After the closure of a landfill, the biogas generation gradually decreases, thus the use of the methane is more and more difficult. Although the amount of methane produced every year decreases, the generation may continue decades after the closure and thus generate notable cumulative GHG emissions in the long term [31].
As biogas collection and use becomes more difficult and the rate of produced methane decreases, systems of waste stabilization are needed.

The waste stabilization on site can be reached by means of landfill aeration or leachate recirculation, namely landfill bioreactors [37–39]. Hybrid Bioreactor stimulates an early semi-aerobic phase able to enhance the methane production during the subsequent anaerobic phase, thus the methanogenesis is accelerated and the release of biogas is faster. Hybrid bioreactor induces a forced aeration step aimed at reducing the residual gaseous emissions [40]. At the end of the anaerobic step, semi-aerobic conditions are restored and flushing applied for leaching residual non-biodegradable compounds. Moreover various systems of landfill capping can be used to prevent long-term emissions, however these systems must be controlled and maintained [41]. The best practice to apply is the stabilization of waste before landfill disposal. The BMT has been tested and its capacity of waste stabilization has been evaluated. The BMT has been proven to be effective in the reduction of biogas production, however the stabilization is not complete (still some biogas is produced in the landfill body) and the stabilization depends on the duration of the biological treatment [6]. A longer time of biostabilization reduces the landfill gas produced during the landfill lifetime, but it increases treatment costs, because of the energy consumed to blow air into the waste.

1.5 Leachate

1.5.1 Formation, characteristics and evolution

Leachate is a liquid produced as water, such as rain or surface water, pass through a landfill, transferring pollutants from the solid waste to the liquid phase. Leachate carries pollutants into groundwater, representing a risk to adjacent ecosystems and human health. Aged landfills are the most affected by leachate production and penetration into groundwater, as the waterproofing is weak and the geomembranes are often broken or even missing in case of open dumps.

The leachate pollutants can be categorized into four groups: dissolved organic matter, inorganic macropollutants, heavy metals, and xenobiotic organic compounds [42]. The major pollutants in landfill leachate are usually represented by the basic parameters Chemical Oxygen Demand (COD), Biochemical Oxygen Demand (BOD), BOD/COD ratio, pH, Total Suspended Solids (TSS), Total Ammonium Nitrogen (TAN), Total Kjeldahl Nitrogen (TKN) and metals. Leachate characteristics depend on the landfill age, the net infiltration of water into the landfill body, the composition and type of collected waste [43].

The volumetric flow rate of the leachate influences the pollutant

Table 1.1: Typical contaminants in landfill leachate [44]

Parameter	**Average value**	**UM**
COD	5000	mg/l
BOD	250	mg/l
TAN	2000	mg/l
pH	8	-
Total phosphate	15	mg/l
TSS	250	mg/l
Chloride	1400	mg/l
Sulfate	200	mg/l
Alkalinity	14000	mg/l

concentration as well, as a higher flow rate dilutes the pollutant in the leachate [43]. A. Barmi, and A. Bennett reported typical contaminants data for landfill leachate [44]. The values for a raw leachate are shown in Table 1.1 .

The COD and BOD are good indexes of organic pollution, as proven by D. Kulikowska and E. Klimiuk, the COD in leachate decreases over time. On the other hand, the same study reports that ammonium nitrogen concentration increases over time: ammonia increased to 370% of the initial level in four years. The other pollutants were more affected by the seasonal variations than by the landfill age [1].

1.5.2 Leachate treatment

The leachate treatments can be distinguished between the type of treatment (physical, chemical, biological or combinations of those) and the localization of the treatments (in situ or ex situ) [43]. The leachate can be treated in the landfill body by means of technologies such as recirculation or landfill body aeration, it can be pumped and treated on site or it can be pumped and treated off site.

As the leachate pollutes the aquifer, large amounts of contaminated water must be treated or contained.
The plume of contamination can be stopped by means of hydraulic barriers. Hydraulic barriers are hard to design and implement, as the plume of contamination is usually wide and the barriers are costly to operate and sometimes not completely effective, because of the heterogeneity of the aquifer. Moreover, this technology is not able to reduce and treat the contamination plume but it transfers the contaminated groundwater to an external treatment plant, with further costs and residues to manage.
The treatment of contaminated groundwater in on-site or off-site treatment plants become more and more expensive as the amount of groundwater pumped out increases. The on-site/off-site treatment applied can be physical, chemical, biological or a combination of those.
As far as ammonia removal is concerned either biological (with external carbon addition) or physical-chemical processes (stripping process or chemical precipitation, namely struvite precipitation) can be applied as on-site/off-site treatment solution. The stripping process can reach efficiencies of ammonia removal up to 90% in one-day treatment with a flow rate of 5 L/min, at room temperature (20°C) and with the addition of 10000 mg/L of calcium hydroxide [45]. D. Kim et al reported nearly 65% of ammonia removal by means of struvite precipitation on average; the efficiency depends on experimental conditions and can reach 90% under optimal conditions [46]. Physical-chemical treatments reach high efficiencies of ammonia removal, although the cost of chemicals makes these treatments economically unsustainable. Moreover, low or none organic matter reduction is reported.

The leachate can also be transferred and treated in a domestic wastewater treatment plant. The wastewater treatment plant is usually located in a separate area. Therefore, this treatment needs a proper transport of the leachate in order to avoid spills. The transport introduces a further treatment cost. Another problem with this technique is the inhibitory effect of some leachate compounds on activated sludge; therefore many studies up to now identify the best sludge-to-leachate ratio [47–49]. Applying a sludge-to-leachate ratio of 9 F. Cecen and O. Aktas obtained a nitrogen removal of above 50% [47].
Biological treatments are quite effective in treating organics and

ammonium nitrogen. Moreover they are cost-effective and easy to manage. Among the biological treatments there are activated sludge, biofilters, Moving-Bed Biofilm Reactors (MBBR), Anammox and partial nitritation (SNAP), and constructed wetlands. Activated sludge treatment proved to be unsuitable to treat leachate without addition of domestic wastewater [47]. Thus, this method is applied after leachate transportation at domestic wastewater treatment plants. MBBR with an anaerobic/aerobic configuration removed 97% of the ammonium nitrogen by applying a Hydraulic Retention Time (HRT) higher than 1.25 days to the aerobic stage [50]. SNAP reached an ammonium conversion efficiency of 98% under the condition of Dissolved Oxygen (DO) 1.0 mg/L, HRT 12 h, pH 7.5– 7.8, and loading rate of 1.2 kg-N/(m^3 d) [51]. Constructed Wetlands (CWs) are a simple and cost-effective technology that can be applied to remove ammonium nitrogen. CWs with Horizontal Subsurface Flow reached 38.7% of average ammonia-removal efficiency [52]. Vertical Flow Constructed Wetlands (VF-CW) achieved complete ammonia removal in a five-day treatment at the lower flow rate tested (40 mL/min) and recirculation ratio of 1:3 [53]. However CWs need a huge surface to treat the leachate and this aspect is important in urban areas. Moreover, the leachate can damage the plants of the wetlands.
Biofilters reached a total ammonium nitrogen reduction of 89% by loading the biofilter, packed with aged refuse, with 20 L/(m^3 d) of leachate [54]. This technology could be applied directly in the landfill body, however the leachate must be pumped and recirculated.
The biosparging technology will be plenty disclosed in chapter 3. This is an in-situ treatment option that is able to remove ammonia and eventual organic pollution. It also reduces the metals present in the liquid phase. This technology is able to treat wide plume of contaminations.

1.6 References

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Table 1.2: Landfill leachate treatment options

Treatment	**Ammonia reduction**	**Organics reduction**	**Advantages and Drawbacks**	**References**
Combined treatment with domestic sewage SBR	50%	95% BOD5	-Inhibitory effect of some leachate compounds Transportation	[55]
Landfill aeration	NA	40% BOD5	-Energy consumption +In situ technology	[39]
Biofilters	89%	80% COD	+Low cost	[54]
Biocovers	-	-	+Low cost	[56]
Constructed wetlands	51%	50% COD	-Long treatment time +Low cost	[57]
MBBR	97%	91% COD	-Energy consumption	[50]
Ammonia stripping	80%	36% COD	-ReagentsEnergy consumption	[45]
Ozonation	25%	40% COD	-Reagents consumption	[58]
Fenton oxidation	88%	55% COD	-Reagents consumption -High chloride and sulfates concentration left	[54]
Coagulation–flocculation	-	79% COD	-Reagents consumption -Energy consumption	[59]
Chemical precipitation	98%	-	-Reagents	[60]
Adsorption	78%	49% COD	-Pollutant transfer Adsorbent cost and disposal	[61]
Reverse osmosis	99%	99% COD	-Not able to treat the whole leachate -Membrane fouling	[62]

Chapter 2

Illegal landfills in Italy (EU)– a multidisciplinary approach

This Chapter is a review on harmful effects of improper waste landfilling; such problems are amplified by illegal behaviour. The sustainable landfill aims at reducing the harmful effects of waste deposition that are extensively described in the following article.

Illegal landfill in Italy (EU)– a multidisciplinary approach

A. Limoli, E. Garzia, A. De Pretto, C. De Muri
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2.0 Abstract

Illegal dumping of solid waste is a matter of recent concern because of its numerous impacts. The regulatory framework of the European Union and Italy is disclosed. Degradation of air, bodies of water, groundwater and soil can occur in the illegal landfill and surrounding sites causing e.g. acidification, eutrophication global warming and photochemical smog. Animals and plants are subjected to landfill borne pollutants that can damage them seriously. Landfill pollutants have harmful effects on human beings raising the risk of illness such as cancer and cardiorespiratory disease. Finally illegal landfilling practices lead to economical, aesthetic impacts and sociological discomfort that leads to multiple collective actions, blockages, and

information campaigns, rising sense of belonging.

Keywords: Illegal dump, environmental impact, social impact, health problems, regulatory framework.

2.1 Introduction

The illegal dumping is the illicit action of uncontrolled discharge of waste into the environment. The illegal dumping can be restricted to a small amount, produced by a single illicit event (uncontrolled disposal of waste), or it can be caused by repeatedly taken actions, covering larger-scale sites, namely illegal landfills. The environmental, health and social impacts of illegal landfilling are numerous and severe and they are even higher in case of uncontrolled combustion of the dumped waste. Therefore a multidisciplinary approach is needed to study this phenomenon.
The harmful effects of illegal landfills are matter of recent concern as they affect the environment and the people safety. The rising concern at impacts of illegal landfilling led the research to analyze the problem and its possible solutions [1]–[3]; however multidisciplinary studies on the topic involve just the relation between environmental pollution and human health [4], [5] with low concern for sociological economical issues and regulatory framework.
Many efforts have been recently spent on the identification and the localization of illegal landfill sites by means of software and geolaocalization [6]–[11].
This paper fit well into the topic of forensic engineering since the forensics science is defined as a multidisciplinary area that applies scientific methods for the purposes of law [12]. The novelty of this study resides on the fact that the effects of illegal landfills will be globally evaluated and identified for different topics of study. The paper aims at defining and reviewing the potential impacts of illegal dumping on the environment, the public health and the society. Moreover the regulatory framework of the European Union and Italy will be discussed.

2.2 Regulatory framework

In this section a brief description of the European Regulations and the Italian Law concerning waste management and disposal will be given. The study focuses on the uncontrolled illicit disposal of solid waste and illegal landfills.

2.2.1 European Union

The General Framework Directive 2008/99/EC was adopted in 2008 by the Council and the European Parliament [13]. The directive aims at reducing waste production, recycling and using the waste as a resource to avoid or postpone landfill disposal. Another key goal is to minimize environmental and health impacts of waste.
The General Framework Directive imposes, on Member States, to establish local

waste prevention programs. It defines the principle that those who pollute must pay and extends the responsibilities of waste prevention to the producers [14].
The Landfill Directive (1999/31/EC) aims at reducing and preventing harmful effects of landfilling on the environment end human health. Both the Landfill Directive and the Water Framework Directive (2000/60/EC) push for the protection of bodies of water and groundwater preventing leachate contamination [15].
The European Union regulates, as well, the closure and the rehabilitation of illegal landfills, since the uncontrolled discharge of large amounts of waste represents a serious risk for the human health and the environment [16].
The Waste Shipment Regulation (1013/2006) aims at supervising and controlling shipments of waste in order to avoid illegal trafficking. The regulation applies to all international waste shipments into or out of the European Union.

2.2.2 Italy

Italy has implemented the European Directive 2000/60/EC by a Legislative Decree 152/2006. The 6th section of the Italian decree defines the crimes against the environment and subjects the polluters to sanctions. The 4th section, namely norms concerning waste management and protection of contaminated sites, sets the proper parameters and virtuous behavior that should be respected in solid waste management and pollution prevention [17].

Table 2.1: Regulatory framework in Italy

Constitution	Art 9 and 32	The environment establishes life quality for the community and for every individual. The environmental damage is the modification, the deterioration, the destruction of the Human Habitat caused by the violations of ordinary laws and regulations.
Ordinary laws	Legislative decree n.152/2006, Law n. 86 /2015, Legislative Decree n. 231/2001, Criminal code	Govern proper waste management (collection, transport, treatments, Disposal, Aftercare of treatments sites). Punish irregular waste management.
Regulations	Decisions of the Regional Councils	The State and the Regions specify the general principles established in ordinary laws (limits, methodologies).

The table provides the pyramid structure of the legal powers related to waste management in Italy. The purpose of that law system is to guarantee that the waste management should be efficient effective transparent and cost- effective according to the principles of Italian Constitution.

2.3 Environmental impacts

The uncontrolled disposal of waste generates fluxes of contaminants in the adjacent areas. The width of the contaminated area depends on the hydrology of the site and on the type of pollutant. The soluble pollutants are more likely to be carried into groundwater and therefore they spread out into the environment easily. The gaseous pollutants, generated from illegal combustion and from degradation processes in the uncontrolled landfill, spread out and are carried by the wind to the surrounding areas. In this study it is hypothesized that pollutants detected in regular landfill are equivalent to the ones emitted by regular landfill and the dose exposure is higher because of the absence of barriers and emissions treatment plants.

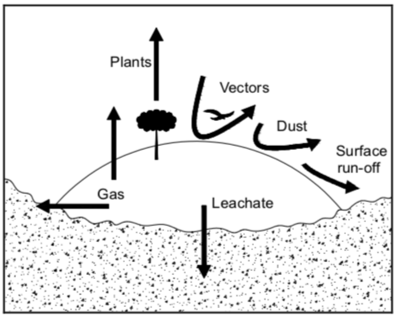

Figure 2.1: Illegal landfill: fluxes of emissions

Figure 2.1 shows the fluxes of contamination from an illegal landfill. The liquid emissions are generated by surface run-off and leachate percolation; these emissions compromise the quality of groundwater and body of water. The harmful effects on bodies of water are described for each pollutant in the following paragraphs. The main effects are: acidification, eutrophication and oxygen depletion, moreover the organoleptic qualities of groundwater can be compromised.
The gaseous emissions spread out in the atmosphere or infiltrate into the ground, leading to global warming, acidification, photochemical smog and formation of ground level ozone. Persistent pollutants bioaccumulate in animals and crops, however this kind of pollutants have usually higher impact on the health of the people and other living beings than on the whole environment.
The emissions generated by hazardous waste will be described separately.

2.3.1 Liquid emissions

The water (usually rain), that contacts the refuse, percolates into ground or flows on the surface of the landfill. Soluble pollutants are leached and move from the solid phase to the liquid one. The leachate can reach the groundwater carrying the pollutants and compromising the quality of the fresh water. Some pollutants show other severe impacts to the environment: a brief description of each substance and

their effects are discussed in the following paragraphs.
The site hydrology plays a big role in leachate production and groundwater pollution. Regular landfills are located in naturally impermeable sites or have a system of waterproofing in order to reduce this risk of contamination [18], however illegal landfills are situated in any kind of site and have no waterproofing systems. The risk of contamination and fast diffusion of the pollutant is very high.
The liquid emissions affect negatively the environment reducing groundwater quality; causing eutrophication and acidification. Liquid pollutants can be toxic to animals and plants and, in some cases; the risk of bioaccumulation and biomagnification can be high.

2.3.1.1 Ammonium nitrogen

Ammonium nitrogen does not decrease with the landfill age and therefore can be considered as a long-term pollutant [19], ammonium nitrogen is a nutrient that can cause eutrophication, as the algae find a favorable environment for growing and the massive bloom of algae causes oxygen depletion of the body of water.
High amounts of free ammonia cause soil and water acidification [20]. Ammonium nitrogen is also toxic to aquatic organisms.

2.3.1.2 Chloride

The chloride modifies the reproduction rate of aquatic organisms and plants. When the leachate percolate into groundwater the quality of the drinking water is compromised [21].

2.3.1.3 Organic matter

As the waste decomposes, the soluble organic matter is leached by the water, which comes into contact with the refuse. The organic matter can be quantified as Chemical Oxygen Demand (COD) or Total Organic Carbon (TOC), volatile fatty acids and more refractory compounds [19].
The degradation of the soluble organic matter requires large amounts of oxygen. This phenomenon causes oxygen depletion in the root zone of the ground compromising the flora wellbeing [22].
The soluble organic matter is rapidly degraded and this kind of pollution decrease in aged landfills.

2.3.1.4 Persistent Organic Pollutants (POP)

Persistent organic pollutants (POPs) are found in leachates from municipal landfill [23]; their effects on the environment are linked to the animal toxicity and the bioaccumulation. These substances accumulate in the food chain and high concentration of them can be toxic to animals. The POPs are rapidly transported by waterbodies, reaching faraway areas [24].

2.3.1.5 Sulfates

The sulfates enhance the release of nutrients in waterbodies that lead to eutrophication; moreover the sulfates stimulate the production of methylmercury by bacteria. The methylmercury is the most bioaccumulative and toxic form of mercury [25].

2.3.2 Gaseous emissions

A distinction should be done, in this section, between the gaseous pollutants of an illegal landfill and the gaseous pollutants generated by the uncontrolled combustion of waste.
The landfill generates greenhouse gases such as carbon dioxide, nitrous oxide and methane from anaerobic decomposition processes [26]. The wind can enhance the volatilization of dust (particulate matter).
The open burning of waste is a matter of concern to the environment. It causes the release in the atmosphere of several pollutants and products of incomplete combustion [27]. The emissions produced by MSW combustion are mainly sulfur oxides, nitrogen oxides, persistent organic pollutants, carbon monoxide, polycyclic aromatic hydrocarbons, Particulate Matter (PM) and greenhouse gases [28]. Several substances are freed in the burning process; the following paragraphs analyze the major pollutants and their effects on the environment, which can be summarized as:

- Greenhouse effect and global warming
- Photochemical smog
- Acidification
- Phytotoxicity and animal toxicity (bioaccumulation)
- Oxygen depletion in the root zone

2.3.2.1 Greenhouse gases (GHGs)

The major emission of GHGs in an open landfill is the release of methane and carbon dioxide generated by the anaerobic processes. Total GHGs emission are estimated to be around 1000 kg CO2-eq tonne^{-}1, more than three times higher than the emissions in conventional landfills sites [29]. MSW combustion causes uncontrolled emissions of carbon dioxide, oxides of nitrogen and nitrous oxide. Total average GHGs emission of MSW combustion is 902 kg CO2-eq tonne^{-}1 [30]. GHGs contribute significantly to the greenhouse effect and the global warming.
Methane also affects negatively the vegetation in the surrounding areas, by substituting the oxygen in the root layer of the ground [22].

2.3.2.2 Nitrogen oxides (NOx)

Nitrogen oxides are precursor of photochemical oxidant; they react with the volatile organic compounds generating secondary pollutants, such as ground level ozone. The secondary pollutants are phytotoxic by reducing the photosynthesis process [31]. They can produce acid rain and acid precipitation that can damage plants and acidify the bodies of water [32].

2.3.2.3 Particulate Matter (PM)

PM concentration is proven to exceed the health protection standards even in the case of regular landfills with proper cover soil [33]. Environmental effects of PM are acid rain formation, lowered visibility and PMs are a precursor of photochemical oxidation [34]. The effects of PM emissions impact on the health of the plants by reducing the photosynthesis process and destroying the leaf tissue [35].

2.3.2.4 Persistent Organic Pollutants (POPs)

POPs are freed in the MSW combustion process. Among these pollutants the polycyclic aromatic hydrocarbons are generated by incomplete combustion. The illegal burning of MSW occur under uncontrolled temperature conditions, therefore a high amount of products of incomplete combustion are expected in the gases [36]. POPs can damage the flora and fauna of surrounding areas and accumulate in the food chains.

2.3.2.5 Sulfur Oxides (SOx)

Sulfur oxides cause acidic precipitation and acid rain affecting negatively the flora and the bodies of water [32].

2.3.2.6 Volatile Organic Compounds (VOC)

Volatile organic compounds are precursor of photochemical production of ground level ozone in the presence of sunlight and NOx [22], and contribute to the formation of secondary organic aerosols [37].

2.3.3 Animals and plants

The flora and the fauna living in the landfill or in the surrounding areas come into contact with the pollutants described in chapter 3.1 and 3.2; therefore they may suffer from diseases induced by the pollutants. Secondary products of photochemical oxidation such as ozone and peroxyacetyl nitrate (PAN) can have harmful effects on plants. These substances decrease growth in plants by reducing photosynthesis [38].

Some pollutants, such as POPs, accumulate in the food chain becoming more toxic at higher levels of the food chain (biomagnification) [39]. Plants accumulate heavy metals and radionuclides as well and this pollutants risk to affect the population in case of crops cultivation nearby the illegal landfill [40]–[42].
Plastic debris has been proven to enter the animal food chain [43]; the effect of plastic ingestion in many marine species has been studied: e.g. seabirds suffer of obstruction of the gastrointestinal tract and lowered feeding stimulus that can lead to starvation [44].

2.3.4 Hazardous waste

The hazardous wastes are a potential hazard to the environment, affecting soil, air and water quality, when improperly treated, stored, transported or disposed [45]. In this chapter we evaluate the environmental impacts of industrial and medical waste pollutants that are usually less abundant in MSW. These pollutants are highly harmful and can cause groundwater quality degradation; phytotoxicity; aquatic and animal toxicity accompanied by possible bioaccumulation.

2.3.4.1 Heavy metals

Heavy metals are a threat to the environment and the species living nearby the landfill, as other persistent pollutants they bioaccumulate in the food chain [46], as leached into the ground heavy metals reduce the groundwater quality and are toxic to aquatic organisms [47].

2.3.4.2 Radionuclides

Radionuclides are a class of chemicals where the nucleus of the atom is unstable and emits radioactive radiation. When a radioactive waste is properly disposed it is put into storage containers made of steel that are then placed inside a further cylinder made of concrete. The protective layers prevent the radiation from spilling and harming the environment; illegal landfills lack in these protection systems, thus the radionuclides spread out. Uncontrolled disposal of radionuclides cause long-term cancerous or genetic problems for many animal. Nuclear waste will remain hazardous for hundreds of thousands of years and the ingestion of food is the most common root of exposure for animals and human beings [48].

2.3.4.3 Pathogens and biohazardous waste

Insects and animals can carry diseases caused by wastes and infect other animals or humans by contact or food ingestion [49]. The toxicity level depends on the type of biohazard. Above 150 known enteric pathogens may be present in the wastes, and new enteric pathogens continue to be discovered [50].

2.4 Health impacts

The health hazards from waste are linked to the toxicity of some pollutants (freed in the combustion process, released into groundwater or accumulate in the food chain) and to the infectious disease carried by insects and animals that can act as vectors (see chapter 3.3).

Children are more vulnerable to pollution than adults, as their immune system and lungs are not fully developed; this condition increases the possibility of more severe responses in children than in adults [51]. In addition, children spend more time outside; therefore children living nearby illegal landfills are more exposed to waste borne pollutants. Exposure to some chemicals, such as heavy metals and polychlorinated biphenyls, during early fetal development can cause neurodevelopmental disorders and subclinical brain dysfunction [52].

Illegal landfills emit several different kinds of pollutants that can be harmful even at low concentrations. The multiple chemical sensitivity is a condition that is caused by the exposition to many different pollutants; individuals affected by multiple chemical sensitivity show an acute hypersensitivity to low levels of chemicals that can be found even in everyday substances [53].

The following paragraphs will identify the major health hazards caused by illegal landfilling and uncontrolled waste burning.

2.4.1 Acute intoxication

Some contaminants can be released in high amounts, affecting people that, at the time of the pollution event, find themself on the surrounding areas of the contaminated sites. The acute intoxication is usually generated by combustion products and it is confined to the landfill and the nearby surroundings.

Carbon monoxide can cause fatal poisoning because it displaces the oxygen in the hemoglobin [54].

Irritation of the eyes and the mucosa can be due to high amounts of ammonia, nitrogen oxides, sulfur oxides and particulate matter [55].

2.4.2 Carcinogenicity

The pollutants released from illegal landfilling can be divided into classes of potential carcinogenicity. Marfe and Di Stefano gathered evidences on the relationship between illegal dumping waste and illegal dumping sites [56]. Other evidences of possible carcinogenicity are given by epidemiological and long-term exposure studies [57]. The International Agency of Research on Cancer classifies the pollutants into [58]:

- Group 1: carcinogenic to humans
- Group 2A: probably carcinogenic to humans
- Group 2B: possibly carcinogenic to humans
- Group 3: Not classifiable as to its carcinogenicity to humans

- Group 4: Probably not carcinogenic to humans

Cogliano et al. review the carcinogenicity of various agents and the site of associated cancer, e.g. benzene is proven to cause leukemia, cadmium affects the lung and, probably, the kidney as the prostate [59]. Benzene and/or vinyl chloride, both carcinogenic substances, have been found in the gaseous emission of 90 percent of analyzed municipal landfills [60].
Some POPs and VOCs as polychlorinated biphenyl (PCB), dioxins and Dichlorodiphenyltrichloroethane (DDT) belong to group 1 and 2; heavy metals as well are classified as group 2 [58].
The asbestos is a construction material banned in Italy in 1992; even if its proper disposal is crucial and prescribed by the law (D M. 29/7/2004 n. 248) large amounts of this pollutant can be found in illegal landfill [61]. Asbestos is carcinogenic to humans; it belongs to group 1 and is proven to affect larynx, lung, mesothelium and ovary [59].
Sulfur oxides and fine particulate matter cause lung cancer; the smaller the particles are the higher is the risk of cancer. The size of the particles defines the site in which they set down: PM10 particles deposit mainly in the upper respiratory tract while fine and ultra fine particles are able to reach lung alveoli [62]. Each 10 $\mu g/m^3$ increase in fine particulate air pollution is associated with approximately 6% increased risk of lung cancer mortality [63].
Persistent organic pollutants and heavy metals accumulate in the food chain and in the organisms; they increase the risk of carcinogenicity as this disease is linked to usual exposition even to low amounts of pollutants [64]. These pollutants can travel far away from the source, as many of them are volatile or leach into groundwater.
Radiations emitted by nuclear waste can be transferred, as well, in any location with no limit to their diffusion. E.g. nuclear waste illegally disposed in Tunisia emits nuclear carcinogenic radiations that pollute local crops. These crops are finally shipped even to countries distant from the radiation source and sold in the supermarkets, spreading the impact profusely [65]. Ingestion of food is the main exposure route for humans and nuclear waste will remain hazardous for hundreds of thousands of years affecting also future generations [48].

2.4.3 Endocrine-related toxicity

Chronic exposure to heavy metal such as copper, zinc and nickel can lead to adverse effects on the function of the endocrine system, by mimicking the activity of steroid hormones [66]. Many persistent organic pollutants are endocrine disruptors; they may be more hazardous during fetal, neonatal, and childhood development and their effect can be transgenerational [67]. Exposition even to low- level of endocrine disruptors is hazardous to humans and animals [68]. Adverse consequences of exposition to endocrine disruptors are: reduced reproductive function, disorders in pediatric patients, and, probably, type 2 diabetes and some types of cancer [64].

2.4.4 Genotoxicity and mutagenicity

Chlorinated hydrocarbons and aromatic amines, benzene, nitric oxide and radioactive radiations have damaging effects on chromosomes [69]–[71]. Global DNA methylation levels, which are indicators of DNA protection, have been reported to be inversely associated with blood levels of persistent organic pollutants [72]. PMs as well have been found to cause toxicity to mammalian and lung cells [73]. Genotoxic pollutants can damage the DNA of somatic cells causing alterations, which may lead to cancer. If the alterations affect germ cells the effects can be transgenerational. Mutagenicity tests are proof of the mutagenicity potential of MSW leachate, other waste such as e-waste, waste from vinyl-chloride industries and waste combustion gases may have even higher mutagenic potential [70].

2.4.5 Infectious disease and biohazards

Direct contact, water contamination or vectors spread biohazards and infectious disease. Microorganisms (bacteria, viruses, parasites and fungi), found in the waste, can affect the human health by means of direct action or toxins formation [64], [74]. Vectors are small animals or insects that carry infectious disease outside the illegal landfill area causing more widespread contamination. Some pathogens can pollute the waterbodies and be accidentally ingested by animals and people.
Common vector-borne diseases associated with waste disposal are: leptospirosis, salmonellosis, amoebiasis, dysentery, cholera, toxoplasmosis and teniasis [75]. Infectious wastes are all materials employed for the treatment and examination of patients affected by infectious disease. In 2010, unsafe handling of infectious syringes was responsible for 33800 new HIV infections, 1.7 million hepatitis B infections and 315000 hepatitis C infections [76]. People living nearby illegal landfills are more subjected to this kind of adventitious infections.

2.4.6 Neurotoxicity

Neurotoxic pollutants have the ability to damage the brain and/or the peripheral nervous system. Neurotoxicity may cause abnormal neuronal development, destruction of neurons and related mental disorders with behavioral changes [77].
Exposure to POPs, such as PCBs and polybrominated diphenyl ethers (PBDEs), has been associated with neurotoxic effects. POPs neurotoxicity has been observed in humans even at low environmental concentrations [78].
Some industrial chemical wastes and heavy metals, namely lead, methylmercury, arsenic, and toluene, are recognized to cause neurodevelopmental disorders and subclinical brain dysfunction [52].

2.4.7 Respiratory and cardiovascular problems

Fine particulate and sulfur oxide are associated with cardiopulmonary mortality. Each 10 $\mu g/m^3$ elevation in fine particulate air pollution increase mortality risk of 8% [63].
Photochemical smog pollutants (nitrogen oxides, VOCs, ozone and PAN) have been proven to cause respiratory problems [62]. Epidemiologic studies have linked dioxin and PAN exposure to increased mortality caused by ischemic heart disease [62], [79].
Health problems such as tachycardia, increased blood pressure and anemia, which is due to an inhibitory effect on hematopoiesis, are proven to be caused by heavy metal pollution [80].

2.5 Social impacts

In this section the aspects related to social and economical issues of illegal landfilling will be discussed, namely local mobilizations, discomfort and change in habits of local populations, economical issues and aesthetic degradation of the landscape.

2.5.1 Local mobilizations

In the last decades the conflict situations related to illegal dumping rose markedly: conflict situations that aim at highlighting a disagreement among a part of the civil society.
The opposition is expressed by the contestation of a particular case or event of illegal dumping, regarding those places that the local population considers as “family places”. Inhabitants, local deputies, trade unions, experts and even environmental activists commit to contesting the situation in which they are.
Local communities organize themselves into unions of inhabitants and organize multiple collective actions, blockages, and information campaigns, denouncing the irregularities and strongly supporting the need to protect the territory and the environment. Thus, “local mobilizations” appear: they emerge to oppose a transformation of the physical, natural, rural or urban environment considered harmful or to claim an improvement of the area where the illegal landfill is.
Di Toro highlight that “Citizens are trying in every way to make themselves heard”, however nothing has been done yet. The sit-in requested that all waste should be removed and cameras installed in order to monitor the territory [81].
These local mobilizations, developing spontaneously, constitute a group of individuals who express a collective purpose. They do not have a formalized organizational structure, thus no role is rigidly defined. They do not have formal and detailed action programs, statues, rules that define the belonging or not to the movement. The cohesive force that holds together the members of a movement is to believe in an idea and the enthusiasm of sharing this idea.
Local mobilizations are directed towards those who should prevent these events of illegal landfill. Therefore the conflict situation proceeds through a questioning of the

legitimacy of public authority, as the newspaper "il centro" states "above all on the initial stretch of the road, it is full of landfills ignored by all the institutions" [82].
Local mobilizations arise in response to a need that is not satisfied by the normal functioning of the society. So these movements bear a need for change, compared to the ordinary situation. They express this need through forms of solicitation that are not institutionalized, namely admitted and codified by the law [83].
The issue of preserving practices, values or ways of life that are linked to these places, damaged by illegal dumping, is of major interest to the collective mobilization. Places, which individuals or social groups invest with sentimental values, can be degraded by such an event of illegal dumping, causing a feeling of irreversible loss. This loss predisposes people to a collective mobilization in order to defend and to protect those places. E.g. in Sardinia the community waited the remediation of a contaminated site that never happened. "Now they cannot even open the windows because of the nauseating smell coming from the landfill under their houses", thus the residents took to the street to protest [84].
Fear and anguish, experienced or supposed, underpin the debates and the action of the mobilizations. The fear of the risk of accidents, of illnesses, the discovery of a threat to their lives, of a danger (real or supposed) of death, and the feeling of impotence turn into a physical, emotional and social discomfort. Fear and anguish are manifested through a malaise experienced by individuals in society. There is therefore a malaise concerning these illegal dumping: the social discomfort involves the part of the population that is directly concerned and more broadly a part of the public opinion. The feeling of malaise becomes a reason for the mobilization [85].
Provenza shows how people feel threaten by the illegal dumping stating "this phenomenon in addition to poisoning the air, pollutes our territory, damages our fruit and vegetables and consequently we are damaged too".
Opponents strongly support the need to monitor and to safeguard the territory and the surrounding environment; they mobilize themselves having as objective the protection of the territory. The threat, represented by the risk of accidents, of illness and of dangers caused by the illegal landfilling, presents itself as a lived abstraction that circumscribes spaces and encourages people to 're-appropriate' them. In this case, the territory appears as an inseparable context from the various strong moments of the conflict.
The conflict becomes an intermediary in the relationship between the actors and the territory [86] and constitutes the founding element of the process of strengthening territorial identities. People recognize and strongly show their belonging to the territory: identities linked to the territory are strengthened. Therefore, at this stage of mobilization, the group seeks the attributes that participate in the construction of its territorial identity. In return, this territorial construction wins in external visibility, which favors the defense of the related territory [87].
People have a relationship with the surrounding territory: "to live" means to belong to a place and to relate to that place. The territory is a living space: a practiced space where people physically inhabit the place; and a social space, a place of creation of social bonds. But the territory is also a lived space: a coherent system of symbols, of thought and reflection. They inhabit and 'live' the territory through their material and symbolic practices. There is indeed a link between the living space and the lived

space [88].
The common territory and the sense of belonging form the basis of the territorial community; the existence of the groups cannot be imagined without a social-emotional link between individuals. Thus, people defend their territory through their material and symbolic practices. Through the conflict, the territory becomes a place of life, action and thought (space of life). The individual recognizes and reinforces his belonging to the territory; in this way, giving a meaning to the territory, he gives himself a meaning. In this regard, an identifying process is generated: the social territory becomes a product of imagination and a product of social relations [89].
Opponents move from the defense of the territory damaged by the illegal dump to the revaluation of this territory. Living the territory, or simply being present on the ground, represents the first step towards identification. The territory participates in building the identity of the group and consolidating the feeling of belonging to a symbolic and material sense. Opponents think themselves as a coherent whole fighting against a common enemy. People want to defend the place where they live; they want to preserve what they create every day. Opponents slip into a process of territorialisation; social geography defines this territorialisation as a double movement of material and ideal appropriation of a portion of space by a social group [87]. It is then necessary for the group to construct and re- construct what surrounds the social actor, both materially and in its representations [89]. The social territory becomes a product of imagination and a product of social relations.

2.5.2 Discomfort

The main discomforts due to MSW illegal landfilling and incineration are odors and decreased visibility.
The main sources of odors from a contaminated site are compounds that are formed during the processes of organic waste decomposition and combustion. Although odorous compounds are not necessarily toxic or hazardous for the environment and the human health, it has been proven that the psychophysical well-being is negatively influenced by exposure to odors [90].
The reduced visibility nearby contaminated sites is due to aerosol particles and photochemical smog [38], [91].

2.5.3 Change in habits

The contamination of local crops and animals can change the eating habits of the population. E.g. the newspapers reported in winter 2007-2008 that in the Naples surroundings polycyclic aromatic hydrocarbons (PAHs), mainly formed by illegal landfilling and uncontrolled incineration, pollute the "Mozzarella di bufala" cheese [92]. These pollutants migrate through the human food chain affecting human health. The awareness of the population to this problem has led to a substantial reduction in sales (-30% in three months) [93].
More severe impacts are due to the establishment of exclusion zones. High levels

of hazardous pollutants can lead to close a site to the public. The case of nuclear pollution around the Chernobyl nuclear plant is a well-known event of establishment of exclusion zones [94]. This event was due to a nuclear accident; however uncontrolled disposal of nuclear waste can cause similar problems. Excessive phosphorus levels in bodies of water can contribute to cyanobacterial growth [95]. Many authors report bathing bans due to cyanobacteria because of its potential toxicity and odors generation [96]–[98].
In 2008 the U.S. Navy began testing the pollution in Naples and Naples surroundings, as many events of MSW illegal combustion were reported. The study led to the definition of a map of lease suspension zones in witch American officials and their family are forbidden to live [99].

2.5.4 Economicalissues

When illicit burning or dumping is an actual disposal option it cannot be taxed directly. The European and Italian law prescribe that who pollutes must pay; however it is usually difficult to identify the polluter. Therefore illegal landfilling is a cost to the State and the population. Some economic studies identify the optimal fee structure as a deposit-refund system, in order to minimize illegal landfilling. Theoretical models have proven that other fee structures, such as virgin materials taxes, advance disposal fees, recycled content standards, and recycling subsidies are less effective than the deposit-refund system [100], [101].
Other economical impacts are linked to negative effects on trade and tourism at a local economy level and to the costs due to environmental remediation, when the polluter is not identified and punished [102]. Several studies proved that the concentration of pollutant in agricultural land near illegal dumpsites is comparable with the level of natural geochemical background, however the legal inquiry caused a severe economic loss because of the concern of the buyers [103]–[105].

2.5.5 Aesthetic degradation of the landscape

Illegal landfills reduce the aesthetic appeal of the landscape and its value of the nearby areas [106]. The surroundings bodies of water can suffer from eutrophication that degrade aesthetic properties of lakes and ponds [107]. Eutrophication is a process due to nutrients pollution that induces algal bloom, thus the body of water becomes cloudy and coloured.

2.6 Conclusions

The rising concern about the impacts of illegal landfilling led to specific law regulations that prescribe duties and sanctions. Recent changes to legislation concern the protection of the environment through criminal law introducing the crimes against the environment. The illegal landfills and uncontrolled waste combustion affect negatively the groundwater quality: the soluble pollutants leach freely into the

ground and accumulate in the aquifer. The surrounding environment can be harmed by eutrophication and acidification. Moreover some pollutants are toxic to living beings and can be accumulated into the food chain. On a larger scale pollutants released by illegal landfilling and burning increase the global warming and may lead to photochemical smog formation.
It appears clear that the pollution generated by these illicit actions affects the air, the bodies of water, the groundwater the soil the flora and the fauna.
Landfill pollutants have also harmful effects on human beings raising the risk of illness such as: acute intoxication, cancer, infectious disease, respiratory and cardiovascular problems. Some pollutants show, as well, toxic effects on endocrine, reproductive and nervous system.
The local communities are also affected by discomfort and change in habits, economical issues and aesthetic degradation of the landscape. The tangible environmental damage due to the illegal landfill raises awareness among the population. This awareness takes the form of social mobilizations and collective actions.

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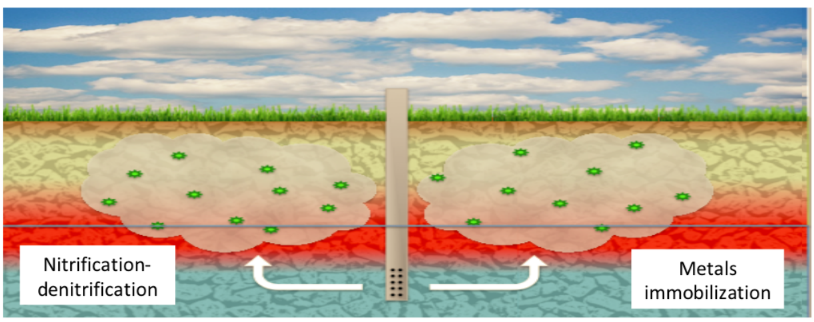
Chapter 3

Leachate contamination on groundwater: a lab-scale study on biosparging remediation and scale-up

A. Limoli; L. Dallago; M. Gabrielli; G. Andreottola

Graphical abstract



3.0 Abstract

In this study the biosparging technology has been applied to remediate an aquifer polluted by leachate percolation. The biosparging stimulates the growth of indigenous bacteria able to convert pollutants, such as ammonium nitrogen, in harmless compounds. The technology shows high efficiency in ammonium nitrogen removal via nitrification processes. The biosparging remediation technology immobilizes metals and removes nearly completely the nitrates accumulated in the nitrification process when the organic carbon source is conveniently dosed. The application of the biosparging on site is feasible and its application in leachate-polluted aquifers shows several advantages.

Keywords: Leachate; Biosparging; heavy metals; ammonia.

3.1 Introduction

Leachate is a liquid produced as rain and other water pass through a landfill, transferring pollutants from the solid waste to the liquid phase. Leachate carries pollutants into groundwater, representing a threat to adjacent ecosystems and human health [1]. Old landfills are the most likely to produce groundwater pollution, as the waterproofing is weak and the geomembranes are broken or even missing. Therefore many studies have been conducted on leachate treatments to reduce the pollutant content. The leachate treatments can be distinguished between the type of treatment (physical, chemical, biological or combinations of those) and the localization of the treatments (in situ or ex situ) [2].

This study focuses mainly on the removal of ammonium nitrogen as the case-study site shows strong ammonium nitrogen contamination and its content in a landfill increases over time [3]. The biosparging can therefore be applied to any aged municipal solid waste landfill. Biological treatments are quite effective in removing organics and ammonium nitrogen.

A well-established physical-chemical treatment namely the air sparging has been applied in several studies to reduce pollutants such as petroleum contaminants [4] and chlorinated solvents [5] by volatilization. The air-sparging technology volatilizes pollutants by injection of air into a polluted area through vertical or horizontal wells [6]. Limiting the airflow it has been established that biological processes are favored to the detriment of volatilization, therefore the process becomes biological instead of physical-chemical [7], [8]. Biosparging stimulate the growth of indigenous microorganisms that decompose the pollutants and has been applied to reduce hydrocarbon [4], [9], moreover the process stimulates the immobilization of metals by oxidation and precipitation. The biosparging is a new technology that has yet to be studied in detail and its effect on ammonia removal has not been established yet.

Among the alternative technologies for the remediation of landfill leachate contaminated aquifers, biosparging is a sustainable technical solution, which shows several advantages; the aim of this study is to analyze the process of nitrification-denitrification stimulated by air and methanol injection focusing on removal kinetics and processes of metal mobilization. The biosparging technology has been applied in a lab-scale plant; the tests were aimed to evaluate the removal efficiencies of several pollutants and to define the scale-up parameters of the system.

3.2 Material and Methods

3.2.1 Contaminated groundwater

The groundwater fed to the system was sampled from a piezometer located near an old landfill in the north of Italy. The samples were stored in a refrigerator to prevent biodegradation. The characteristics of feeding substrate are shown in table 3.1. The groundwater shows a high Total Ammonia Nitrogen (TAN) concentration that exceeds the law limits of drinking water [10]. Manganese as well exceeds the law limits (50 μ g Mn/L).

Table 3.1: Feeding groundwater characteristics

Parameter	**Average concentration**	**Law limits**	**UM**
TAN	65.7	0.5	mg N-NH_3/L
Nitrates	0.8	11.3	mg N-NO_3/L
Iron	22.5	200	μ g Fe/L
Manganese	228.8	50	μ g Mn/L
Arsenic	2.0	10.0	μ g As/L
Nickel	14.5	20.0	μ g Ni/L
COD	39.0	N.A.	μ g gO_2/L

3.2.2 Experimental apparatus

The lab scale treatment plant, shown in figure 3.1, consist of:

- Column 1: down flow aerobic conditions
- Column 2: up flow anaerobic conditions
- Aeration system
- Hydraulic system

Both the columns were filled with the packing material described in paragraph 3.2.2.1 and the feeding groundwater disclosed in paragraph 3.2.1. Methanol was injected in the second column to favor heterotrophic processes.

3.2.2.1 Packing material

The packing material of the two columns is gravel extracted by core sampling. The gravel was sampled in the area of a landfill in the north of Italy at a depth of 50-70m,

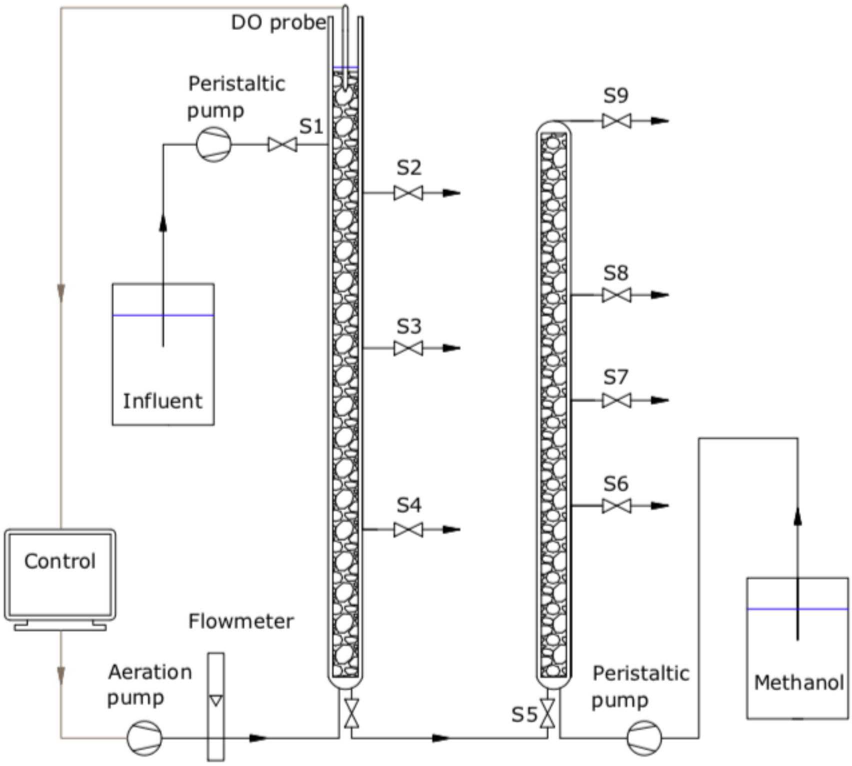


Figure 3.1: Biosparging reactor scheme

corresponding to the saturated layer of the ground.

3.2.2.2 Column 1

In the first column the nitrification process takes place. The bacteria oxidize the ammonium nitrogen to nitrite and nitrate. This process is aerobic, thus injection of air is needed (2.2.4). The diameter of the column is 82 mm and the height is 2.16 m. The sampling procedure in the first column occurs through four faucets. The faucets are located at 0, 180, 576, 1084 and 1678 mm from the influent injection point. The injection point is at the top of the column and the influent moves down flow. The effluent flows to the bottom of the second column.

3.2.2.3 Column 2

The second column hosts the denitrification process that reduces nitrates to gaseous nitrogen N2. To favor this process anoxic conditions and an additional carbon source (2.2.5) are provided. The diameter of the column is 82 mm and the height is 1.82 m. The sampling procedure in the first column occurs through four faucets. The faucets are located at 0, 560, 905, 1250 and 1820 cm from the influent injection point. The injection point is at the bottom of the column and the influent moves up flow. The effluent flows to a storage tank in witch it is collected. The last sampling point (S9) is located in the effluent tank.

3.2.2.4 Aeration system

The software OUR.net allow to set the range of dissolved oxygen required. The lower limit was set to 2 mg/L and the upper one to 4 mg/L. The software controls the blower that pumps the air at the bottom of column 1. The airflow is limited by a fluximeter to 1 L/min in order to avoid the volatilization of some compounds and limit the turbulence [7].

3.2.2.5 Hydraulic system

A peristaltic pump feeds the first column the influent providing a water velocity in the column similar to real groundwater speed. The first column Hydraulic Retention Time (HRT) is 4.9 days. For carbon source a second pump injects the methanol solution at the bottom of the second column. The HRT of the second column is 2.89 days.

3.2.2.6 Carbon source

The feeding groundwater has low organic matter content. In order to avoid process limitation by organic carbon lack, an external source has been added to the denitrification process. The feeding solution has been prepared mixing methanol to tap water. The methanol added was calculated stoichiometrically in order to reduce the nitrates produced in the first column, however earlier experimentations showed that the stochiometric amount was not enough to complete denitrification. Therefore the quantity of methanol was increased by 50% and 25%.
The three concentration tested were:

- 243.6 mg/L stoichiometric C/N ratio
- 356 mg/L overdosing 50%
- 300.6 mg/L overdosing 25%

3.2.3 Analytical methods

Analyses of ammonium nitrogen, nitrates, nitrites, nickel and manganese were performed with a spectrophotometer HI 83206 Hanna Instruments and the relative Hanna Instruments kits. The ammonium nitrogen concentration has been verified using APAT CNR IRSA Met 4030 A2 Man 29/2003. The Chemical Oxygen Demand (COD) analysis was performed according to APAT CNR IRSA Met 5130 Man 29/2003. The analyses of other metals were performed according to UNI EN ISO 17294-2 2005. Redox potential and pH were monitored with specific probes.

3.3 Results and discussion

3.3.1 The nitrification process

After an 18-day acclimatization period, in witch the liquid phase was recirculated in a closed loop, the polluted groundwater started to be fed to the plant.

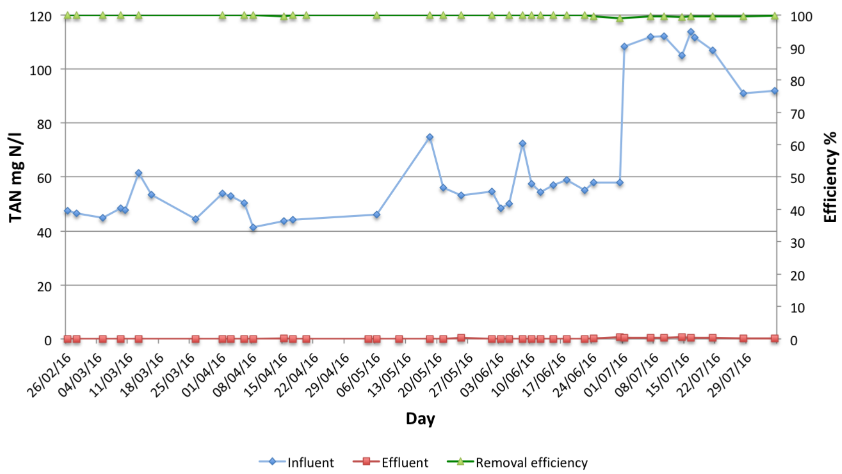


Figure 3.2: Ammonia removal: influent, effluent and removal efficiency

Experimental evidences figure 3.2 show that the Total Ammonium Nitrogen (TAN), at the bottom of the column 1, was almost completely removed. The average ammonia removal in the whole system is 99.8%. The TAN concentration exceeded the law limits after the first column in just two cases out of 40 measurements and never after the second column.

3.3.2 The denitrification process

The denitrification process (column 2) is proved to remove 99% of the nitrate produced in the nitrification process. This process, however, depends on the organic carbon content: in the middle of April the methanol was increased of 50% (356 mg/L) with a subsequent improvement of the nitrate removal efficiency, as shown in figure 3.3. On the 22nd of July the methanol was reduced to 300.6 mg/L. The experimental results show that this quantity is enough to ensure complete denitrification.
The law limits of 11.3 mg N-NO3 were never exceeded even in the initial phase.

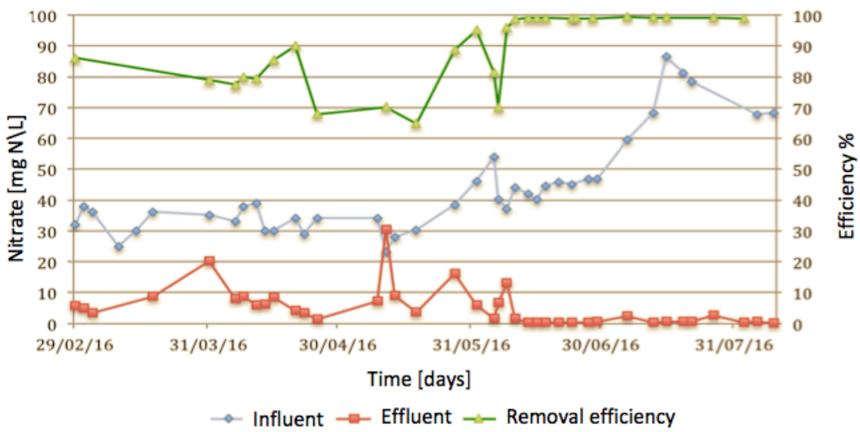


Figure 3.3: Nitrate removal: influent, effluent and removal efficiency

3.3.3 The mobilization/immobilization process

The mobilization and immobilization of metals are processes governed by oxidation and precipitation of metal ions. Arsenic and manganese reached respectively 86% and 97% removal efficiencies in the fist column, while iron shows a lower removal due to its reduced initial concentration: this concentration was probably lowered by the precipitation of iron oxides and hydroxides in the storage tank. Manganese spatial profiles are shown in figure 3.4; Mn ions in solution oxidizes and precipitates as manganese hydroxide in column 1 where the Oxidation Reduction Potential (ORP) is above 100 mV and the pH is above 7 [11]. During the denitrification process (column 2) the ORP drops at levels lower than 100 mV, favoring a partial mobilization of manganese. Mn does not reach the law limits of 50 μg Mn/L in column 2 remaining below 20 μg Mn/L even at the minimum ORP.

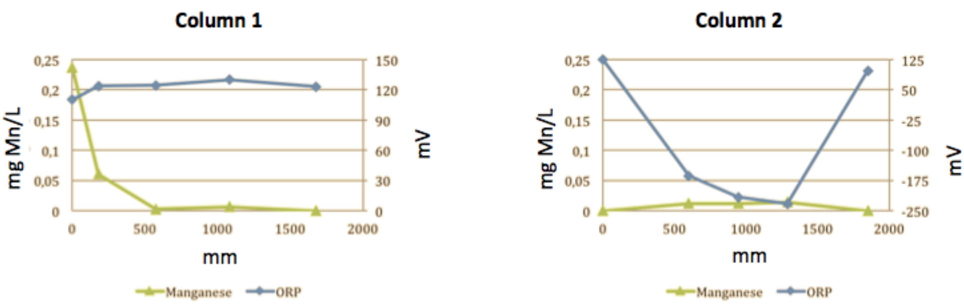


Figure 3.4: Mn mobilization and immobilization processes and ORP trend.

Nickel shows an anomalous behavior: it increases in the first column and decreases in the second column; however, at the end of the treatment, the Ni exceeded the law limits. The enrichment of nickel in the liquid phase is probably due to sorption equilibrium of nickel in the presence of competing cations and organic matter. The pH changes given by the processes of nitrification and the intermittent aeration could influence, as well, the Ni mobilization.

3.3.4 The scale-up

The results show that the process studied is able to efficiently remove the total ammonia nitrogen; therefore a scale-up of the system is needed in order to apply the technology at a real case. To apply the technology to a real case it is necessary to evaluate the speed of the groundwater at the site; the flow rate has to be changed according to the groundwater speed [12].

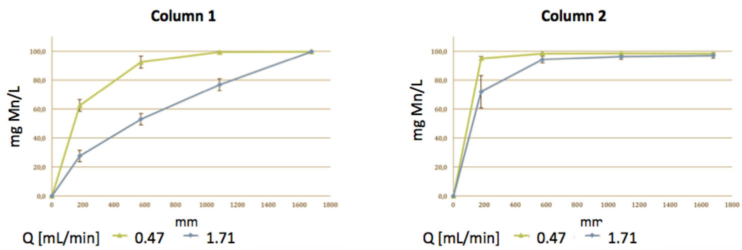


Figure 3.5: Ammonia and nitrate trend at different flow rate

The plant scale-up parameters have been calculated on the hypothesis that the process has reached dynamic stability. Two flow rates have been tested: a flow of 0.47 mL/min that was set in most of the experimentation and a flow rate of 1.71 mL/min deduced from the hydraulic gradient of the site. The increase of the flow rate causes a reduction of the HRT to 1.3d in column 1 and 0.8d in column 2. The results show that the ammonia was successfully removed, however the trend of ammonia removal in column 1 changed (figure 3.5). The trend with lower flow rate is nearly exponential, but with rising flow rates the trend becomes linear. The TAN was below the law limits just in the lower faucet at the exit of the nitrification column. Despite a further increase of the flow rate could lead to incomplete ammonia removal, the process have been proved to be suitable at a real scale in the studied site.

The denitrification process was faster and it occurs in the first part of the column even at higher flow rates. The denitrification efficiency shows a stronger dependence on the organic carbon than on the HRT. The average removal efficiencies of TAN and nitrate are above 99% and 97% respectively. The average rate of TAN removal is approximately 2.2 mgN/(L*h). Figure 3.6 shows the scheme of the injection well. The length of the well depends on the site and the aerobic area is calculated using the TAN removal rate and the ammonia pollution of the site.

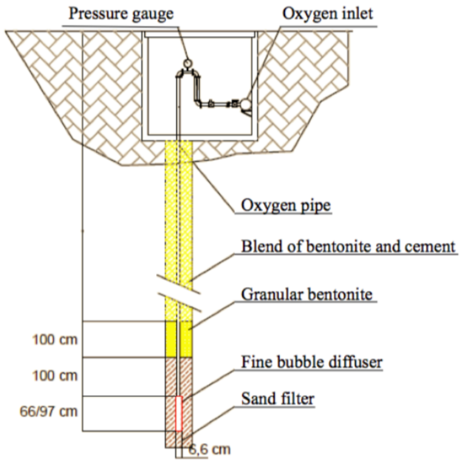


Figure 3.6: Biosparging Injection well scheme

3.4 Conclusions

This study proved the efficiency of the biosparging process as an in- situ technology for bioremediation of a leachate-contaminated site. The advantages of the process can be summarized as follows:

- Biosparging stimulate the growth of indigenous microorganisms able to convert pollutants into harmless substances
- The average ammonia removal in the whole system is 99.8%
- With a proper organic carbon dosage the nitrates, produced in the nitrification phase, are almost completely removed
- Biosparging favor the immobilization of some metals such as manganese and arsenic preventing heavy metal contamination in groundwater
- The process can be applied at a real scale on site, without transporting large quantities of waste off site: procedure that arise the costs and the potential of spills, with a threat to the human health and the environment.

AKNOWLEDGEMENTS

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Chapter 4

Solidification and stabilization of municipal solid waste

A. Limoli; G. Andreottola

4.1 Abstract

This paper aims at defining a cost-effective solution to minimize volume of waste into landfill and reduce environmental pollution caused by municipal solid waste. The S/S technology has been applied to build waste slabs. Chemicals improve physical and mechanical characteristics of the waste. The best chemical recipe has been defined with lab scale experiments and on field applications. Leaching test shows that gaseous and liquid pollutant decreased strongly in case of S/S treatment with cement and lime.The methanogenesis is strongly inhibited as the respirometric index test assess .

Keywords: Sustainable landfill; weste solidification; waste stabilization; cement; lime

4.2 Introduction

The Solidification/Stabilization (S/S) technology is commonly applied a a waste pretreatment process, before landfilling. It has been used for different types of hazardous wastes, since it has proven to be effective in heavy metal immobilization [1]. The S/S has been widely applied to hazardous wastes and municipal solid waste (MSW) incineration ashes [2–10]; however, this technology has never been applied to MSW treatment.

The S/S process uses chemically reactive formulations that, together with water, form stable solids; it also insolubilizes, immobilizes, encapsulates, destroys, sorbs, or

otherwise interacts with selected waste components. The stabilization is the process in which hazardous compounds are complexed or bound in a stable insoluble form. The solidification process consists in encapsulating the waste in a solid matrix, improving the physical characteristics of the waste and reducing the mobility of the hazardous compounds; thus less contaminants are freed into the environment. The result of this process is a less hazardous solid. Two parameters, strength and the leach resistance, define the degree of effectiveness of the final product [10]. This study will focus on these aspects and will prove that greenhouse gases emissions are blocked.
Solidification results in changes in primarily physical properties of the waste material so that a well-solidified waste will contain less free liquids and will have improved strength. Furthermore, a solidified waste will have less impact on the environment when disposed. It will contain less free liquids that can be easily transported to contaminate the environment. It will usually be formed into solids of larger size than the untreated waste. This will result in smaller area/volume ratios that will result in lower rates of release of contaminants with the exception of S/S by means of granulation. Granulation increase the surface area of the waste mixture, thus it can lead to higher pollutant emission into leachate.
The solidify waste in slabs or cubes will typically have decreased permeability, which reduces the flow through the waste. If the treated material has a substantially lower permeability than the material surrounding the disposal area, fluids will flow around rather than through it, resulting in substantially reduced release of contaminants [11].
S/S pre-landfill treatment inhibits the biogas production. The pH has a fundamental role in methane production and it has been proven that above pH 8.8 the methane production is almost completely inhibited [12]. This study will prove this effect by means of respirometric index analyses.
Compared to other remediation technologies, S/S shows the following advantages [1]:

- Relatively low cost solution,
- Easy to process,
- High repeatability,
- Good long-term stability, both physical and chemical,
- Good impact and comprehensive strength,
- Non-toxicity of the chemical ingredients,
- High resistance to biodegradation,
- Relatively low water permeability.

Furthermore this study proved the S/S technology to be effective in waste volume reduction, which is crucial for extending the lifespan of landfills.
The S/S of wastes, to date, has been mostly applied to treat hazardous waste [8–10,13] and incineration fly ashes [2–6] and its application to municipal solid waste treatment is new and needs experimental studies able to define the feasibility of this method.
The S/S can also enhance the traceability of the waste depending on the technology adopted to perform the stabilization. The slabs or cubes of waste can be confined and marked in order to obtain complete and enduring traceability.

This paper aims at defining the best chemicals to use (cement and/or calcium oxide) and the optimum quantity of each chemical in MSW slabs production. Furthermore the biological activity and liquid emissions of both the untreated and treated waste are analyzed. This technology is proven to be low-cost, it reduces pollutant emitted into leachate and blocks methanogenesis preventing greenhouse gases emissions.

4.3 Materials and methods

4.3.1 Chemicals and waste

Analytical grade calcium oxide (99.9% minimum on weight, CaO) was used in the laboratory experiments while a less expensive and less pure (90% CaO w/w) lime was used to build the slabs on field. Portland cement with limestone CEM II/B-LL 32,5 R was used both on field and in the laboratory. Tap water was added to enhance pozzolanic reaction.

The waste was sampled from a Mechanical Biological Treatment (MBT) plant, which treats the residual waste of the province of Trento. The amount of the recycled material in relation to the residual waste increased over years reaching 81.32% in 2017. The residual fraction (18.68%, 60Gg in 2017) reaches the treatment plant; it is ground and sieved using a 50 mm sieve. The fraction >50mm goes to a waste-to-energy plant while the fraction <50mm is stabilized in the MTB plant (7.2Gg in 2017). The stabilized waste is than sieved using a 20 mm sieve. The fraction >20mm goes to a waste-to-energy plant while the fraction <20 is used in the experiments. The preliminary compressive strength tests and the leaching tests were performed on cubic samples (150x150x150mm) at least 28 days after manufacture.

4.3.2 Solidification/stabilization experimental setup and procedure

As a first step, preliminary tests were carried out to investigate optimal dosage of chemicals (cement and/or calcium oxide) at a lab scale. Then, slabs of waste with and without chemicals were built in order to analyze physical properties. Finally samples were collected from the slabs to perform leaching tests and greenhouse gases emissions.

4.3.3 Preliminary tests

The first test was aimed at assessing the lime quantity to enhance physical properties of the waste. The test was performed according to ASTM C 977/95 guideline that defines the minimum lime amount for organic soils. The pH of the mixture of waste and lime should reach at least pH 12 for 2 hours. At first several amount of lime were tested and the pH was measured according to EPA Standard Method 9045D. The samples that showed an initial pH higher than 12 were selected and their pH was measured each 15 minutes to verify that the pH does not drop under 12 in the 2 hours of test, the test ended each time pH dropped under 12. For reference the pH of the tap water used to perform the analyses was detected. The pH values were assessed by a pH-meter with a resolution of 0.01. The probe was cleaned and calibrated before using. The second test determined the quantity of cement needed to have a good compressive strength. The compressive strength tests were performed in a Zwick/Roell Z250 apparatus applying increasing load by steps of 75N each 30s.

Table 4.1: Slabs composition

Slab n	**1**	**2**	**3**	**4**	**UM**
Waste	5100	5700	4760	5100	kg
Water	27.8	27.4	34.4	26.8	%wet waste weight
Lime	5.0	5.0	15.0	0.0	%wet waste weight
Cement	10.0	5.0	0.0	0.0	%wet waste weight

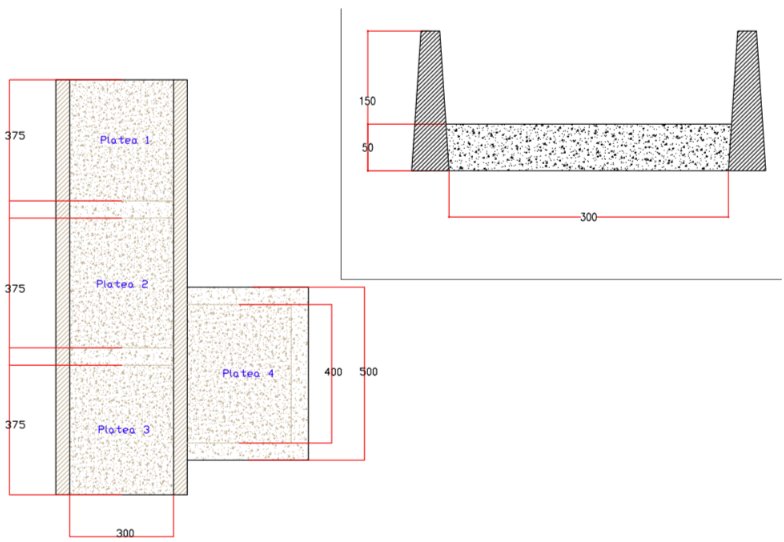

Figure 4.1: Waste slabs geometry

4.3.4 Waste slabs building

The waste slabs were built as described in table 4.1 and set as shown in figure4.1. The water added was higher in slab 3 due to the high temperature reached. Lime and water produce an exothermic reaction that induces water evaporation thus increasing amount of lime requires higher loads of water.

4.3.4.1 Waste slabs physical properties

The bearing capacity and compaction of the slabs were tested by means of dynamic plate load test employing the Light Drop-Weight Tester; the tests were performed in triplicate. This instrument facilitates a quick assessments, however it is usually less accurate than static plate load test. Static plate load test is more accurate but compromise the structure of the slabs permanently, therefore this test was performed after 55 days from building when dynamic plate loads showed a good performance. The tests was performed according to the standard procedure ASTM D1195 / D1195M - 09(2015).
The permeability of the slabs was detected by falling head permeability tests in boreholes each performed in duplicate after 50-55 days from building. We prepared a hole with constant diameter of 120 mm, where a PVC tube was pushed down and fixed with bentonite clay. The tube was filled with water and a water level sensor, which records levels each 30s, was immersed. The formulas for calculating permeability from these tests are assessed by the British standard Code of practice for site investigations BS 5930:1999 Method 1 (after Hvorslev) configuration D, where the intake factor is chosen as described in equation 4.3.1

$$F = 2\pi d + h \tag{4.3.1}$$

The geometry of the test is shown in figure 4.2.
The specific weight was measured 55 days after building of the slabs. Holes were

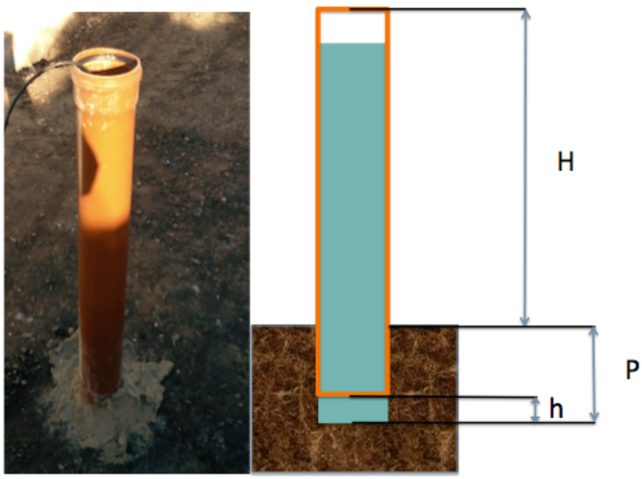

Figure 4.2: Infiltration test geometry

dug into the slabs; the waste mixture was collected and weighted; than the hole was filled with sand which density was previously calculated in the laboratory by filling 1-liter jar and measuring the weight. The sand was put in a bucket and weighted; the amount of sand needed to fill each hole was calculated by subtracting the weight of the bucket after filling the hole to the initial bucket weight.

4.3.4.2 Leaching tests

Tank leaching tests were performed on cubic samples made of the materials used to build the slabs. This test is aimed at comparing the liquid emissions of the best performing slabs described in the previous paragraphs and the emissions of untreated waste slabs. The leaching apparatus is composed by a tank, in which a cubic sample (150x150x150 mm) is immersed in distilled water, a recirculation pump and a tripod to hold the sample (figure 4.3).

The samples of leachate were collected at the end of the experiment after 24h. The

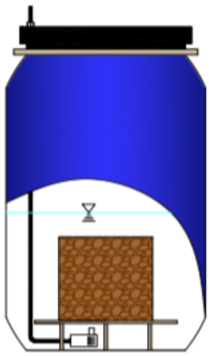

Figure 4.3: Leaching test apparatus

samples of the leaching test were filtered through 0.45 mm Millipore filter papers before being analyzed. Analyses of metals were performed according to EPA Compendium Method IO- 3.5. Total Ammonium Nitrogen (TAN) and nitrates analyses were performed with a spectrophotometer, according to, respectively, EPA method 350.1 and 352.1. Chemical Oxygen Demand (COD) was analyzed by titration following EPA Standard Method 410.3. Inorganic ions were determined by ion chromatography according to EPA Standard Methods 300.0.
The pH values were assessed by a pH-meter with a resolution of 0.01. The probe was cleaned and calibrated before using.

4.3.4.3 Respirometric index

The respirometric index can be applied as a tool to define potential biogas generation [14]. To analyze the bacterial activity an AIR-A respirometer (Open Respirometric Index Analyzer) was used [15]. The experimental parameters are shown in table 4.2, this analysis was applied to the best performing S/S slab (slab 2) and to untreated waste (slab 4). The samples were collected from the slabs after 55 days of hardening.

Total solids and volatile solids were detected according to EPA Method 1684: Total, Fixed, and Volatile Solids in Water, Solid, and Biosolids.

Table 4.2: Respirometric Index parameters

Slab n	2	4	UM
Temperature	30.8	30.9	° C
Natural humidity	46.4	32.7	%w w
Revised humidity	46.4	44.0	%ww
Sample weight	1.0	1.0	kg
Total Solids	53.6	67.6	%ww
Volatile Solids	30.5	27.3	%STww

4.4 Results and discussion

4.4.1 Preliminary tests

The first test was aimed at assessing the lime quantity to enhance physical properties of the waste; results are shown in Fig. 4.4. The tap water pH increased instantly to the pH lime-saturated water of 12.4 [16]. The water has proven to have no buffering capacity to lime thus it is possible to claim that the pH depends on the lime content and on the composition of the waste. This experiment should be repeated in case the characteristics of the waste change drastically.

Samples with lime content higher than 1.5% on wet weight of refuse reached pH

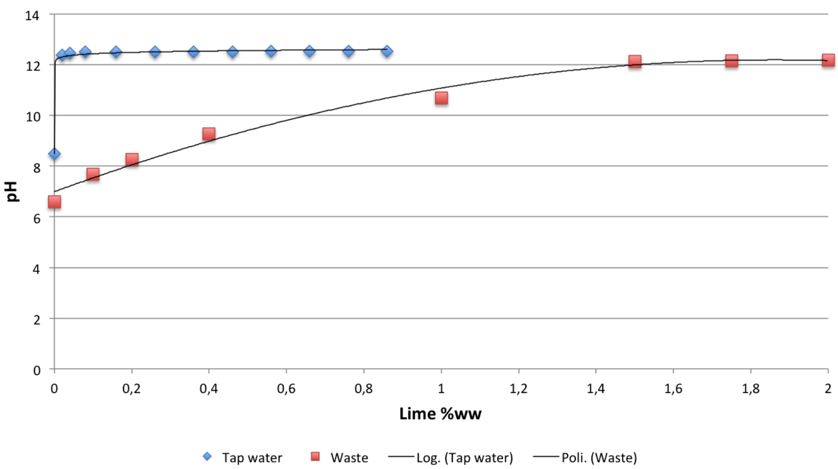

Figure 4.4: pH increase over lime content

12 at the beginning of the experiment. The sample having 1.5% of lime reached pH 12.02 but in less than 30 minutes the pH of this mixture dropped under 12; 1.75% CaO sample maintained a pH higher than 12 for approximately 50 minutes; just the

sample containing 2% CaO maintained a pH higher than 12 for enough time, figure 4.5.

The minimum lime content is 2% on wet weight of waste. However, on-field

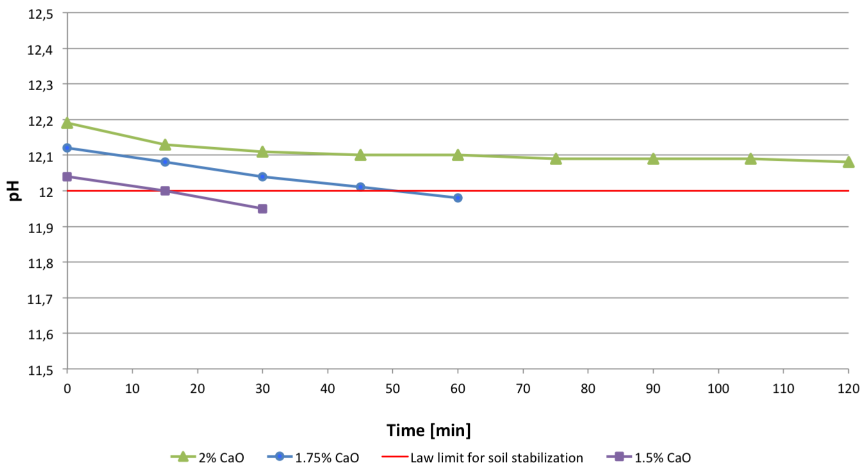

Figure 4.5: pH trend over time of waste-lime mixtures

applications need a higher amount of lime because the calcium oxide used for these applications is less pure and mixing procedures are less effective on field than in the laboratory. In order to ensure a pH higher than 12 for at least 2 hours 5% of lime was used on field.

The second test determined the quantity of cement needed to have a good compressive strength. The tests were performed at a lab- scale on cubic samples of 150x150x150mm. The results of the experiments are shown in table 4.3. The load was applied by steps of 75N each 30s, column 4 of table 4.3 shows the number of steps applied before sample collapse.

For the purposes of the study even the lower value reached ensure slabs collapse

Table 4.3: Compressive strength of waste-cement-lime cubic samples

Cement	**Lime**	**Peak load**	**Steps n**
5 %ww	2 %ww	855	3
8 %ww	2 %ww	1265	4
13 %ww	2 %ww	2565	8
18 %ww	2 %ww	3295	10

resistance. In order to lower costs, a range of cement between 5 and 10% is chosen to build the slabs.

4.4.2 Waste slabs physical properties

Table 4.4: Outputs of the plate load tests

Slab number	**1**	**2**	**3**	**4**
Days of hardening		**Dynamic**	**modulus**	**[MN/m^2]**
7	7.06	7.76	5.88	3.45
14	8.54	8.85	6.36	3.30
23	8.24	7.95	5.91	3.15
50	14.04	12.07	7.99	3.54
		Static	**modulus**	**[MN/m^2]**
55	14.36	10.49	8.29	3.05

The bearing capacity and compaction of the slabs described in the previous chapter (table 4.1) were tested by means of dynamic plate load test and static plate load test. The results of the experiments are provided by table 4.4; static plate load test is more accurate but compromise the structure of the slabs permanently, therefore this test was performed just once after 55 days from the building process, after dynamic plate loads began to record a good performance.

Slab 3 composed by waste and lime (15%w/w) shows lower bearing capacity than

Table 4.5: Permeability tests

Slab n	**1**	**2**	**3**	**4**	**UM**
Lime	5.0	5.0	15.0	0.0	%ww
Cement	10.0	5.0	0.0	0.0	%ww
h	70	90	60	70	mm
H	877	850	870	880	mm
P	150	250	200	200	mm
h	$2.42*10^-6$	$2.12*10^-6$	$2.12*10^-6$	$2.44*10^-6$	cm/s

the other slabs containing cement. The slab of untreated waste reaches the minimum value. In this last case it was visible that the slab was unable to bear the weight of a truck or other vehicles in the landfill, generating management problems. Both slab 1 and 2 showed good bearing capacity. The permeability of the slabs is shown in table 5.5; the best performing slabs are number 2 (5% cement and 5% CaO) and number

3 (15% CaO). The recorded data of the water level and its trend on time clarify the

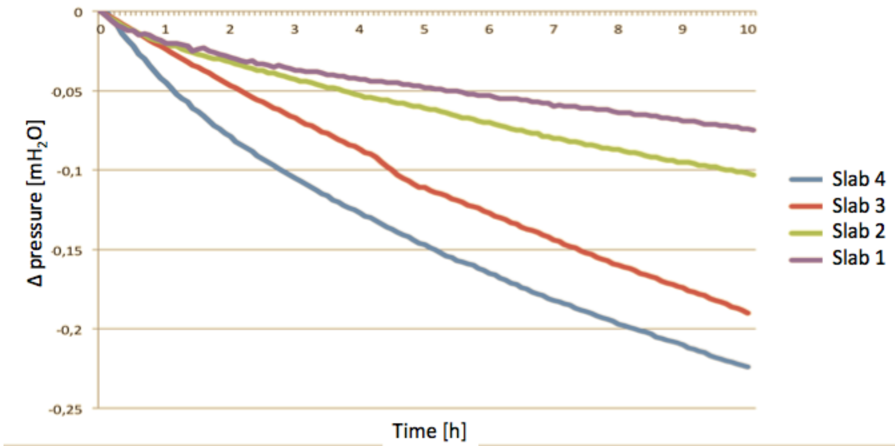

Figure 4.6: Outputs of the permeability tests

results of the permeability test (figure 4.6); the level of the water decreased much faster in slabs 1 (no reagents) and 4 (10% cement and 5% CaO). Water pass through fast in untreated waste because waste particles are less cohesive thus in this case we have the higher coefficient of permeability k than in other cases.
The specific weight of the slabs shown in table 4.6 proves that a volume up to 15% can

Table 4.6: Outputs of specific weight tests

Slab n	**1**	**2**	**3**	**4**	**UM**
Lime	5.0	5.0	15.0	0.0	%ww
Cement	10.0	5.0	0.0	0.0	%ww
Sand weight	2465	1556	2144	1718	g
Sand volume	1611	1017	1401	1123	mL
Refuse volume	2152	1385	1746	1332	g
density	1.34	1.36	1.25	1.19	cm/s

be spared in the landfill in case of S/S technology implementation. The best results are achieved by slab 1 and 2. As landfills are gradually phased out all over Europe, volume reduction is a topic of major interest and it is one of the most important purposes of this study. The costs of S/S chemicals can be covered with the saving of volume into the landfill and related disposal costs.
This set of experiments proved that the mechanical characteristics of the waste are improved by the S/S treatment. The slabs that better fit the technology and the purposes of the study are number 1 and 2, respectively 10% cement, 5% CaO and 5%

cement, 5% CaO. Therefore leaching tests will be performed on samples of slab 1 and 2.

Table 4.7: Outputs of the leaching tests

Parameter	*UM*	*Law limits**	*0%lime 0%cement*	*5%lime 5%cement*	*5%lime 10%cement*
pH		5.5 ÷ 9.5	7.7	9.45	9.9
DOC	mg/l O_2	160	349	437.5	1685
Aluminum	mg /l Al	1	0.864	0.268	0.3085
Arsenic	mg/l As	0.5	<0.08	<0.08	<0.08
Barium	mg/l Ba	20	0.0268	0.02015	0.0785
Boron	mg/l B	2	0.2446	0.04725	0.0524
Cadmium	mg/l Cd	0.02	<0.005	<0.005	<0.005
Total chromium	mg/l Cr	2	0.0095	0.00765	0.01185
Iron	mg Fe/l	2	1.468	0.253	0.158
Manganese	mg Mg/l	2	0.099	0.00865	0.01195
Mercury	mg/l Hg	0.005	<0.003	<0.003	<0.003
Nickel	mg/l Ni	2	0.082	0.06405	0.26025
Lead	mg/l Pb	0.2	<0.05	0.07705	0.1832
Copper	mg/l Cu	0.1	0.2218	0.39895	1.9489
Selenium	mg/l Se	0.03	<0.01	<0.01	<0.01
Tin	mg Sn/l	10	<0.03	<0.03	<0.03
Zinc	mg/l Zn	0.5	0.2923	0.06735	0.05445
Sulfates	mg/l SO_4	1000	322.4	90.45	190.9
Chloride	mg/l Cl	1200	337.6	157.95	292.95
Fluoride	mg/l F	6	<1	<1	<1
Ammonia	mg/l N	11.7	34.8	6.3	7.1
Nitrates	mg/l N	20	3.3	2.26	8.875

* Surface water law limits: table 3 attachment 5 D. Lgs 152/06

4.4.3 Leaching tests

Tank leaching tests were performed on cubic samples of slabs 1 and 2. Moreover a sample of untreated waste was analyzed for comparison to slabs 1 and 2. This test is aimed at comparing the liquid emissions of the best performing slabs described in the previous paragraph and the emissions of untreated waste slabs. Table 4.7 shows the results of the leaching tests and compares them to the Italian law limit for surface water discharge [17].
The sample of untreated waste exceeds law limitations for DOC, copper and ammonia; furthermore aluminum, iron and zinc are high and in case of even small changes of the substrate the threshold could be exceeded. Slab 3 exceeded the threshold of pH. All the samples exceeded law limitation for DOC and copper. Copper in leachate

is mostly bond to organic substances [18] thus, reducing DOC content in the waste will bring both copper and DOC under the threshold. Further investigation proved that the biological stabilization process in the waste treatment line is not effective as supposed. Therefore the administration chose to build a new biological stabilization plant. This should ensure the complete respect of law limits in case of slab 2.
The test outputs prove that the most performing mixture is given by 5% cement and 5% lime on wet weight, thus respirometric index analyses were performed for slab 2 and for slab without reagents to have a comparison.

4.4.4 Respirometric index

The Respirometric Index (RI) can be applied as a tool to define potential biogas generation, a high RI24max (higher value over 24 hours testing) results in high biogas production [14]. The experimental parameters are shown in Table 4, this analysis was applied to the best performing S/S slab (slab 2: 5% cement and 5% lime) and to untreated waste (slab 4). The flat line Fig. 5 represents the RI of the treated waste sample, it shows that there is no bacterial activities when a waste is treated with 5% cement and 5% lime. Therefore the methanogenesis in this slab is blocked and greenhouse gases emissions are stopped or at least drastically lowered.

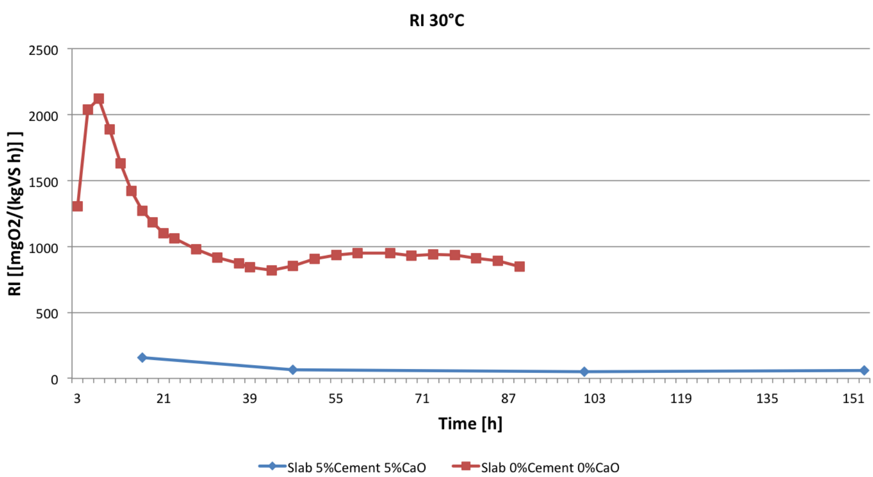

Figure 4.7: Respirometric Index of waste slabs

4.5 Conclusions

S/S technology has been applied to treat MSW in landfill slabs. Preliminary tests proved that calcium oxide is needed to improve physical and mechanic characteristics of the waste. Minimum lime quantity is proven to be 2% on wet weight of waste using RPE calcium oxide in the laboratory. The experimentation continues with the building of slabs on field. The lime used on field was less pure and mixing process was less effective; thus an amount of at least 5%w/w was added to the waste. The slabs having both cement and calcium oxide showed the best physical properties. Leaching tests demonstrated that the best S/S mixture is 5%w/w calcium oxide and 5%w/w cement in order to minimize liquid emissions. The S/S technology is suitable for minimization of GHG emissions as the RI test can be used to estimate biogas production potential.

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Chapter 5

Trentino Sustainable Landfill (TSL) model

A. Limoli; G. Andreottola

5.1 Abstract

The Solidification and Stabilization (S/S) technology studied in the previous chapter has been applied to build a model of sustainable landfill. This model is named after the region, in which it has been applied at a full scale. The TSL model includes a Mechanical Biological Treatment (MBT) step and a S/S step performed using cement and lime; it provided a better chemical physical structure and reduced environmental impacts. The leachate is produced in lower amount and it is less polluted in comparison to a waste treated by only MBT. Moreover the methanogenesis in TSL is drastically lowered by the addition of cement and lime.

Keywords: Sustainable landfill; weste solidification; waste stabilization; cement; lime

5.2 Introduction

The landfill can be defined as an "engineered deposit of waste onto and into land in such a way that pollution and harm to the environment is prevented and through restoration, land provided which may be used for another purpose" [1].
In order to minimize the pollution leaking into the environment three different approaches for landfill management have been proposed. Landfill disposal in uncontrolled sites defined as open dump has the advantage of a fast self-depuration, however the risks to the environment, human health and society are high and can be compared to the ones analyzed for illegal landfill in chapter 2. This landfill practice

is nowadays obsolete. Therefore in more recent years, the driving principle of landfill management has been to prevent emissions into the environment and to reduce the rate of leachate leaking. Entombment landfills fulfill this objective by means of compacted soil and plastic sheets; containment landfills have physical barriers and collection system for leachate and biogas [2]. This has resulted in low rates of waste degradation, thus the complete stabilization time reaches hundreds of years [3]. Moreover the containment and entombment approaches are no longer effective in case of damaged shells. These types of landfills can show a high pollution rate after closure. Therefore, the need of a new landfill type has been developed by Allen in 2001. A sustainable landfill "should encompass the following basic principles:

- Reduction in the generation of waste.
- Waste streaming at source.
- Recycling and reuse.
- Pre-treatment of waste to minimize quantity and volume." [4]

The Solidification/Stabilization (S/S) technology is a pre-landfill waste treatment process, which uses chemically reactive formulations that, together with water, form stable solids. The S/S process consists in encapsulating the waste in a solid matrix, improving the physical characteristics of the waste and reducing the mobility of the hazardous compounds, thus less contaminants are freed into the environment. The result of this process is a less hazardous solid, which shows also a higher density. S/S pre-landfill treatment inhibits the biogas production.

The S/S pre-treatment fulfill the objective disclosed by Allen. Therefore this study focuses on the comparison of different pre- landfill treatment scenarios to enhance waste stability and volume reduction. The best performing pre-treatment is the Mechanical Biological Treatment (MBT) coupled with S/S technology, this treatment scheme is defined as Trentino sustainable landfill (TSL) model. A sustainable landfill pre-treatment, which has been widely applied, is Mechanical Biological Treatment (MBT) [2,5–7]. However this study shows how coupling MBT and S/S pre-treatment improves the reduction of volume occupied in the landfill, moreover it drops the biogas production at negligible levels and significantly reduces leachate emissions into groundwater.

The emissions of bio-stabilized waste slabs and the enhanced process by means of S/S technology will be analyzed and the positive effect S/S on waste stabilization will be proven. The TSL model is as well cost-effective and easy to put into practice..

5.3 Materials and methods

5.3.1 Chemicals and waste

90% CaO w/w lime was used to build the S/S treated slabs on field, 5% of lime on waste wet weigh. 5% on waste wet weigh of Portland cement CEM II/B-LL 32,5 R was used. Tap water was added to enhance pozzolanic reaction.

The waste was sampled from a Mechanical Biological Treatment (MBT) plant, which treats the residual waste of the province of Trento. The amount of the recycled material in relation to the residual waste increased over years reaching 81.3% in 2017. The residual fraction (18.7%, 60Gg in 2017) reaches the treatment plant; it is ground and sieved using a 50 mm sieve. The upper-grid goes to a waste-to-energy plant while the lower-grid is stabilized in the MTB plant (7.2Gg in 2017). The stabilized waste is than sieved using a 20 mm sieve. The fraction > 20mm goes to a waste-to-energy plant while the fraction < 20 is used in the experiments.

5.3.2 Waste slabs building

The waste slabs were built as described in table 5.1 and set as shown in figure 5.1.
For the purpose of this paper, only the slab 4 (no reagent) and the slab 2 (best

Table 5.1: Slabs composition

	TSL	MBT	UM
Waste	5700	5100	kg
Water	27.4	26.8	%ww
Lime	5.0	0.0	%ww
Cement	5.0	0.0	%ww

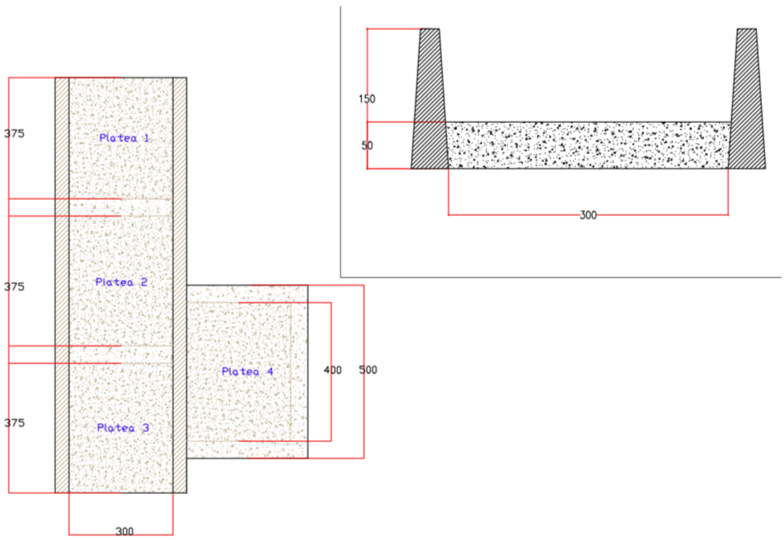

Figure 5.1: Waste slabs geometry; TSL slab 1, MBT slab 4

performing slab, as proven in the previous chapter) will be tested.
The comparison of the effectiveness will be performed on TSL model (MBT+S/S)

and on MBT lower-grid waste.

5.3.3 Waste characterization

The waste has been sieved at a 4.75mm mesh: the fraction smaller than 4.75mm has been weighted while the fraction >4.75mm has been sorted according to classes defined by the EPA municipal waste characterization procedure [8].

5.3.4 Leachate composition

The liquid emissions have been estimated by means of leaching tests comparing emissions of slabs with and without S/S treatment. Moreover permeability tests have been performed to compare the potential amount of leachate produced.
MSW landfilled without pre-treatment causes twice to four times as strong as equivalent leachates from MBT sustainable landfills, and high organic strengths can persist for several decades [9]. Efficient MBT can considerably reduce the organic strength of leachates, however the reduction of pollutant is not complete and S/S technology can improve both the encapsulation of pollutants and the impermeability of the waste producing less leachate in the landfill body.
The permeability of the slabs was detected by falling head permeability tests in boreholes each performed in duplicate after 50- 55 days from building. We prepared a hole with constant diameter of 120 mm, where a PVC tube was pushed down and fixed with bentonite clay. The tube was filled with water and a water level sensor, which records levels each 30s, was immersed. The formulas for calculating permeability from these tests are assessed by the British standard Code of practice for site investigations BS 5930:1999 Method 1 (after Hvorslev) configuration D, where the intake factor is chosen as described in the following equation 5.3.2:

$$F = 2\pi d + h \tag{5.3.1}$$

Tank leaching tests were performed on cubic samples made of the materials used to build the slabs with and without S/S pre-treatment. This test is aimed at comparing the liquid emissions of the TSL slabs and the emissions of MBT waste slabs. The leaching apparatus is composed by a tank, in which a cubic sample (150x150x150 mm) is immersed in distilled water, a recirculation pump and a tripod to hold the sample (figure 5.2)..

The samples of leachate were collected at the end of the experiment after 24h. The samples of the leaching test were filtered through 0.45 mm Millipore filter papers before being analyzed. Analyses of metals were performed according to EPA Compendium Method IO- 3.5. Total Ammonium Nitrogen (TAN) and nitrates analyses were performed with a spectrophotometer, according to, respectively, EPA method 350.1 and 352.1. Chemical Oxygen Demand (COD) was analyzed by titration following EPA Standard Method 410.3. Inorganic ions were determined by ion chromatography according to EPA Standard Methods 300.0 [10].
The pH values were assessed by a pH-meter with a resolution of 0.01. The probe was

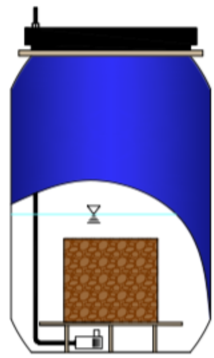

Figure 5.2: Leaching test apparatus

cleaned and calibrated before using.

5.3.5 Use of respirometric test for the assessment of biogas production potential

The respirometric index can be applied as a tool to define potential biogas generation [12]. To analyze the bacterial activity an AIR-A respirometer (Open Respirometric Index Analyzer) was used [13]. The experimental parameters are shown in table 5.2, this analysis was applied to the waste treated by TSL model and to MBT waste (lower-grid of MTB plant). The samples were collected from the slabs after 55 days of hardening.

The potential biogas production of the MBT waste has been estimated as Scaglia et al. proposed in 2009 using a respirometric approach and compared to the emissions of the TSL waste slab. The linear regression model, obtained by a jackknife approach

Table 5.2: Respirometric Index parameters

Slab n	**2**	**4**	**UM**
Temperature	30.8	30.9	° C
Natural humidity	46.4	32.7	%w w
Revised humidity	46.4	44.0	%ww
Sample weight	1.0	1.0	kg
Total Solids	53.6	67.6	%ww
Volatile Solids	30.5	27.3	%STww

is shown in equation 5.3.2:

$$ABP = (34.4 \pm 2.5) + (0.109 \pm 0.003) * DRI \qquad (5.3.2)$$

Where ABP is the estimated anaerobic biogas potential and DRI is the dynamic respiration index defined as

$$DRI = \frac{\sum_{h=0}^{24} DRI_h}{24}; [11] \qquad (5.3.3)$$

The total solids (TS) has been measured in agreement with UNI EN 14346:2007 and the volatile solids (expressed in percent of dry matter) have been detected by means of the procedure described in UNI EN 15403 2011.

5.3.6 Scale-up

The TSL model will be evaluated and compared to MBT waste deposition in landfills. The TSL model will be explained; the costs and technology used will be set. The prices are derived from the Italian statistics institute (Istat), which sets price for cement about 100€/t and lime 95€/t [14]. The prices of landfill discharge per cubic meter is about 110€ in Trentino Alto Adige (a region in the north of Italy in which the study took place) and 89€ in Italy [15]. In order to compare costs for both treatment options (MBT and TSL) the density of TSL slabs and MBT waste slabs was measured 55 days after building of the slabs. Holes were dug into the slabs; the waste mixture was collected and weighted; than the hole was filled with sand which density was previously calculated in the laboratory by filling 1-liter jar and measuring the weight. The sand was put in a bucket and weighted; the amount of sand needed to fill each hole was calculated by subtracting the weight of the bucket after filling the hole to the initial bucket weight.

5.4 Results and discussions

5.4.1 Waste characterization

The influent waste to biological aerobic treatment plant is the fraction < 20mm of the sieved raw waste. The result of the characterization analysis is shown in figure 5.3.

The main fraction of the sieved waste has a size smaller than 4.75 mm; this fraction is visually identified as mixed organic fraction. Summing this fraction with paper, textiles, wood and organic waste the total biodegradable waste fraction is about 82%. The waste organic content is high, thus the potential greenhouse gases (GHGs) emission is sizeable. The main objective of a sustainable landfill is to lower GHGs emissions and to have a stable substrate into the landfill body, thus the raw sieved waste follows a biological aerobic treatment [9,16,17].

An accelerated bio-drying process able to reduce humidity will treat the fraction $<$ 20 mm. The major components of this fraction have a high calorific value (plastic and paper). Therefore it is used as refuse-derived fuel (RDF).

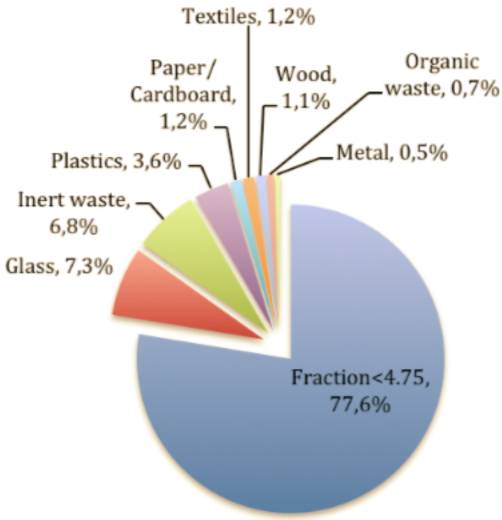

Figure 5.3: Waste characterization

5.4.2 Leachate composition

The leachate produced by the two waste slabs with and without S/S pre-treatment has been analyzed, both permeability and leaching tests results show that the performance of the S/S pre-treated slab is higher than the slab of MBT lower-grid waste. Water pass through fast in MBT waste because waste particles are less cohesive thus in this case we have a high coefficient of permeability k: the coefficient of S/S pre-treated slab is of 15% lower than the one for MBT waste slab, see table 5.3.

The leaching test is aimed at comparing the liquid emissions of the TSL (MBT+S/S)

Table 5.3: Outputs of the permeability tests

	TSL model	MBT pre-treatment	UM
h	70	90	mm
H	880	850	mm
P	200	250	mm
h	2.44*10⁻6	2.12*10⁻6	cm/s

treated waste slabs and the emissions of MBT waste slabs. Table 5.4 shows the results of the leaching tests and compares them to the Italian law limit for surface water discharge [18]. The sample of MBT waste exceeds law limitations for DOC, copper and ammonia; furthermore aluminum, iron and zinc are high and in case of even small changes of the substrate the threshold could be exceeded. Both the samples

exceeded law limitation for DOC and copper. Copper in leachate is mostly bond to organic substances [19] thus, reducing DOC content in the waste will bring both copper and DOC under the threshold. Further investigation proved that the biological stabilization process in the waste treatment line is not effective as supposed. Therefore the administration chose to build a new biological stabilization plant. This should ensure the complete respect of law limits in case of TSL (MBT+S/S) pre-treatment. However even the best MBT have significant DOC levels [9], thus a S/S treatment is needed in order to reduce organic pollution in liquid phases.
The leachate produced in the landfill body can be used as hydration in the S/S phase and/or discharged into the environment after a phytodepuration step because of low pollutant quantities.

Table 5.4: Outputs of the leaching tests

Parameter	*UM*	*Law limits**	*0%lime 0%cement*	*5%lime 5%cement*	*5%lime 10%cement*
pH		5.5 ÷ 9.5	7.7	9.45	9.9
DOC	mg/l O_2	160	349	437.5	1685
Aluminum	mg /l Al	1	0.864	0.268	0.3085
Arsenic	mg/l As	0.5	<0.08	<0.08	<0.08
Barium	mg/l Ba	20	0.0268	0.02015	0.0785
Boron	mg/l B	2	0.2446	0.04725	0.0524
Cadmium	mg/l Cd	0.02	<0.005	<0.005	<0.005
Total chromium	mg/l Cr	2	0.0095	0.00765	0.01185
Iron	mg Fe/l	2	1.468	0.253	0.158
Manganese	mg Mg/l	2	0.099	0.00865	0.01195
Mercury	mg/l Hg	0.005	<0.003	<0.003	<0.003
Nickel	mg/l Ni	2	0.082	0.06405	0.26025
Lead	mg/l Pb	0.2	<0.05	0.07705	0.1832
Copper	mg/l Cu	0.1	0.2218	0.39895	1.9489
Selenium	mg/l Se	0.03	<0.01	<0.01	<0.01
Tin	mg Sn/l	10	<0.03	<0.03	<0.03
Zinc	mg/l Zn	0.5	0.2923	0.06735	0.05445
Sulfates	mg/l SO_4	1000	322.4	90.45	190.9
Chloride	mg/l Cl	1200	337.6	157.95	292.95
Fluoride	mg/l F	6	<1	<1	<1
Ammonia	mg/l N	11.7	34.8	6.3	7.1
Nitrates	mg/l N	20	3.3	2.26	8.875

* Surface water law limits: table 3 attachment 5 D. Lgs 152/06

5.4.3 Respirometric tests for the assessment of biogas production potential

The Respirometric Index (RI) can be applied as a tool to define potential biogas generation, a high RI24max results in high biogas production [12]. The experimental parameters are shown in table 5.4, this analysis was applied to the TSL slab (5% cement and 5% lime) and to MBT slab (0% cement and 0% lime).

The ABP calculated for the MBT slab is related to the average RI24 1470

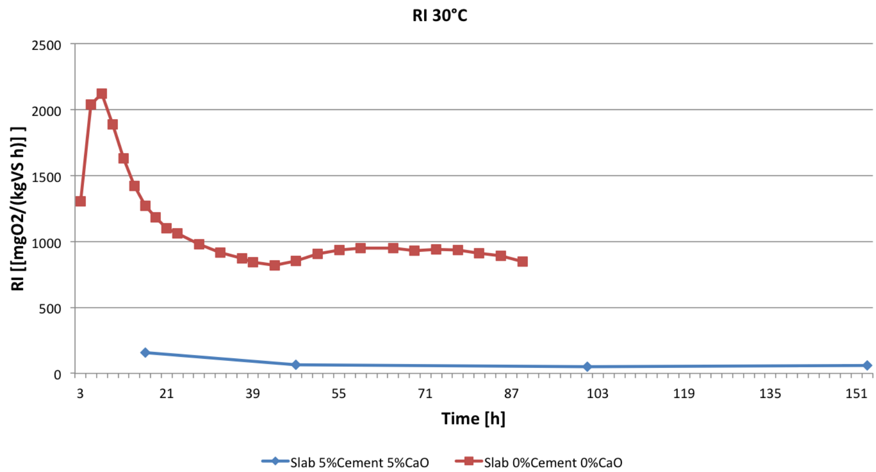

Figure 5.4: Respirometric Index of waste slabs

mgO2/(kgVS h) and reach a value of 78.1 ± 3.7 Nl/kgTS or 0,078 ± 0,004 Nm3/kgST was found to lie within the range recommended by Bogner and Spokas [20]. This amount of biogas is not produced in case of S/S pre-treatment. The pH recommended range for methanogenesis is 6.5-8 [21], the basic environment of S/S slab prevent biogas formation. Moreover the flat line in figure 5.4 represents the RI of the treated waste sample, it shows that there is no bacterial activities when a waste is treated with 5% cement and 5% lime. Therefore the methanogenesis in this slab is blocked and greenhouse gases emissions are stopped or at least drastically lowered.
The pH has been found to be the strongest predictor of the methane production rate, strong alkaline pH reduces strongly the methanogenesis [22]. Amoozegar et al. tested waste reactor provided with covers containing various amounts of lime; significant volumes of methane were found to be produced only in the reactor without lime in the cover material [23]. The tests were performed for over 250 days, in order to verify this result in the study case a further experimentation should be performed. The application at site of TSL will provide the informations about the real methane production on a longer time scale.

5.4.4 Scale-up

The TSL model has been applied at a full-scale in order to evaluate reduction of the volume occupied in the landfill body. The bulk density of the MBT and TSL slab was measured and the values are respectively of 1.19 and 1.26 t/m3, thus reduction was about 15%. This reduction lead to a decrease of landfilling costs. Both lime and cement content was set to 5% in weight of wet waste.
The comprehensive costs of the MBT and TSL treatment schemes are compared in table 5.5, the costs of sieving and biological aerobic treatment are not reported because they are used in both schemes. The equipment needed to perform the solidification and stabilization process is low cost and easy to handle, instead of a truck to move the waste from the MBT to the landfill site a truck mixer is used, with negligible cost increase.

It is interesting to evaluate that the cost of reagents is completely covered by the

Table 5.5: Treatment costs

	TSL model	**MBT pre-treatment**	**Unit cost**
Landfilling	80.80€/twaste	92.73€/tww	110€/m^3
Lime	4.45€/twaste	0.00€/twaste	89€/t
Cement	5.00€/twaste	0.00€/twaste	89€/t
TOTAL	90.25€/twaste	92.73€/twaste	

reduction of volume occupied in the landfill site as settled in table 5.5.
The Trentino sustainable landfill (TSL) model involve both MBT and S/S pre-treatment as shown in figure 5.5. The TSL model has the advantage to be low-cost, to have a good performance in volume saving, to reduce landfill gas emissions at negligible levels and to form less leachate that is proven to be less polluted as well. The leachate can be used as hydration water in the S/S step and/or discharged after a potential phytodepuration step. The mechanical performance of the slabs proves that the slabs can easily carry the weight of the machines needed to perform the treatment (truck mixer and steamroller).

5.5 Conclusions

TSL model has been defined as the coupling of MBT and S/S pre- treatment (5%w/w calcium oxide and 5%w/w). The MBT alone is not able to stop the landfill gas emissions and the leachate produced shows higher pollutant concentration. Moreover the leachate is produced in higher amount because of the waste permeability; the coefficient of permeability k of TSL slab is 15% lower than the one for MBT waste slab. Leaching tests demonstrated that the TSL treated waste has lower liquid pollutant

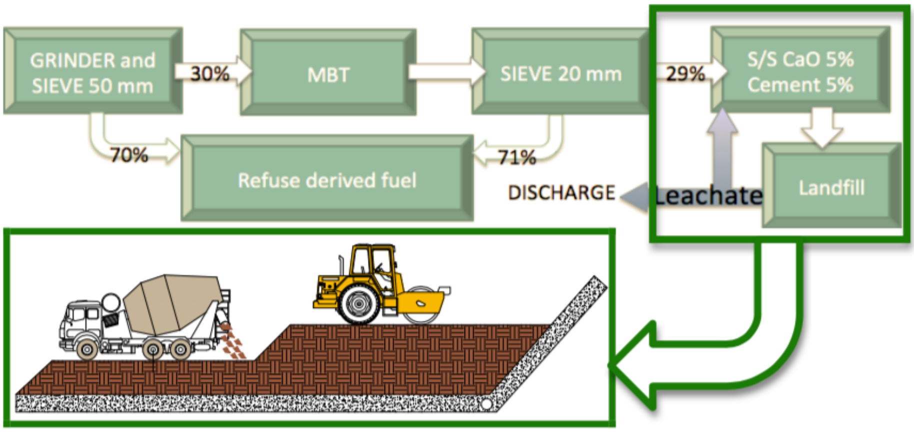

Figure 5.5: TSL plant scheme

emissions. RI tests proved that TSL model is suitable for minimization of GHG emissions. The ABP calculated for the MBT slab is 0,078 ± 0,004 Nm^3/kgST. This amount of biogas is not produced in case of TSL treatment. The TSL treatment plant is proven to be low cost as the volume reduction of waste slabs pays off the costs of lime and cement. The volume reduction was about 15%. The TSL is easy to implement and it is proven to have a very high performance in GHG and leachate emissions reduction.

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Chapter 6

Conclusions and future developments

6.1 Conclusions

To date municipal solid waste landfilling is a big issue, because of the harmful effects of the pollutant emitted from the landfill body. The rising concern about the impacts of landfilling led to specific law regulations, that prescribe duties and sanctions. Recent changes to legislation concern the protection of the environment through criminal law introducing the crimes against the environment. The duties and sanctions prescribed by the Italian law can be summarized as follows:

- Criminal or administrative sanctions for illegal landfilling
- Duty of environmental remediation of the contaminated sites
- Compensations to the aggrieved parties
- Duty to check the conformity of waste with the leaching thresholds for MSW landfilling

The main environmental impacts related to liquid and gaseous emission in landfill sites can be:

- Groundwater quality degradation
- Eutrophication
- Acidification
- Phytotoxicity and aquatic toxicity
- Oxygen depletion in the root zone
- Animal toxicity and bioaccumulation
- Greenhouse effect and global warming.

It appears clear that the pollution generated by MSW landfills can affect the air, the bodies of water, the groundwater the soil the flora and the fauna. The risk depends on the design of the landfill. The most harmful are illegal landfills and open dumps as these designs lack of pollution containment systems. Entombment and containment landfills can show similar impacts in case of problems with the isolation systems. Landfill pollutants have also harmful effects on human beings raising the risk of illness such as:

- Cancer
- Infectious diseases
- Toxic effects on endocrine, reproductive and nervous system
- Respiratory and cardiovascular problems

The main environmental problem of aged landfills is the leachate pollution as the GHGs emissions decrease over time. Into the leachate ammonia increases over time and metals are usually present for several decades. The leachate may rich the aquifer spreading pollutants in large areas. To date technologies able to remove or contain ammonia and metals pollution into groundwater are expensive (i.e. pump and treat) or hard to perform (hydraulic barriers). This work proved the efficiency of the biosparging process as an in- situ technology for bioremediation of a leachate-contaminated site. The advantages of the process can be summarized as follows:

- Biosparging stimulates the growth of indigenous microorganisms able to convert pollutants into harmless substances;
- The average ammonia removal in the whole system is 99.8%;
- TWith a proper organic carbon dosage the nitrates, produced in the nitrification phase, are almost completely removed;
- Biosparging favors the immobilization of some metals such as manganese and arsenic, preventing heavy metal contamination in groundwater.

The process can be applied at a real scale on site, without transporting large quantities of leachate off site: procedure that arise the costs and the potential of spills, with a subsequent threat to the human health and the environment. Landfills that lack of waste stabilization pre-treatment can cause longer and more severe pollution. Open dumps, containment and entombment landfills can be a threat to the environment and human health, as they can cause severe and extended impacts. Therefore, new types of landfills are studied; to date sustainable landfilling of municipal solid waste appear to be the most effective in waste stabilization and reduction of volume. Mechanical biological treatment (MBT) is used as a pre-landfill step, in order to lower emissions in sustainable landfill design. This pre-treatment reduces environmental pollution; however, MBT does not block completely the green house gases production and leachate emissions. The purpose of this work was to analyze and minimize the harmful effects of landfills. Therefore a further step of the experimentation was the definition of a new model of sustainable landfill. The model, namely Trentino Sustainable Landfill (TSL), involves a step of mechanical biological treatment followed by a treatment of solidification and stabilization (S/S). S/S technology has been applied to treat MSW in landfill slabs. Preliminary tests proved that calcium oxide is needed to improve physical and mechanic characteristics of the waste. Minimum lime quantity is proven to be 2% on wet weight of waste using RPE calcium oxide in the laboratory. The experimentation continued with the building of slabs on field. The lime used on field was less pure and the mixing process was less effective thus an amount of about 5%w/w was added to the waste. The slabs having both cement and calcium oxide showed the best physical properties. Leaching tests demonstrated that the best S/S mixture is 5%w/w calcium oxide and 5%w/w cement, in order to minimize liquid emissions. The Trentino sustainable landfill model involves both MBT and S/S pre-treatment as shown in figure 6.1. The TSL model shows the following advantages:

- Methanogenesis inhibition resulting in landfill gas emissions at negligible levels,
- Leachate is produced in lower quantities and it is proven to be less polluted than in case of MBT waste landfilling.
- Low-cost treatments and reagents,
- Good performance in volume saving,

The leachate can be used as hydration water in the S/S step and/or discharged after a potential phytodepuration step. The mechanical performance of the slabs proved that the slabs could easily carry the weight of the machines needed to perform the treatment (truck mixer and steamroller). The TSL model has been tested on field

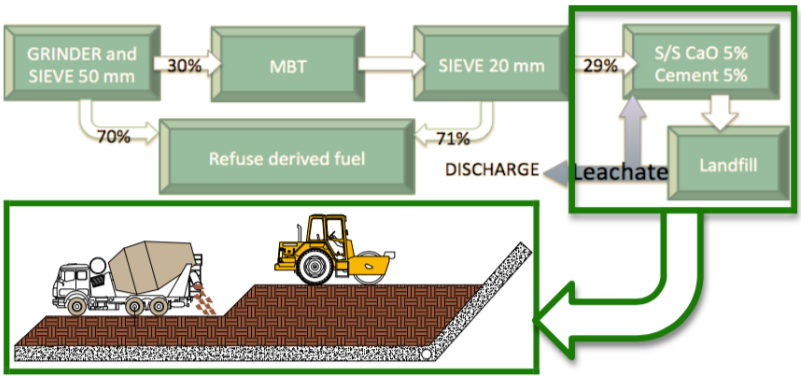

Figure 6.1: TSL plant scheme

in waste slabs and it has been compared with a similar model involving the MBT process but without the S/S step (namely MBT slab).
The MBT alone is not able to stop the landfill gas emissions and the leachate produced shows higher pollutant concentration. Moreover the leachate is produced in higher amounts because of the waste permeability; the coefficient of permeability k of TSL slab is 15% lower than the one for MBT slab. Leaching tests demonstrated that the TSL treated waste shows lower liquid pollutant emissions. RI tests proved that TSL model is suitable for minimization of GHG emissions. The ABP calculated for the MBT slab is 0,078 ± 0,004 Nm3/kgST. This quantity of biogas is not produced in case of TSL treatment. The TSL treatment plant is proven to be low cost, as the volume reduction of waste slabs pays off the costs of lime and cement, the volume reduction was about 15%. The TSL is easy to implement and it's proven to have a very high performance in GHG and leachate emissions reduction, thus it is proven to

be an effective model of sustainable landfill.

6.2 Future developments

The TSL model will be applied at a full scale in a landfill of the autonomous province of Trento. A slot of the Trentino sustainable landfill will be set and prepared for a verification of the thesis outputs. The leachate produced in the slot will be analyzed and the amount of leachate produced will be measured. The slot will be, as well, useful to verify the physical property and the methane production of the S/S waste slabs. The trend over time of these properties will be analyzed in a medium/long term. The application of TSL on site will provide informations about the quality of the real leachate produced on a time scale longer than 24 hours. The design of landfill leachate-collection filters will be dependent on the characteristics of the leachate.

Finally the stage of MBT will be optimized in order to reduce the organic contamination. The optimization of this treatment step will reduce copper leaching, as copper is bond to organic substances.The new MBT plant is already under construction.

The optimization of MBT and the medium/long term analysis of leachate properties and methane production will guarantee the effectiveness of the TSL model.

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The deposition of waste in a landfill can be a threat to the environment and human health; in spite of their potential pollution, landfill are still of grate use, thus efficient and cost effective technologies need to be studied in order to minimize aqueous and gaseous emissions. The present work focuses on the evaluation of the remediation of old landfill sites that pollutes groundwater and on the determination of a new treatment of fresh waste upstream of landfilling. The biosparging technology has been applied to remediate an aquifer polluted by leachate; it stimulates the growth of indigenous bacteria able to convert pollutants in harmless compounds. The technology shows high efficiency in ammonium nitrogen removal via nitrification processes and in the immobilization of metals. The application of the biosparging on site has proven to be feasible. Later a technology able to reduce fresh residual waste has been studied. The Solidification/Stabilization (S/S) technology is a pre-landfill waste treatment process, which has been applied to different types of hazardous wastes since it has a proved efficiency on heavy metal immobilization. The S/S process blocks methanogenesis reducing GHG emissions into the atmosphere. The S/S process improves the physical characteristics of the waste and reduces the mobility of the hazardous compounds, thus the waste leaches less contaminants into the environment. The result of this process is a less hazardous solid. The experimental evidences proved that this technology reduces volumes and costs. The reduced permeability and the leaching test results show that the leachate produced is of a smaller amount and less polluted. The enhanced mechanical properties and the reduced emissions both in bodies of water and atmosphere have proven the worth of this technology. Therefore the Trentino Sustainamle Landfill (TSL) model is proposed. The TSL involves a mechanical biological treatment and a S/S step.

Alice Limoli is a PhD student at the Department of Civil, Environmental and Mechanical Engineering (University of Trento, Italy) where she received her MSc in environmental engineering (2014). She was visiting student at the University of Innsbruck where she contributed to waste management projects (2012-2013). She worked as research assistant in industrial and environmental engineering (2015). She studied chloroform removal techniques from wastewater and published an article on the topic of ammonia stripping. Her research interests include solid waste management and wastewater treatment; she published an article on the topic of environmental forensics about illegal landfills.