

COMPREHENSIVE META ANALYSIS OF WORLDWIDE LANDFILL
LEACHATE

A THESIS SUBMITTED TO
THE GRADUATE SCHOOL OF NATURAL AND APPLIED SCIENCES
OF
MIDDLE EAST TECHNICAL UNIVERSITY

BY

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IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR
THE DEGREE OF DOCTOR OF PHILOSOPHY
IN
ENVIRONMENTAL ENGINEERING

JUNE 2022

Approval of the thesis:

**COMPREHENSIVE META ANALYSIS OF WORLDWIDE LANDFILL
LEACHATE**

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ABSTRACT

COMPREHENSIVE META ANALYSIS OF WORLDWIDE LANDFILL LEACHATE

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June 2022, 257 pages

Landfill leachate data compiled from 220 different landfills from 46 countries in Europe, Middle East, Asia, Africa, and America were analysed by multivariate statistical approach for the assessment of main properties as well as variation of leachate characteristics with respect to landfill age, climate, and development status. Results showed that inorganic content of leachate has the highest correlations with each other as well as accounting for the majority of variation in leachate. Changes in leachate composition were observed in statistically significant manner with respect to climate, landfill age, and development status only for some parameters including pH, BOD, COD, and BOD/COD ratio. On the other hand, most of the parameters particularly inorganics did not show any statistically important difference between impact groups. Change in leachate quality with time was found significant only in relatively low precipitation climates. Landfill age effect was observed on the organic content of leachate while most of the inorganics were not found statistically different with time. On the contrary, in high precipitation climates, organic content and biodegradability were found low without any time effect, whereas inorganic content was found fluctuating in high quantities. In that respect, leachate age definition in current literature as young, medium, and old needs to be re-evaluated by taking into consideration the climate impact with particular attention to precipitation rates.

Comprehensive leachate database and results of statistical analyses suggest that leachate management methods should be evaluated differently for landfills in high and low rainfall climates as well as in developed and developing countries.

Keywords: Landfill Leachate, Regression Modeling, Multivariate Analysis, PCA, ANOVA



ÖZ

DÜNYA GENELİNDE KATI ATIK DEPOLAMA SAHASI SIZINTI SUYUNUN KAPSAMLI META ANALİZİ

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Haziran 2022, 257 sayfa

Avrupa, Orta Doğu, Asya, Afrika ve Amerika'da yer alan 46 ülkedeki 220 farklı katı atık depolama sahasından derlenen sızıntı suyu verileri, temel özelliklerinin yanı sıra depolama sahası yaşı, yerel iklim ve ülkenin gelişme durumu faktörlerine göre çok değişkenli analiz yöntemleri ile incelenmiştir. Sonuçlar, sızıntı suyunun inorganik içeriğinin birbirleriyle en yüksek korelasyona sahip olduğunu ve sızıntı suyundaki değişimin çoğunluğunu açıkladığını göstermiştir. Sadece pH, BOİ, KOİ ve BOİ/KOİ oranı gibi bazı parametreler için sızıntı suyu içeriğindeki farklılıklar iklim, depolama yaşı ve gelişme durumu faktörlerine göre istatistiksel olarak anlamlı bulunmuştur. Öte yandan, özellikle inorganik parametrelerin çoğu, etki grupları arasında istatistiksel olarak önemli bir fark göstermemiştir. Sızıntı suyu kalitesindeki zamana göre değişim, yalnızca nispeten düşük yağışlı iklimlerde istatistiksel olarak anlamlı bulunmuştur. Sızıntı suyunun organik içeriği depolama yaşına göre değişiklik gösterirken, inorganiklerin çoğu zamana bağlı istatistiksel olarak anlamlı bir farklılık göstermemiştir. Ancak, yüksek yağışlı iklimlerde, organik içerik ve biyo-çözünürlük zamana bağlı olmaksızın düşük bulunurken, inorganik içeriğin yüksek konsantrasyonlarda dalgalandığı görülmüştür. Bu bağlamda, mevcut literatürde

sızıntı suyunun genç, orta ve yaşlı olarak tanımının, iklim etkisi ve özellikle yağış oranları dikkate alınarak yeniden değerlendirilmesi gerekmektedir. Kapsamlı sızıntı suyu veri tabanı ve elde edilen istatistiksel sonuçlar, sızıntı suyu yönetiminin, yüksek ve düşük yağışlı iklimlerde ve gelişmiş ve gelişmekte olan ülkelerde farklı değerlendirilmesi gerektiğini göstermektedir.

Anahtar Kelimeler: Sızıntı Suyu, Regresyon Modelleme, Çok Değişkenli Analiz, PCA, ANOVA





Bahar...

ACKNOWLEDGEMENTS

I wish to express my deepest gratitude to my supervisor Prof. Dr. F. Dilek Sanin and co-supervisor Prof. Dr. Ayşegül Aksoy for their guidance, advice, invaluable comments, and patience throughout the research.

I am also thankful to my thesis monitoring committee members: Prof. Dr. İpek İmamoğlu and Assoc. Prof. Dr. Güray Doğan for their supporting manners and valuable comments. Moreover, I wish to thank my other Defense Juri Members Prof. Dr. Müfide Banar and Assoc. Prof. Dr. Emre Alp for providing fruitful comments/evaluations to improve my dissertation.

I would like to express my sincere gratitude to Mrs. Buğçe Doğan Çimentepe from Ministry of Environment Urbanization and Climate Change for her support and understanding to complete my thesis. Also, I would like to mention my colleague Ms. Selen Gültekin for offering her helps whenever I needed. They are valuable engineers graduated from METU and deserve my sincere gratitudes.

Finally, I wish to thank my family for their support and for sacrificing their time to finish my thesis.

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LIST OF ABBREVIATIONS

ABBREVIATIONS

EPA	Environmental Protection Agency
EU	European Union
COD	Chemical Oxygen Demand
BOD	Biological Oxygen Demand
TOC	Total Organic Carbon
NH ₄ -N	Ammonium Nitrogen
TKN	Total Kjeldahl Nitrogen
TN	Total Nitrogen
SS	Suspended Solids
TDS	Total Dissolved Solids
TS	Total Solids
EC	Electrical Conductivity
XOCs	Xenobiotic Organic Compounds
EDCs	Endocrine Disrupting Compounds
TA	Total Alkalinity
PCBs	Polychlorinated Biphenyls
PAHs	Polycyclic Aromatic Hydrocarbon
MSW	Municipal Solid Waste
PCA	Principle Component Analysis
ANOVA	Analysis of Variance
SPSS	Statistical Package for Social Sciences
VIF	Variance Inflation Factor
HCA	Hierarchical Cluster Analysis
KMO	Kaiser-Meyer-Olkin
PI	Prediction Interval

MSA	Kaiser's Measures of Sampling Adequacy
PC	Principal Components
FAO	Food and Agricultural Organization
CI	Confidence Interval
MBT	Mechanical biological treatment



LIST OF SYMBOLS

SYMBOLS

μ	Mean of a sample
σ	Standard deviation
z	Z-score
H_0	Null hypothesis
H_1	Alternative hypothesis
p	Significance level
R^2	Coefficient of determination
S	Standard error of the estimate
r	Pearson's product moment correlation coefficient
N	Number of sample data
R	Coefficient of multiple correlation

CHAPTER 1

INTRODUCTION

Solid waste disposal in sanitary landfills or uncontrolled dumpsites is a widespread method in the World due to being an affordable and convenient solution in comparison to other disposal methods (Luo et al., 2020; Vaccari et al., 2019). This is mainly due to some problems associated with other waste disposal and management methods, especially for developing countries (Mmereki et al., 2016). Even in developed countries, issues related to current and emerging waste management alternatives may necessitate the continuation of the use of landfills. For example, combustion may provide significant benefits such as considerable mass and volume reduction, possible recovery of energy, less retention times, etc. (Robinson et al., 2004). In developing countries generally organic fraction and moisture content of the waste are high that results in difficulty in combustion without an external fuel (Nanda and Berruti, 2021; Ye et al., 2011). For such wastes, composting and digestion methods can be employed. However, management and implementation may require improved waste collection, handling, and pre-treatment which may not be applicable in many developing countries (Mmereki et al., 2016; U.S. EPA, 1995).

In Europe, significant variations are observed in the approaches used for waste management and disposal. For example, waste incineration and composting rates are increasing in Denmark, Sweden, France, Germany, Belgium, and Austria. On the other hand, many other European countries do not have any incineration and composting facilities at all (EU, 2017; Karak et al., 2012). Therefore, landfilling is still employed. It was indicated that around 70% of the total municipal waste collected in the world is landfilled (Ma et al., 2022). In many developing countries, municipal solid waste (MSW) is disposed mainly in sanitary landfills or uncontrolled dumpsites. Vaccari et al. (2019) stated that about 40% of the World's waste goes to the uncontrolled

dumpsites serving around 3-4 billion people. Most of the dumpsites and unsanitary landfills are located in Africa, Latin America, and Asia. In Turkey, 69% of the municipal waste goes to sanitary landfills while 17% to uncontrolled dumpsites (TUIK, 2021).

Leachate production is one of the consequences of waste disposal in landfills/dumpsites and should be controlled to prevent adverse impacts on the environment. Leachate is a highly polluted wastewater composed of high concentrations of organic and inorganic compounds including humic acids, ammonium nitrogen, heavy metals, and inorganic salts. Many of those are stated as hazardous for environment and human health (Mukherjee et al., 2015; Öman and Junestedt, 2008). Leachate characteristics and amount depends mainly on waste constituents, decomposition activities, moisture content of waste, and percolated rainwater as well as the detention time of waste in the landfill (Adhikari et al., 2014; Vaccari et al., 2019).

It is well documented that leachate composition correlates well both with the landfill age and BOD/COD ratio which have been usually employed by the authors to define their leachate type for deciding the proper treatment method (Lee et al. 2010; Reinhart and Grosh 1998). Therefore, leachate classification has been emerged from the necessity of comparison leachate samples in other studies including mainly the treatability investigations. Leachate age/type was an important parameter for the researchers to plan their study for treatment alternatives and then compare their results with others studying different or the same type of leachate. In the literature, there are reference tables for leachate type/age definition purposing the same idea that if the landfill age is lower than 5 years it is accepted as young, between 5 to 10 years it is medium, and if it is higher than 10 years it is old (Chian and DeWalle, 1977; Gao et al., 2015; Luo et al., 2020). However, this classification is very approximate so that it cannot be applied properly to every landfill. Given proportions for COD and BOD values according to the current leachate age definition in the literature are not valid in real life cases. There are many factors affecting the leachate composition such as solid

waste content, income levels of the countries, climatic conditions, operation type of the landfills (being sanitary or unsanitary), waste pre-treatment, leachate recirculation, etc. For these reasons, there is a need for a better classification of leachate type where only landfill age may not be the primary basis. Moreover, if the landfill age will be of concern for categorization of leachate, then leachate parameter value ranges should be reevaluated.

As stated by Castrillon et al. (2010), suitability of the treatments for landfill leachate depends on its composition which is highly related whether the leachate is classified as “young” or “old”. Therefore, leachate age is an important parameter for providing guideline to plan and design the treatment alternatives through the landfill life span. In literature review, it is seen that for young leachate where COD and BOD concentrations are very high and BOD/COD ratio is high, biological treatment alternatives are prevailing. For old leachate cases where COD and BOD amount is relatively low and having mostly refractory compounds, chemical and physical technologies are preferred. For this reason, estimating the leachate composition throughout the landfilling time would be very valuable in the planning phase of leachate management alternatives. Especially COD, BOD and BOD/COD ratio showing the biodegradability would be important. For this purpose, a high quality leachate database would be very valuable in order to predict the future composition of leachate. This prediction is important especially for cases where leachate treatment is to be designed prior to landfill construction (Lo, 1996).

Literature studies shows that application of multivariate statistical techniques on landfill leachate is very limited (Mishra et al., 2016). Some applications on landfill leachate data includes classifying leachate samples or landfill cells, investigating differences in leachate quality among landfill sites in the same area, searching relations between leachate parameters and evaluation of toxicological tests in leachate studies (Modin, 2012; van Praagh, 2007). Principal Component Analysis (PCA) as well as cluster method are powerful multivariate statistical methods that are commonly used in many research fields including complex multivariate data (Adelopo et al., 2018; De

et al., 2017; Rinaldi et al., 2014; Talalaj et al., 2016). PCA method has been used in the literature for some landfill leachate studies as well (Adelopo et al., 2018; Galvez et al., 2010; Mishra et al., 2016). PCA helps to group variables with similar characteristics into the same components. Hence, variables within each component are highly correlated with each other, but slightly correlated with variables in other components (Liu et al., 2003; Pastore et al., 2018). Large data sets could be simplified into a few components which hold majority of the information in the original data (Bro and Smilde, 2014; Pastore et al., 2018; Wuensch, 2016).

With all these evaluations of literature, it is clear that there is a need for landfill and leachate characteristic analyses over a much larger data set, with the inclusion of major influencing factors to identify the most prevalent parameters. Additionally, no previous research has done multivariate statistical analysis using more than one tool on a big data set obtained from over the Globe. Besides these, existing leachate classification methods in terms of landfill age, organic content, and biodegradability criteria need further assessment with a larger data set since the first proposal was done with limited number of leachate samples in the past. Therefore, the purpose of this study is to conduct various statistical analyses and modeling for predicting leachate characteristics based on extensive data collected from landfills around the World. Correlations between leachate pollution parameters were investigated and analyzed statistically and variations according to the age of the landfill as well as climate conditions and development status (income based) were evaluated. In this study, there are three main research topics for which clarifications were sought:

- Assessment of the main properties of landfill leachate by various multivariate statistical techniques,
- Variation of leachate characteristics with respect to mainly landfill age, climate, and development status factors,
- Validity of leachate classification concept in the literature.

For the research topics given above, analyses were conducted in three different concepts. The first one is correlation analysis and regression modeling. The second

one is grouping of data into clusters and components. The final analysis is the comparison of leachate composition according to selected factors. Structure of this dissertation is formed as a collection of articles. There are also other parts including literature review and methodology. Therefore, inevitable repetitions especially in the methodology parts of the chapters would be possible. Name of the chapters are sequenced as follows: Chapter 1 provides introduction about solid waste management and landfill leachate, and explains the purpose of the study as well. Chapter 2 presents literature review and Chapter 3 introduces methodology. Chapter 4 provides historical and current review of the leachate classification practice. Chapter 5 deals with complex statistical analysis of the leachate data by employing correlation analysis, regression modeling, cluster and PCA tests. Chapter 6 includes comparison tests for studying the impact of landfill age, climate, and development status on the leachate characteristics. Chapter 7 presents the results of all the analyses by discussion of the findings with relevant literature. Chapter 8 states the main conclusions of the overall study, and final chapter provides recommendations for future studies.

CHAPTER 2

LITERATURE REVIEW

2.1 From Waste through Leachate: Formation and Composition

2.1.1 How is Leachate Formed?

It is a well presented fact that the most common method for MSW disposal in the World is still landfilling, being mostly in two forms: (I) unplanned and uncontrolled open dumpings without any leachate or gas collection and monitoring, (II) engineered/controlled/sanitary landfills with sophisticated gas and leachate collection and treatment systems that are planned and controlled from the start-up till the post-closure period (Vaccari et al., 2019). Regardless of which method is used, leachate formation is one of the most undesirable consequence of solid waste landfilling.

Leachate is formed by degradation of waste supported by initial moisture content (inherent water present inside the waste) and to a large extent by percolated rainwater through the waste storage area. When the field capacity (ability of waste to store water) is reached, water starts to move down through the waste deposits and organic and inorganic pollutants formed by various physical, chemical, and microbiological processes are transferred into the leaching wastewater (Kamaruddin et al., 2017; Lu et al., 1984) (Figure 2.1).

Leachate quality and quantity in landfills fluctuate significantly due to composition of waste, disposal techniques, landfill operations, physical properties of the site, environmental conditions within the storage area (i.e., temperature, moisture, pH), landfilling time (generally termed as landfill age), climate and rainfall patterns, hydrogeological conditions and sampling techniques (Adhikari et al., 2014; El-Fadel et al., 2002; Schiopu and Gavrilescu, 2010; Vaccari et al., 2019). Details of the major

factors affecting leachate quantity and composition are presented in Table 2.1 with relevant references.

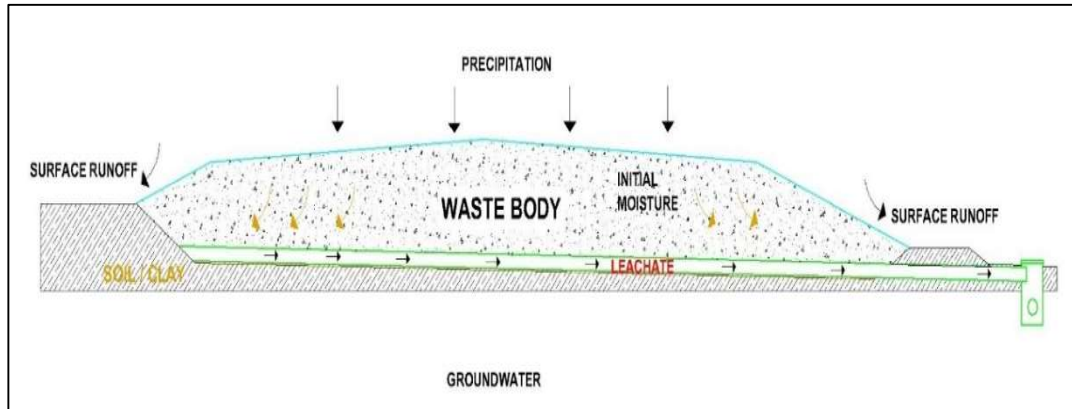


Figure 2.1. General view of leachate formation and collection in landfills

As seen in Table 2.1, there are many different as well as interrelated factors affecting leachate properties. Moisture content is known as the most important element in waste biodegradation process such that in the case of its absence, degradation activities will stop or be very limited (Adhikari et al., 2014). Initial moisture is important for the start-up of degradation of the deposited waste. However, water from rainfall, which is directly related with climatic conditions, is essential for maintaining further activities and is the principal contributor to the leachate volume (Komilis and Athiniotou, 2014; Zegzouti et al., 2019). Meanwhile, rainwater infiltration into a landfill depends on many factors such as precipitation rate, depth of waste, density and compaction of waste, having a landfill cover or not, landfill cover properties, runoff amount, evapotranspiration etc. (Yang et al., 2015). Depth of waste is one of the key factors affecting the leachate quality as it impacts waste amounts and travel times of pollutants (Zacharof and Butler, 2004). The rate of infiltration affects initial production of leachate, rate of leachate generated, and the extent of the biodegradation within the waste (Armstrong and Rowe, 1999).

Table 2.1 Factors affecting landfill leachate quality and quantity

Factors	Description	References
Waste properties	composition of the waste	Adhikari et al. (2014); Kamaruddin et al. (2017)
	age of the waste	Reinhart and Grosh (1998); Vaccari et al. (2019)
	density, particle size, and permeability	El-Fadel et al. (2002)
	initial moisture content	Kamaruddin et al. (2017)
Disposal methods	processing of waste	Adhikari et al. (2014); Reinhart and Grosh (1998)
	design of a landfill	Adhikari et al. (2014); Lema et al. (1988)
	cover soil	Schiopu and Gavrilescu (2010)
	degree of compaction	Scott et al. (2005)
Landfill operations	waste pre-treatment	Trankler et al. (2005); Scott et al. (2005)
	liquid waste/ sludge co-disposal	El-Fadel et al. (2002)
	co-disposal with ash	Reinhart and Grosh (1998)
	leachate recirculation	Ozkaya et al. (2006); Reinhart and Yousfi (1996)
	depth of waste	Schiopu and Gavrilescu (2010)
Climate and hydrogeology of the area	climatic and hydrogeological conditions	Lema et al. (1988); Vaccari et al. (2019)
	site hydrology, infiltration rate	Armstrong and Rowe (1999)
Environmental conditions in waste piles	temperature, moisture content, pH	Adhikari et al. (2014); Reinhart and Grosh (1998)
	decomposition stage	Bhalla et al. (2013); Schiopu and Gavrilescu (2010)
Sampling/collection	place of sampling (ponds, pipes, surface flows)	Reinhart and Grosh (1998); Durmusoglu and Yilmaz (2006)
	no standard protocols for leachate	Adhikari and Khanal (2015); Kjeldsen et al. (2002)

Waste properties are among the most important factors as well since they have direct effect on leachate composition. High organic content, protein rich and more biodegradable waste means concentrated leachate whereas low organic content waste leads to low strength and dilute leachate. Age of the waste or landfill is related with the phases of waste stabilisation process which has great impact on leachate composition (Adhikari et al., 2014; Reinhart and Grosh, 1998).

Environmental conditions inside the waste piles are important indicators for a balanced system for stabilisation of the waste. In each decomposition stage, leachate composition fluctuates due to the changing environment (pH, substrates, nutrients, etc.). Therefore, in every landfill, and even in different sections of the same landfill, waste undergoes various biochemical and physical transformations resulting in different leachate quality (Armstrong and Rowe, 1999; Bhalla et al., 2013).

Impact of leachate collection and sampling methods was not mentioned adequately in the literature although it is a very important factor in the characterization study. Taking representative samples from landfills enables the researchers to make healthy comparisons and evaluations of the leachate quality from different sites. Sampling, preservation, and analytical methods influence leachate properties (Chian and DeWalle, 1975; Kjeldsen et al., 2002). For example, it was stated that collected leachate from ponds or surface flows (open dumpings) can undergo some physical, chemical, and biological processes upon exposure to the air (i.e., nitrification, precipitation, etc.) which results in different leachate characteristics from the actual (Chu et al., 1994). Also, it was stated that leachate collected from landfills without a liner system can have lower pollutant concentrations due to dilution and sampling errors (Durmusoglu and Yilmaz, 2006; Reinhart and Grosh, 1998).

2.1.2 Phases of Waste Decomposition in Landfills

Leachate quality and quantity varies considerably with time or more accurately as a function of stabilisation stages of the waste (Kamaruddin et al., 2017; Reinhart and

Grosh, 1998). Therefore, it should be explained firstly how waste decomposition occurs in landfills which produces highly polluted leachate.

When waste is buried in a landfill, decomposition processes start immediately in the form of various complex biological and chemical reactions. Many field and laboratory-scale studies show that the stabilization of waste proceeds in five sequential and distinct stages as depicted in Figure 2.2 through which different characteristics of leachate is produced (Adhikari et al., 2014; Pohland and Harper, 1985; Reinhart and Yousfi, 1996). In some studies, initial and transition phases are combined in one stage (Kjeldsen et al., 2002; Mohammad-pajoooh et al., 2017). Characteristics of the five stages are explained below with focus on produced leachate composition.

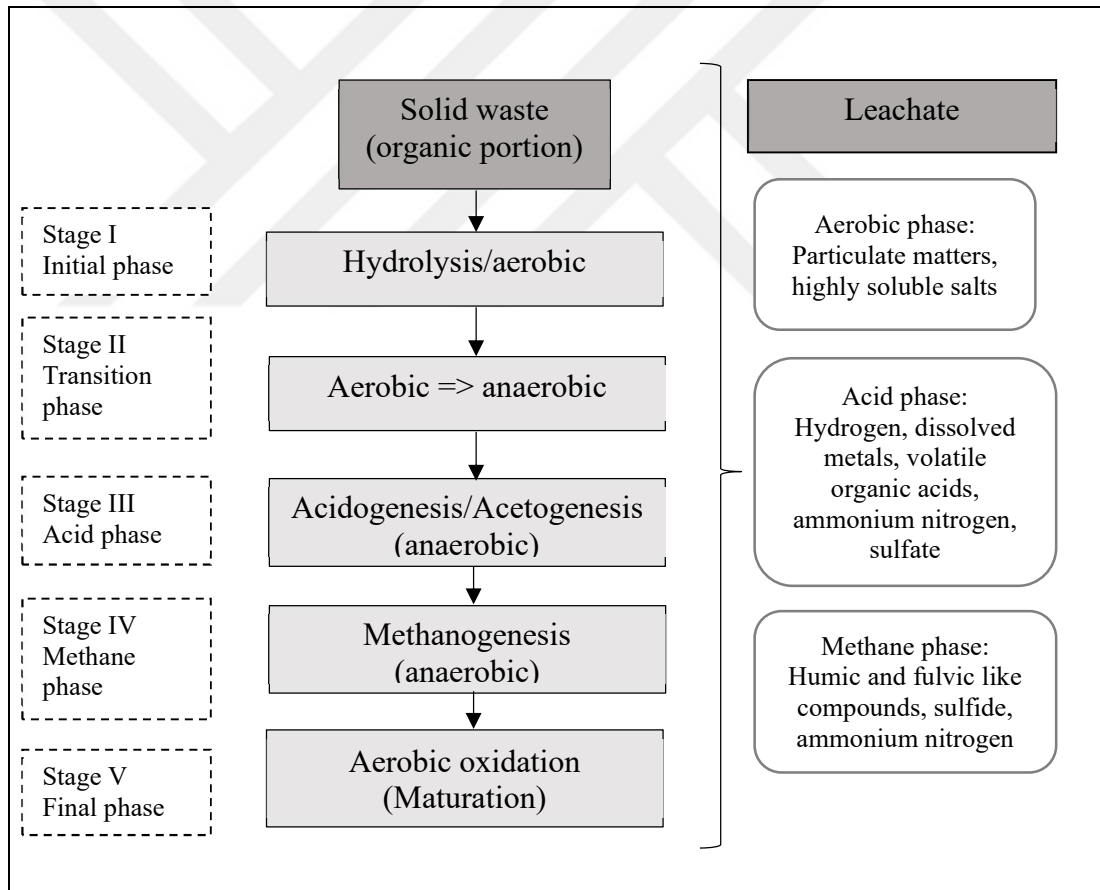


Figure 2.2. Waste decomposition stages in a landfill and related leachate composition

Stage I – Initial/Aerobic Phase

This phase involves initial placement of waste and accumulation of moisture. Stage I and II are both aerobic phases since there is some oxygen present within the deposited waste. Oxygen is quickly consumed and CO₂, H₂O, and heat are produced (Heyer and Stegmann, 2001). This phase lasts for less than a month following waste placement (Lo, 1996) and leachate is just formed from the initial moisture of the waste (Zegzouti et al., 2019) due to compaction and biological activities. Leachate produced in this stage is composed of entrapped particulate matters, dissolution of some soluble salts, and small amounts of organic substances from aerobic decomposition (Adhikari and Khanal, 2015; Reinhart and Grosh, 1998).

Stage II – Transition Phase

In this phase, interior of waste deposits is slowly transferred from an aerobic to an anaerobic environment. Reducing conditions are established and electron acceptors become nitrates and sulfates instead of oxygen. At the end of this phase, considerable amount of volatile organic acids are formed resulting in high concentrations of COD and BOD in the leachate (Adhikari et al., 2014; Scott et al., 2005).

Stage III – Acid Phase (Acidogenesis/Acetogenesis)

When all the oxygen is used up, the acid phase starts and anaerobic and facultative microorganisms (acidogenic/acetogenic bacteria) become dominant in the waste to hydrolyze and ferment cellulosic and other biodegradable materials and proteins. As by-products, ammonium nitrogen is produced and concentration of acids increases which decreases the leachate pH (Figure 2.2). In low pH values, solubility of many compounds increases and leachate would have the highest BOD, COD, and electrical conductivity values for the next few years. Leachate also contains high amounts of calcium, magnesium, iron, zinc, and manganese (Ehrig, 1983; Kjeldsen et al., 2002).

Stage IV - Methane Phase (Methanogenesis)

High quantities of methane and carbon dioxide are produced at this stage which is released as landfill gas. This phase occurs generally in between 4 to 10 years after

waste placement and may last for several years. In this stage, methanogenic bacteria start to consume accumulated acids from continuing acetogenic processes (Figure 2.2) and reduction of sulfate and nitrate into sulfide and ammonia takes place (Scott et al., 2005). As a result BOD and COD concentrations start to decrease while leachate pH increases. Inorganic compounds like chloride, sulfate, sodium, potassium, and iron continue to leach for many years and ammonium nitrogen concentration remains at high levels. Some of the organic matters like lignin-type aromatic compounds cannot be degraded in anaerobic conditions, hence remain in the landfill. These play an important role in complexation and precipitation of heavy metals (Adhikari et al., 2014; Reinhart and Grosh, 1998).

Stage V - Final Phase (Maturation)

In the final stage of decomposition process, biological activity weakens due to the deficiency of nutrients and organics. Leachate strength remains constant at very low values having mostly recalcitrant compounds. Oxygen could reappear slowly due to intrusion of air into the decomposed waste and this could help for degradation of recalcitrant organics producing humic-like substances (Reinhart and Yousfi, 1996).

All of the stages above are distinguished from each other according to the variations in the concentrations of specific compounds in leachate like nitrogen, organics, and heavy metals as well as gas quality and quantity (Mohammad-pajooch et al., 2017). It is a well known subject that landfilling of waste is a long lasting process that could last for 20-30 years or more so that separate parts of a landfill can be in different phases of waste decomposition. Therefore, it is emphasized in many sources that landfills cannot have just a single stage but different phases in various parts of the site and time required for each stage to be completed can be different depending on the prevailing physical, chemical, and biological conditions within the waste (Kjeldsen et al., 2002; Scott et al., 2005). Furthermore, only the most distinct phases, namely stages III and IV, could be differentiated truly due to specific characteristics of produced leachate and gas (Adhikari et al., 2014; Pohland and Harper, 1985). For that reason, it is noticed that in some references leachate is defined as acidogenic/acetogenic or methanogenic

leachate (Gao et al., 2015; Hussein et al., 2019; Mishra et al., 2016; Wijesekara et al., 2014).

2.1.3 Leachate Properties and Characteristics

Characteristics of leachate are generally represented by pH, chemical oxygen demand (COD), biological oxygen demand (BOD), suspended solids (SS), ammonium nitrogen ($\text{NH}_4\text{-N}$), total Kjeldahl nitrogen (TKN) and heavy metals (Mukherjee et al., 2015; Renou et al., 2008). It could be said that main pollutants in leachate are organics and nitrogen (mostly in the form of $\text{NH}_4\text{-N}$). Some heavy metals could be slightly higher in early stages due to dissolution at low pH (Kulikowska and Klimiuk, 2008), otherwise they are not found in major quantities (Kjeldsen et al., 2002). Main pollutant groups in leachate are provided in Table 2.2 and explained below in detail.

Table 2.2 Main pollutant groups in landfill leachate (Christensen et al. 2001; He et al., 2009)

Groups	Constituents
Organic compounds	Dissolved organics measured as COD/TOC including volatile fatty acids and refractory compounds like fulvic and humic-like substances.
Inorganic compounds	Sulfate, phosphate, chloride, nitrogen species, calcium, magnesium, sodium, potassium, carbonate, iron, and manganese.
Heavy metals	Nickel, zinc, copper, lead, cadmium, mercury, arsenic, chromium.
Specific organic compounds	Xenobiotic Organic Compounds (XOCs) such as aromatic hydrocarbons, phenols, pesticides etc., endocrine disrupting compounds (EDCs).

Organic compounds

Majority of the biodegradable portion of the waste consists of food, yard and wood waste, paper, textile, and some inorganics (Bhatt et al., 2016). Organic composition of

leachate (mostly measured as COD, BOD₅ (simplified as BOD throughout the text), and TOC) comes from the soluble parts of those wastes and decomposition by-products. The highest amounts of organic compounds found in leachates are generally volatile fatty acids like acetic, butyric, valeric, and propionic acids. This composition could change and shift into other constituents like fulvic and humic-like compounds with time due to microbial and physical/chemical processes in a landfill (Adhikari and Khanal, 2015; Chian and DeWalle, 1977).

COD is used for measuring all oxidizable matters in leachate and BOD is for the biodegradable portions only (Lee and Nikraz, 2014). Therefore, BOD/COD ratio is related mostly with the organic composition of leachate and it could be a good indicator for showing the phases of waste stabilisation. It can also be considered as showing the maturity of leachate since it decreases considerably with time (El-Fadel et al., 2002). Very low values of BOD/COD ratio mean that most of the biodegradable waste has been stabilized. Therefore, it is one of the best indicators for understanding the degree of waste stabilization as well as leachate biodegradability (Bhatt et al., 2016; Zakaria and Aziz, 2018).

Inorganic compounds

A great variety of inorganic compounds are detected in landfill leachate including NH₄-N, chloride (Cl), sodium (Na), potassium (K), calcium (Ca), magnesium (Mg), manganese (Mn), iron (Fe), bicarbonate (HCO₃), and sulfate (SO₄) at considerable concentrations (Renou et al., 2008). Concentrations depend on the waste stabilization stages as explained in Section 2.1.2. For example, in the methanogenic stage, concentrations of calcium, magnesium, iron, and manganese are lower since higher pH enhances precipitation and sorption activities inside the waste. Similarly, SO₄ levels are low in the methanogenic stage as SO₄ concentration can decrease with time due to the reduction to sulfide (SO₂) under anaerobic conditions in landfills (Akyol, 2005; Renou et al., 2008). The resulting SO₂ ions react with metals to precipitate as metal sulfides (Kjeldsen et al., 2002; Talalaj et al., 2016).

Mg and Ca are not major pollutants in leachate. Decay of plant and animal tissues can contribute to high levels of Ca levels beside some industrial wastes. Therefore, this sort of waste in landfills can contribute to high concentrations of Ca and Mg in leachate (Oketola and Akpotu, 2015).

The salt content of leachate is mainly due to Na, K, Cl, SO₄, and NH₄-N ions. They mainly come from the degradation of organic matters in waste (Naveen et al., 2014). Conductivity (EC) of the leachate consists of major ions like Ca, Mg, Cl, Na, and K and it can be extremely high due to high levels of anions and cations (De et al., 2016; Naveen et al, 2017). Chloride is a conservative parameter so that its concentration in leachate does not change notably with time (Reinhart and Grosh, 1998; Tatsi and Zouboulis, 2002). Its concentration change is mainly related to dilution effect due to rainfall (Kulikowska and Klimiuk, 2008; Statom et al., 2004). In some studies, variations were shown to increase with landfilling time (Chu et al., 1994). But, Ehrig (1983) indicated no significant change in chloride concentrations in acidogenic and methanogenic stages.

Nitrogen in leachate can be found in many forms like organic nitrogen, NH₄-N, nitrite (NO₂) and nitrate (NO₃) ions. It originates from the biodegradation of proteins and amino acids in waste and measured mostly as NH₄-N and TKN. TKN is the sum of organic nitrogen and NH₄-N while total nitrogen is calculated as the summation of TKN and nitrite/nitrate concentrations. TKN and NH₄-N values are found to have very similar magnitudes and most leachate samples have no or very low nitrate values (Statom et al., 2004). Kim and Lee (2009) stated that NH₄-N concentration is generally more than 90% of the total nitrogen (TN) in leachate. NH₄-N/TN ratio decreases as landfill ages, indicating increase in other N species like NO₂ and NO₃ (Ziyang et al., 2009).

NH₄-N is one of the most important polluter in leachate due to its relatively high concentrations, toxic effect and having no mechanism of removal inside the landfill in the long term. Since anaerobic hydrolysis of proteins is slower than other compounds, nitrogen is released gradually as NH₄-N. Therefore, its concentration does not decrease

with time but could increase to very high levels as landfill ages and remains at those levels for decades (Chu et al., 1994; Kim and Lee, 2009; Kjeldsen et al., 2002).

Alkalinity in leachate is composed of mainly bicarbonate, carbonate and hydroxyl ions. Total alkalinity (TA) in landfill leachate is generally very high due to microbial decomposition and dissolution activities in landfills. Biodegradation of organics generates considerable amounts of carbon dioxide which is also a source of alkalinity (Naveen et al, 2017).

Heavy metals

Heavy metals originate mainly from ash and construction & demolition wastes (Durmugoglu and Yilmaz, 2006) as well as from discarded materials like batteries, bulbs, paints, pipes, chemicals (Mor et al., 2006), electronics, motor oils, inks, foils, plastics, glass, rubber and other scrap materials (Kjeldsen et al., 2002; Naveen et al., 2014). Heavy metals can arise as the by-products of corrosion and complexation activities or dissolved components from waste (Durmugoglu and Yilmaz, 2006). A wide range of heavy metals can be found in landfill leachate such as nickel (Ni), zinc (Zn), chromium (Cr), copper (Cu), lead (Pb), aluminium (Al), and cadmium (Cd) (Kjeldsen et al., 2002; Renou et al., 2008).

Heavy metal release depends mainly on the presence of complexing agents and pH of the medium. In anaerobic conditions, most heavy metals such as Ni, Zn, Pb, Cu, and Cd could form insoluble metal sulfides and precipitate. High pH also decreases the solubility of metals and precipitation of metal hydroxides (i.e., chromium) may occur (Kjeldsen et al., 2002; Renou et al., 2008).

Specific organic compounds

Xenobiotic organic compounds (XOCs) may be found in leachate at slightly low concentrations which may originate from industrial and household chemicals. Aromatic hydrocarbons, phenols, chlorinated aliphatics, pesticides and plastizers like PCBs, dioxins, and PAHs are common in XOCs compounds. Many XOCs could be observed in leachate depending on landfill technologies used, waste composition, and

landfill age. The most frequently seen XOCs are the halogenated hydrocarbons and monoaromatic hydrocarbons like benzene and toluene (Kjeldsen et al., 2002; Renou et al., 2008).

Beside XOCs many endocrine disrupting compounds (EDCs) have started to appear in landfill leachate in very small concentrations (i.e., ppm) as detection is enabled with the progress in measurement techniques (He et al., 2009). Their sources are mainly industrial chemicals, pharmaceuticals, and pesticides present in the waste (Scott et al., 2005).

As explained above, leachate contains many different and complex compounds based on the type of waste landfilled. There are many other compounds in landfill leachate. Up to now, more than 200 hazardous compounds have been measured (Khalil et al., 2018; Luo et al., 2020). For example, sulfide, barium, mercury, borate, arsenate, lithium, selenate, and cobalt can be found in leachate but in very low quantities so that could be taken with little importance (Kjeldsen et al., 2002).

2.2 Leachate Characterization Studies

In literature, there are extensive studies on landfill leachate providing valuable information about the composition, transport, and treatment alternatives. Leachate characterization data mostly comes from the treatability studies including operating or closed landfills as well as pilot scale or laboratory lysimeter studies. The main problem with landfill leachate can be stated as its composition such as strength, biodegradability, and toxicity changes with time as the waste decomposes in the landfill. Although leachate content fluctuates considerably between the aerobic, acetogenic, and methanogenic phases during waste stabilization, according to literature mainly three categories of leachate have been defined with respect to landfill age (Baig et al., 1999). Leachate produced in young landfills, generally less than 5 years old, are typically high strength wastewaters characterized by low pH, high organic content, high BOD/COD ratio (>0.6), and the presence of several hazardous compounds. Leachates from medium aged landfills, generally 5-10 years, mainly have

refractory organic compounds, low BOD/COD ratio (<0.3), and high concentration of $\text{NH}_4\text{-N}$ (Neczaj et al., 2008). On the other hand, in leachates from old landfills (age >10 years) mainly compounds such as humic and fulvic acids were found with low COD (<5000 mg/L), low BOD/COD ratio (<0.1), and high pH (>7.5) (Ding et al., 2018).

Ehrig (1983) studied quantity and quality of leachate for 3 years at 20 landfills in the west of Germany. The age of the studied landfills were up to 15 years. It was stated that concentrations of some parameters were changing with landfill age (i.e., pH, COD, BOD, Fe and Ca). As the pH was increasing, Fe and Ca were decreasing due to decreasing solubility. Many of the inorganics were found varying considerably with a low increase with time (i.e., Cl, $\text{NH}_4\text{-N}$, K and Na). Heavy metals were found in low levels (Pb, Ni, As, Cu, Cr).

Tatsi and Zouboulis (2002) studied the composition of leachates depending on landfill age and seasonal changes. It was stated that high rainwater percolating through the waste piles dilutes leachate while extracting several constituents, but during dry months, concentrations of some parameters would be higher. They studied the main physico-chemical parameters of leachate samples on a seasonal basis taken from two different sites (old and fresh part) in the same sanitary landfill. Relationships between those parameters were investigated and findings are summarized as follows:

- Fresh (young) leachate is generally characterized by higher pollutant concentrations. The concentration of total dissolved solids (TDS) fluctuates too much and conductivity is higher showing the high amounts of soluble inorganics. On the other hand, organic and inorganic contaminants are found in decreasing levels in older leachate (stabilized). BOD/COD ratio decreases as well, being 0.5 for a young leachate and 0.2 for an old (more stabilized) one.
- $\text{NH}_4\text{-N}$ concentrations remained high despite decreasing trend of the organic compounds (as COD and BOD) in leachate with landfill age.
- Chloride concentration in young and old leachate was found to be similar without any decreasing or increasing trend with landfill age (between 3200 and 4100 mg/l).

- Total phosphorus (TP) was found in lower concentrations in old leachate (around 9 mg/l) while relatively higher (around 170 mg/l) in young leachate.
- Due to the anaerobic reactions within the landfill, leachate pH increases in old leachate (6.2 to 7.9) showing the decrease in the free volatile acids concentration.

Statom et al. (2004) investigated the leachate chemistry from lysimeter tests and pilot/full-scale landfills. Concentrations of organic and inorganic parameters were changing in large ranges. Majority of the parameters were found in decreasing concentrations in older landfills compared to younger ones. Similarly, Ding et al. (2018) stated that leachate composition from young landfills (age < 5 years) contains high organic substances (COD > 10,000 mg/L) and BOD/COD ratio (> 0.3) with low pH (< 6.5).

Calli et al. (2005) presented leachate characteristics of a landfill site in Turkey. It was shown that BOD/COD ratio is generally above 0.6 indicating high biodegradability. Close values for TKN and NH₄-N concentrations show the absence or very low amounts of organic nitrogen in young landfill leachate. Heavy metal concentrations were found very low except for iron. Mertoğlu and Çallı (2011) performed physical and chemical analyses of leachate samples for two years obtained from eight landfills in Turkey. It was stated that BOD/COD ratio is more than 0.5 in active landfills. In young landfill sites, COD and TKN values were more than 70,000 mg/L and 5000 mg/L, respectively.

Robinson (2005) presented detailed data for leachate composition from landfills in different countries and climates. It was found that it takes up to 3 years to achieve methanogenic leachate composition for landfills in moderate climates, whereas at warmer climates (tropical sites), very rapid transition occurs in only 12 to 18 months.

Fan et al. (2006) studied three different types of landfills in Taiwan, namely a closed MSW landfill, an active mixed landfill (co-disposal of MSW with bottom ashes of MSW incinerators) and an active MSW landfill. Seasonal effects, landfill age, pH, organic content (BOD, COD, fulvic acid, humic acid, and non-humic substances),

solids, and metals (Pb, Ca, Cd, Hg, Cr, Cu, Fe, K, Mg, Mn, Na, Ni, and Zn) in leachate have been analyzed. It was shown that active landfills have considerably higher COD, total solids (TS), TOC and conductivity values. Furthermore, COD, SS, TS, color, TOC, BOD and conductivity were found having a decreasing trend with increasing landfill age.

Jianguo et al. (2007) investigated the impacts of leachate recirculation on waste stabilization, landfill gas production, and leachate composition. It was stated that recirculation results in accumulation of ammonium, phosphorus and some persistent organics in the leachate. Recirculated leachate composition is similar to the old leachate.

Öman and Junestedt (2008) studied leachates from 12 Swedish municipal landfill sites and detected in total 140 organic, metal organic and inorganic compounds. Many of those compounds are stated as hazardous to the environment and human health. Li et al. (1999) studied leachates from 13 landfills in Hong Kong and very high concentrations of ammonia, low BOD/COD ratio (0.19), and high conductivity and alkalinity values were found.

Galvez et al. (2010) studied the leachate from a landfill in Spain for 3 years. The analyzed parameters were mainly pH, COD, BOD, nitrogen, solids, conductivity, major anions and cations. The leachate was characterized as partially stabilized because of existence of both active and closed cells at different decomposition phases and leachate recirculation at the site. Landfill site accepted waste fractions that cannot be recycled or recovered in the nearby waste composting and recovery plant (approximately 59% of the total waste). Therefore, organic content of the waste was reduced that lowers biodegradable organics in the leachate. They stated that conductivity values were high in leachate indicating the presence of dissolved inorganic materials. The highest ion concentrations were chloride, sodium and potassium which are part of the total dissolved solids. TKN and $\text{NH}_4\text{-N}$ concentrations were similar while nitrate and nitrite levels were negligible showing that organic nitrogen was very low and most of the nitrogen (over 90%) was in ammoniacal form.

Castrillon et al. (2010) analysed the variations in the physico-chemical composition of the leachate in a landfill with different ages. Organic content of the leachate reached maximum values during the first year of operation (up to 80,000 mg/L) and gradually decreased. On the other hand, the concentration of $\text{NH}_4\text{-N}$ has been increased continuously to around 2000 mg/L. Old landfill leachate was characterized by higher $\text{NH}_4\text{-N}$ content and lower BOD/COD ratio.

Mohammad-pajooch et al. (2017) studied the leachate composition by a five-year survey in different states of Germany. Major contaminants were stated as COD, Cl, Na, K, and $\text{NH}_4\text{-N}$, but heavy metal concentrations were not found significant. They also stated that generation of landfill leachate could be reduced to some point by waste pre-treatment which has been widely applied in developed countries. Pre-treatment of waste can be done by mechanical biological treatment (MBT) plants where paper, metals, plastics, etc. are recovered by mechanical processes. Then, biological processes (composting, digesting, etc.) applied for stabilization of the waste before landfilling. Those processes affect incredibly the content of the landfilled waste and hence composition of the leachate. After MBT processes, leachate organics and heavy metal concentrations are reduced significantly but nitrogen concentrations does not change too much and remain at high levels.

2.3 Studies Regarding Statistical Analysis and Modeling Approaches for Landfill Leachate

Due to high number of parameters, sampling, preservation and analysis of leachate samples is a time consuming and expensive process. Therefore, different statistical approaches and models can be used to show the significant similarities and differences between leachate samples from different landfills. Although site specific conditions have great impact on the leachate composition, common trends and properties can be found through data analysis.

Usage of simulation programs for design, operation and monitoring of landfills is not widespread due to difficulty in modeling of landfill processes. Local factors such as

composition of waste, disposal method as well as several physical and biochemical factors should be considered such as climate, liquid and gas transfers, biological and chemical processes etc. (de Cortazar and Monzon, 2007). It is possible to find many studies about leachate characterization and treatment methods in the literature as well as modeling of landfills, mass transfer calculations, etc. However, only limited information was found regarding modeling/statistical analysis of leachate properties. Some of those studies are provided in this section.

Tatsi and Zouboulis (2002) studied leachate samples taken from two different sites (old and fresh part) in the same sanitary landfill. Pearson correlation analysis was performed to show the possible relationships between the parameters. Good correlation was found between those:

- Conductivity with nitrate, $\text{NH}_4\text{-N}$, TKN, solids, alkalinity, color;
- pH and solids;
- BOD and COD, TDS, sulfate;
- $\text{NH}_4\text{-N}$ and TKN, solids, alkalinity, color;
- COD and solids.

Fan et al. (2006) conducted Pearson correlation analysis for some leachate parameters and found that landfill age is inversely correlated with pH, COD, SS, TS, TOC, BOD, and conductivity (EC). Pearson correlation results are presented for other parameters as follows:

- pH is correlated with EC, COD, TS, TDS, TOC;
- SS is positively correlated with COD, TS, TOC, EC, TDS;
- COD is positively correlated with pH, SS, TS, TOC, BOD, EC, TDS;
- TS is positively correlated with pH, COD, TDS, TOC, BOD, EC.

Ziyang et al. (2009) studied leachate samples with different ages by principle component analysis (PCA). Results indicated that leachate could be categorized mainly into three phases by degradation in the southern areas of China (humid and warm climate). Phase I included the first 4 years as fast degradation stage, Phase II

was called as transitional stage between 5 to 7 years, and Phase III was accepted as consolidated stabilization stage after 8 years.

Galvez et al. (2010) studied leachate from a landfill in the proximity of a composting and recovery plant in Spain for 3 years. Leachate data were evaluated statistically using various statistical tools. With Pearson analysis, a good correlation was found between COD and total solids, especially with volatile total solids indicating the contribution of organic solids into the COD. $\text{NH}_4\text{-N}$ showed significant correlation with BOD. Correlation between dissolved solids, anions and cations (Cl, Na, K, Mg), and conductivity shows that these ions are dissolved in the leachate. It also confirms the well-known relationship between conductivity and dissolved solids.

Wijesekara et al. (2014) stated that pH has an inverse correlation with heavy metals in leachate like Mn and Cr showing dissolution of these metals at low pH. TDS has a significant correlation with COD and heavy metals like Pb, Ni, and Cr showing high mobility of these pollutants in leachate.

Bhatt et al. (2016) studied models to estimate BOD and COD concentrations with time in MSW landfills depending on the parameters such as precipitation rate, temperature, and type of waste. The models obtained showed that high rainfall and temperature rates result in higher BOD and COD concentrations in leachate.

Mishra et al. (2016) stated that some statistical methods have been utilized in the past for landfill leachate. However, application of multivariate data analysis technique on leachate data is very limited. For example, PCA was performed by only a few researchers for leachate data (Adelopo et al., 2018; Boateng et al., 2018). Some applications of PCA on landfill leachate data includes classifying leachate samples or landfill cells, investigating differences in leachate quality among landfill sites in the same area, and searching relations between leachate parameters (Modin, 2012). Adelopo et al. (2018) analysed leachate data from closed and active landfills and showed the existence of significant differences between those landfills. These differences were apparent especially for the concentrations of EC, COD, TKN, solids, chloride, sodium, potassium, calcium, magnesium, and nickel. No significant

differences were found between the concentrations of other heavy metals (Cu, Zn, Cr, Pb, Cd).

Mishra et al. (2016) studied leachate samples from a landfill site in India. They performed PCA to examine the correlation between large number of leachate parameters. EC was found positively correlated with Cl, COD, BOD, Mg, and Fe. COD content of the leachate was correlated positively with SS, BOD, EC, Ca, Fe, Ni, Mn, and Cl. Also, seasonal variations were observed in landfill leachate regarding COD, BOD, alkalinity, pH and conductivity. Durmusoglu and Yilmaz (2006) conducted PCA for leachate samples from a landfill in Turkey. It was stated that Cu, TP, Zn, and COD are the most essential elements in their leachate samples for explaining the variation in the data set.

Mohammad-pajooch et al. (2017) studied the leachate composition by a five-year survey in different states of Germany. Major contaminants were stated as COD, Cl, N, K, and NH₄-N, but heavy metal concentrations were not found significant. It was found that COD and NH₄-N were highly correlated with EC values. For this reason, possibility of prediction COD and NH₄-N values from EC measurements was investigated. A linear regression model was used and equations were found highly significant.

CHAPTER 3

METHODOLOGY

To achieve the objectives of this study, leachate characterization data belonging to different landfills in different countries have been collected to conduct various statistical analyses and regression modeling studies. As a first step, data were collected from different types of literature including published articles, theses, and available site data from operating landfills. Collected data includes leachate pollution parameters (i.e., organic content, inorganics, heavy metals, etc.), landfill properties (operational years, status, etc.), climate and development status of the landfill location (Figure 3.1).

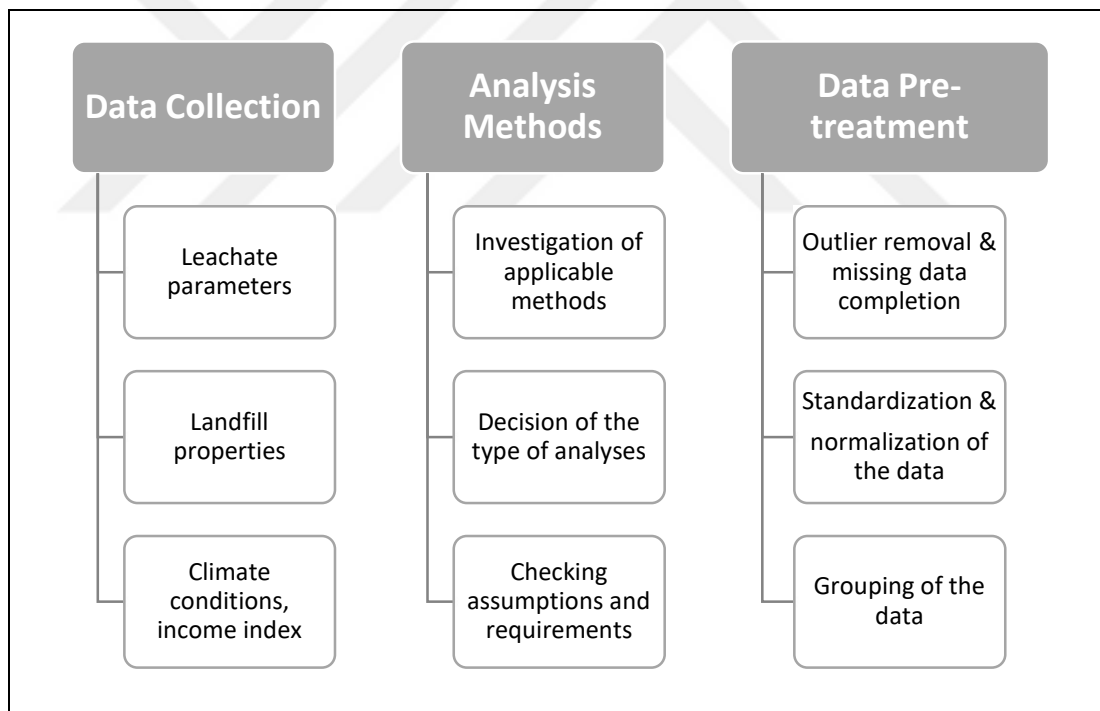


Figure 3.1. Main activities conducted for analysis of leachate data

After extensive data collection phase, analysis tools were investigated from current literature. It was observed that there have been limited studies for some of the complex statistical methods given in Section 3.2. Those methods were investigated further for

their applicability to collected leachate data. The main assumptions and requirements were studied in order to figure out the data pre-treatment needs. Mainly, outlier and missing data problems emerged as important factors. Furthermore, standardization and normalization as well as grouping of the data were arisen as the main steps to be taken prior to the analyses. A simple view of the main activities conducted before any detailed analysis is presented in Figure 3.1 and explained in detail in the following sections. Detailed flowcharts are provided in Section 3.1 and Chapters 5 and 6 with the results of selected analyses.

3.1 Data Collection

Available leachate data were collected from national and international references as well as from operating landfill sites in Turkey. Leachate composition data were gathered mostly from treatment and pollution monitoring studies. In most of the researches, composition of the landfill leachate is presented by some basic parameters such as pH, COD, BOD, $\text{NH}_4\text{-N}$, Cl, solids, electrical conductivity (EC) and alkalinity (as CaCO_3).

Some difficulties were faced during collection of the proper data to be used for regression modelling and statistical analyses. Some references provided only limited leachate parameters which caused discontinuity in the data set. For example, TOC, solids, alkalinity, sulfate, phosphorus, Na, K, and Cl as well as some heavy metal measurements were absent in some studies. On the other hand, pH, COD, BOD, and $\text{NH}_4\text{-N}$ parameters were measured in most of the studies.

For the aim of collecting leachate data in relatively similar conditions in terms of landfill operations, leachate handling methods, etc., references satisfying the below criteria were selected:

- municipal solid waste is the major waste type landfilled which includes the commercial waste, yard waste and domestic waste.
- no leachate recirculation at the site

- no pre-processing of the waste before landfilling
- no disposal of specific waste like ash, sludge, etc. in considerable amounts
- no mixing of landfill leachate with any other wastewaters

Furthermore, landfill properties like being closed or active (operational) during sampling period as well as being sanitary (engineered/controlled) or unsanitary/uncontrolled landfill/dumpsite was indicated in the data matrix accordingly.

Landfill/leachate age definition was absent in many references. Furthermore, since landfills operate continuously until the closure, it was difficult to attribute a definite number for the leachate age. As stated by Ziyang et al. (2009) waste disposal occurs by multi-layers in most of the landfills in considerable heights. Therefore, it is not possible to collect leachate samples with a certain disposal time/age. Leachate comes from older and freshly deposited parts together and generally mixed in collection ponds. In majority of the references, leachate age is equal to the landfill age. Leachate age is calculated specifically as explained below.

- Leachate age is calculated as the difference between start date of the landfill and sampling date of leachate. Sampling time is determined by some assumptions if not provided clearly in the reference. For example, if sampling year is absent, 2-3 years back is assumed from the article submission date (relevancy of this assumption was observed in many articles during data collection and considered that it is quite acceptable).
- If sampling time is given as a time interval, the date corresponding to the mid-point of the interval is taken as the sampling time in the landfill/leachate age calculation. For example, if leachate average data is given for 2010-2012 period, then 2011 is taken as the average sampling year.
- If a landfill name is known but its age is not given in the reference clearly, extensive web search was done to find out the landfill operation dates (start/closure). For some cases, search was done for some landfills in Korea, Spain, Norway, Sweden, Brazil, France, Russia, etc. in their original language

and translation programs were used. Some of the absent data about a few landfills were obtained in this way.

- If sampling period is several years and leachate composition is given for each year separately, then the data corresponding to different years are included in the data set for different landfill ages of the same landfill. For example, for 3 years of sampling time of a given landfill, 3 data sets were included in the analyses as 1-year, 2-years, and 3-years old landfill ages. For that reason, the number of leachate data sets (511) is greater than the number of total landfill sites (330) in the study.

All relevant data collected from references, including specific information like names of the landfills, location as province and country, relevant dates (i.e., sampling time, landfill start year and closure year), being sanitary or unsanitary, closed or active landfills, etc. were entered into an Excel spreadsheet. Furthermore, countries' development criteria as income levels (high, lower middle, and upper middle) have been determined according to the World Bank list of economies (June 2020). Climate data of landfill locations (annual average temperature and precipitation rates) was taken from the articles if given, otherwise a web link was used (Climate-data.org).

It is worth to note that below issues were taken into account during data collection:

- There were inconsistencies in collected data for some leachate parameters. For example, if the TKN was lower than $\text{NH}_4\text{-N}$, if TOC value was higher than COD, and so on. For those cases, data was considered as incorrect and eliminated.
- If a parameter concentration is given as “under detection limit” in the form of less than a given number, then half of that number (detection limit) is taken as the quantity (i.e., if concentration is <0.04 mg/L, in the analysis it was considered as 0.02 mg/L). This procedure was applied generally for very low heavy metal concentrations declared as under detection limit.
- Mathematical calculations were done to complete missing data during data collection step. For example, TN was calculated from given TKN and

nitrate/nitrite values, or TKN from given organic nitrogen and $\text{NH}_4\text{-N}$ values and vice versa. Similarly, TDS was calculated from given SS and TS or TS from SS and TDS and so on.

- Some leachate parameter values were read from the charts given in relevant studies if not tabulated (encountered only in a few cases).
- If leachate parameters are provided in value ranges, then averages were calculated. Similarly, if several months of data are given within a given year, again averages were taken.
- Care was given to use leachate data having relatively similar conditions in all the landfills. For example, leachate data for any indicated specific site properties like soil cover with moulding sludge, clay layer with high calcium content etc. were not used. Therefore, landfills where leachate quantity and quality might have been impacted considerably by site-specific conditions were eliminated during data collection phase.
- In many references landfill names are given, but in many others it is not. In those cases where the landfill name is absent but the specific data exists (start date, operation type), then the leachate data were used under the name of unknown landfill.
- Unsanitary landfills are the ones with no gas and leachate collection mechanisms and generally named as open dumps, uncontrolled landfills, unengineered or unlined landfills, etc. in different articles (Vaccari et al., 2018). At those sites, leachate samples are generally collected from surface flows, seepages, and self-formed small ponds. This individual non-homogeneous leachate deposits cannot properly reflect the leachate properties of the whole site. Therefore, their indication was done clearly on the data sheet as landfill type and not included in the analyses.
- For the climate classification, climate data of the nearest location was taken according to the available information given in the reference such as map of the site or name of the landfill place.

- Data sheet has been checked several times. If landfill operational dates and sampling dates were absent or incorrect dates were given as realized from cross-checks with other relevant references or web search, they were corrected as much as possible.

Approximately 850 references constituting published articles, reports, and theses have been reviewed. A total of 511 leachate data have been generated, 87 of which belongs to unsanitary landfills and 64 being from closed sites while the remaining ones are still active, operating sanitary landfills. As a result, an initial raw input leachate data matrix of 360 rows (landfills/leachate samples) was obtained from different parts of the World such as countries from Europe, Middle East, Asia, Africa, USA, and Canada (Appendix A). In some references, it was not possible to obtain or estimate the real landfill or leachate ages so those data had to be removed from the analysis related with age.

Main statistical representation of the leachate pollution parameters according to the collected data were given in Table 3.1. It is seen that there are high variability in concentrations of many parameters. Beside high concentrations of organics, $\text{NH}_4\text{-N}$, chloride and alkalinity, there are much smaller concentrations of heavy metals and other inorganic compounds in landfill leachate.

Table 3.1 Descriptive statistics for physico-chemical characteristics of leachate

Parameters*	Sample N	Minimum	Maximum	Mean	Median	Std. Deviation
pH	344	5.2	10.0	7.6	7.8	0.7
COD	345	117	80750	9535	4800	11780
BOD	300	9	49400	4521	1106	7427
TOC	89	32	19800	2726	1430	3742
BOD/COD	293	0.01	0.94	0.33	0.31	0.22
NH ₄ -N	314	10	5931	1359	1157	1096
TKN	144	75	6335	1769	1556	1397
TN	107	56	6370	1846	1370	1592
TP	156	0.2	149	24	15	25
PO ₄ -P	86	0.0	349	21	9	42
TSS	184	10	21000	1051	530	1854
TDS	92	2000	36222	14098	12812	8308
TS	80	292	43840	15869	13754	9395
Cl	222	3	11443	2808	2331	2195
SO ₄	152	0	2250	291	88	458
EC	190	0.5	68.1	19.8	17.1	12.7
TA	159	567	23250	7638	7094	4551
Na	121	128	11850	1938	1445	1740
K	106	117	10252	1340	959	1371
Fe	209	0	810	57	11	114
Ca	133	7	7413	477	184	888
Mg	128	2	1963	219	142	278
Ni	158	0.0	9.8	0.7	0.3	1.4
Zn	207	0.0	76.7	3.2	0.7	8.9
Cu	173	0.0	15.2	0.9	0.1	2.5
Cr	177	0.0	13.8	1.0	0.3	2.0
Pb	175	0.0	56.7	0.8	0.1	4.4
Cd	156	0.0	3.7	0.1	0.0	0.4
Mn	128	0.0	49.0	4.2	0.8	8.6

*Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

Algorithm for the steps taken during the data collection and analysis is presented in Figure 3.2. The first and substantial step was the data collection. The data on both leachate parameters and other variables such as landfill properties, socio-economic conditions of the places (as income index), and climatic conditions were compiled at this step. After data collection was completed, controls and checks were done for the missing or insufficient data. As mentioned before some references provide values only for some of the leachate parameters. For example, organic and ammonia content measured mostly while some inorganics (phosphorus, sulfate, calcium, and magnesium) and heavy metals were absent. Therefore, other available references about the same landfill were searched to have more data or for consistency check. Furthermore, for the missing information in the references, extensive search was conducted to complete the required fields in the data sheet. For example, if landfill operational dates and sampling dates are absent, cross references were checked or web search was done to be able to calculate landfill/leachate age properly. In the data analysis part, references were required to be reviewed many times due to outlier or missing data. If necessary information cannot be found within the article or other cross-checked references, then data removal was done accordingly.

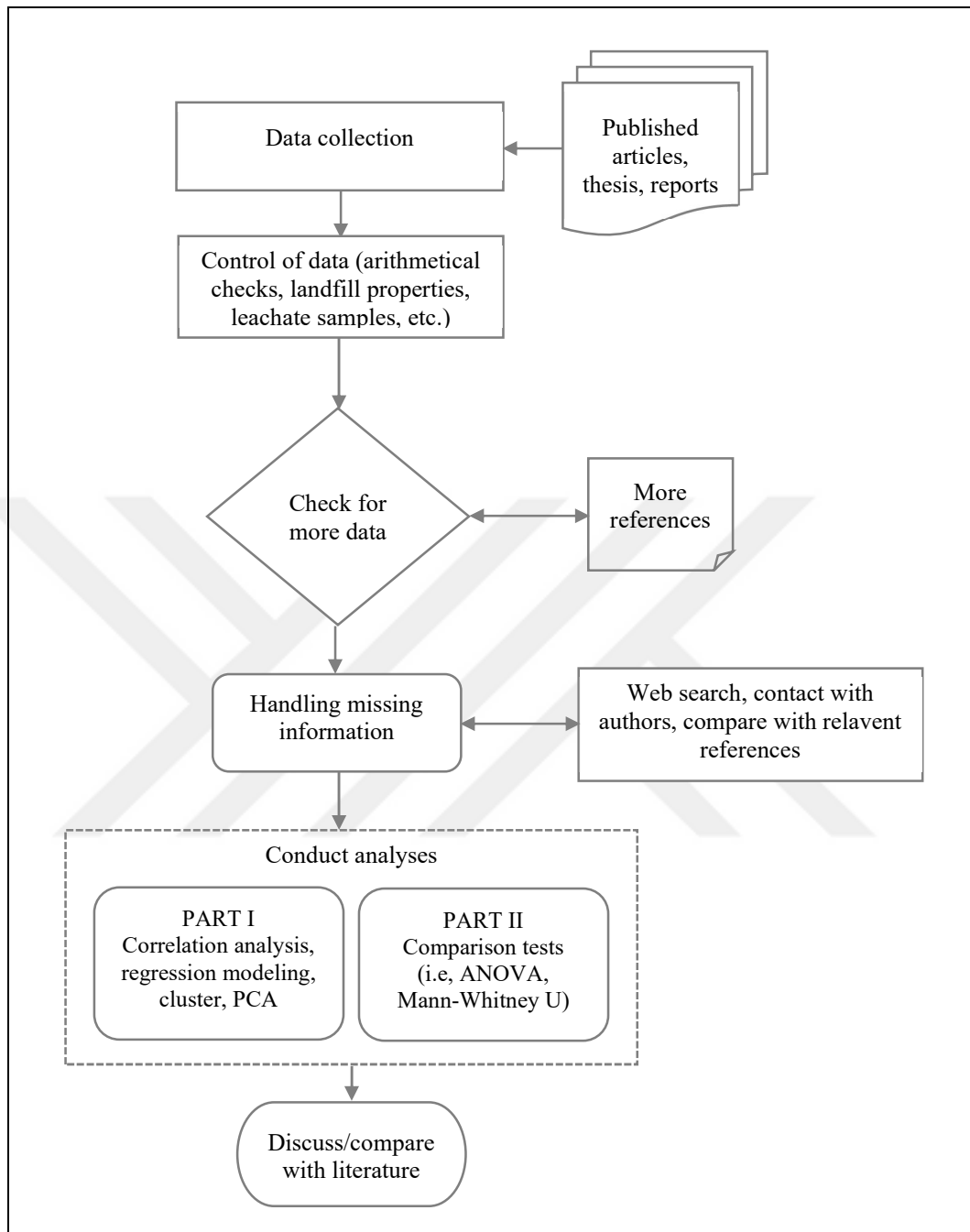


Figure 3.2. Algorithm of major data collection and analysis activities

3.2 Analysis Methods and Assumptions

After extensive data collection phase, various multivariate statistical techniques such as principle component analysis (PCA), regression analysis, and cluster analysis were investigated for application to the landfill leachate data set obtained from different municipal solid waste landfills. For comparison of groups, parametric and non-parametric statistical methods were selected including analysis of variance (ANOVA), Kruskal-Wallis and Mann-Whitney U Tests. Objectives of each analysis are presented in Figure 3.3. Details and assumptions about the applied statistical methods are explained in the relevant sections below. IBM SPSS software for Windows version 25 was used for all the statistical analyses with the confidence level of 95% ($p < 0.05$).

Correlation Analysis	• For definition of the degree of correlation between leachate parameters
Regression Modeling	• For modeling of the leachate data having strong correlation with each other (simple/multiple)
Cluster Analysis	• For classification of leachate parameters into groups according to similarities and dissimilarities between them
Principal Component Analysis (PCA)	• For explaining much of the variance in leachate data with relatively few components
ANOVA/ Kruskal-Wallis test	• For making comparisons of leachate parameters between groups (more than 2) and to find out the significant differences
ANOVA Post-Hoc/ Mann-Whitney U test	• For making multiple (pairwise) comparisons of leachate parameters in each pair of groups

Figure 3.3. Objective of each multivariate analysis

3.2.1 Pearson Correlation Analysis

Pearson correlation analysis is used to show the possible relationships between leachate parameters. Parameters having Pearson Correlation Factor (r) > 0.7 are considered to be strongly correlated and others having r between 0.5 and 0.7 are accepted as moderately correlated with a significance level of $p < 0.05$ (Adams et al., 2001; Banar and Özkan, 2008). On the other hand, Wdowczyk and Pulikowska (2020) assumed strong correlation if correlation coefficient r is between 0.7 and 0.9 and moderate correlation if $0.4 < r < 0.7$. This means, if r is less than 0.4 there is a weak correlation and if r is greater than 0.9 there is a very strong correlation with a significance level of $p < 0.05$.

3.2.2 Regression Data Modeling and Multicollinearity

Regression modelling was conducted for modeling of leachate parameters which have strong correlation with each other in order to predict some of the missing data in leachate data matrix. In literature, it is stated that “If a variable has no place in the theory, it should not be included in the regression analysis” (Schroeder et al., 2018). To select the variables for the regression modeling, Pearson correlation analysis was done in the SPSS to see the main relationships between leachate parameters. Only highly relevant parameters were regressed according to the results of correlation analysis ($r > 0.7$). Furthermore, scatter plots were checked for the ones having high correlation. It was clearly observed that linear relationship exists between many leachate parameters. Therefore, simple/multiple linear regression modeling was applied to leachate parameters accordingly.

Multiple linear regression analysis is used to estimate the effects of several variables at the same time. More than two independent variables can be used simultaneously to test the variations in the dependent parameter. On the other hand, if variation in the dependent parameter is associated to changes in only one independent variable, then it is called as bi-variate or simple linear regression (Schroeder et al., 2018). In this study

both methods were employed depending on the correlation status of leachate parameters.

The R in the regression analysis is stated as “coefficient of multiple correlation” and it is similar to r in the coefficient of correlation. Both parameters measure the degree of association between variables. Similarly, R^2 which is the “coefficient of determination”, measures the percentage of the variation occurs in the dependent variable due to the variations in the independent variables. Higher R^2 means that predicted data will be closer to the observed data. Higher R^2 values are good, but they don’t show how far the observed values are from the regression line. It is the standard error of estimate (S) that shows the distance between the observations and the regression line (Schroeder et al., 2018).

Another important indicator used to find the best model is checking the residuals. For that, average and maximum % errors are calculated with below formula:

$$\% \text{ Error} = \frac{\text{observed data} - \text{predicted value (y)}}{\text{observed data}} * 100$$

Consequently, various regression models with different leachate parameters were conducted in the SPSS and the best one was selected for predictions of the missing data according to those conditions of: the lowest S, the highest R^2 , and the lowest % error values. Precedency was given to the S and average % error. Generally, if those are lower, the R^2 would be sufficiently high too. Another important point in selecting the best model was the number of missing data that can be calculated from available data points with the selected model. If only a few number of missing data could be calculated with the best model, then the other models should be selected which provide similar or reasonable S, R^2 , and % error values.

Collinearity is a correlation between two independent variables in a statistical analysis and multicollinearity is when more than two independent variables are associated (SPSS guide, 2021). Collinearity or multicollinearity condition should be checked in

the multiple linear regression analysis. Collinearity was tested in SPSS according to the below criteria (Regorz, 2020):

1. "VIF" value in the table of "Coefficients" is checked. If it is less than 10 for all predictors (independent variables), there is no collinearity problem.
2. If there are only a maximum of two values of the VIF above 10, then it could be assumed that the collinearity problem exists between these two values and do not interpret the "collinearity diagnostics" table.
3. If there are more than two variables with a VIF above 10, then collinearity diagnostics table should be controlled.
4. If the Condition Index is above 15, then on the same lines Variance Proportions are checked. If there are two or more variables above 0.90 in one line, a collinearity problem can be assumed between those.
5. However, if only one variable in a line has a value above 0.90, no collinearity problem is assumed.

3.2.3 Cluster Analysis

Cluster analysis is a method that classifies variables (cases) into groups (clusters) according to similarities within a group and dissimilarities between other groups (Akbal et al., 2011). The degree of relationship is the largest among variables which are in the same group (Talalaj et al., 2016). In a Dendrogram, the appropriate number of clusters can be seen clearly (Bhuiyan et al., 2011; Rinaldi et al., 2014).

In this study, cluster analysis was applied to identify the relationship among leachate pollutants. It helps to visualize possible groupings of leachate parameters according to the correlation status between each other. The order of magnitudes of the values for different variables may be different from each other. For example, COD, BOD, and TDS can be in thousands of mg/L whereas pH and EC are in much lower numbers (i.e.,

between 1-100 max). Therefore, standardization of the data is very important since clustering methods are highly influenced by large variances of the data (Li et al., 2012). This subject is elaborated in the data pre-treatment section.

In this study, hierarchical cluster analysis (HCA) was selected for grouping of leachate pollutant parameters. HCA uses the distance between samples for the measure of similarity (Akbal et al., 2011). In HCA, each case is a cluster of its own at the beginning. Then, the distance between each case is compared with the next one and the closest cases are clustered together. This continues until all the cases are grouped into one big cluster (Wuensch, 2016).

For clustering method, Ward linkage was used to link the clusters and Euclidean distance was used as the interval of clustering (measure of distance) for the leachate parameters. Ward's method employs an analysis of variance approach to assess the distance between clusters and try to minimize the sum of squares of any two clusters formed at each step (Kim et al., 2005). The Ward's method with squared Euclidean distance as a measure of similarity was proven to be very powerful grouping mechanism which uses more information about cluster contents than other methods (Akbal et al., 2011). In fact, they are the ones frequently applied in the literature for leachate as well as other water pollution related studies (Adelopo et al., 2018; De et al., 2017; Liu et al., 2008; Talalaj et al., 2016).

3.2.4 Principal Component Analysis (PCA)

Factor analysis, which includes principle component analysis (PCA) as well, is a multivariate data analysis method used to extract smaller number of factors that explains most of the variation in the original data (Liu et al., 2003; Pastore et al., 2018). The aim is to reduce the high number of variables into a smaller number of factors or components that explain most of the variance in the data. The first factor or component explains the highest variance, the second one explains the next largest variance, and so on. In this study, PCA was performed to find out the possible similarities or differences between high number of leachate samples originated from different aged

landfills as well as climatic conditions. PCA can also help to distinguish between leachate samples/landfills for prevalent transport and biodegradation mechanisms for the tested parameters. PCA was used jointly with cluster analysis to supplement the results of each other as they both show the possible groupings of leachate samples depending on the analysed parameters.

Factor analysis and PCA provide similar results. In this study, PCA method was selected for the analysis of leachate data since it has been used by some researchers in the past for landfill leachate studies (Adelopo et al., 2018; Galvez et al., 2010; Mishra et al., 2016). As said, PCA helps to group variables with similar characteristics into the same components. Hence, variables within each component are highly correlated with themselves but slightly correlated with variables in other groups. It is stated that each variable group has a single reason for the related correlations (Akyol, 2005). In general, the first component is more correlated with the variables than the others. This is expected since each component is extracted successively accounting for as much of the remaining variance as possible (Liu et al., 2003).

In this study of PCA, the variables are the leachate parameters and the objects are the samples from different landfills. In the new system formed by PCA, the coordinates of the objects are called as scores, and the contribution of each parameter to each component is named as loadings (Indelicato et al., 2018). Score plots help to identify groups of objects (i.e., different landfills in this case) or observations (leachate samples) with similarities. On the other hand, loading plots help to identify leachate parameters responsible for similarities or differences among the leachate samples (as given in the score plot). The loading plot also gives information about the correlation structure of the variables (Andreas et al., 1999). The larger the loading value the more that variable contributes to that component. In the loading plot, variables highly contributing to the related component are located far from the origin. Conversely, variables locating close to the origin do not contribute much to these components. Furthermore, variables that are located opposite to each other have a negative covariance (Modin, 2012).

There are some output tables/parameters in PCA that require controlling at each run whether they are within the pre-defined limits or not. The most important parameter that should be checked is the Kaiser-Meyer-Olkin (KMO) value that shows the sample number adequacy for PCA. High value of KMO (>0.6) indicates sufficient number of samples to conduct the PCA (Durmusoglu and Yilmaz, 2006; Zhao and Cui, 2009).

In a PCA, correlations between analysed variables should be checked as well. The variable having no correlation with any other variable should be removed. Variables with correlation of 0.9 or above with another variable (multicollinearity) should be excluded as well. On the other hand, the correlations between variables should be above 0.3 to avoid weak relationships (Galvez et al., 2010). To confirm the absence of multicollinearity, Determinant score could be checked ($>1.0 \times 10^{-5}$) in the PCA outputs (Yong and Pearce, 2013). Since pairs of variables with large partial correlations share variance with one another but not with the remaining variables, this should be checked as well by Anti-image Correlation Matrix in PCA outputs. Variables having small Kaiser's Measures of Sampling Adequacy ($MSA < 0.5$) should be removed from the parameter list (Durmusoglu and Yilmaz, 2006).

Another item in the PCA outputs is the communalities table showing the amount of variance in the variables accounted by the extracted components. Variables with low communalities (<0.2) need to be eliminated from the analysis. Finally, Reproduced Correlation Matrix should be checked to see if the model is a good fit or not. If the model is a good fit, percentage of the non-redundant residuals with absolute values greater than 0.05 would be less than 50% (Yong and Pearce, 2013).

3.2.5 Comparison of Means Tests

Analysis of Variance (ANOVA) was used to find out the differences in the leachate parameters with respect to different conditions mainly landfill age, climate, and development status. ANOVA is a parametric test for comparison of sample (population) means between different groups (Torrance et al., 2009; Treister et al,

2015). The null hypothesis (H_0) is tested in ANOVA against the alternative one (H_1) about the population means as follows:

H_0 : There is no significant difference between means of the leachate samples

H_1 : The mean of at least one group is significantly different from others

The logic of the ANOVA is to compare the variances of groups as between-groups and within-groups. Therefore, total variance between variables is divided into within-group and between-group variances. If the between-group variance is more than within-group one, it can be accepted that at least one group mean is different from the others and H_0 is rejected. For that, calculated p-value should be lower than the significance level which is generally accepted as 0.05. If the p-value is found less than 0.05, the null hypothesis of equal population means (H_0) is rejected and the alternate one (H_1) for a significant difference (statistically) between the population means is accepted (Nist, 2022). In this study, significance level was set at 0.05 as well for the probability of rejecting H_0 when it is true. Hence, the hypothesis of unequal means (H_1) was accepted at the 95% confidence level for leachate samples in different groups.

ANOVA results may suggest that group means are significantly different from each other, but it does not indicate which pair of groups are different. Therefore, ANOVA Post-Hoc tests were conducted as well for pairwise comparisons. Which Post-Hoc test should be used is based on the homogeneity or non-homogeneity of variances among data groups. In this study, Benferroni method was selected for homogeneous variances and unequal sample sizes and Games-Howell method was used for non-homogeneous variances and unequal sample sizes for multiple comparison of means (Post-Hoc, 2016). In the non-parametric tests, equivalent methods were used as Kruskal–Wallis (named as K independent samples test in SPSS) to compare all the groups, and Mann-Whitney U (named as 2 independent samples test in SPSS) for pairwise comparisons (Torrance et al., 2009). Therefore, in the non-parametric tests, at first leachate parameters were compared between all the groups by using Kruskal-Wallis test. If

significant difference was found ($p < 0.05$), then Mann-Whitney U test was applied for comparisons between pair of groups one by one (Torrance et al., 2009).

Main effect of factor groups on leachate parameters can be found by one-way ANOVA. However, there could be interaction effect of different groups on the same parameter (Yusof et al., 2009). Therefore, it should be investigated that how precipitation values in the landfill area interacts with the effect of landfill age on the leachate parameters, or how development status interacts with landfill age and so on. Interaction effect is mainly the influence of one group on the leachate parameters depending on the other groups. Two-Way ANOVA (Factorial ANOVA) was used to find out possible interaction effects of the groups on leachate parameters by employing SPSS Univariate function and relevant plots. Interaction effect is very important in this study because leachate samples come from different sites in different ages and climates. It should be analysed how climate variable interacts with the landfill age effect. For example, interaction effect could tell that the variation of a leachate parameter between climate factors is much higher in one or more age categories. It is relevant for development status and age effects as well. Instead of two as in one-way ANOVA, there are three hypotheses to be tested (Yusof et al., 2009): the main effects of two groups (i.e., climate and landfill age) and the interaction effect. Interaction effect should be tested for significance before the main effects.

There is an important drawback with two-way ANOVA that it is not possible to get any conclusive finding about the interaction effect. It is not clear between which groups there is an interaction effect. In that respect, profile plots help to get some understanding about the trend in parameter values between groups. Profile plots show how the related parameter changes between different groups. If the gap between lines is large, it refers to a considerable difference between groups (i.e., climates). If this gap is bigger in one or more groups it shows that there is an interaction effect. Furthermore, in two-way ANOVA marginal means are used in some outputs and results which could create different results than one-way ANOVA. Marginal means are calculated assuming all groups have an equal number of parameters. But in many cases, group numbers are not equal resulting in slight differences between the real and

marginal means. Therefore, according to the results of two-way ANOVA and profile plots, one-way ANOVA was conducted between individual groups in order to have a deeper perspective on the findings. This was achieved by isolating the data in SPSS (by select cases function) for a specific group and then performing the one-way ANOVA for other groups. Those issues are elaborated accordingly in the results section of Chapter 6.

There are two major requirements of ANOVA:

1. Leachate data in each group should be normally distributed. Transformation of the data (by Log10, Ln etc.) could be done to achieve normal distribution if required.
2. Group variances should be equal. If not Welch's Test could be used to test the differences between groups and relevant Post-Hoc multiple comparison test can be used.

It is highly recommended to check the data for normal distribution before a parametric test is applied. If the data are not normally distributed, then the parametric test should not be applied (Treister et al, 2015). It was stated that most of the data sets achieve normal distribution when data transformation is applied (Akyol, 2005; Brennan et al., 2016; Kjeldsen and Christophersen, 2001). Therefore, in this study normal distribution was checked and if violated, transformation of the data was applied. Whether the data is normal or not was checked by using descriptive statistics/explore function of the SPSS. The Kolmogorov-Smirnov and Shapiro-Wilk tests were used to assess the normal distribution of leachate data set (both raw and transformed data). If $p > 0.05$ was achieved in either of those tests, data was accepted to be normally distributed (Treister et al, 2015).

If neither the original nor the transformed data (commonly Log10) have a normal distribution then other methods that does not require normal distribution of the data could be applied like non-parametric tests of Kruskal-Wallis and Man Whitney U. Advantage of the non-parametric tests is small number of samples would be enough for the required analysis beside being distribution free (Treister et al, 2015; Wdowczyk and Pulikowska, 2020).

In some references it is indicated that both parametric tests without normal distribution of the data and non-parametric tests could provide the same results (Torrance et al., 2009). However, this could depend on many properties of the analysed data including complexity of the data, adequacy of the number of samples, and degree of variations between the data. Therefore, in this study parametric tests were tried to be used as much as possible if the conditions stated above are met. If not, non-parametric tests were conducted.

3.3 Data Pre-treatment and Outlier Handling

For the statistical analysis of data, there are number of methods that may require different conditions as a pre-requisite for obtaining accurate results. Therefore, it is a very crucial step to prepare the data set for a proper analysis with the right approach (van den Berg et al., 2006). Especially, collection of the leachate data had some restrictions in this study due to availability of data for each pollution parameter. Also, existence of outlier data due to special characteristics of landfill leachate in its own merit was an important problem. Outlier data can influence the results considerably so should be treated properly (Stanimirova et al., 2007; van den Berg et al., 2006).

To overcome those problems about the data structure, outlier and missing data subjects should be meticulously handled. Unfortunately, it is not possible to complete whole missing data due to the limitations or to remove all the outliers to prevent the data loss. For that reason, some relevant parameters in the leachate composition cannot be included in all of the analyses. This situation is hard to avoid and prevents to unveil the whole picture with full list of critical leachate parameters in order to be able to make comparisons with other studies (Galvez et al., 2010; Modin, 2012).

3.3.1 Data Pre-treatment

Data pre-treatment issue for the complex statistical techniques is very important and this subject was not handled sufficiently in other studies which could prevent to make

proper comparisons. Therefore, a detailed investigation was done for necessary data pre-treatment methods and application results are presented accordingly for guidance to future studies.

Each statistical method can have some pre-conditions before conducting the analysis to get accurate results. For example, it is stated that parametric tests such as ANOVA can be assumed only accurate when the data is normally distributed. Therefore, it is required to check the data for normal distribution before conducting parametric tests (Treister et al., 2015).

Furthermore, a uniform variance is required in the ANOVA test since it is tried to find the differences between the groups having more or less similar variances. Only some leachate parameters follow a normal distribution with uniform variance. Others need to be transformed by a suitable method. Generally, when the data is transformed and normal distribution is obtained, the variances become uniform as well (Bland and Altman, 1996).

Transformation of the data could be done by taking the logarithm, square root, or some other calculations of the data. After transforming the data, they are analysed instead of the raw data. The logarithmic (log) transformation is the most common method that generally makes the data normally distributed (Brennan et al., 2016; Wallace et al., 2015). By log transformation the most similar variances could be obtained to achieve the most valid test of significance. Also, log transformation allow interpretable results after transforming back (antilog) in relation to the original data (Bland and Altman, 1996).

Although it is an accepted approach that large samples will follow a normal distribution, in this study normal distribution was checked and if violated, transformation of the data was applied. Whether the data is normal or not was checked by the statistical assessment tests in SPSS. The Kolmogorov-Smirnov and Shapiro-Wilk tests were used to assess the normality of leachate data set (both raw data and the transformed data). If $p > 0.05$ was achieved in either of those tests, data was accepted to be normally distributed (Treister et al, 2015). If transformation of data results in

normal distribution in some groups only, then outlier check was done for other noncomplying groups. Removal of some outliers could help to achieve normality in those groups as well. If normality of the data cannot be achieved for some parameters in any condition then non-parametric tests were applied for them.

It is stated by Bland and Altman (1996) that while other transformations (such as taking square roots or reciprocals) could be used in principle, only the log transformation permit the results to be interpreted in relation to the original data. If it is essential to express the results in relation to the actual measurements, then it is better not to use other transformation methods in this context. Therefore, Log10 transformation was used in this study even if it requires some data elimination to achieve normality.

PCA and cluster analyses are required to be conducted in the proper data format such as in a standardized format. It is seen that some leachate parameters such as COD, BOD, and TDS are measured in values that are much higher than pH, EC, Fe, and other heavy metals. This huge variation in scale should be handled before the PCA and cluster tests. Therefore, data could be standardized to enable all variables are taken equally in the model (Bro and Smilde, 2014).

The standardization procedure helps to eliminate the influence of different units of measurement and makes the data dimensionless so that influence of different units and order of magnitudes for different parameters would be eliminated (Akbal et al., 2011; De et al., 2017; Liu et al., 2008). Standardization also helps to increase the influence of variables having small variance, and decrease the influence of variables having large variance (Liu et al., 2003). The most common way of standardization is to convert variables into z-scores. In this method, all data is converted into a distribution with an arithmetic mean of "0" and a standard deviation of "1". Thus, data in different measures are brought into the same scale (De et al., 2017; Pastore et al., 2018).

Another important problem in PCA and cluster analysis is the missing values for some parameters in the analysed data list that could be faced in most of the sample data (Mishra et al., 2016). There are 29 different leachate parameters in the leachate data

matrix including conventional leachate parameters (COD, BOD, TOC, BOD/COD, pH, alkalinity, conductivity, NH₄-N, TKN, TN, TSS, TDS, TS, SO₄, TP, PO₄-P, Cl, Na, K, Fe, Mn, Ca, Mg) as well as heavy metals (Ni, Zn, Cu, Pb, Cd, Cr). However, values for all those parameters were not given in all the references. Therefore, there is a discontinuity problem in the whole data set due to those missing values. When a number of leachate parameters is selected for the PCA and cluster analyses, cases with missing data were excluded automatically. Hence, total number of data to be analysed (sample N) decreases considerably depending on the number of missing data.

Dealing with missing data is a challenging issue in statistical analysis. Many authors use mean or median replacement for the missing data not to lost the data by elimination method (Modin 2012; Civan et al., 2015; Stanimirova et al., 2007). It was stated by Modin (2012) that acceptable amounts (<10%) of missing data were replaced by parameter mean in their study. Also, exceptionally they accepted 12.5% missing data substitution for some parameters as required in one of their studies. For the sake of increasing the data numbers to a satisfactory level in the statistical analyses like ANOVA, cluster analysis, and PCA, missing data should be completed with sufficient accuracy. Therefore, regression data modelling was preferred for that in this study as there is a good correlation between many leachate parameters. As an acceptable limit, 15% missing data completion ratio was accepted reasonable due to the high prediction levels in many leachate parameters with the regression modeling.

3.3.2 Handling Outlier Data

Leachate data used are contributed from around the World from different types of climates and landfill ages, therefore many outlier data could be observed. Outliers can negatively affect regression line, correlation, means, and standard deviations. It was shown that elimination of the outlier data can improve the prediction performance of the regression model (Yuzugullu and Aksoy, 2014). The outlying data can easily be detected on the scatter plots and they can be eliminated from the data set (Stanimirova et al., 2007). However, not all the outliers have been removed due to possible loss of

valuable data. Instead, only a few data were removed with the use of Prediction Intervals (PI) in scatter plots. The Prediction Interval (PI) shows the likelihood of a future observation being in a given value range for the similar settings of predictors (Forthofer et al., 2007; Frost, 2021a). A PI of 99% was selected in SPSS to limit the data loss by outliers. Data that falls out of the PI range was first checked within the data matrix against the quantities reported in the literature for a given leachate parameter. Only if it was not in the expected range, and somehow created divergence from the common pattern, then it was excluded from the regression modeling study as well as from the data set permanently. The list for removed references is provided in Appendix B (Table B.1).

General approach used for elimination of outlier data from the relevant regression models (i.e., Na vs Cl, EC vs alkalinity etc.) can be explained as follows:

- To minimize the data loss only ones that lie behind the 99% prediction interval of the regression line and that result in low R^2 (<0.7) were eliminated from the data set permanently.
- Minimum number of data were eliminated for R^2 to approach and take a value around 0.7. Then it was stopped for that case and no more data was eliminated even if they stand outside the 99% prediction interval (Table 3.2).
- Outliers were checked for other cases as well to decide which parameter is problematic and should be removed. For example, for the same data points (i.e., 277 in Figures B.2 and B.3) EC vs Na and EC vs K plots were checked to see which parameter (Na, K, or EC) is an outlier. If in both cases the same data is an outlier then common parameter (in that case Na and K) were deleted. Otherwise, outlier data was deleted accordingly by checking their general pattern through the data set. For example, it was checked if the data is the highest or the lowest value amongst data set, or whether it is outside the observed range when compared with other parameters in similar conditions (landfill age, climate etc.) and so on.

Table 3.2 Details of the removed outlier data in the leachate data set

Landfill ID	Outlier parameter	Regression model	R ² (new*)	Explanation**
150, 151, 168	Alkalinity	EC vs Alkalinity NH ₄ -N vs Alkalinity	0.486 (0.605) 0.565 (0.658)	Alkalinity is too high with respect to low EC and NH ₄ -N values
196	Alkalinity	EC vs Alkalinity NH ₄ -N vs Alkalinity	0.605 (0.752) 0.658 (0.686)	Alkalinity is too low with respect to high EC and NH ₄ -N values
37	Cl	Cl vs EC	0.620 (0.709)	Cl is too high with respect to low EC value
81	EC	EC vs NH ₄ -N	0.682 (0.720)	EC value is too low with respect to high NH ₄ -N value
277	Na	Na vs Cl Na vs EC	0.601 (0.821) 0.489 (0.695)	Na is too high with respect to low Cl and EC values
277	K	K vs EC	0.404 (0.726)	K is too high with respect to low EC value
294	Na	Na vs TDS	0.609 (0.819)	Na is very high with respect to low TDS value

*new R² after removal of outlier(s)

**related scatter plots are given in Appendix B

The only data treatment done for PCA and cluster tests were standardization into Z-scores in most of the studies (De et al., 2017; Indelicato et al., 2018; Kim et al., 2005; Liu et al., 2008; Pastore et al., 2018). However, there could be some outliers due to the nature of data set having leachate data from different landfills. Therefore, outlier check is required for leachate parameters in Cluster analysis and PCA parts as well. For that purpose box plots were drawn to see the possible outliers according to the selected parameters for analysis. There was not any permanent removal of references but only elimination of some extreme outliers from the data set or removal of parameter itself from the data list of analysed parameters. The former was applied generally as presented in Section 5.3.4.

CHAPTER 4

A REVIEW FOR LANDFILL LEACHATE CLASSIFICATION

4.1 Introduction

Leachate management can be a complicated task due to leachate composition which is often expressed in terms of its strength, biodegradability, and toxicity which can change over time as waste decomposes in a landfill (Neczaj et al., 2008). Variations in the amount of pollutants and specification of leachate are correlated mostly with the age of the landfill. It was shown in a four-year leachate monitoring study that the age of a landfill has a significant effect on leachate composition (especially organics and ammonium nitrogen concentrations). On the other hand, no significant variations were stated for other parameters like phosphorus, chlorides, magnesium, calcium, sulphates, dissolved solids, and suspended solids with respect to landfill age (Kulikowska and Klimiuk, 2008). Conversely, Andreottola and Cannas (1992) indicated that some main inorganics such as heavy metals, chloride, sulfate, etc. gradually decrease with time beside the organics. It was also stated that although landfill age affects leachate characteristics significantly, changes in leachate composition do not only depend on that but also on the stabilization degree of buried waste and infiltration of water (Lu et al., 1984). Lu et al. (1984) emphasized that landfill age is just a convenient way of evaluating removal of pollutants from the waste and related changes in the composition of produced leachate. Beside the age of the landfill, seasonal factors and waste management applications, for example previous recycling and separation of waste in the composting or recovery plants, storage of leachate in ponds and recirculation back to the landfill, the presence of active and closed cells within the same landfill could affect the leachate quality and quantity (Galvez et al., 2010).

Leachates generated in new landfills are typically high strength wastewaters characterized by low pH, high organic content, high BOD/COD ratio, and presence of

several hazardous compounds. On the other hand, leachates from older landfills mainly contain refractory organic compounds, have low BOD/COD ratio, and high concentrations of $\text{NH}_4\text{-N}$ (Adhikari and Khanal, 2015; Neczaj et al., 2008). As a result, due to high fluctuations in leachate composition, monitoring is required for an extended period of time for representative results and better characterization that may aid in proper leachate management. Operation of a leachate treatment plant can be modified/upgraded based on these variations such that at some times high volumes of diluted leachate should be treated while in other times only a small amount but high-strength leachate. Therefore, leachate treatment plants need to be designed and upgraded accordingly to handle those variations (Galvez et al., 2010). This requires an estimation about expected variations in leachate quality and quantity.

Leachate classification is important in landfill management, including closure (Hussein et al., 2019; Kim and Lee, 2009), planning and design of treatment alternatives through the landfill life span (Lo, 1996). It was observed that for some type of leachate where COD and BOD concentrations and BOD/COD ratio are relatively high, biological treatment alternatives are prevailing. For other types of leachate cases where COD and BOD amount is lower and mostly refractory compounds exist, chemical and physical treatment technologies are preferred (Renou et al., 2008). For this reason, estimating the leachate composition according to the landfill age would be valuable for the planning phase of leachate management. Especially BOD/COD ratio showing the biodegradability as well as being the indicator of leachate age would be important (Alvarez-Vazquez et al., 2004).

A matter of debate was raised by Kamaruddin et al. (2017) that it is the decomposition process of waste that actually affects leachate quality but not the landfill age itself. It is indicated that many researchers are prone to relate the landfill age with leachate quality instead of biodegradation of waste. The reason for this conduct is linked to the fact that it is very hard to estimate leachate quality in relation to the waste stabilisation stage. Therefore, it is found practical to employ the landfill age based on the operation time of a landfill for that purpose. Yet, leachate characteristics based on landfill age

have not been defined precisely in the literature. As is discussed in this chapter, there are different approaches for the classification of leachate in the general and scientific literature. Researchers try to identify their studied leachate according to common understanding and some basic considerations that generally arise from the past experiences and/or relevant published studies in waste management. It would be helpful to remark here that landfill or waste age is the principal factor for establishing leachate age terminology. It means when the leachate is termed as young or old it is the landfill or waste age. Most of the researchers prefer to use this leachate age term to define their leachate type.

The main objective of this review is to have a critical look at leachate age/type concept which has been evaluated differently in the literature. Historical developments are presented about leachate age classification to emphasize the ambiguity and differences between approaches and to highlight the need to be cautious for researchers in using each other's work. In the following chapters more detailed discussion is presented for the impact of landfill age on leachate characteristics by evaluating the results of performed multivariate statistical analyses.

4.2 History of Leachate Studies and Appearance of Leachate Types

Concerns about the leachate dates back to 1940s when the wells in the vicinity of disposal sites/ponds of both solid and liquid wastes got polluted. As stated by Steiner et al. (1979), one of the earliest studies on leachate was in 1940 in which boreholes were drilled throughout a landfill to collect and analyse leachate samples. Later on, in 60s and 70s, adverse effects of leachate on water supplies like surface and groundwaters were detected. Chian and DeWalle (1977) and Englehardt et al. (2006) reported that leachate treatment studies started in the early 1970s.

One of the first studies on composition of leachate from sanitary landfills was conducted in 1954 and after that lysimeter studies for representing landfill conditions and leachate productions increased accordingly (Chian and DeWalle, 1977; Lu et al.,

1984). In 1970, three landfills in Northern Illinois were investigated and leachate characteristics were provided for different years of landfill operation of 0.5, 6, and 17 years which showed a decreasing trend in leachate pollutant concentrations with time such as organics and solids (Steiner et al., 1979).

As far as seen from the reviewed literature, the first structured classification of landfill age was done by Chian and DeWalle (1977). They introduced the concept of age such that if the landfill is less than 5 years old, it is young, if between 5 to 10 years, it is medium-aged and if higher than 10 years it is an old landfill. Chian and DeWalle are the first researchers indicating that age of a landfill represents the degree of waste stabilisation, it affects leachate composition significantly, and it is best measured by the COD content, BOD/COD and COD/TOC ratios as presented in Table 4.1. It was emphasized that usage of COD/TOC ratio instead of BOD/COD ratio could be better since BOD analysis maybe more susceptible to variations and mismeasures than TOC analysis (Chian and DeWalle, 1977). Similar to BOD/COD ratio, COD/TOC ratio decreases as well with increasing landfill age.

Table 4.1 Leachate characteristics as defined by Chian and DeWalle (1977)

Age of a landfill	BOD/COD	COD/TOC	COD (mg/L)
< 5 years	>0.5	>2.8	>10,000
5 - 10 years	0.1-0. 5	2.0-2.8	500-10,000
> 10 years	<0. 1	<2. 0	<500

Chian and DeWalle (1977) proposed to use ratios of different parameters instead of their absolute values (i.e., no COD or BOD but BOD/COD ratio) because concentrations of those pollutants can vary considerably by time. They stated that ratios of chemical components like COD/TOC and BOD/COD are the best parameters for representing the characteristics of leachate and indicating the amount of organic compounds present in leachate, as well as relating to the age of a landfill. Chian and DeWalle (1977) selected those leachate parameters to predict the stabilisation status of a landfill for the purpose of identifying proper leachate treatment options. They declared at least four parameters: landfill age, BOD/COD and COD/TOC ratios, and

COD content should be known to determine the most appropriate treatment method. Chian and DeWalle (1977) also stated that having only a limited number of samples being investigated, it was difficult to provide corresponding ages for young, medium, and old landfills. With the available data they assumed that the first 5 years of a landfill life indicates a young phase, five to ten years corresponds to intermediate phase, and higher than 10 years indicates an old phase. Yet, they stated that those landfill age ranges are very tentative and many more leachate samples have to be analysed from different aged landfills to find out more accurate ranges for a full-classification. They also recommended further research for correlation of leachate characteristics with landfill age due to considerable variations observed between samples from different landfills. The reasons for those variations were connected to different climatic conditions and specific characteristics of each landfill which complicates further the correlation of measurements with the age of landfills. Steiner et al. (1979) referenced Chian and DeWalle in their report as well for bringing an order to the disagreement found in the literature about the relationship between landfill age and characteristics of leachate as presented (Table 4.1).

Leachate age classification according to whether being in methanogenic stage (old field) or acidogenic stage (new field) was done the first time by Cord-Landwehr et al. (1982). After that in a US EPA report prepared by Pohland and Harper (1985), landfill age was linked directly to the degree of waste stabilisation as well as to the quantity and quality of leachate and gas productions. Leachate was classified into three categories for comparison of treatment performances. Influent concentrations as COD and BOD, and biodegradability ranges as BOD/COD ratio were provided in Table 4.2.

Table 4.2 Leachate categories defined according to Pohland and Harper (1985)

Leachate type	COD (mg/L)	BOD (mg/L)	BOD/COD
Low-strength	<1,000	220-750	<0.50
Medium-strength	1,000-10,000	750-1,500	0.50-0.75
High-strength	10,000	1,500-36,000	>0.75

Jasper et al. (1985) stated leachate organic composition changing according to the time spent in the landfill, water content, and leachate collection system. They also explained the status of leachate as “young” and “old” according to the state of methanogenesis. In old leachate there is an active methanogenesis, hence low COD. In young leachate, on the other hand, high COD concentrations prevail due to lack of methanogenesis. Harmsen (1983) confirmed the relationship of leachate organic composition with methanogenesis conditions but not with the age of a landfill. Similarly, Heyer and Stegmann (2001) defined three distinct periods for leachate according to the BOD/COD ratio: (I) acid phase ($\text{BOD/COD}=0.4$), (II) intermediate phase ($0.2<\text{BOD/COD}<0.4$), and (III) methanogenic phase ($\text{BOD/COD}=0.2$).

Henry et al. (1987) demonstrated general characteristics of leachate with respect to different landfill ages as given in Table 4.3. Unlike Chian and DeWalle (1977) they proposed four different landfill ages (young, mature, ageing, and old) without assigning any specific years. They also named leachate types differently than other researchers mentioned above. They stated the difficulty of designating meaningful ages for landfills due to continuous waste disposal activities over a long period of time. Baccini et al. (1987) remarked the same problem indicating that each part in a landfill has a different residence time and this temporal diversity makes it complicated to define a specific age for a landfill.

Table 4.3 Leachate characteristics depending on landfill ages by Henry et al. (1987)

Landfill age	Leachate type	BOD/COD
Young	Raw, undegraded	0.7
Mature	Partly degraded	0.5
Ageing	Partially stabilized	0.3
Old	Well stabilized	0.1

Lema et al. (1988) employed the terminologies young, medium, and old to define a landfill or waste age and provided leachate characteristics accordingly. However,

specific operation times to identify a landfill as young, medium, or old were not provided.

Forgie (1988) stated that due to leachate strength changing with time, it is common to mention about high, medium, and low strength leachates as well as young-acid-phase and old-methanogenic leachate. It was also indicated that time required for transition from young to old leachate would require a time period from 3-5 years to 6-10 years following landfill start-up. However, it was said that a shorter time period (i.e., 2 years) would be observed in some specific cases as well.

4.3 Effect of Landfill Age and Other Parameters on Leachate Classification

For the sake of a standard comparison, most of the researchers utilized a common definition as given in the literature so that leachate is generally classified as one of those: young/fresh/recent, medium/intermediate, or old/mature/stabilized leachate. This is done mainly according to the landfill or waste age which is defined as the time passed between the first placement of waste (landfill start-up date) and leachate collection (sampling time) (Brennan et al., 2016; Reinhart and Grosh, 1998). Beside the landfill age, the most commonly used criteria for leachate classification is the BOD/COD ratio. If the BOD/COD ratio is high, leachate is accepted as young. If it is very low then leachate is referred to be old or stabilized leachate. However, it was emphasized by some authors that BOD/COD ratio of an older landfill site (i.e., 18 years) could be higher than a younger site (i.e., 8 years), therefore stabilisation is not always directly linked with the chronological age of the landfill due to the complications mentioned in this chapter (Henry et al., 1987; Rowe 1995). Moreover, if the pH is very low (5-6) then acidogenic conditions (stage III) are foreseen and leachate is defined as young, if it is higher (8-9) then methanogenic conditions (stage IV), hence old/stabilized leachate is assumed. This is again not relevant for landfills in some countries depending on high alkalinity conditions in the waste (i.e., high ash content) so that even in the initial phases acidic pH may not be observed.

A number of different ranges of years has been used in classification of landfill/leachate age. These are presented in Figure 4.1a as collated from 42 studies in the literature. It is seen that more than half of the reviewed studies (57%) employed the range band of less than 5 years, 5 to 10 years, and more than 10 years for young, medium, and old landfill age definition, respectively (Table 4.4, Figure 4.1a). The ranges for BOD/COD ratio used frequently for classification of leachate type are provided in Figure 4.1b .

As given in Table 4.4, numerous authors classified leachate according to landfill age with similar terminology as young, medium, and old; while some others defined a little bit differently. Especially medium and old leachate were named interchangeably with other terms such as for medium leachate “intermediate” and for old leachate “mature” or “stabilised” terms are used frequently. Accepted ranges for parameters like BOD/COD ratio and pH showed variances between references as well. On the other hand, age was assumed greatly in the same way in many references as had been first proposed by Chian and DeWalle (1977) decades ago (i.e., less than 5 years, more than 10 years, and in between); though there are other references as well in which different values were accepted (Figure 4.1a as compiled from references in Table 4.4).

As aforementioned, many more studies classified leachate type as young, medium/intermediate, or old/mature/stabilized according to one of the parameters given in Table 4.4, mostly using BOD/COD ratio (Bakraouy et al., 2017; Berthe et al., 2008; Chofqi et al., 2004; Comstock et al., 2010; da Costa et al., 2018; El-Fadel et al., 2002; Filho and Miguel, 2017; Lopez et al., 2004; Ozkaya et al., 2006; Ren et al., 2017; Zamora et al., 2000), age of the landfill (Calace et al., 2001; Lee et al., 2010; Pereira et al., 2018; Sruthi et al., 2018), or COD concentration (Chofqi et al., 2004; Lopez et al., 2018; Zamora et al., 2000). The number of references, as compiled from Table 4.4, employing different ranges of BOD/COD ratio for classification of leachate is presented in Figure 4.1b. It is seen that although different ratios are used, leachate classification has been concentrated mostly on BOD/COD ratio of greater than 0.5 or 0.3 and less than 0.1.

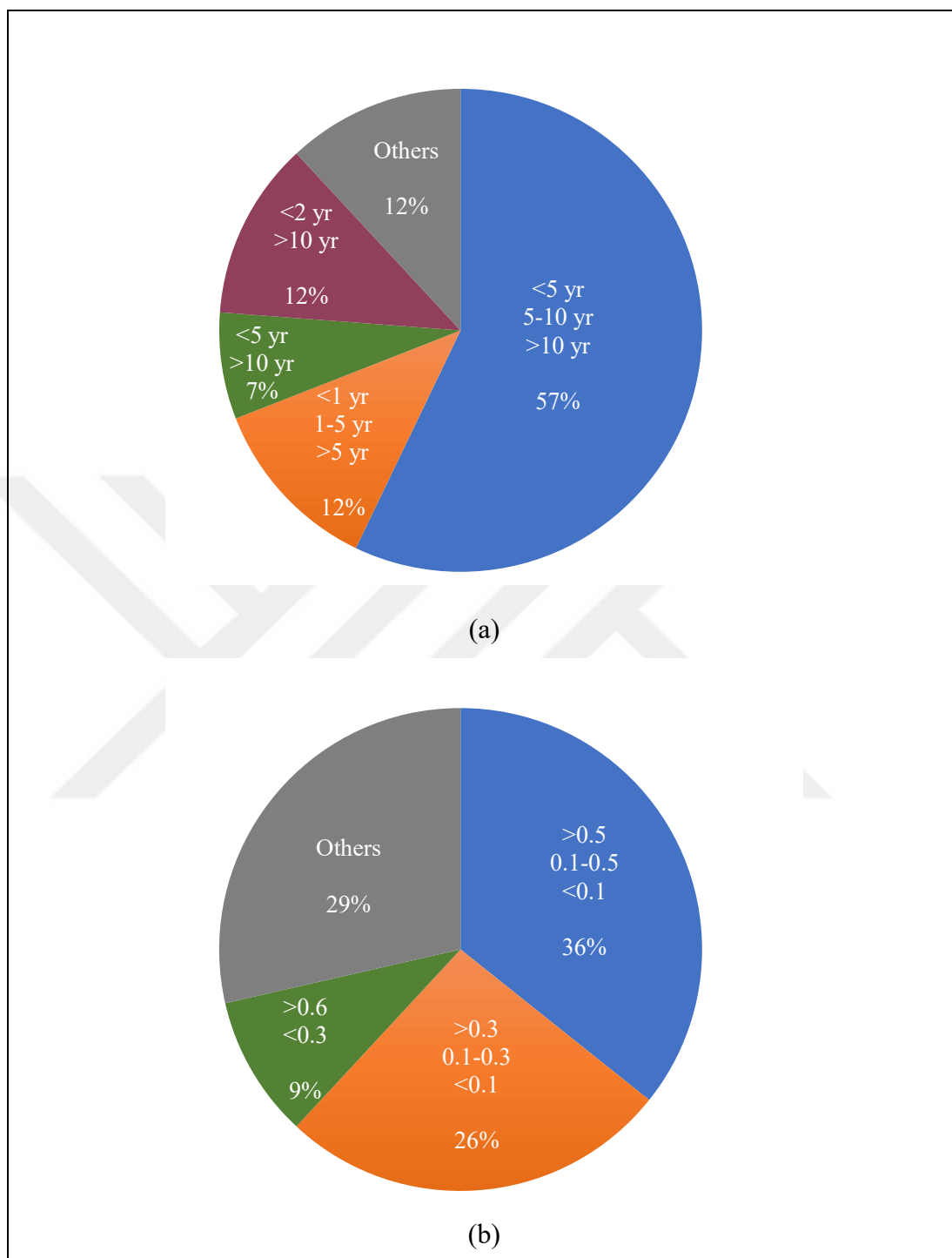


Figure 4.1. Distribution of (a) landfill age criteria (b) BOD/COD ratio used for leachate classification in 42 studies collated from literature

Table 4.4 Different leachate classification terminologies as given in the literature depending on landfill age and other parameters

Leachate Definition	Landfill Age (years)	BOD/ COD	COD/ TOC*	pH	References	Leachate Definition	Landfill Age (years)	BOD/ COD	COD/ TOC*	pH	References
Young Intermediate Stabilized	<5	>0.3	3.3	<6.5	Schiopu and Gavrilescu (2010); Baig et al. (1999)	Young Medium Old	<1	>0.6	NG	NG	Alvarez-Vazquez et al. (2004)
	5-10	0.1-0.3	-	7.0			1-5	0.3-0.6			
	>10	<0.1	2.5	>7.5			>5	<0.3			
	<5	>0.3	NG	<6.5	Zainol et al. (2012); Zakaria and Aziz (2018)		<1	>0.5	<3.3	<6.5	Gao et al. (2015)
	5-10	0.1-0.3		6.5-7.5			1-5	0.1-0.5	2.0-3.3	6.5-7.5	
	>10	<0.1		>7.5			>5	<0.1	>2.0	>7.5	
	<5	>0.5	<3.3	<6.5	Talalaj et al. (2019)		<5	>0.5	>2.7	<6.5	Amokrane et al. (1997)
	5-10	0.1-0.5	2.0-3.3	6.5-7.5			5-10	0.1-0.5	2.0-2.7	6.5-7.5	
	>10	<0.1	>2.0	>7.5			>10	<0.1	<2.0	>7.5	
	<5	>0.5	NG	<6.5	Ahmed and Lan (2012)		<5	>0.5	>2.8	NG	Chian and DeWalle (1977)
	5-10	0.1-0.5		6.5-7.5			5-10	0.1-0.5	2.0-2.8		
	>10	<0.1		>7.5			>10	<0.1	<2.0		
<5	>0.5	<3.3	<6.5	Foo and Hameed (2009)	<5		0.4	NG	NG	Corsino et al. (2020)	
5-10	0.1-0.5	2.0-3.3	6.5-7.5		5-10		<0.2				
>10	<0.1	>2.0	>7.5		>10		0.1				
<1	>0.5	<3.3	<6.5	Liu (2013); Kurniawan et al. (2006)	<5		≥0.5	NG	NG	Naveen et al. (2014)	
1-5	0.1-0.5	2.0-3.3	6.5-7.5		5-10		0.1-0.5				
>5	<0.1	>2.0	>7.5		>10		<0.1				

Table 4.4 (continued)

Leachate Definition	Landfill Age (years)	BOD/ COD	COD/ TOC*	pH	References	Leachate Definition	Landfill Age (years)	BOD/ COD	COD/ TOC*	pH	References
Young Intermediate Old	<5	>0.3	NG	6.5	Bhalla et al. (2013); Mishra et al. (2018); Yildirim et al. (2018); Ngo et al. (2008)	Young	<5	0.4-0.8	NG	NG	Costa et al. (2019)
	5-10	0.1-0.3		6.5-7.5		Medium	5-10	0.1-0.4			
	>10	<0.1		>7.5		Old/Mature	>10	<0.1			
	<5	NG	NG	6.5	Sil and Kumar (2017)	Young	<1	>0.5	NG	NG	Karimipourfard et al. (2019)
	5-10			6.5-7.5		Medium	1-5	0.1-0.5			
>10	>7.5			Mature		>5	<0.1				
<5	>0.3	NG	<6.5	Aziz et al. (2018)	Young Intermediate Mature	<5	≥0.5	NG	NG	Luo et al. (2020)	
5-10	0.1-0.3		6.5-7.5			5-10	0.1-0.5				
>10	<0.1		>7.5			>10	<0.1				
<5	0.7	NG	NG	Brennan et al. (2017)	<5	>0.3	NG	6.5	Adhikari and Khanal (2015)		
5-10	-				5-10	0.1-0.3		6.5-7.5			
>10	<0.2				>10	<0.1		>7.5			
Recent Intermediate Old	<5	>0.3	NG	6.5	Renou et al. (2008)	Early	<5	0.5-0.7	NG	6.5-7.5	Wang et al. (2018)
	5-10	0.1-0.3		6.5-7.5		Medium	5-10	0.3-0.5		7.0-8.0	
	>10	<0.1		>7.5		Old	>10	<0.3		7.5-8.5	
Young (fresh) Intermediate Mature	<2	>0.5	<3.3	4.5-7.5	Kamaruddin et al. (2017)	Fresh	<5	>0.5	NG	<6.5	Ye et al. (2020)
	2-10	0.1-0.5	2.0-3.3	6.5-7.5		Intermediate	5-10	0.1-0.5		6.5-7.5	
	>10	<0.1	>2.0	6.6-7.5		Mature	>10	<0.1		>7.5	
Young Mature	<2	0.4-0.6	NG	4.5-7.5	Noerfitriyani et al. (2017)	Young Old/Stabilized	NG	>0.5	NG	<6.5	Ghafari et al. (2010)
	>10	0.02-0.5		6.6-7.5				<0.1		>7.5	

Table 4.4 (continued)

Leachate Definition	Landfill Age (years)	BOD/ COD	COD/ TOC*	pH	References	Leachate Definition	Landfill Age (years)	BOD/ COD	COD/ TOC*	pH	References
Young Stabilized	<5	0.4-0.7	>2.8	4.5-6.5	Kurniawan et al. (2010)	Young Old	<5	NG	NG	NG	Robinson et al. (2004)
	>10	<0.1	<2.0	7.5-9.0			>5				
	NG	0.4-0.5 -	NG	<6.5 >7.5	Umar et al. (2010)		<1–2 >5–10	>0.6 <0.3	NG	6.0 6.6-7.5	Deng and Englehardt (2007)
	<5 >10	0.4-0.7 <0.1	NG	<6.5 -	Aziz et al. (2011)		<1-2 >10	>0.6 <0.3	NG	4.5-7.5 6.6-7.5	Englehardt et al. (2006)
Old	>10	<0.1	NG	NG	Kulikowska and Klimiuk (2008)		<1-2 >10	>0.6 <0.3	NG	NG	Hermosilla et al. (2009)
Young	<5	>0.5	NG	NG	Bourechech et al. (2018)		<5 >10	>0.5 <0.1	NG	NG	Fernandes et al. (2015)

* In some references TOC/COD ratio was given, so it is reversed accordingly. NG: Not Given

Some of the authors just mention leachate type as young or old according to one of the parameters given in Table 4.4. For example, if the BOD/COD ratio is greater than 0.3, leachate is defined as young (Lak et al., 2018). Pereira et al. (2018) stated that if landfill age is less than 2 years, leachate is young and if it is bigger than 10 years then leachate is old. Zegzouti et al. (2020) classified their leachate samples as young, intermediate, and old according to their landfilling period (<5, 5 to 10, and >10 year). Su et al. (2016) defined their leachate according to landfill ages as young (<3 years), intermediate (3 to 10 years), and old (>10 years). Yong et al. (2018) accepted the leachate as young if BOD/COD is greater than 0.5 and classified their leachate sample as intermediate since BOD/COD was around 0.2. Similarly, Yilmaz et al. (2010) defined their leachate as young considering the high BOD/COD ratio (0.6) and COD value (≈ 40 g/L); Ferraz et al. (2014) classified the leachate used in their study as old due to very low BOD/COD ratio (0.1) and high $\text{NH}_4\text{-N}$ concentration; Kheradmand et al. (2010) characterized their leachate sample taken from a landfill less than 5 years of age as a young leachate due to high values of COD, BOD, and BOD/COD ratio and so on.

Some authors defined leachate age in their studies as young, medium or old without specifying the parameter quantities used for the classification such that landfill age or values of leachate parameters like COD or BOD/COD ratio were not provided (Kabdaslı et al., 2000; Lema et al., 1988; Zhang et al., 2005). Moreover, some authors employed two terminologies at the same time for the same leachate in their articles, for instance old and stabilized leachate (Ghafari et al., 2010; Umar et al., 2010); old, mature, and stabilized leachate (Costa et al., 2019; Karimipourfard et al., 2019); mature and old leachate (Englehardt et al., 2006).

El-Fadel et al. (2002) and Scott et al. (2005), opposed to the common applications in the literature, presented leachate types in four landfill age groups such as 0–5 years, 5–10 years, 10–20 years, and >20 years. However, they have not attributed any names for leachate types corresponding to a specific year. On the other hand, Mukherjee et al. (2015) used the same four landfill age groups to classify the leachate as young (0–5 years), intermediate (5–10 years), stabilized (10–20 years), and old (>20 years).

Zakaria and Aziz (2018) categorized their leachate in a different manner based on the stabilization stages as well as BOD/COD ratio such as young leachate from acid phase, partially stabilized leachate from intermediate phase and stabilized leachate from methanogenic phase. Other researchers also mentioned similar terminology for leachate classification. For example, Adhikari and Khanal (2015) used acid-phase leachate and methanogenic-phase leachate terms according to organic matter content; Gao et al. (2015) presented leachate types according to the landfilling phases such as young leachate for aerobic and acidic phases, old leachate for a methanogenic phase; Hussein et al. (2019) mentioned acid/acidogenic and methanogenic phase leachates.

4.4 Why Leachate is Classified Differently?

4.4.1 Differences in the Used Criteria

As inferred from the extensive literature survey, although there are few parameters commonly used to define the leachate age, such as landfill age, BOD/COD ratio, or pH, there is no standard approach on how to determine it. Parameters that should be employed and value ranges for those parameters in classifying the leachate into a leachate age category (i.e., young, old, etc.) were not set yet. Most of the studies defined leachate type according to the landfill age as first proposed in 70s by Chian and DeWalle (1977) (i.e., 5 and 10 years). Actually, they have done this specification (Table 4.1) according to their investigation of various landfills in the USA of different ages that function in different climates. Although similarities cannot be overlooked, today there have been many changes from then on and advancements in the solid waste management area such as disposal techniques, waste compositions, and waste production capacities of communities, etc. (Modin, 2012). All of those as well as other factors presented in Table 2.1 have some effects on the leachate quality and quantity. For instance, Kulikowska and Klimiuk (2008) stated that in recent landfills, leachate properties become “methanogenic” in a shorter period of time. Therefore, even young landfill leachates can have low levels of COD and BOD/COD ratio (Chen, 1996; Lo,

1996; Rivas et al., 2003). Similarly, some authors defined their leachate as young due to having high COD and BOD values or BOD/COD ratios in spite of landfill age being greater than 5 or 10 years. Examples from some of the references are provided in Table 4.5 accordingly. Therefore, using the same value ranges for defining a landfill age (i.e., 5 to 10 years scale) for specification of the leachate as young, medium or old could be misleading for the conditions of today after almost 50 years from the first proposal.

Table 4.5 Examples for usage of leachate age/type terminology from different aged landfills

Landfill age (years)	Leachate Type*	BOD/COD	COD (g/L)	pH	References
<5	Young	0.4	0.1	5.8	Asibor et al. (2016)
<5	Young	-	11	7.5	Corsino et al. (2020)
5-10	Young	0.3	3.6	6.5	Lee et al. (2010)
>10	Young	-	2.6	6.6	Kalyuzhnyi et al. (2003)
>10	Young	0.5	19	8	Insel et al. (2013)
>10	Young	0.4	17.2	7.1	Lak et al. (2018)
>10	Young	0.6	20.5	8.0	Kabdaşlı et al. (2008)
>10	Young	0.7	5.9	7.9	Rani et al. (2020)
5-10	Intermediate	0.4	5.5	8.2	Baettker et al. (2020)
5-10	Medium	0.1	10.4	7.4	Naveen et al. (2014)
5-10	Medium	0.4	14.6	8.1	Zegzouti et al. (2019)
<5	Intermediate/stabilized	0.4	2.3	8.3	Rivas et al. (2003)
5-10	Intermediate/Old	0.4	6.7	8.1	Barnes et al. (2007)
5-10	Stabilized/mature	0.1	2.4	8.2	Jia et al. (2011)
>10	Old/mature	0.1	40.7	9.1	Vakilabadi et al. (2017)
<5	Old	0.2	8.3	8.8	Mishra et al. (2018)
>10	Old	0.2	9.5	9.1	Zegzouti et al. (2019)
5-10	Mature	-	5	7.9	Chen et al. (2020)
>10	Mature	0.1	41	9.1	Vakilabadi et al. (2017)
>10	Mature	0.4	2.6	8.0	Karimipourfard et al. (2019)
>10	Mature	0.2	0.9	5.7	Lee et al. (2010)

*Leachate type was defined so by the cited authors as given in the table

In Table 4.5, some examples from the literature were provided to show the incompatible uses of leachate age/type terminology. As seen from the table, every author used their own assessment for the definition of leachate partially according to the parameters given in Table 4.4. However, it was done in a way that, while the range of one parameter is obeyed, the others are disregarded or overlooked. For example, despite the landfill age is more than 10 years old, a leachate was classified as young according to the pH as being low (Kalyuzhnyi et al., 2003), or although the landfill age is less than 5 years old, leachate was defined as intermediate or old due to high pH and lower COD or BOD/COD values (Mishra et al., 2018; Rivas et al., 2003). On the other hand, leachate was classified according to the landfill age concept, but other parameters were not within the limits of the commonly defined literature values by other researchers (i.e., Asibor et al., 2016; Vakilabadi et al., 2017; Zegzouti et al., 2019).

4.4.2 Leachate Collection and Sampling Methods

Landfilling is a very long lasting process taking about 20 to 30 years. Hence there will not be a single age of a landfill but combination of different ages related with different cells or sections (Adhikari and Khanal, 2015; Reinhart and Yousfi, 1996). As the waste disposal continues, newly deposited wastes will barely produce any leachate while the older portions will produce stronger leachate and the firstly placed waste piles would be at the end of their stabilisation period and may no longer produce any leachate at all. In most of the landfills, leachate is collected and mixed together from all the landfill units, regardless of the waste age and leachate concentrations. Hence, leachate with lower concentrations and biodegradability from the landfill sections of older waste age may be mixed with the leachate from other parts with younger age (Youcai et al., 2000). For that reason, it becomes very difficult to assign a leachate an exact age due to this temporal and spatial heterogeneity in most of the landfills (Baccini et al., 1987; Reinhart and Grosh, 1998). There could be some exclusive ones that leachate produced from wastes deposited in small sized cells is collected separately, but those are not

very common. Instead, in majority of the landfills, leachate collection system works to collect composite samples into a pond or pumped to a point of discharge from various waste compartments having different ages (Robinson et al., 2004).

In fact, the waste placement method can be an important factor for the leachate quality (Yıldız et al., 2004). Armstrong and Rowe (1999) stated that when fresh waste is deposited on older waste lifts, old waste becomes like a bioreactor for the leachate coming from the fresh parts. Therefore, upper parts of the landfill may show acidogenic characteristics due to fresh waste layers, while leachate collected from the bottom layers with older waste may display methanogenic conditions. Furthermore, when the leachate from the acid phase sections pass through the already decomposed waste parts, it could present the characteristics of methanogenic leachate due to consumption of high COD of the acid phase leachate at the carbon deficient medium below (Kjeldsen et al., 2002). This, hence results in wrong assessment of the landfill age and related leachate characteristics.

It is also worth to state that before late 1980's, landfills did not have any liner systems. Therefore, collection of representative leachate samples was not possible and due to sampling errors and/or groundwater dilution, it was probable to have lower concentrations (Reinhart and Grosh, 1998). Those factors might have caused the variability in leachate quality data. Only after the development of lined landfills, leachate quality data became less variable and so more useful. Furthermore, it is worth to mention that there are still no standard protocols for sampling, filtration, and storage of leachate samples (Adhikari and Khanal, 2015; Chian and Dewalle, 1977; Kjeldsen et al., 2002).

4.4.3 Other Factors

In the past 30 years, significant changes have occurred in solid waste management area especially in response to EU Landfill (EC, 1999) and Waste (EC, 2008) Directives impacting the composition, quantity, and treatability of landfill leachate (Brennan et

al., 2016). For example, the amount of waste going into landfills in Sweden has decreased significantly from 35 % (in 1995) to 1 % in 15 years. Due to the Landfill Directive, similar changes are expected in other member countries as well. Also, the composition of waste has changed in a way that organic matter, water content and biodegradability of the waste were reduced because of pre-treatment applications (Modin, 2012). Similarly, it was stated by Lo (1996) that in Hong Kong landfills, methanogenic conditions are established very quickly and three stages of the waste decomposition are not very distinctive so that aerobic and acid phases are completed almost in a year.

Heyer and Stegmann (2001) found significant differences between the studies of Kruse (1994) and Ehrig (1990) regarding the organic content of leachate. It was stated that in a ten year period COD, BOD and TOC content of leachate from younger landfills become lower than those given by Ehrig (1990). This was linked to the improvements in the landfill technology.

Kjeldsen et al. (2002) pointed out that in arid regions, waste could remain dry due to low rainfall amount and little infiltration so that landfills would stand in the acid phase or at the beginning of methane phase for many years. On the contrary, in tropical regions weather is more favorable for a faster and better decomposition of the waste (Adhikari and Khanal, 2015; Trankler et al., 2005). According to the analysis of Chen (1996), the time required for the stabilisation of leachate in Taiwan is much shorter than 10 to 20 years as given in the literature. Reinhart and Grosh (1998) stated that Florida climate with heavy rainfalls causes production of dilute leachates what makes it uncomparable with the literature values.

Finally, landfills being active (operational) or closed affect the leachate quality regardless of the waste age and it is suggested that those should be evaluated differently in the leachate classification. In closed landfills there will be no additional waste input and water content will decrease considerably due to the final cover layer preventing rainfall intrusion considerably. This issue is consistent with the study of Yıldız et al. (2004) who stated in their work that when no fresh waste was added into

the landfill for a long time, leachate BOD values decreased to very low values as expected, but if new waste piles were placed, BOD values increased sharply. Similarly, Rowe (1995) stated that the BOD/COD ratio and pH data of an active landfill leachate after 14 years of operation shows that the leachate mix (which includes leachate from different parts of the landfill developed at different times) is still acetogenic.

4.5 Conclusions

Historical records show that the need for classifying the leachate has emerged from the treatability studies of landfill leachate. Most of the researchers in that area tried to define their leachate type for the purpose of determining proper treatment method as well as to enable comparison with other treatability studies with different type of leachates. Many reviews have been done in leachate treatment showing the most appropriate treatment methods and their efficiencies with respect to the leachate age category.

The most important problem in classification of leachate is the use of different criteria as well as varied range of parameters. Some researchers use mostly BOD/COD ratio while many others still employ landfill age. However, there is not any common acceptance for the values of those criteria so that every author may employ different numbers. Especially, leachate classification according to the landfill age has not been defined in a standard way in the literature, hence it should be used by other researchers by caution. This is because, waste is landfilled generally in cells until the cell becomes full and then a new cell is started. In most of the landfills, leachate is collected in a combined system constituting of all cells. Therefore, it is very difficult and can be very deceptive to assign a single age to a landfill or leachate.

On the basis of reviewed literature, it could be remarked that leachate classification is better to be done according to both organic content (COD, BOD, or TOC) and stabilisation status (BOD/COD or COD/TOC ratios) of the leachate rather than the landfill age or pH values which would be misleading in many situations. Most

importantly, common naming of the leachate as used in the current literature as young, medium/intermediate, and old/stabilized/mature can be better replaced with more representative terminologies such as high strength, medium or low strength leachate which have been already employed by some researchers. Besides, authors could continue to define their studied site as young or old landfills but it could be better to provide the age of the landfill as operation years (i.e., 3 years, 8 years, etc.) in order to make rational comparisons with other studies including different aged landfills.

All of the issues explained in this chapter regarding the classification of leachate are required to be further analysed with real site data. For example, leachate from landfills located in different countries with different development status and climates as well as landfills with different ages and operational conditions (active/closed and sanitary/open etc.) need to be taken into consideration to clarify the leachate age/type ambiguity presented here.

CHAPTER 5

COMPREHENSIVE ANALYSIS AND MODELING OF LANDFILL LEACHATE

5.1 Introduction

Although landfilling is the least preferred alternative in the municipal waste management hierarchy, it is still used around the World for being a comparatively convenient and economic solution (Vaccari et al., 2019). Leachate, which is a highly polluted wastewater composed of high concentrations of organic and inorganic compounds, is produced as one of the consequences of waste disposal in landfills (Naveen et al., 2017). Therefore, it should be monitored and treated accordingly.

In literature, there are extensive studies on individual landfills, each providing valuable information about the specific properties of leachate in terms of its composition, transport, and treatment alternatives. However, there is an important shortcoming about the possibility of generalizing those findings for every landfill. In order to come up with a general model, detailed modeling studies are required. Besides, due to high number and variety of parameters, analysis of leachate is a time consuming, complex, and economically intense process. Being able to define leachate with as small number of parameters as possible is desirable. Understanding relationship between parameters through statistical analysis can help reduce this burden. Unfortunately, statistical analysis of leachate characteristics is limited in number, as well as scope. Earlier studies mainly focused on classifying leachate samples, investigating differences in leachate quality among few landfills, and evaluating the relationships between leachate parameters (Indelicato et al., 2018; Pablos et al., 2011). Although site specific conditions have great impact on the quality and quantity of leachate, common trends and properties can prevail for the majority of landfills which can be perceived through data analysis.

Statistical approaches and models can be used to show the significant similarities and differences between leachate samples from different landfills provided that critical data pre-treatment methods are employed. Principal Component Analysis (PCA) and cluster analysis are among powerful multivariate methods commonly used in many research fields involving complex multivariate data (Rana et al., 2018; Rinaldi et al., 2014). PCA helps to express the information given in a data set by grouping the variables with similar characteristics into specific components. Hence, variables within each component are highly correlated with each other, but slightly correlated with variables in other groups (Pastore et al., 2018). PCA was used for a limited number of leachate studies (Adelopo et al., 2018; Boateng et al., 2018; Mishra et al., 2016), even though it can provide valuable information.

The purpose of this study is to evaluate the leachate characteristics of a high number of landfills around the World to uncover possible similarities or differences, conduct statistical modeling for prediction of leachate characteristics, and contribute to the limited examples of multivariate analysis particularly on landfill leachate. For this purpose, leachate data belonging to different landfills from 46 countries were compiled. Data was analysed by statistical tools following data pre-treatment including outlier removal, missing data substitution, and data standardization. Correlation analysis and regression modeling were applied to find out the type and degree of associations between leachate parameters. PCA and cluster analysis were conducted to classify leachate data into groups for the assessment of main properties. This is one of the most comprehensive studies on the statistical analysis of leachate data from different parts of the World.

5.2 Materials and Methods

5.2.1 Data Collection

Leachate characterization data from many landfills were compiled to conduct the regression modeling and statistical analyses. During data collection, care was given to

select active landfills with similar operations and leachate handling methods such as landfills accepting mainly municipal solid waste (MSW), no leachate recirculation, no pre-processing of waste (i.e., no shredding or bailing), no considerable disposal of specific waste like ash or sludge, no mixing of leachate with wastewater from composting plants or another facility/location. A total of 511 leachate data (some belonging to given landfills at different operation times/years) were compiled based on the collected data. Eighty seven of those belonged to unsanitary landfills and 64 to closed sites. The remaining 360 data sets were for active sanitary landfills, which were used in the analyses. Related data sheet is provided in Appendix A (Table A.1) with their references (Table A.2). Some important checks were made to validate the data as presented in Section 3.1.

The data set for the active landfills constituted leachate parameters including conventional parameters (COD, BOD, TOC, BOD/COD, pH, alkalinity, conductivity, NH₄-N, TKN, TN, TSS, TDS, TS), inorganics (SO₄, TP, PO₄-P, Cl, Na, K, Ca, Mg), and heavy metals (Fe, Mn, Ni, Zn, Cu, Pb, Cd, Cr). Some difficulties were faced during data collection such that some references provide limited leachate parameters causing discontinuity in the data set. For example, measurements for TOC, solids, alkalinity, total phosphorus (TP), Na, K, and majority of heavy metals were absent in many studies, while quantities for COD, BOD, and NH₄-N were reported in most of them. Nevertheless, leachate data including 29 parameters from 220 different landfills of 46 countries in Europe, Middle East, Asia, Africa, and America were compiled. Some landfills had temporal observations, therefore more than one data set/point existed for those in the data matrix. As a result, an initial input leachate data matrix of 360 rows (landfills or leachate samples) and 29 columns (pollution parameters) were obtained.

5.2.2 Data Pre-treatment

Data pre-treatment was applied to remove outliers and estimate missing data (Stanimirova et al., 2007). A flowchart showing the phases of data collection, data treatment, and analyses is presented in Figure 5.1. As elimination of outlier data can

improve the prediction performance of a regression model (Yuzugullu and Aksoy, 2014), outlier detection and removal were done concurrently with regression analysis. Correlation analysis was conducted to identify potential relationships between leachate parameters. Then, data scatter charts were plotted and linear relationships between parameters were scrutinized. Removal of outliers may also lead to potential loss of valuable data, so to minimize that, data prediction intervals were used in scatter plots to detect extreme outliers. The Prediction Interval (PI) shows the likelihood of a future observation being in a given value range for the similar settings of predictors (Forthofer et al., 2007; Frost, 2021a). In this study, a PI of 99% was selected in SPSS to minimize data loss. Ma et al. (2022) selected it as 97.5% for removal of outlier data.

Relationships exhibiting Pearson Correlation Factor (r) values greater than 0.7 were considered as strongly correlated, while values between 0.5 and 0.7 were accepted as moderately correlated with a significance level of $p < 0.05$ (Adams et al., 2001). Excluding exceptional cases, up to 15% of data was completed using the regression models for given parameters. This value was in accordance with Modin (2012) who accepted this ratio as high as 12.5%.

As the last step of data pre-treatment prior to PCA and cluster tests, a standardization procedure was used to eliminate the influence of different measurement units and make the data dimensionless (Pastore et al., 2018) through conversion into z-scores (Figure 5.1). Detailed information about data pre-treatment is provided in Section 3.3.

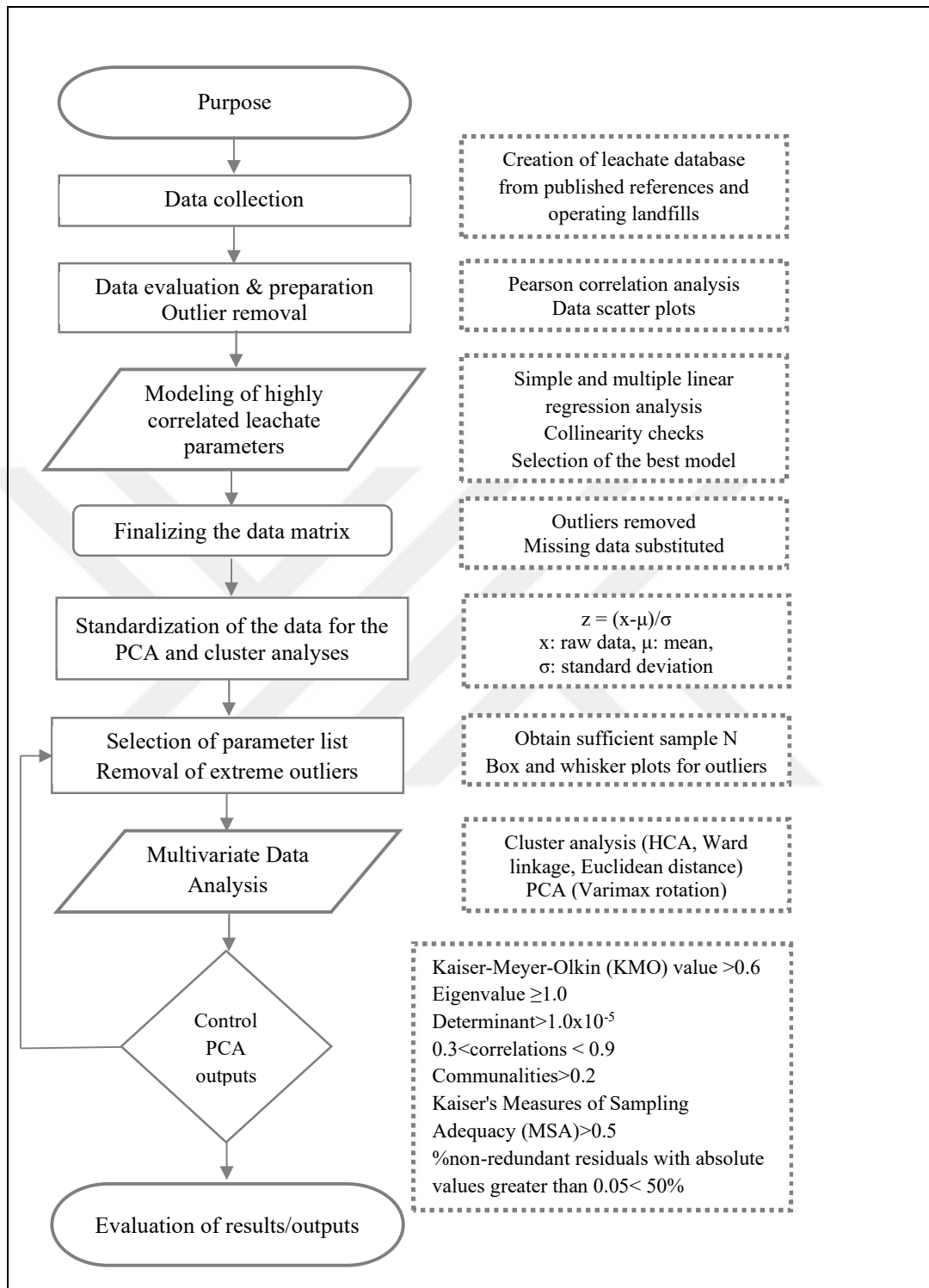


Figure 5.1. Flowchart for data collection, preparation, and analysis steps

5.2.3 Data Analysis

IBM SPSS software (version 25) was used for statistical analyses. The confidence level was set at 95%. Both simple and multiple linear regression models were derived depending on the correlation status of leachate parameters. Automatic Linear Modeling tool of SPSS was employed to evaluate the importance of a predictor and find out the independent variables of a regression model having a better fit of the observation data. The standard error of estimate (S) and coefficient of determination (R^2) were used as indicators of goodness-of-fit. The R^2 measures the variance in a dependent variable that can be explained by an independent variable. The S provides the absolute measure of the average distance of the data points from the regression line. It shows how precise the model predictions are in the units of the dependent variable (i.e., error in mg/L for most of the leachate parameters) (Schroeder et al., 2018; Frost, 2021b). Also, minimum, average, and maximum % errors were calculated.

In development of the regression models, multicollinearity check was done. Multicollinearity (also collinearity) is having high correlation between predictor variables (independent variables) in a regression analysis so that they exhibit a strong linear relationship among each other. Collinearity condition was checked in multiple linear regression analysis by selecting “Collinearity Diagnostics” in SPSS. Variance Inflation Factor (VIF) and tolerance values were checked in the output table “Coefficients” in which the VIF value is to be less than 10, and the corresponding tolerance should be above 0.1 for not having collinearity. Detailed information about regression modeling is provided in Section 3.2.2.

Cluster analysis was used to classify variables into groups (clusters) according to the similarities within a group and dissimilarities among other groups. Hierarchical cluster analysis (HCA) was applied for the grouping of leachate parameters. Ward linkage was used to link clusters and Euclidean distance was used as the interval of clustering (measure of distance) of leachate data (Rinaldi et al., 2014; Zhao and Cui, 2009). Detailed information about cluster analysis is provided in Section 3.2.3.

Large data sets can be simplified into few components which hold the majority of the information in original data with PCA (Bro and Smilde, 2014; Wuensch, 2016). In PCA, each component would represent some feature as a latent variable that explains why a group of variables is in that component. PCA results were visualized by loading tables, plots, and score charts. Loading of a variable represents the correlation between the parameter and the component. Loadings are classified as “strong”, “moderate”, and “weak” according to absolute values of >0.75 , $0.75-0.5$, and <0.5 respectively (Liu et al., 2003; Zhao and Cui, 2009). On the other hand, Boateng et al. (2018) accepted loadings of <0.5 as poor, 0.5 as moderate, and >0.5 as high loadings. According to the Rinaldi et al. (2014) loadings greater than 0.6 indicate significant correlations between the original variables and the related components. Similarly, Durmusoglu and Yilmaz (2006) selected loadings greater than 0.5 for the PCA interpretation in their study. Rotation is used to maximize the loading of each parameter on the related component while minimizing its loading onto others. For a better interpretation of results, Varimax rotation (Akyol, 2005; Pastore et al., 2018) was selected in this study. Also, to manage the number of components that will be extracted, eigenvalue greater than or equal to 1 was considered as being statistically significant (Boateng et al., 2018; Yong and Pearce, 2013). Basic requirements of the PCA are provided in Section 3.2.4.

Apart from the outlier treatment applied to individual leachate parameters for the whole leachate data set during regression analysis, another outlier removal step was conducted for the smaller scale data matrix of selected parameters for PCA and cluster analysis. Outliers have a strong effect on the variance as well as on the correlation status of the data. Therefore, PCA is strongly affected by the outliers in a way that extracted components may not be able to represent the dataset very well. For detecting the outliers, box and whisker plots were applied to the list of parameters used in the analysis. Only extreme outliers were removed to prevent data loss.

5.3 Results and Discussion

5.3.1 Handling Outlier Data

Correlation analysis was conducted on raw data without removal of any outliers to identify potential relationships between leachate parameters (Table B.2 in Appendix B). Strong correlations were found between several leachate parameters including organic compounds (BOD, COD, TOC), nitrogen group (TKN, TN, $\text{NH}_4\text{-N}$), and other parameters. One can see that COD is highly correlated to BOD and TOC, and then to TDS and TS, as expected, as organic fraction of leachate originates from dissolved particles of solid waste as well as by-products of biodegradation processes within a landfill. Another example for relatively strong correlations is the correlations of Na and Cl to conductivity (EC). Sum of the concentrations of mainly chloride (Cl), sodium (Na), alkalinity, and ammonia forms the major portion of the EC (Statom et al., 2004). Similarly, Indelicato et al. (2018) stated a good correlation of EC with Cl.

For detection and removal of outliers, scatter plots were drawn for correlated parameters. A sample scatter plot with prediction intervals (PI) is presented in Figure 5.2 to illustrate the approach and the remaining plots are provided in Appendix B. An example for outlier data is the alkalinity measured in four landfills. As alkalinity is comprised of three ions, it is expected to correlate with EC to a certain level. This correlation was nicely observed for the majority of the landfills, except for four landfills as indicated in Figure 5.2. These four alkalinity values that fall out of the PI range were first checked within the data matrix against the quantities reported in the literature. It was observed that three alkalinity values were very high in comparison to low EC and $\text{NH}_4\text{-N}$ concentrations, while one value was low against very high EC and $\text{NH}_4\text{-N}$ values. Since they were not in the expected range and somehow created divergence from the common pattern, they were excluded from the regression study as well as from the data set. In this manner, a total of nine outlier data were eliminated permanently from the data matrix: four alkalinity data (Figure 5.2), one Cl data (Figure

B.1a), one EC data (Figure B.1b), and one Na (Figure B.2) data from different landfills, and one Na (Figure B.3a) and K (Figure B.3b) data of a landfill.

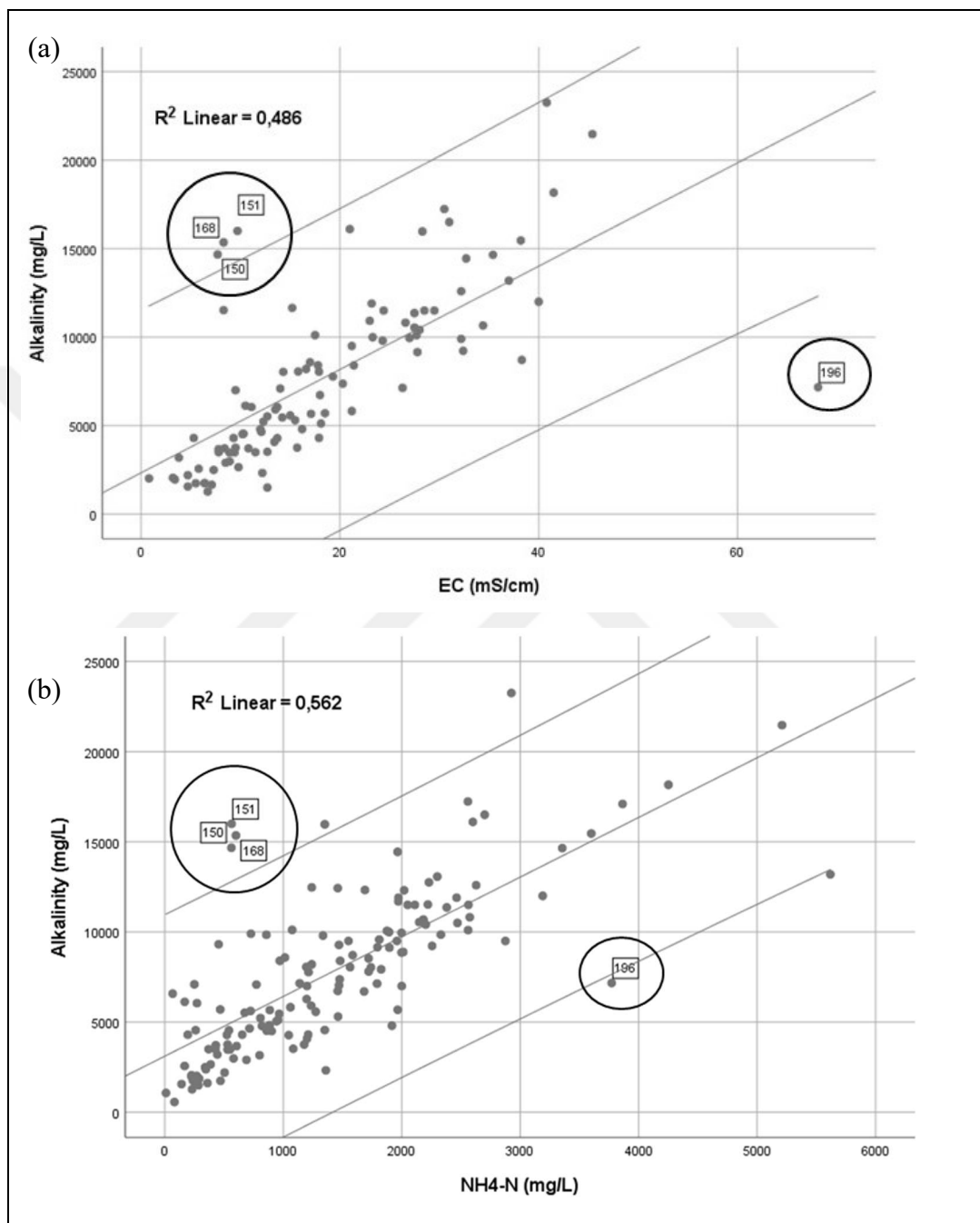


Figure 5.2. Example of outlier data (circled) with scatter plots for (a) EC vs Alkalinity and (b) $\text{NH}_4\text{-N}$ vs Alkalinity (the upper and lower lines show 99% prediction interval; the numbers within boxes indicate landfill ID)

5.3.2 Regression Modeling

Discontinuity problem in data set due to missing values in some parameters was handled by regression data modeling. It emerged as a proper tool due to strong correlation between many leachate parameters. In addition to simple linear regression, multiple regression modeling was conducted in a number of cases due to high correlation of one dependent parameter to more than one independent variable. Collinearity checks were done and independent parameters exhibiting collinearity were not included in regression models.

The Automatic Linear Modeling tool of SPSS was used in order to get the predictor importance for a multiple regression model. Figure 5.3 shows example predictors for some leachate characteristics. For example, instead of Na and K, interestingly $\text{NH}_4\text{-N}$ was found the most important predictor in EC modeling. $\text{NH}_4\text{-N}$ was found highly correlated with EC in other studies as well (Chu et al, 1994; Mohammad-pajoooh et al., 2017). This could stem from very high $\text{NH}_4\text{-N}$ concentrations in leachate that may constitute a major portion of the conductivity. For prediction of K, Na has the highest importance as a predictor. Since Na and K are both conservative parameters like Cl, they are not involved in any precipitation and complexation reactions, therefore their concentrations show the same trend (Erses et al., 2008). Predictor importance for all leachate parameters considered in regression modeling is detailed in Table 5.1.

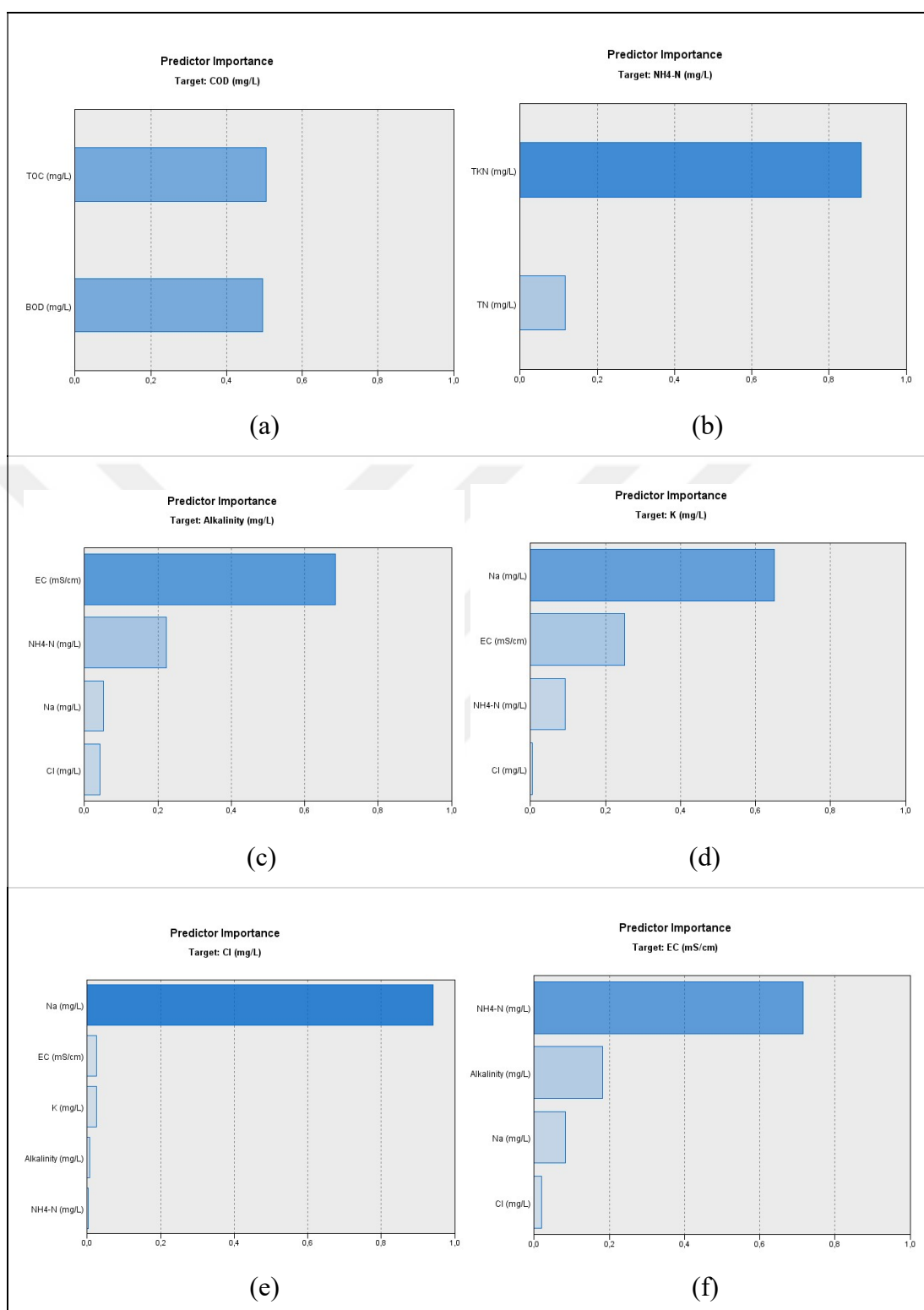


Figure 5.3. Predictor importance charts for the regression model variables as (a) COD (b) NH₄-N (c) Alkalinity (d) K (e) Cl and (f) EC

Table 5.1 Predictor importance of the model variables for highly correlated leachate parameters

Dependent parameter*	The most important predictor(s)	Less important predictor(s)
BOD	COD	TOC
COD	TOC	BOD
TKN	TN	NH ₄ -N
NH ₄ -N	TKN	TN
NH ₄ -N	EC	K>Na>Cl>TA
EC	NH ₄ -N	TA>Na>Cl or TDS>TA>Cl**
TA	EC	Na>Cl>NH ₄ -N>K or NH ₄ -N>Na>Cl***
Na	Cl	K>NH ₄ -N>TA>EC or EC>NH ₄ -N>TA***
K	Na	EC>NH ₄ -N>Cl
Cl	Na TDS	EC=K>TA>NH ₄ -N or K>EC>Na**
TDS	Cl	EC>K>Na or EC>NH ₄ -N>Na***

* Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

** Case if TDS inserted into the model (less important variables were excluded to achieve sufficient data numbers)

*** Case if K is excluded from the model due to low number of observed data for that parameter

Regression models derived are presented in Table 5.2. They were selected among a number of models based on the lowest S, highest R^2 , and the lowest average % error values. As seen, majority of the selected models for missing data prediction have a R^2 between 0.80 and 0.99. Kylefors (2003) stated that a good model generally has $R^2 > 0.8$. Regression model details are provided in Figure 5.4 and Table 5.3 for relevant parameters and a brief summary is presented in the following paragraphs.

Table 5.2 Details of selected regression models for calculation of missing leachate data

Dependent variable*	Model formulation	Sample N**	R ²	% error (average)	S***	Limiting conditions for the model
BOD	0.61[COD]–1207	294	0.899	22	2380	BOD>3000 mg/L
COD	1.49[BOD]+2741	294	0.899	22	3729	COD>3000 mg/L
COD	3.10[TOC]–552	81	0.933	30	3243	COD>2000 mg/L
TKN	0.99[TN]–16	66	0.998	3.5	78	None
NH ₄ -N	0.83[TKN]–33	133	0.948	16	277	NH ₄ -N>50 mg/L
NH ₄ -N	0.81[TN]–51	96	0.925	20	376	None
TDS	0.94[TS]–161	71	0.969	8	1527	None
TDS	1.54[Cl]+326.5[EC]+1618	61	0.869	21	2992	None
TA	0.52[Cl]+[NH ₄ -N]+252[EC]–13	80	0.814	22	2118	TA>2000 mg/L
Cl	179.7[EC]–0.14[NH ₄ -N]–331	134	0.745	25	1092	Cl>1500 mg/L
Na	0.40[Cl]+48.8[EC]–117	89	0.877	20	557	Na> 400 mg/L
K	0.52[Na]+0.25[NH ₄ -N]–30	92	0.789	29	505	K>250 mg/L
EC	10 ⁻³ [2[Cl]+4[NH ₄ -N]+[TA]]+3	80	0.889	15	3.5	EC>3 mS/cm

* Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

** Number of data used for model building

*** Standard error of estimation (S) is in mg/L for all parameters except EC which is mS/cm.

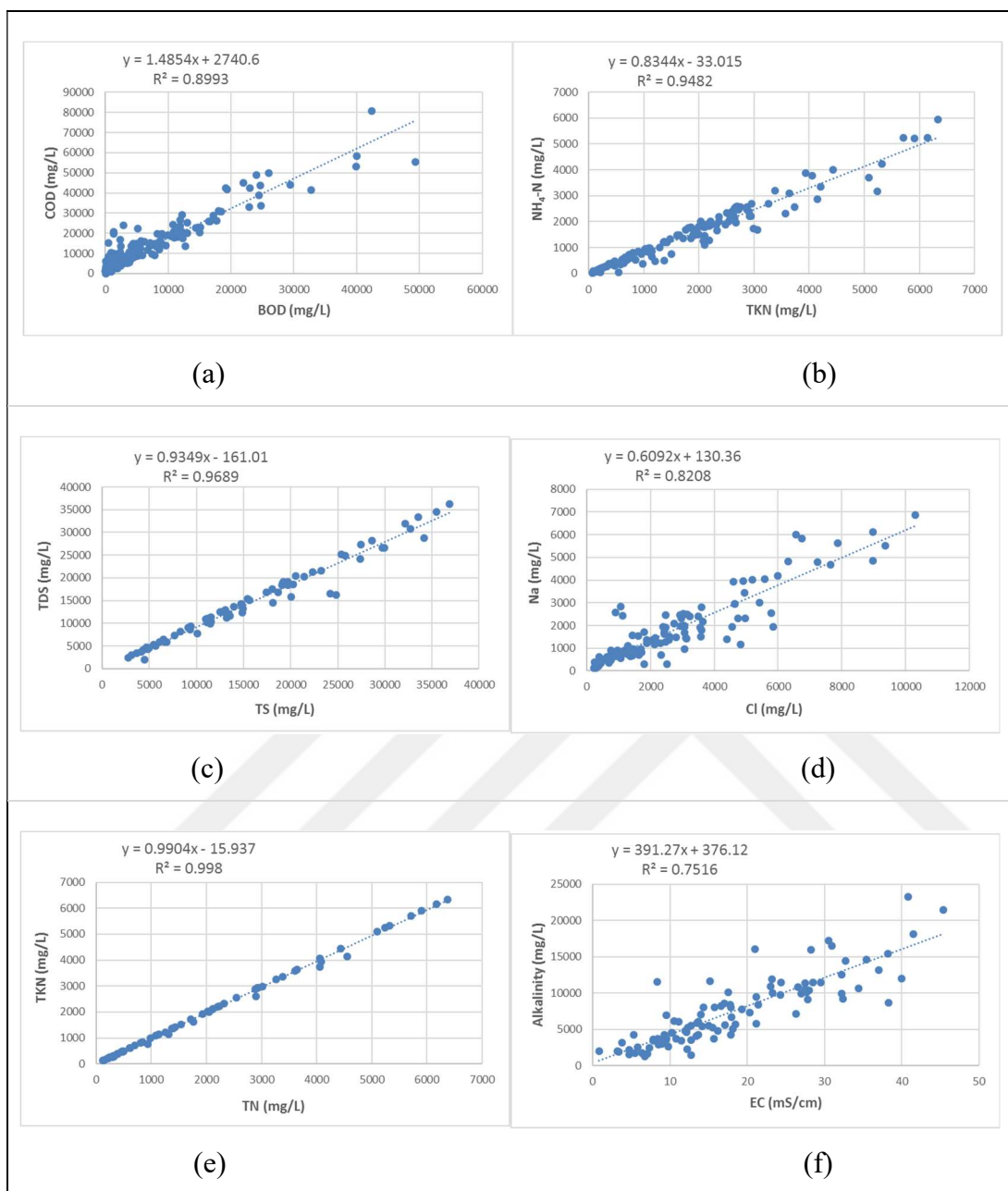


Figure 5.4. Sample regression plots for (a) BOD vs COD (b) TKN vs $\text{NH}_4\text{-N}$ (c) TS vs TDS (d) Cl vs Na (e) TN vs TKN (f) EC vs Alkalinity

Leachate BOD can be predicted based on available COD data with 22% error on average and R^2 of 0.90. Yet, there is some limitations of the models. For example, in the modeling of BOD using COD as the independent variable, the case of $\text{BOD} < 3000$ mg/L caused negative and erroneous BOD predictions (Figure 5.5). Similarly, while

modeling COD with TOC data, COD was predicted mostly negative or lower compared to the observed COD values when COD is less than 2000 mg/L (Figure C.1). Therefore, limitations were applied such that, if a predicted parameter value is outside of a stated limit, then that predicted value was not used in missing data substitution. Limiting conditions for all regression models are given in Table 5.2 and relevant plots are provided in Appendix C.

Table 5.3 Details of the multiple regression modeling results (shaded row indicates the selected best model)

Dependent (y)*	Independent (x)	Sample N	R ²	S*	avg % error
TDS	Cl, EC, and NH ₄ -N**	57	0.887	2869	21
	EC and Cl	61	0.869	2992	21
	NH ₄ -N and Cl	64	0.839	3326	24
	EC and NH ₄ -N	66	0.801	3905	25
Na	Cl and EC	89	0.877	557	19.8
	Cl	115	0.821	632	23.7
	EC	90	0.797	709	29.6
K	Na and NH ₄ -N	92	0.789	505	29.3
	EC and Na	76	0.780	555	32.2
	Na	103	0.745	547	32.1
Cl	Na***	115	0.821	939	19.2
	TDS***	69	0.781	1185	20.5
	EC and NH ₄ -N	134	0.745	1092	25.0
	EC	143	0.705	1214	25.2
EC	NH ₄ -N, TA, and Na	54	0.901	3.3	17.3
	NH ₄ -N, Na, and Cl	81	0.898	3.8	20.1
	NH ₄ -N and Na	81	0.890	3.9	20.9
	NH ₄ -N, TA, and Cl	80	0.889	3.5	15.2
TA	EC, NH ₄ -N, and Na	54	0.820	2201	23.6
	EC, NH ₄ -N, and Cl	80	0.814	2118	21.9
	EC and NH ₄ -N	93	0.792	2178	21.1
	EC	99	0.752	2347	22.8

* Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

** There is a collinearity problem for the best model (TDS-Cl-EC-NH₄-N), so the 2nd best model was selected.

*** Due to low number of observed data for Na and TDS, the other best model was selected.

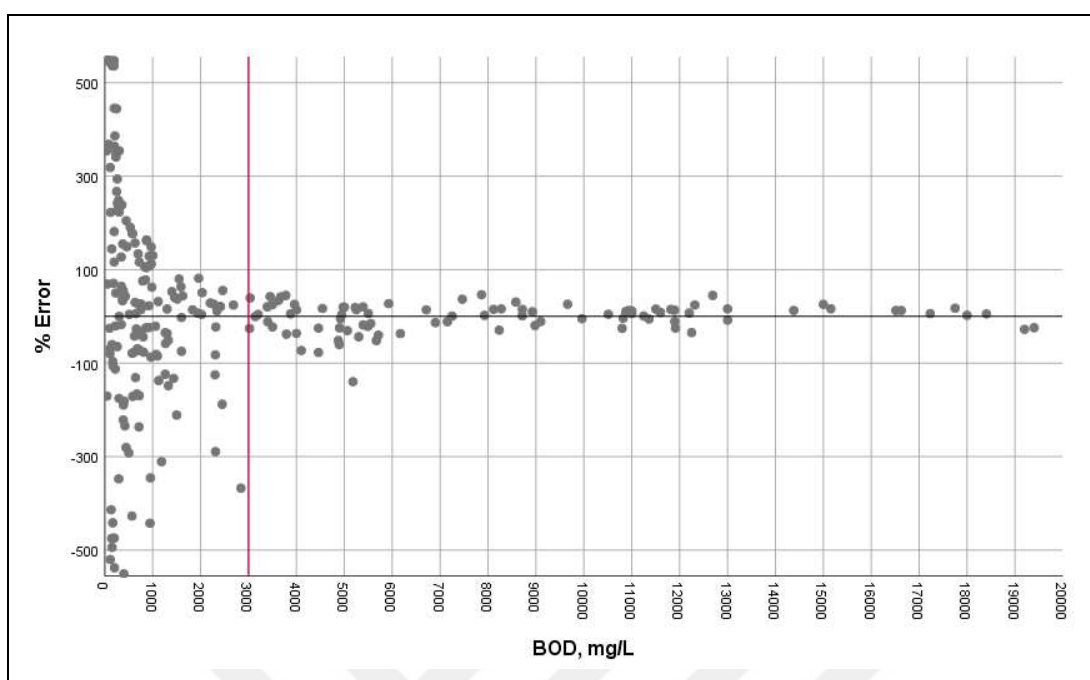


Figure 5.5. Sample % error chart for BOD-COD regression model

In a number of data prediction cases, multiple linear regression models were used. For TDS, firstly available TS data was used, as there was perfect linear relationship between TDS and TS (Figure 5.4c). However, due to lack of sufficient TS data in the data matrix to calculate all missing TDS data from the TDS-TS model, other regression models were studied (Table 5.3). A multiple linear regression model of TDS-Cl-EC was deemed as the best option for TDS predictions.

Regarding monovalent ions, Na could be estimated with multiple regression model of Na-EC-Cl with the condition of $\text{Na} > 400 \text{ mg/L}$ (Figure C.6). Although K is the most important predictor after Cl for Na estimation, it could not be used due to lack of observed K values in the data matrix. Similarly, Na and TDS are the most important predictors for Cl (Table 5.1), but due to the deficiency of observed data for both parameters, as well as for K, other independent parameters were used in modeling (Table 5.2).

As mentioned before, numbers of observed data in relevant references were very low for some parameters like TDS, Na, and K. Yet, the percentage of predicted data was

targeted to be at most 15% in this study. This target was satisfactorily met for all parameters, except for TKN and TDS. These in fact have very strong correlation with TN (TN=TKN+Nitrogen Oxides) and TS (TS=TDS+SS), respectively. Therefore, for the TDS case, 15% limit was applied only for the second model. A similar approach was used to complete most of the missing TKN data from available TN values with high accuracy (Table 5.2). Following data substitution, missing data still existed in the data matrix (Table 5.4). This problem was handled to some degree by changing the parameters analysed in the PCA and cluster tests to observe the important features as much as possible.

Table 5.4 Status of missing data in the data set for modeled leachate parameters

Dependent (y)*	Number of observed data**	Total number of missing data	% of missing data	Number of predicted data	Number of missing data remained	% of missing data substitution
COD	345	15	4%	9	6	3%
BOD	300	60	17%	23	37	7%
NH ₄ -N	314	46	13%	17	29	5%
TKN	144	216	60%	42	174	23%
Cl	221	139	39%	29	110	12%
EC	189	171	48%	24	147	11%
TA	155	205	57%	28	177	15%
Na	119	241	67%	21	220	15%
K	105	255	71%	14	241	12%
TDS	92	268	74%	25***	243	21%***

* Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

** In total 360 data points exist in the leachate data matrix

*** 9 data from the model TDS-TS and 16 data from the model TDS-Cl-EC (15%)

5.3.3 Correlation Analysis

Correlation analysis was repeated after the removal of outliers and substitution of missing data. Results are presented in Table 5.5. Outlier removal and missing data substitution increased the correlation status of many parameters considerably. For example, correlations increased between alkalinity and EC (from 0.7 to 0.9), alkalinity and K (from 0.4 to 0.8), Cl and EC (from 0.8 to 0.9). Regarding heavy metals, only

moderate correlation was observed between heavy metals and other leachate parameters, for example between Cr and TP, Na, K, Cu; between Mn and pH, Fe, Zn. Besides these, only some heavy metals were moderately ($0.5 < r < 0.7$) correlated with each other.

In Table 5.5, it is possible to see the highest correlations belong to organic parameters (BOD and COD), as well as to TKN and $\text{NH}_4\text{-N}$. Alkalinity had a strong correlation with many monovalent ions in the landfill leachate. Furthermore, EC and K emerged to be correlated very well with $\text{NH}_4\text{-N}$, TKN, and alkalinity. Similar to this, findings of multivariate statistical techniques can be used to plan monitoring activities. Some mostly correlated parameters can be excluded from sampling to minimize the number of analyses and save considerable cost and time as also suggested by Klylefors (2003), Galvez et al. (2010), and Modin (2012). For example, TKN analysis would be difficult and time consuming due to measurement of organic portion, while $\text{NH}_4\text{-N}$ measurement can be done easily even at site by portable devices. Similarly, TOC measurement would need high technological equipment with detailed sample preparation, while COD and BOD would be cheaper and COD measurement even requires less time and effort. Also, it would be sufficient to measure only one of the salt parameters as Cl, Na, or K to estimate the others.

Table 5.5 Pearson correlation coefficients matrix for different leachate parameters

	pH	COD	BOD	NH ₄ -N	TKN	TDS	Cl	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
pH	1																				
COD	-0.45	1	0.96(323)			0.67(116)															
BOD	-0.50	0.96(323)	1																		
NH ₄ -N	0.30	0.26	0.17	1	0.97(184)		0.68(240)	0.85(201)	0.84(174)	0.69(132)	0.72(112)										
TKN	0.28	0.33	0.23	0.97(184)	1	0.72(74)	0.73(128)	0.90(108)	0.84(98)	0.81(65)	0.76(53)										
TDS	-0.05	0.67(116)	0.58	0.64	0.72(74)	1	0.82(95)	0.83(98)	0.70(74)	0.85(61)	0.76(45)										
Cl	0.26	0.34	0.25	0.68(240)	0.73(128)	0.82(95)	1	0.87(195)	0.76(151)	0.91(136)	0.83(113)										
EC	0.18	0.42	0.32	0.85(201)	0.90(108)	0.83(98)	0.87(195)	1	0.89(150)	0.87(120)	0.86(99)										
TA	0.22	0.35	0.27	0.84(174)	0.84(98)	0.70(74)	0.76(151)	0.89(150)	1	0.81(100)	0.78(81)										
Na	0.31	0.37	0.22	0.69(132)	0.81(65)	0.85(61)	0.91(136)	0.87(120)	0.81(100)	1	0.87(117)										
K	0.34	0.37	0.17	0.72(112)	0.76(53)	0.76(45)	0.83(113)	0.86(99)	0.78(81)	0.87(117)	1										
Fe	-0.65	0.46	0.49	-0.17	-0.22	0.15	-0.14	-0.09	-0.08	-0.14	-0.13	1									
Ca	-0.49	0.48	0.48	-0.11	-0.10	0.25	0.07	0.20	0.09	0.22	0.20	0.49	1	0.84(120)							
Mg	-0.38	0.56	0.59	0.07	0.08	0.34	0.14	0.28	0.30	0.27	0.27	0.42	0.84(120)	1							
Ni	0.00	0.10	0.06	0.01	0.02	0.21	0.10	0.03	0.16	0.16	0.10	0.07	0.14	0.04	1						
Zn	-0.26	0.28	0.29	-0.08	0.02	0.09	0.06	0.15	-0.05	-0.11	-0.12	0.33	0.15	-0.02	0.18	1					
Cu	0.04	0.15	0.14	0.05	0.35	0.23	0.19	0.23	0.09	0.28	0.16	0.00	-0.05	-0.11	0.34	0.65	1				
Cr	0.04	0.34	0.26	0.20	0.36	0.44	0.42	0.29	0.30	0.52	0.63	0.04	0.21	0.04	0.19	0.47	0.57	1			
Pb	-0.07	0.18	0.22	-0.09	0.05	0.09	0.04	0.16	0.16	0.14	0.12	0.05	0.08	0.16	0.03	0.53	0.59	0.35	1		
Cd	-0.14	0.15	0.11	-0.02	0.27	-0.16	-0.03	-0.07	0.21	0.08	0.02	0.07	0.36	0.07	0.26	0.20	0.30	0.27	0.58	1	
Mn	-0.58	0.36	0.36	-0.15	-0.20	0.07	-0.17	-0.09	-0.12	-0.21	-0.23	0.64	0.32	0.13	0.08	0.54	-0.08	-0.05	0.09	0.29	1

Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

Values in the brackets show the number of data used in the pairwise comparisons. Bold values represent correlations with $r \geq 0.7$

Many authors studied possible relationships between leachate parameters, generally for the leachate of a single landfill. The novelty of this study is that analyses were conducted considering the leachate data from numerous landfills around the World (Table 5.6). Therefore, obtained results can be helpful in the assessment of leachate parameters in various landfills, although minor differences are possible due to fluctuating leachate characteristics. Similar correlations were observed for TDS, COD, NH₄-N, and EC (Table 5.7). However, for heavy metals, there were different results in different studies. Possible differences and similarities can be associated with leachate type depending mostly on the landfilling period. For example, if a leachate study was conducted in early times of landfilling, heavy metal concentrations would be higher as with other parameters, which could impact the correlation status.

Table 5.6 Locations of MSW sanitary landfills included in the leachate data set

Country	No of landfills*	Country	No of landfills	Country	No of landfills	Country	No of landfills
Algeria	3	Ghana	1	Mexico	4	South Korea	16
Australia	1	Greece	1	Morocco	3	Spain	30
Brazil	18	Hong Kong	8	Nepal	2	Sweden	2
Canada	30	India	3	New Zealand	5	Taiwan	11
China	40	Indonesia	5	Norway	2	Thailand	4
Colombia	1	Iran	2	Palestine	2	Tunisia	4
Croatia	1	Ireland	3	Poland	6	Turkey	55
Denmark	7	Island Of Mauritius	3	Portugal	2	UK	9
Egypt	2	Italy	12	Russia	2	Uruguay	1
Finland	2	Lebanon	2	Saudi Arabia	1	USA	17
France	10	Lithuania	1	Slovenia	1		
Germany	2	Malaysia	15	South Africa	8		

* Total number of landfills in the data set from that country (includes the same and different landfills)

Table 5.7 Comparison of correlation results of leachate parameters with literature (common well correlated parameters ($r \geq 0.5$ cases) are indicated as bold)

Modeled Parameter	Parameters exhibiting correlation	Reference	Remarks
EC	COD, TDS , TS, Cl , Na , K , and Mg	Galvez et al. (2010)	Electrical Conductivity (EC) is well correlated with many parameters especially with TDS and salts.
	Cl , COD, BOD, Mg, Fe, and Ni	Mishra et al. (2016)	
	TDS , COD, Cl , Na , K , SO ₄ , NH₄-N , alkalinity , Fe, Cd, Pb, and Zn	Naveen et al. (2017)	
	TDS , Cl , Na , K , alkalinity , NH₄-N , TKN	Present Study	
pH	SS, K, and Mg	Galvez et al. (2010)	pH is only moderately correlated with a few parameters.
	Cl, Mg, and Cr	Mishra et al. (2016)	
	Fe, Mn, Ca, and BOD	Present Study	
COD	BOD , solids , EC, Cl , Na , K , and Mg	Galvez et al. (2010)	COD can be correlated with various parameters including BOD and solids.
	SS, BOD , Cl , EC, Ca , Fe, Mn, and Ni	Mishra et al. (2016)	
	Cl, SO ₄ , NH₄-N , Ca , alkalinity , Zn, Ni, Na, Cd, Cr, Pb, and Fe	Naveen et al. (2017)	
	BOD , TDS , Ca , and Mg	Present Study	
TDS	TS , EC , Cl , Na , K , Ca and Mg	Galvez et al. (2010)	TDS has strong correlation with TS, EC, and salts.
	Alkalinity , SS, P, Ca, and Mg	Mishra et al. (2016)	
	EC , COD , Cl , SO ₄ , alkalinity , Fe, Pb, Zn, Na , K , and NH₄-N	Naveen et al. (2017)	
	TS , COD , BOD, NH₄-N , TKN, alkalinity , Cl , Na , K , and EC	Present Study	
NH ₄ -N	BOD, Cl , Na , and SO ₄	Galvez et al. (2010)	NH ₄ -N is well correlated with many dissolved ions.
	EC , TDS , COD, SO ₄ , Cl , alkalinity , Ca, Fe Cd, Cr, Pb, Zn, and Ni	Naveen et al. (2017)	
	TKN, TP, TDS , EC , Cl , Na , alkalinity , K	Present Study	
Heavy metals	Fe with Cl, EC, SS, COD, and BOD Mn with COD and SO ₄ Cu with Cl Cr with pH, Cl, BOD, and Ni	Mishra et al. (2016)	Correlation status of heavy metals with each other and with other parameters are different in each study depending on specific leachate type.
	Pb and Zn with EC, TDS, COD, SO ₄ , Fe, alkalinity , Cl , NH₄-N , Cd, Cr, Zn, Ni, Na Cr with COD, Cl, SO ₄ , alkalinity , Fe, Cd	Naveen et al. (2017)	
	Fe with pH, BOD, Ca, and Mn Mn with pH, Fe, and Zn Zn with Cu, Pb, and Mn Cu with Cr, Pb, and Zn Pb with Zn, Cu, and Cd	Present Study	

5.3.4 PCA and Cluster Analysis

Results of the PCA and cluster analysis provide similar outcomes (Bhuiyan et al., 2011; Liu et al., 2008). They both show the possible grouping of leachate parameters according to their correlation status and used jointly to supplement the results of each other. However, PCA could present more information about the data matrix including the possible reasons behind groupings and some important findings about data structure and content as well. In earlier studies where PCA was applied, generally leachate samples of one landfill were used which could create difficulty to generalize the results for different landfills (Adelopo et al., 2018; Mishra et al., 2016; Ziyang et al., 2009). This study overcomes that shortcoming and aims to provide more generalized outputs for landfill leachate. Unfortunately, since multivariate statistical tools are not employed frequently in landfill leachate studies, there is only limited literature data for comparison, hence only few examples are presented.

5.3.4.1 Cluster Analysis

Based on the standardized (z-scores) data of leachate samples from different landfills, hierarchical cluster analysis (HCA) was run. HCA was performed in two parts due to the missing data problem. Parameters were selected accordingly to obtain sufficient number of data at each step. Parameters with high level of missing data, like TDS and TKN, were not included in the PCA and cluster analysis. However, this is not crucial, as EC and NH₄-N were included instead, which are highly correlated with TDS and TKN, respectively.

Initially 12 leachate parameters were selected including pH, COD, BOD, EC, NH₄-N, alkalinity, K, Cl, Na, Fe, Ca, and Mg. Before conducting cluster analysis, box and whisker plot of the selected parameters were drawn to see possible outliers. It was observed that a number of outlier data existed for some of the parameters (Figure 5.6a). For the purpose of not decreasing the data number further, only some extreme outliers (in total 9) were removed from parameters like COD, BOD, and Fe (i.e., 362, 308,

282, 230 in Figure 5.6a). Even if only COD data is removed from a leachate sample (i.e., landfill with ID 362), whole data line including other tested parameters like Fe, Mg, Ca was taken out of the analysis. The box plot was then re-checked (Figure 5.6b).

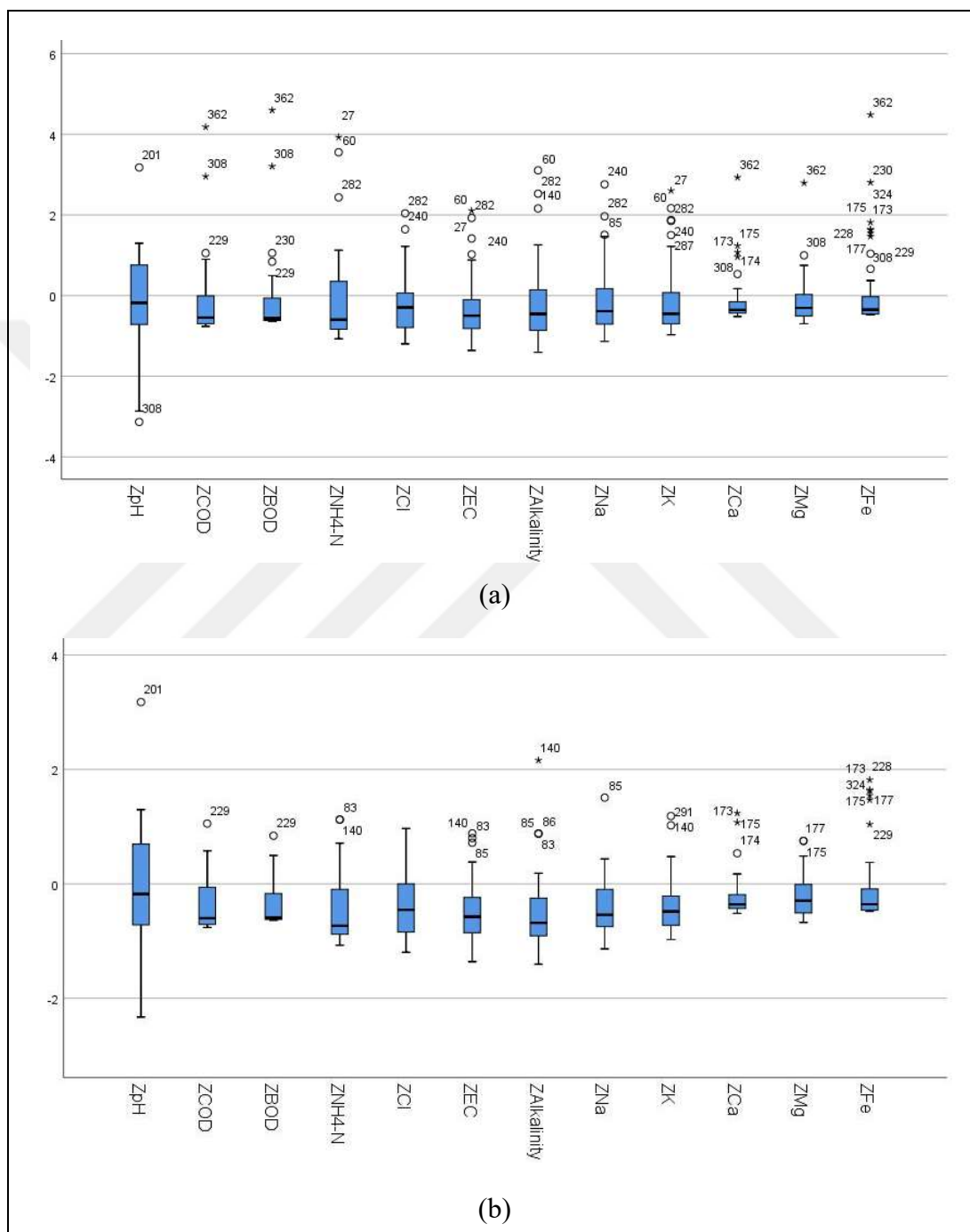


Figure 5.6. Box and whisker plots for the selected 12 leachate parameters (a) with outliers (b) after removal of extreme outliers

HCA was performed separately for two different parameter groups. Resulting dendrograms are shown in Figure 5.7. The number of clusters were selected based on visual examination of the dendrogram. More or fewer clusters could be defined by moving the phenon line/cut off on the dendrogram (De et al., 2017; Rinaldi et al., 2014). For the first case, three main clusters were identified when a phenon line was placed around 10% linkage distance. Cluster 1 contained COD, BOD, Ca, Mg, and Fe; cluster 2 had Cl, Na, K, alkalinity, EC, and NH₄-N, and cluster 3 had only pH.

The Dendrogram in Figure 5.7a shows that the dissolved ion content of leachate forms a separate class with two main sub-groups as Na-Cl-K and EC-alkalinity-NH₄-N. The first cluster having organic content (COD, BOD) and cations (Ca, Mg, Fe) as sub-groups is linked to the other clusters at the highest linkage distance indicating the lowest similarity with them. Clusters were in conformity with the related regression models such that sub-groups in the dendrogram are composed of the most correlated parameters like EC-alkalinity, COD-BOD and so on.

Cluster analysis was performed by another parameter group including heavy metals and organic content. Inclusion of heavy metals increased the possibility of having outliers. Therefore, some of the extreme outliers were removed (8 outliers). In total, 11 leachate parameters were considered (COD, BOD, pH, Fe, Mn, Zn, Cr, Cu, Cd, Pb, and Ni). Some parameters having a small number of data like salts, Ca, and Mg were not included in this trial. Resulting dendrogram is presented in Figure 5.7b. Four main clusters could be identified including some sub-groups as well. The first and biggest cluster included all heavy metals. The second cluster had Fe and Mn linked to the biggest cluster, the third cluster captured COD and BOD. pH stood alone in the fourth cluster.

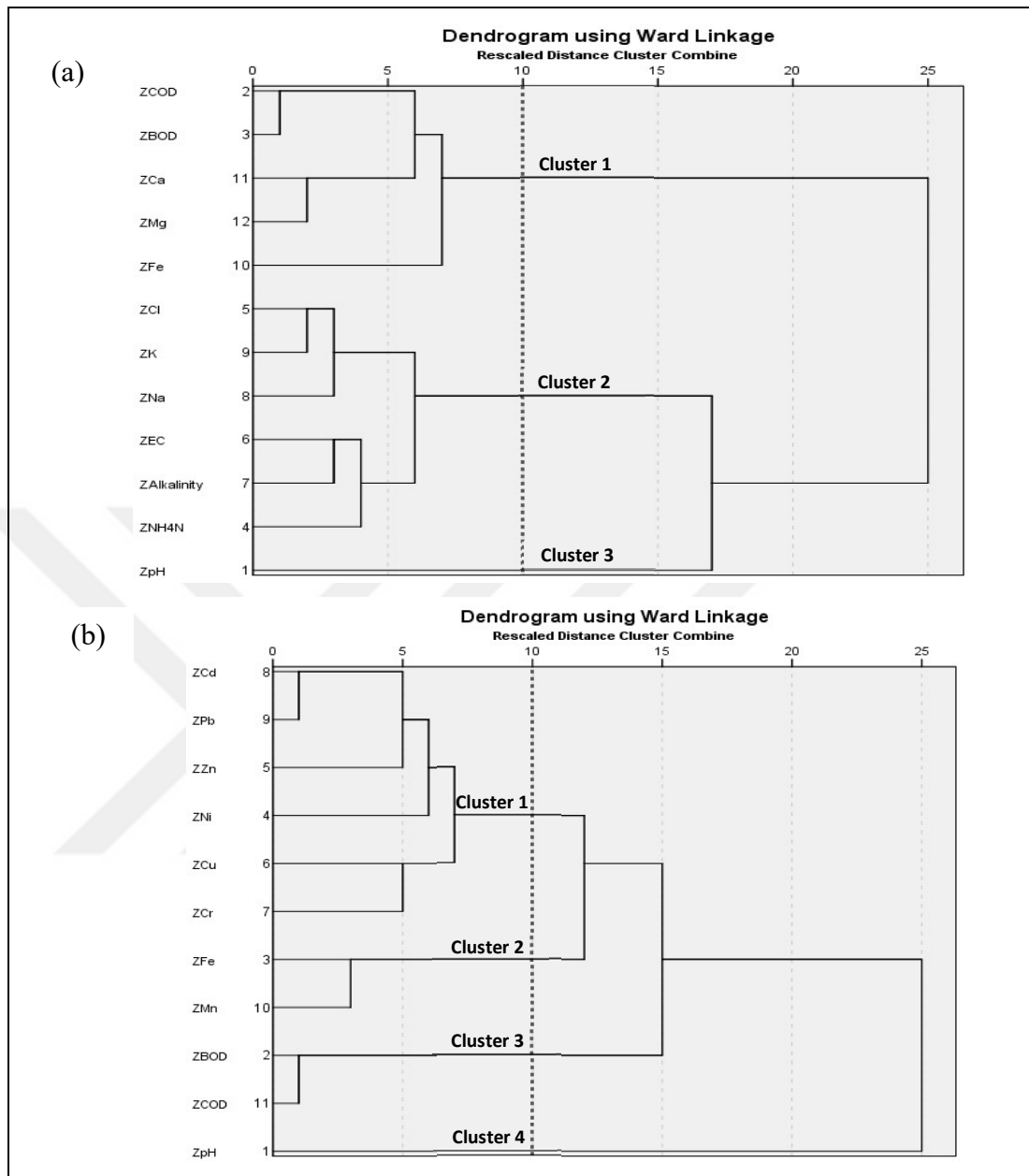


Figure 5.7. Dendrograms showing the clustering of standardized (a) 12 leachate parameters (Sample N=36) (b) leachate parameters including heavy metals (Sample N=63)

There are few studies in the literature about cluster analysis of landfill leachate particularly. The most relevant study was done by Talalaj et al. (2016). However, they did not mention about any data standardization step which is crucial in cluster analysis (De et al., 2017; Liu et al., 2008). They had 27 samples having mainly TOC, pH, EC,

Pb, Cd, Cu, Zn, and Cr measurements for a landfill in Poland covering a monitoring period of 7-years. They obtained two groups, one of which has only TOC and the other the rest of parameters. This clustering was due to higher TOC values in comparison to lower concentrations of other parameters, showing the importance of data pre-treatment for certain statistical tests.

5.3.4.2 Principal Component Analysis (PCA)

For the PCA study, initially the same leachate parameters used in the cluster analysis were selected for comparison. However, when PCA was applied for 12 parameters, multicollinearity condition was violated ($\text{determinant} < 10^{-5}$). Therefore, BOD was dropped due to high correlation ($r \geq 0.9$) with COD. Moreover, Na was excluded due to the high correlation with Cl and K. Therefore, PCA was re-run for a data set of 10 variables. All checks (Figure 5.1) were done. KMO value was 0.756 indicating suitability to conduct PCA. Table 5.8 shows the loadings of 10 parameters at each component with the associated cluster groups. High loadings were indicated as bold. All the inorganic parameters within cluster 2 were collected in PC1, while remaining parameters in clusters 1 and 3 were placed in PC2. It must be noted that an additional analysis which kept BOD as a parameter rather than COD revealed similar results in terms of PC formations as in Table 5.8.

PCA results indicated that 10 leachate parameters can be grouped into two main principal components having eigenvalues greater than 1.0 and accounting for 74% of the total variance. The remaining 26% of the variance in data set cannot be explained only with these two components. If higher percentage of variance to be explained, more components can be extracted by increasing the factor numbers in the SPSS. PCA results for three (Table D.1) and four components (Table D.2) are provided in Appendix D. In the two component-case, half of the parameters were gathered in the first component accounting for 41% of the total variance in the data. This component can be characterized by high positive loadings of dissolved ions. The second

component accounting for 33% of the total variance is mainly associated with high positive loadings of COD, Fe, Ca and Mg, with a high but negative loading of pH.

Table 5.8 Principal components (PC) and related cluster groups (Sample N=46, KMO=0.756)

Parameters	PC1	PC2	Cluster groups
ZEC	0.947	-0.065	Cluster 2
ZAlkalinity	0.899	0.052	Cluster 2
ZCl	0.893	-0.069	Cluster 2
ZK	0.877	-0.212	Cluster 2
ZNH ₄ -N	0.752	-0.128	Cluster 2
ZCa	-0.217	0.886	Cluster 1
ZFe	-0.063	0.849	Cluster 1
ZCOD	0.346	0.805	Cluster 1
ZMg	-0.087	0.698	Cluster 1
ZpH	0.315	-0.771	Cluster 3
Eigenvalue	4.4	3.0	
% variance	41	33	
Cumulative %	41	74	

High loadings of monovalent ions in PC1 result from solid wastes and are important constituents of the conductivity in leachate. Therefore, this component can be apportioned to the inorganic portion of leachate. Having most of the ions and conductivity in the same component may indicate that transport mechanism of those parameters from waste to leachate may be similar. Likewise, having organic matter with Fe, Ca, and Mg heavily loaded in PC2 may indicate that those are important parameters in the biodegradation mechanism in landfills. Armstrong and Rowe (1999) stated that COD and Ca concentrations tend to be proportional, and inversely proportional to leachate pH. Since in PCA the parameters in the first component are weakly related to the parameters in the second or other components, it can be argued that conductivity with monovalent ions is present in leachate (PC1) regardless of the presence of organics and associated divalent ions (PC2). In other words, the majority of the inorganic matter in leachate are not mainly related to biological activities, so their concentration in the leachate is not affected by the state of biodegradation but by

other mechanisms such as dissolution, leaching, or dilution. Armstrong and Rowe (1999) indicated a similar finding as the variation in chloride concentration is not correlated well with other leachate parameters (i.e., pH, COD, and Ca), and can increase even after 14 years of landfilling.

Loading plots indicate the importance of parameters being placed in a given component. In Figure 5.8a, PC1, which explains most of the variance, was located at the right side as positively and strongly correlated with EC, NH₄-N, alkalinity, Cl, and K, while COD, Ca, Fe, and Mg were seen at the top having a strong and positive correlation with the second component. PCA results can also be visualized by a score chart as given in Figure 5.8b showing the influence of each variable on each component.

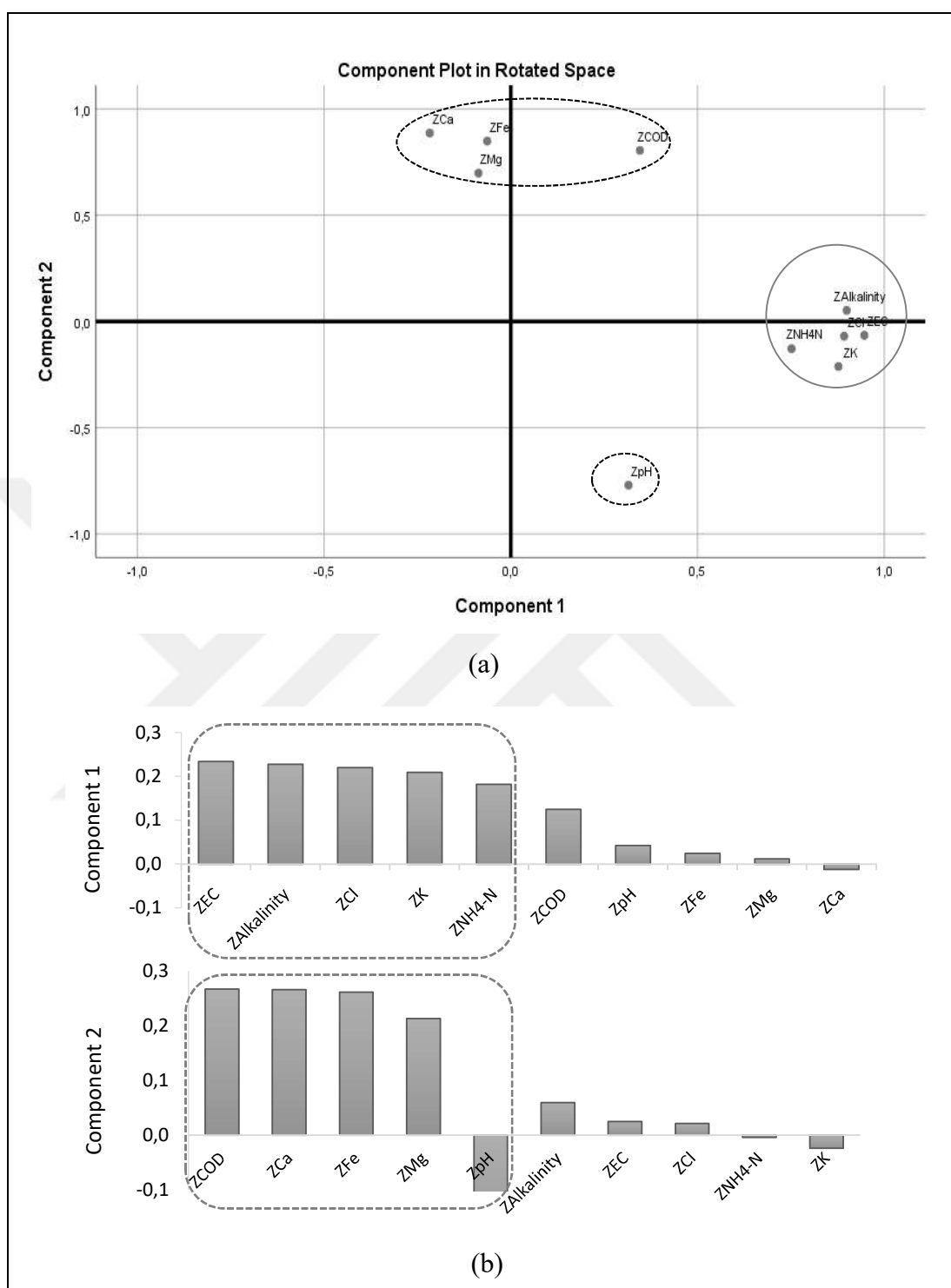


Figure 5.8. PCA results with 10 leachate parameters (a) Component plot (b) Score chart

In the analysis, as data was gathered from different landfills having different operational lives, it was deemed necessary to seek if there is a correlation between the extracted components and samples (landfills). There may be some underlying reasons behind the component formations such that each component is characterized by biological, chemical or physical factors which cannot be measured directly. In that respect, component scores obtained from the PCA analysis were saved for further investigation. It was seen that the highest scores in PC1 corresponded to the high concentrations of inorganic parameters among 46 leachate samples. Similarly, the highest scores in PC2 corresponded to high concentrations of both organics and divalent ions and lower pH values (Figure 5.8b). Furthermore, when the source of leachate samples having the highest scores were cross checked in the data matrix, it was observed that majority of the inorganic parameters listed in PC1 had the highest concentrations in samples coming from aged landfills (>10 years). Similarly, parameters listed in PC2 had the highest concentrations in samples from younger landfills (<10 years). Thus, PCA scores revealed the difference between components as landfilling time (landfill age). As purposed by Modin (2012), extracted components may distinguish between more and less stabilized landfills. Therefore, it is probable that most of the inorganic parameters in leachate will have relatively high concentrations in the long-term operation of landfills (older age), while organics and associated divalent ions will dominate in leachate during the initial and medium terms (younger age).

Component scores were used also to draw a plot showing the locations of the samples along the extracted two components as presented in Figure 5.9. In this score plot of 46 landfill's leachate samples, there are some data points close to each other representing the similar properties in terms of the investigated 10 leachate parameters. For example, on the lower right part (red circled) there are leachate samples coming from places with high and low precipitation rates with different landfill ages. Among them, two samples are from areas where annual average precipitation rates are relatively high (>1400 mm) and landfill age is less than 10 years, while two samples come from landfills with older age (≥ 13 years) and areas with much lower precipitation rates (400-

800 mm). Similarly, on the upper right part (blue circled) there are three samples with similar scores. Two samples come from landfills with younger (<10 years) age in higher annual rainfall rates (1000-1200 mm) and one sample from a lower precipitation area (800-900 mm) with much older age (14 years). Those examples suggest that leachate samples could show similar characteristics depending on climate conditions and landfill ages. In other words, a younger leachate sample could have similar characteristics with a much older landfill leachate if they come from relatively different climates. On the other hand, the leachate samples coming from similar conditions with respect to climate and landfill age could show similar properties as well. For instance, on the right most part of the plot in Figure 5.9 (green circled) there are similar scored leachate samples coming from landfills having similar precipitation rates (700-1000 mm) and landfill ages between 10-15 years. Those findings are supported with comparison tests accordingly in Chapter 6.

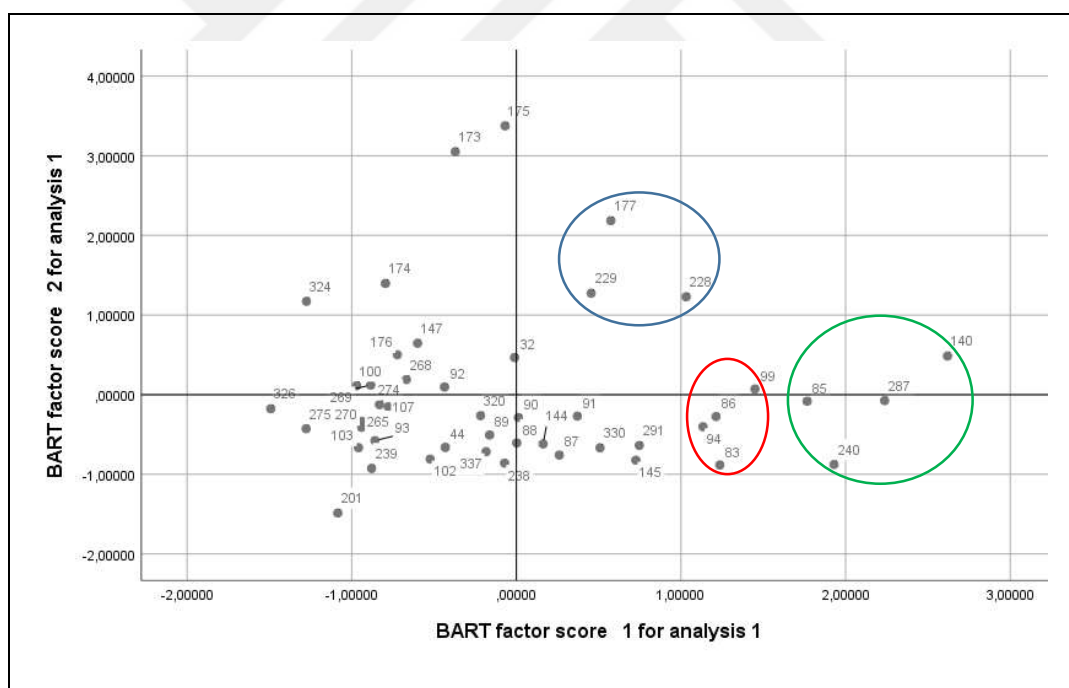


Figure 5.9. Scatter plots of the principal component scores with 10 leachate parameters

Andreas et al. (1999) extracted two components in PCA of leachate samples taken from a German landfill at different operational times. The first component included

mainly COD, BOD, Fe, Zn, and EC, whereas NH₄-N, Cl, TS, and K were placed in the second component. Ca and Mn contributed to both components. In the present work, COD and Fe were included in PC2 together with Ca and Mg, while NH₄-N, Cl, and K were in PC1. The major contrast of Andreas et al. (1999) with the present study is the reverse order of components including the same parameters. This could be explained by the difference in variance status of those parameters. In the study of Andreas et al. (1999), PC1 parameters shared a bigger portion of the variance (52%) in their leachate samples. They stated that degradation phases in landfills generated a major data variation so that PC1 primarily depicted biological conditions.

Results of multivariate statistical methods are strongly dependent on the data set used. Leachate composition generally shows huge variations with respect to time, site conditions, and seasonal differences (Vaccari et al., 2019). Therefore, some variations in results can be expected when applied to different leachate samples. By changing the leachate parameters evaluated in PCA, different outputs could be obtained. To be able to compare PCA results with the literature, as well as to see the segregation of heavy metals between components, PCA was performed for heavy metals with organic and inorganic parameters separately and results are provided below.

Variables of the PCA for organics and heavy metals were similar to the ones used in the cluster analysis of the same parameters (Figure 5.7b). Only, COD and BOD were not included together due to multicollinearity ($r > 0.9$). In many cases, outlier removal does help to meet the conditions of PCA to some extent. Around 21 outliers were removed to meet the requirements for the PCA. KMO value was found as 0.644 indicating sufficiency for PCA application. 10 leachate parameters were grouped into three main components having eigenvalues greater than 1.0, and capturing 70% of the variance in the original data (Table 5.9). Cluster groups for these parameters were well correlated with PCA results. The big cluster having all the heavy metals were divided into two components. For example, PC1 had Pb, Cd, and Ni, and PC3 had Cu, Cr, and Zn. Fe and Mn were combined with pH to form PC2. It was observed that organic content (BOD) had moderate loadings in both PC1 and PC2. This suggests that organic

matter and most of the heavy metals would exist together in leachate. This finding complies well with the literature as many heavy metals like Cd, Cu, Pb, and Ni are complexed strongly with the organic material in leachate (Baun and Christensen, 2004). Relation of heavy metals with organic matter was also indicated by Modin (2012). Yet, types of metals strongly related to organic matter showed differences between landfills. The highest concentrations of Fe and Mn can be observed in acidic conditions in landfill leachate showing a continuous negative correlation with pH as indicated in PC2. The results of the PCA were visualized by score charts as given in Figure 5.10.

Table 5.9 Principal components for heavy metals with the organic (Sample N=50, KMO=0.644) and inorganic (Sample N=46, KMO=0.763) contents of leachate

Parameters	Component Matrix			Parameters	Component Matrix		
	PC1	PC2	PC3		PC1	PC2	PC3
ZNi	0.847	0.010	0.091	ZCa	0.937	-0.026	-0.140
ZPb	0.845	0.017	0.013	ZFe	0.875	0.055	-0.020
ZCd	0.832	-0.025	0.035	ZMn	0.869	-0.194	0.160
ZBOD	0.509	0.489	0.241	ZpH	-0.732	0.269	0.111
(ZCOD)*	(0.505)	0.333	(0.467)	ZCu	-0.101	0.823	-0.075
ZMn	0.055	0.911	-0.077	ZCl	-0.203	0.732	0.369
ZFe	0.086	0.904	-0.046	ZCr	-0.192	0.714	0.450
ZpH	0.128	-0.741	0.034	ZNH ₄ -N	-0.283	0.633	0.375
ZCu	-0.017	-0.086	0.914	ZZn	0.409	0.605	-0.223
ZZn	-0.020	0.071	0.830	ZPb	0.006	-0.149	0.804
ZCr	0.353	-0.108	0.648	ZCd	-0.067	0.189	0.698
-	-	-	-	ZNi	0.028	0.343	0.574
-	-	-	-				
Eigenvalue	2.8	2.4	1.7	Eigenvalue	4.1	2.5	1.5
% variance	25	25	20	% variance	27	23	17
Cumulative %	25	50	70	Cumulative %	27	50	67

*Result of PCA when BOD was replaced with COD

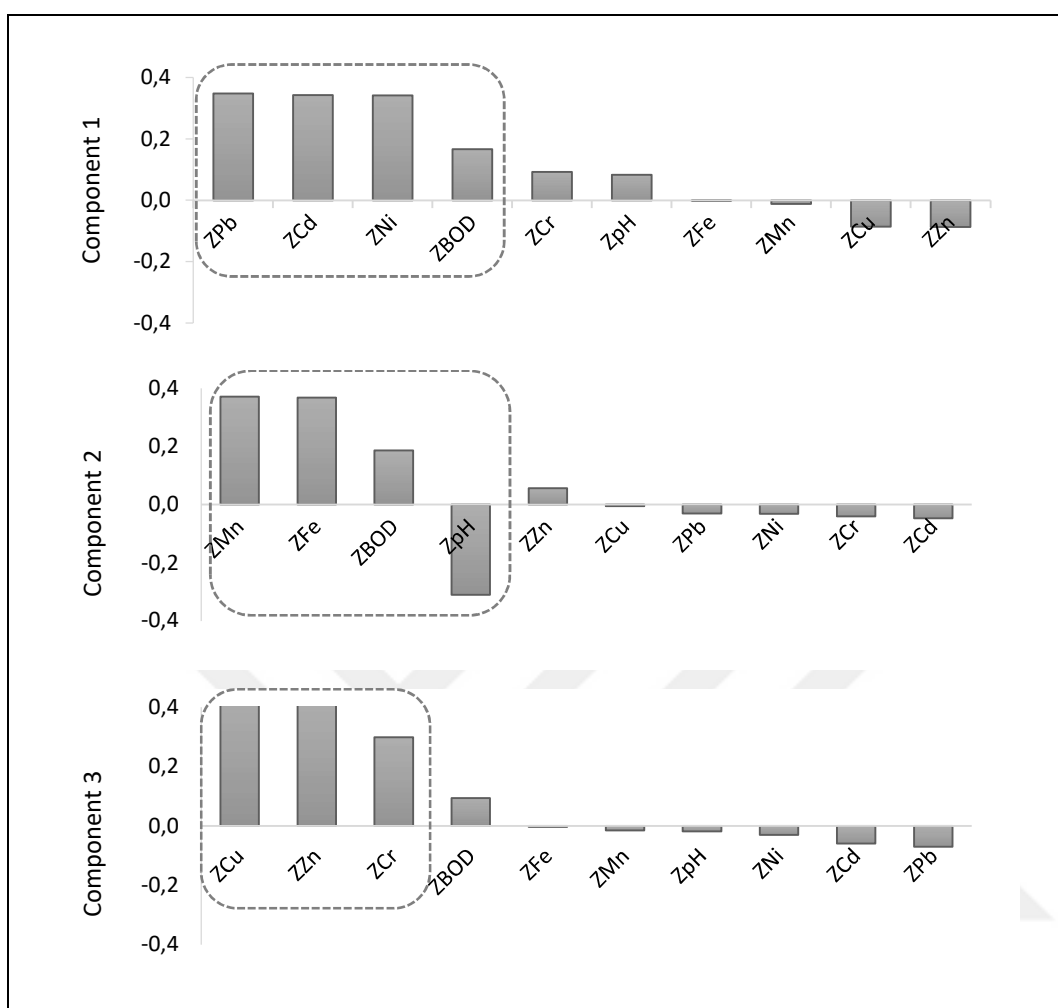


Figure 5.10. Component score charts for the PCA of organics and heavy metal content of leachate

When COD was used as input instead of BOD, similar results were obtained. The only difference was the co-existence of COD parameter in PC1 and PC3, unlike BOD being in PC1 and PC2 (Table 5.9). COD parameter could account for other substances that create additional space for transport of some heavy metals like Cu, Cr, and Zn. In fact, COD measurements include a strong chemical oxidant which can oxidize inorganic matters as well (Moody and Townsend, 2017). Kylefors et al. (2003) stated that inorganic ions such as SO_2 and Fe could be oxidized during the COD measurement. Baun and Christensen (2004) stated that significant amounts of heavy metals could be

found in organic complexes, but also in colloidal matters which mostly consist of carbonates, sulphides, and other inorganic materials besides the organics.

As Indelicato et al. (2018) stated, the higher the correlation between the leachate parameters, the lower the number of components in PCA. This could explain the higher number of components in data sets involving heavy metals (Durmugoglu and Yılmaz, 2006; Wdowczyk and Pulikowska, 2020). The correlation status of heavy metals is not strong. Only a few metals are moderately correlated with each other and only little or no correlation exists with other leachate components (Table 5.5). However, PCA results of some studies including heavy metals indicated strong correlation between some heavy metals and inorganic components. As stated by Modin (2012), higher focus was given to the discussion of heavy metal solubility with organic complexes in the literature, but attention should also be given to the effects of inorganic complexes on heavy metal leaching. For example, Cr was stated as the most related metal with the inorganic parameters and in the majority of leachate samples having high ammonia concentrations, ammonia-metal complexes could be formed as well.

For metals, solubility and hence their dominant forms are the critical determinants controlling their presence in aqueous systems. Therefore, leaching of metals depends strongly on the dissolution/precipitation of their hydroxides, carbonates, or sulfates (Banar et al., 2006; Modin, 2012) which are abundant in leachate during biological decomposition of waste. Some heavy metals are only soluble in their compounds of acetate, chloride, and sulfate (i.e., Fe, Mn, Zn, Cu, Ni, and Cr), while they are insoluble especially with hydroxide, carbonate and sulfide compounds (Angel et al., 2020; Erses et al., 2008). Banar et al. (2006) stated that high correlation exists between Cl and heavy metals. Modin (2012) stated that Cl enhanced solubilization of Cd and Zn by forming soluble complexes. Talalaj et al. (2016) stated majority of Zn seems to be related to inorganic colloids so Zn is more mobile than other metals.

To test above hypotheses, PCA was repeated for the inorganic composition of leachate together with heavy metals. In total 12 leachate parameters were input to PCA including $\text{NH}_4\text{-N}$, Cl, Fe, Ni, Zn, Cu, Cr, Cd, Pb, Mn, pH, and Ca. To comply with the

requirements of PCA, heavily correlated parameters, Na and K (with Cl), were not included and 6 outliers were removed from the data set accordingly. Following those steps, a high KMO value (0.763) was obtained. Three components were extracted explaining 67% of the variance in the data set. Related matrix is provided on the right hand side of Table 5.9. It was observed that some heavy metals (Cr, Cu, Zn) were grouped strongly with Cl and NH₄-N in the second component. Resulting score charts for each component are depicted in Figure 5.11.

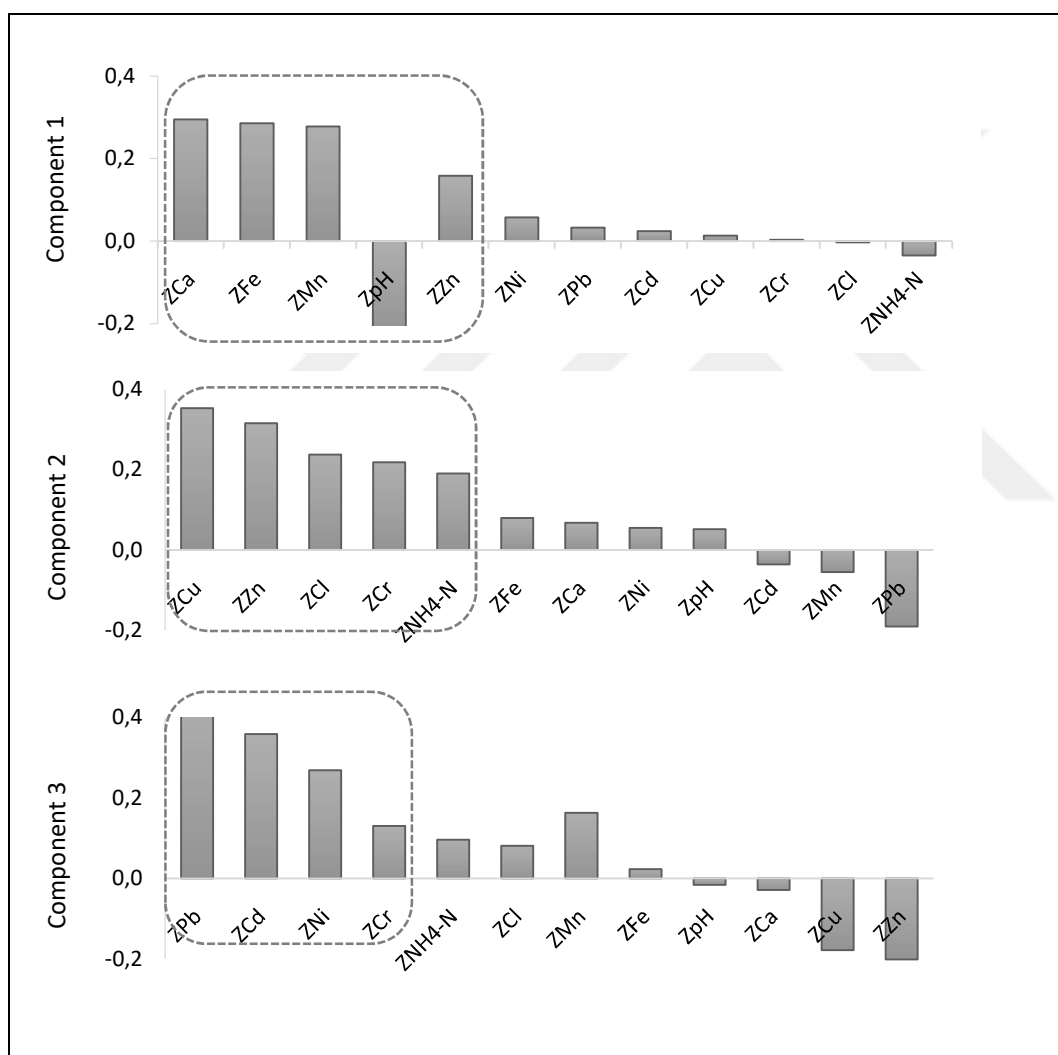


Figure 5.11. Component score charts for the PCA of inorganics and heavy metal content of leachate

As seen from Figure 5.11, PC1 was strongly correlated with Ca, Mn, Fe, and pH, with little contribution from Zn. Similarly, PC3 was strongly correlated with Pb, Cd, and Ni, with little contributions from Cr. As stated by Nyame et al. (2012), PCA results could be interpreted to explain the transport mechanisms of some pollutants from waste to leachate. In that respect, it could be said that majority of inorganic components acts together and heavy metals could be found in leachate mostly bonded in organic and inorganic complexes.

Durmusoglu and Yilmaz (2006) conducted PCA for leachate samples from a landfill in Turkey. Four factors were extracted which explained 75% of the variance in the data. The first component was heavily loaded with Cu, TP, Zn, and COD, with little contributions from Ni. It was stated that Cu, TP, Zn, and COD were the most important elements in their leachate samples for explaining the variation in the data. COD was in the same components (PC1 and PC3) with Cu, Zn, and Ni, as in the present study as well (Table 5.9).

Modin (2012) performed PCA for leachate samples from six Swedish landfills with different waste properties and various operational times. Data pre-treatment was conducted including data standardization, normalization of data (Log10), outlier removal, and missing data replacement by means. For the first MSW landfill case, 21 samples and 20 parameters were evaluated. The first three principal components explained 61% of the total variance. PC1 clustered heavy metals like Ni, Cr, Co, Pb, and Zn. PC2 included EC, Cl, and NH₄-N. COD and BOD were correlated to both PC1 and PC2. In the second MSW landfill case with 42 samples and 15 parameters, three components were extracted capturing 68% of the total variance. PC1 included EC, Cl, TN, and Cr. PC2 had COD, TOC, Ni, Co, and Zn. It was stated that for six landfills, different types of components comprising of different parameters were extracted. However, there were similarities as well such that salts generally dominated the first components, while organics with some heavy metals were included in the others. It was stated that landfilling time and deposited waste properties, as well as operational conditions, resulted in some changes in the correlation status of leachate parameters, hence created those differences.

Mishra et al. (2016) studied leachate samples from a landfill site in India. In the PCA of sixteen leachate samples from a collection tank, four components were extracted explaining more than 70% of the variance. Some parameters such as pH, EC, alkalinity, Cl, SO₄, BOD, COD, and Ca had impact on more than one component. Sample number was low (N=16) and KMO value to check sampling adequacy was not provided. Results of Mishra et al. (2016) are different than ours because of the composition of parameters in their PCA trial. They analysed both the organic and inorganic contents of leachate together with heavy metals, which might have increased the presence of outliers. However, there was no mention about any outlier control or checks for the requirements of PCA. For example, highly correlated parameters like COD and BOD, EC and TDS were inputs to PCA. When heavy metals were tested with both organic and inorganic parameters together in the present study (not shown), similar results were obtained in some components. However, certain parameters like BOD, EC, alkalinity, Na, and K were not included to satisfy multicollinearity condition; therefore, a definitive comparison to Mishra et al. (2016) could not be made.

Wdowczyk and Pulikowska (2020) studied leachate data from closed and active landfills in Poland. In their PCA study on leachate data (7 set of samples) from two active landfills, five components were extracted explaining more than 80% of the variability in their data. The problem of some parameters (pH, SS, TDS, Cd, Mn, Zn, and TKN) being in more than one component was encountered in their study as well. Outliers and requirements for PCA, which was not mentioned anyhow in the article, generally result in some difficulties in component formations. It was observed that there are similar points with the present study in terms of the content of the first components including Ca, Mn, Fe, pH, and Zn (weakly correlated). Furthermore, Pb, Ni, and Cr (weakly correlated) were together in one component.

Both reviewed references and results of the present study indicated that it is important to meet the basic requirements of a particular statistical technique to get accurate and comparable results. Cross checking of the extracted component scores with the related data matrix of leachate samples demonstrated that the highest scores in every

component corresponded to the highest concentrations of each variable in the components. This finding is important for emphasizing the necessity for checking of extreme outliers as they could impact the results significantly. However, because of specific characteristics of landfill leachate and its high variability, data treatment process may have some restrictions such that not all the outliers could be removed to prevent data loss.

An overall comparative summary of the PCA results obtained in relevant studies in the literature and this study is given in Table 5.10. Outputs of a selected statistical method depend highly on the data set used. Therefore, results of multivariate statistical methods of one study may not be the same with other studies conducted on different leachate samples. Nevertheless, comparison of the PCA results of the present study, including leachate samples from different landfills, with the results obtained for mostly single landfills shows that in general similar outcomes can be obtained. This may help to confirm the reproducibility and reliability of the results/outputs of PCA. However, the lack of information in most of the reviewed articles about whether outliers were controlled or how PCA requirements were met was emerged as an important shortcoming for comparisons.

Table 5.10 Comparison of the data analysed and principal components extracted in this study with literature

Leachate content analysed	Parameter list	Principal components (PC) extracted	Reference
Organics (as COD) and inorganics	EC, alkalinity, NH ₄ -N, K, Cl, COD, Fe, Ca, Mg, pH	PC1: EC, alkalinity, Cl, K, NH ₄ -N PC2: COD, Fe, Ca, Mg, pH	Present Study
Organics (as BOD) and inorganics	EC, alkalinity, NH ₄ -N, K, Cl, BOD, Fe, Ca, Mg, pH	PC1: EC, NH ₄ -N, Cl, K, alkalinity PC2: BOD, Fe, Ca, Mg, pH	Present Study
Organics and inorganics	pH, COD, BOD, Fe, Zn, EC, NH ₄ -N, Cl, TS, K, Na, Ca, Mn, SO ₄ , NO ₃	PC1: pH, COD, BOD, Fe, Zn, EC, Ca, Mn, SO ₄ , NO ₃ PC2: NH ₄ -N, Cl, TS, K, Na, Ca, Mn	Andreas et al. (1999)
Organics (as BOD) and heavy metals	BOD, Ni, Pb, Cd, Mn, Fe, pH, Cu, Zn, Cr	PC1: BOD, Ni, Pb, Cd PC2: BOD, Mn, Fe, pH PC3: Cu, Zn, Cr	Present Study
Organics (as COD) and heavy metals	COD, Ni, Pb, Cd, Mn, Fe, pH, Cu, Zn, Cr	PC1: COD, Ni, Pb, Cd PC2: Mn, Fe, pH PC3: COD, Cu, Zn, Cr	Present Study
Organics (as COD) and heavy metals	Cu, TP, Zn, COD, S, Ni, Phenol, Hg, SS, Sb, Pb	PC1: Cu, TP, Zn, COD, Ni, Phenol PC2: Hg, SS, Sb PC3: Pb, Phenol PC4: S, Sb, Ni	Durmusoglu and Yilmaz (2006)
Inorganics and heavy metals	Ca, Fe, Mn, pH, Cu, Cl, Cr, NH ₄ -N, Zn, Pb, Cd, Ni	PC1: Ca, Fe, Mn, pH, Zn PC2: Cu, Cl, Cr, NH ₄ -N, Zn PC3: Pb, Cd, Ni, Cr	Present Study
Organics, inorganics, and heavy metals	COD, BOD, TOC, pH, Ni, Cu, As, Cr, Co, Pb, Zn, EC, Cl, TP, NH ₄ -N, TN, Mn, Fe, Phenol	PC1: Ni, Cr, Co, Pb, Zn, COD, BOD PC2: EC, Cl, NH ₄ -N, COD, BOD PC3: Not provided	Modin (2012)
Organics, inorganics and heavy metals	EC, Cl, TN, Cr, COD, TOC, DOC, Ni, Co, Zn, Cu, Pb, Cd, As, Hg	PC1: EC, Cl, TN, Cr, As PC2: COD, TOC, Hg, Ni, Co, Zn PC3: Not provided	Modin (2012)
Organics, inorganics, and heavy metals	COD, BOD, pH, EC, alkalinity, Cl, TDS, SS, Ca, Mg, Fe, Cu, Al, SO ₄ , Ni, K	PC1: pH, EC, alkalinity, Cl, TDS, COD, BOD, Ca, Mg, Fe, Cu, Al PC2: pH, EC, alkalinity, Cl, SO ₄ , Ni PC3: pH, EC, alkalinity, SO ₄ , TDS, SS, BOD, COD, K, Al PC4: pH, SO ₄ , COD, BOD, Ca	Mishra et al. (2016)
Organics, inorganics, and heavy metals	EC, pH, TDS, SS, TS, Cl, Ca, Fe, Zn, Mn, Cd, Cr, Cu, Pb, Ni, COD, Na, K, TKN, Mg	PC1: EC, pH, TDS, SS, Cl, Ca, Fe, Zn, Mn, Cd PC2: Cr, Pb, Ni, Mn, Zn, Cd PC3: COD, TDS, Na, K, Cu PC4: TKN, Mg PC5: pH, TKN, SS, TS	Wdowczyk and Pulikowska (2020)

5.4 Conclusions

Findings of multivariate statistical analyses supplied valuable information about the specific properties of landfill leachate as well as its source. Co-existence of some pollution parameters in the same or different components in PCA enabled to get insight about their pathways into leachate in terms of transport and biodegradation mechanisms. In that sense, strong statistical evidences were found showing the existence of some heavy metals in leachate bonded not only with organic but also with inorganic complexes. Furthermore, extracted components referred to a notable divergence between inorganic and organic portion of leachate. Organic portion (BOD, COD) and associated parameters like Ca, Mg, and Fe appeared in separate components than the majority of inorganics, indicating their concentrations in leachate are independent from each other. Therefore, it can be concluded that concentrations of particular inorganic substances like salts, $\text{NH}_4\text{-N}$, and alkalinity in leachate may not be directly affected by the state of biodegradation but by other mechanisms (dissolution, dilution, etc.).

Correlation status of parameters played an important role in the component formations in PCA, while the concentrations (especially higher ones) impacted distribution of parameters between components. In that respect, higher concentrations of EC, alkalinity and monovalent ions in the first component were associated with samples from aged landfills. On the other hand, divalent ions and organics located in the second component that had higher concentrations belonged to leachate samples from landfills of early operational life. Thus, PCA partitioned leachate parameters into components according to samples originating from more or less stabilized landfills.

Having found that there are many leachate parameters strongly correlated with each other, it is worthy of recommending that highly correlated parameters can be excluded from sampling and analytical procedures during monitoring activities at landfills. For instance, those could be interchangeably selected for monitoring are $\text{NH}_4\text{-N}$, TN or TKN; Ca or Mg; K, Na, or Cl; EC or TDS.

CHAPTER 6

COMPARATIVE STUDY FOR LANDFILL AGE, CLIMATE, AND DEVELOPMENT STATUS IMPACTS ON LANDFILL LEACHATE CHARACTERISTICS

6.1 Introduction

Landfilling is a worldwide method for the disposal of solid wastes (Luo et al., 2020; Mmereki et al., 2016). Meanwhile, leachate generation is one of the most important problems of waste disposal in landfills due to being highly polluted wastewater with high concentrations of organic and inorganic compounds. In literature, many authors compared their leachate samples with leachates of other studies originating from different landfills having different operational lives (termed generally as landfill age). It appears that this comparison was done regardless of observing the differences in the climate of the region that the given landfill is located (Öman and Junestedt, 2008; Somani et al., 2019). Since leachate is a specific wastewater that shows huge variation in characteristics even in the same landfill from time to time, it is very difficult to make reasonable comparisons between leachate samples from landfills in different geographies. It should also be noted that many leachate characterization studies were restricted to few leachate samples from single landfills.

In many studies, leachate data from a single landfill site in different operational years and seasons were used to show the effect of landfill age and rainfall amounts on the leachate quality and quantity. Therefore, climate related articles are limited to seasonal differences mostly in the same landfill (Khattabi et al., 2002; Mangimbulude et al., 2009; Mohammad-pajooh et al., 2017; Tatsi and Zouboulis, 2002; Tsarpali et al., 2012). In some studies, different landfill sites were compared for age and/or climate impact on leachate (Ma et al., 2022; Mohammad-pajooh et al., 2017; Robinson, 2005; Zegzouti et al., 2019). Only few studies employed a detailed statistical analysis with considerable data including samples from different landfills (Brennan et al., 2016; Kjeldsen and Christophersen, 2001; Somani et al., 2019; Vaccari et al., 2019).

Beside the time and climate impacts, another important factor that could affect the leachate properties is the development status of countries. Solid waste management in developing countries are different compared to developed countries. Sanitary landfilling, incineration of waste, composting, and recycling are common methods in developed countries, whereas illegal dumping and burning of waste are applied mostly in developing countries (Nanda and Berruti, 2021; Vaccari et al., 2019). Regarding waste type, developing countries produce waste with higher organic content causing high density waste and high moisture content in landfills (Karak et al., 2012; Mmereki et al., 2016; Vaccari et al., 2018). Those issues can create considerable differences in landfill leachate properties (Ma et al., 2022).

The main purpose of this study is to find out the variation in leachate characteristics originating from landfills having different operational lives (termed as landfill age hereafter). Beside that, well-known climate effect on leachate composition was investigated in detail together with interaction of climate-landfill age factors. A relatively high number of leachate samples from landfills all around the World with different ages and climates were compiled for detailed statistical analysis to find out the effect of both landfill age and climate factors (in terms of precipitation and temperature) on leachate characteristics. Besides these environmental factors, development status of countries where landfills belong were also studied as a separate analysis. Leachate data were classified into specific groups and leachate pollution parameters between groups (age, climate, and development status) were compared using parametric and non-parametric tests including analysis of variance (ANOVA), multiple comparison Post-Hoc tests, and Mann-Whitney U test.

6.2 Materials and Methods

6.2.1 Data Collection and Classification into Groups

Leachate data collection phase and data pre-treatment procedures in terms of removal of outliers and missing data completion are explained in Sections 5.2 and 5.3. However, data set used in this part differs from that used in Chapter 5 such that in the

original data matrix, there are more than one leachate sample for the same landfill belonging to different times compiled from different studies each providing different number of parameters. In this part, only one set of data including as many parameters as possible was selected in order to have only one leachate sample from the same landfill. Missing data substitution was reconsidered here according to the 15% limit (Ergene et al., 2022; Modin, 2012) at each group. For both data removal and missing data substitution steps, priority was given to keep the samples for parameters having less number of data in certain landfill age or climate groups. For example, sample of a landfill having older age (i.e., greater than 15 or 20 years) was kept or missing data was calculated accordingly for that selection due to having less data in the data matrix for landfills of older age.

This study investigated leachate data collected from numerous landfills with different ages from regions of the World including both developed and developing countries and having different climate conditions (Table 6.1 and Figure 6.1). Data had to be analysed according to the climatic conditions and landfill age, and also for development status in order to find out which factors and how they impact the leachate properties. Therefore, a method was developed to categorize the leachate data into relevant groups as explained below. It is worth to note that, as Wijesekara et al. (2014) indicated as well, there are many characterization studies and reviews about the composition of landfill leachate mainly from developed countries. On the other hand, number of studies on the landfill leachate from tropical climates such as developing Asian countries are very limited. This could be observed from the global distribution of collected leachate data as presented in Figure 6.1.

Table 6.1 Locations and climate conditions of landfills in leachate data matrix

Country	Number of landfills*	Precipitation (mm)**	Temperature (°C)**	Country	Number of landfills*	Precipitation (mm)**	Temperature (°C)**
Algeria	3	575	17.4	Malaysia	9	2744	27.3
Australia	1	536	16.4	Mexico	2	718	20.9
Brazil	10	1348	20.8	Morocco	3	350	18.7
Canada	10	890	6.4	Nepal	2	2490	19.4
China	26	1136	16.0	New Zealand	5	1095	13.3
Colombia	1	1159	23.4	Norway	2	772	5.4
Croatia	1	930	11.0	Palestine	2	318	19.7
Denmark	7	695	7.8	Poland	3	626	7.5
Egypt	1	183	20.6	Portugal	1	1087	14.7
Finland	2	644	4.8	Russia	1	680	4.7
France	7	778	11.5	Saudi Arabia	1	111	25.4
Germany	2	719	9.6	Slovenia	1	1290	10.4
Ghana	1	1145	25.9	South Africa	4	807	19.5
Greece	1	700	17.3	South Korea	14	1284	12.5
Hong Kong	5	2018	22.7	Spain	18	852	14.1
India	2	1695	26.9	Sweden	2	597	7.4
Indonesia	4	2576	24.5	Taiwan	10	1696	22.3
Iran	1	316	16.8	Thailand	3	1463	27.3
Ireland	2	882	9.3	Tunisia	1	466	18.1
Island of Mauritius	1	1916	21.8	Turkey	16	696	14.5
Italy	5	666	15.5	UK	8	872	9.6
Lebanon	2	790	17.9	Uruguay	1	933	16.3
Lithuania	1	632	6.1	USA	10	1320	18.5

* Each is a different landfill (i.e., 26 different landfills from China)

** Annual Average Rates for the landfill location in a given country

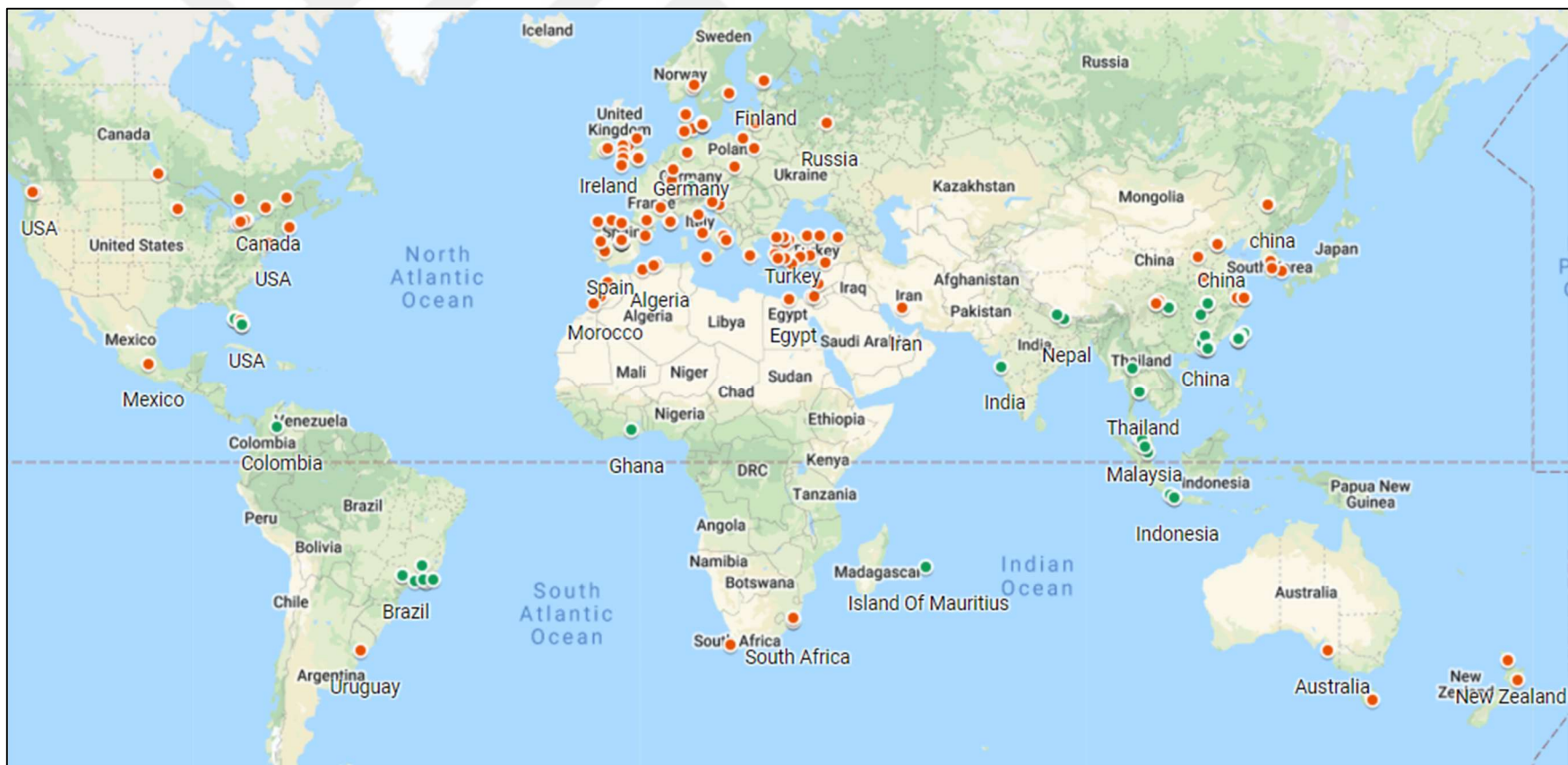


Figure 6.1. Global distribution of the landfills from which leachate samples were acquired (red circles indicate low rainfall & low temperature areas and green circles high rainfall & high temperature areas)

In this study, some approximations were done if needed as explained here. Landfill or leachate age was not provided clearly in many references. In these studies, the age of the landfill at the time of leachate sampling was determined as the difference between initial waste placement time (start date of landfill operation) and sampling time of leachate (Reinhart and Grosh, 1998). If starting time of the landfill was not provided but landfill name is known, web search was done to find out the landfill operation dates (start/closure) from other sources. If sampling time was given as a time interval, the date corresponding to the mid-point of the interval was taken as the sampling time. For example, if leachate average data was given for 2010-2012 period, then 2011 was taken as the average sampling year to calculate the landfill age. Other details of data collection are provided in Section 3.1.

It is not possible to compare the leachate characteristics according to individual landfill ages without any categorization into groups. This is mainly because of the limitations in the data availability. As the number of cases increases, the number of data distributed under each group decreases (Table 6.2). Therefore, data was classified into different age groups for analysis of landfill age effect. Age groups were arranged in a way that differences between younger and older landfills could be detected. Furthermore, for comparison purposes, it was tried to comply with the landfill age concept in literature that was based on generally three age groups as young (<5 years), medium (5-10 years), and old (>10 years) (Amokrane et al., 1997; Gao et al., 2015; Ma et al., 2022) as presented in Chapter 4. Selected age groups are provided in Table 6.3 with the number of data in each group. Number of data stands for the number of landfills corresponding to each age group. For example, there are a total of 59 leachate samples belonging to landfills having operational life up to 5 years. Number of data for each leachate parameter existing in age groups are different depending on data availability.

Table 6.2 Number of landfills corresponding to each landfill age in leachate data set

Landfill age (yr)	Number of Landfills	Landfill age (yr)	Number of Landfills	Landfill age (yr)	Number of Landfills	Landfill age (yr)	Number of Landfills
1	20	10	6	19	8	30	1
2	15	11	3	20	4	33	4
3	8	12	10	21	3	34	2
4	16	13	7	22	2	36	1
5	7	14	11	23	1	41	1
6	12	15	9	24	1	42	1
7	17	16	6	25	1	49	1
8	11	17	9	26	1	57	1
9	9	18	4	27	2		

Table 6.3 Generated landfill age groups with number of landfills in each group

Age group	Landfill age	Number of landfills
1	<5 years	59
2	5-10 years	56
3	10-15 years	46
4	>15 years	54
Total		215

The other data grouping was done for climate factor. Climate data of landfill locations (annual average precipitation rate and temperature) were taken from the articles if given. However, it was not provided in most of the references, therefore a web link was used (Climate-data.org) which provides climate data given in Table 6.1 according to the specified location. Landfill location was found according to the available information given in the reference such as map of the site or province name.

Climatic conditions of a landfill area are treated as environmental factors affecting the leachate characteristics. In many studies it was stated that seasonal variations have great influence on the leachate quantity and quality (Mangimbulude et al., 2009; Talalaj et al., 2016; Fan et al., 2006; Tsarpali et al., 2012). This was generally related

to rainfall amounts (Yang et al., 2015). However, impact of ambient temperature was also indicated in some studies affecting the degradation rate of waste positively thus increasing the COD and BOD in leachate (Ma et al., 2022). In this study, climate impact was analysed in two groups incorporating both temperature and precipitation rates in different scales. It was not possible to investigate climate effect by taking into account only temperature parameter due to lack of climate classification method with respect to only temperature (Pidwirny, 2002). However, precipitation rates could be accepted as the main parameter in climate classification (Table 6.4). Furthermore, the possibility of having sufficient number of leachate data in as many temperature and rainfall cases as is very low (Vaccari et al., 2019). In many places where leachate data belonged, high temperatures were co-existing with high rainfalls (i.e., tropical areas). On the other hand, in many moderate climate zones, low temperatures (i.e., $<10^{\circ}\text{C}$) could accompany with very low (<600 mm, i.e., in Sweden, Poland, Canada) or high (>1000 mm, i.e., in USA, UK, and Canada) precipitation rates.

Climatic zones presented by the Food and Agriculture Organization (FAO) was taken into consideration for grouping of leachate data as annual rainfall amounts are used for specification. A distinction was made by FAO (FAO, 2021) between 6 major climatic zones in terms of precipitation rates as shown in Table 6.4. In the leachate data set, there is no landfill in a desert zone and the number of data is very low for the arid and semi-arid regions. Therefore, arid and semi-arid zones were combined into the same group in terms of precipitation rates.

Table 6.4 Climate zones defined by FAO according to the annual precipitation rates (FAO, 2021)

Climatic zone	Annual rainfall (mm)	Mean precipitation (mm)*	Mean temperature ($^{\circ}\text{C}$)*	Number of leachate data
Desert	<100	-**	-	-
Arid	100-400	282	18.1	10
Semi-arid	400-600	530	12.7	19
Sub-humid	600-1200	832	12.4	101
Moist sub-humid	1200-1500	1347	17.6	51
Humid	>1500	2224	24.1	34

* Mean values were calculated from leachate data matrix under each group.

** There is no landfill/leachate data in desert zone.

According to the defined climate zones, mean temperature values for landfill locations in leachate data matrix are presented in Figure 6.2. As seen from Table 6.4 and Figure 6.2, mean temperature values are close to each other in some climate zones. In addition, in the data matrix, the annual average temperatures in places with high precipitation rates were observed generally above 20°C, and on the contrary, the annual average temperatures are generally below 20°C in places with low precipitation rates. Since there is not any available criteria for the climate grouping with respect to temperature, the FAO's climate groups were modified to include the temperature as well. For aforementioned reasons, it was deemed appropriate to divide the climate factor into 2 groups as given in Table 6.5. It was observed that in the first group (<1200 mm), there are a few places having mean annual temperature values greater than 20°C, and in the second group (>1200 mm) there are some places having mean annual temperature values less than 20°C. Therefore, 22 data in total were removed from the data matrix to comply with the climate conditions given in Table 6.5. Distribution of landfills according to the climate groups is presented in Figure 6.1 as red circles indicating climate group 1 and green circles climate group 2.

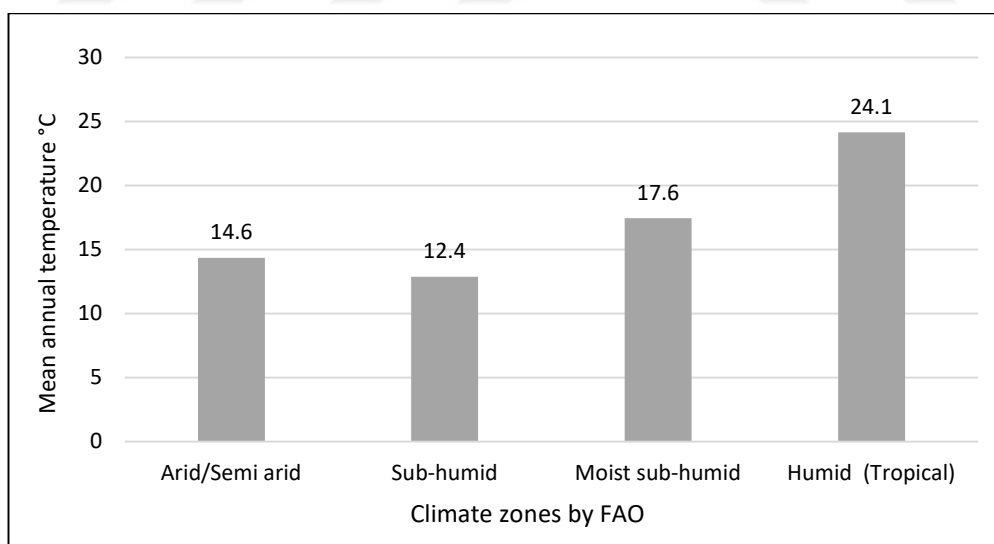


Figure 6.2. Mean temperature values for the landfills included in each climate zone

Table 6.5 Generated climate groups with respect to annual average precipitation and temperature rates

Group number	Climate Groups	Precipitation range (mm)	Temperature range (°C)	Mean precipitation (mm)*	Mean temperature (°C)*	Landfill numbers
1	Arid/Semi arid/Sub-humid	<1200	<20	765	12.4	131
2	Moist sub-humid/Humid	>1200	>20	1833	23.0	62

*Mean values were calculated from leachate data matrix under each group

There are some distinct features of solid waste management methods in developed and developing countries (Hoornweg and Bhada-Tata, 2012; Vaccari et al., 2019). The most important difference is the waste properties sent to the landfills. In developed countries, landfilling of waste has restrictions for organic content, sludge content, and recyclable matter content as well (Mmereki et al., 2016). There are many other differences such as co-disposal with large quantities of ash (mainly waste incineration plants) in developed countries (Modin, 2012; Moody and Townsend, 2017) while co-disposal of large amounts of sludge in developing countries. However, those specific cases were not taken into account in this study because only leachate samples taken from MSW landfills were included in the data matrix with other conditions as stated in Section 3.1. Therefore, it was deemed useful to investigate also the effect of development status of countries on landfill leachate characteristics.

As seen from Table 6.4, most of the landfills belong to sub-humid climate zone with a precipitation rate between 600-1200 mm. For this reason, statistical analysis focused only on data from this precipitation group to analyse the development status effect to eliminate the possible precipitation impact. For comparison purposes, a common practice was followed as using the income criterion to indicate the development status of countries where landfills are located (Ma et al., 2022; Mmereki et al., 2016; Vaccari et al., 2019). Accordingly, World Bank (2020) provided list of economies mainly as high income (H), upper middle income (UM), and lower middle income. In this study majority of leachate samples come from H and UM countries. Therefore, classification of data was done into two groups accordingly and developing countries are defined as

upper middle income (UM) and developed countries as high income (H) (Mmereki et al., 2016; Vaccari et al., 2019).

Due to possibility of leachate parameters showing significant difference with time, for the analysis of development status, landfill age was classified into two groups as <10 years and >10 years. Otherwise, if the landfill age group was taken as four (as <5, 5-10, 10-15, and >15 years) or three in any combination (i.e., <5, 5-15, and >15 years), data distribution between groups for many parameters resulted in less than 10 data which was taken as the minimum number for analysis (Vaccari et al., 2019). Therefore, to evaluate the impact of development status on leachate parameters, groups were selected as given in Table 6.6. As seen, mean values of landfill age, precipitation, and temperature rates are distributed more or less homogeneously between each group. With regard to mean precipitation and temperature values in groups, it is seen that the average data obtained resembles the first climate group (precipitation<1200 mm and temperature<20°C).

Table 6.6 Mean values of the data according to development status and age groups

Development status	Developed countries (H)		Developing countries (UM)	
Age group	<10 years	>10 years	<10 years	>10 years
Landfill age, (yr)	5.4	19.5	4.2	16.2
Precipitation, (mm)*	914	823	891	872
Temperature, (°C)*	11.5	10.1	16.4	15.8
Landfill numbers	36	42	23	14

*Annual average rates as calculated from leachate data matrix under each group

6.2.2 Data Analysis

After completion of the data matrix with the steps as presented in Figure 6.3, comparison tests were performed. Analyses were designed and applied on extensive leachate data set to show the trends, to find out the impacts of climate, landfill age, and development status on leachate composition. Test for comparison of means helps to find out how leachate parameters vary between different groups and if those changes

are significant or not in order to generalize the outcome for the landfills having similar conditions. For comparison of groups, parametric and non-parametric statistical methods could be used including analysis of variance (ANOVA) and Kruskal-Wallis, respectively (Torrance et al., 2009; Treister et al, 2015). Parametric tests have some advantages for data comparison such that it is more straightforward method and easier to apply by just one function in SPSS to find out the differences between multiple as well as pair of groups. Furthermore, mostly ANOVA was used by researchers for analysis of leachate data (Brennan et al., 2016; Mangimbulude et al., 2009; Mor et al., 2006; Somani et al., 2019). Therefore, ANOVA was preferred in this study for comparison of means between groups. IBM SPSS software for Windows version 25 was used for all the statistical analyses. Significance level was set as 0.05 ($p < 0.05$).

Since there are two different groups (climate vs age, and development status vs age), each having sub-groups as well, two-way ANOVA was conducted first to compare the leachate parameters at different climate/development status conditions for each landfill age group due to possible interaction effect. Independent variables were climate factor with two groups (precipitation rate < 1200 mm with temperature $< 20^{\circ}\text{C}$ and precipitation rate > 1200 mm with temperature $> 20^{\circ}\text{C}$) and landfill age with four groups (< 5 years, 5-10 years, 10-15 years, and > 15 years). Similarly, for the second case, independent variables were development status with two groups (high income and upper middle income) and landfill age with two groups (< 10 years and > 10 years). Dependent variables were the leachate pollution parameters. Since there is a problem of missing data for some leachate parameters, analyses were performed on the parameters having sufficient data ($N \geq 10$) at each group (Vaccari et al., 2019).

It is indicated in many studies that if the data are not normally distributed, then parametric tests should not be applied (Treister et al, 2015). Results of many leachate characterization studies indicated that the majority of pollutant concentrations fluctuates in large scales, hence leachate data generally have non-normal distribution (Akyol, 2005). After compilation of leachate data set and categorization of the data into related groups, required procedures were conducted to meet the conditions of ANOVA. Test results revealed that raw leachate data do not have a normal distribution

as expected. Therefore, logarithmic data transformation (Log10) was tried first. Majority of the leachate parameters according to the groups were log normally distributed except pH which already measured on a logarithmic scale (Kjeldsen and Christophersen, 2001; Visconti et al., 2009). For some parameters having log-normal distribution in majority of the groups but not in one or two groups, outlier data were checked from the box and whisker plots. For those as well as for pH, some of the outlier data (mostly having extremely low and high values) were deleted one by one and normality test was repeated till $p > 0.05$ is obtained in these groups as well.

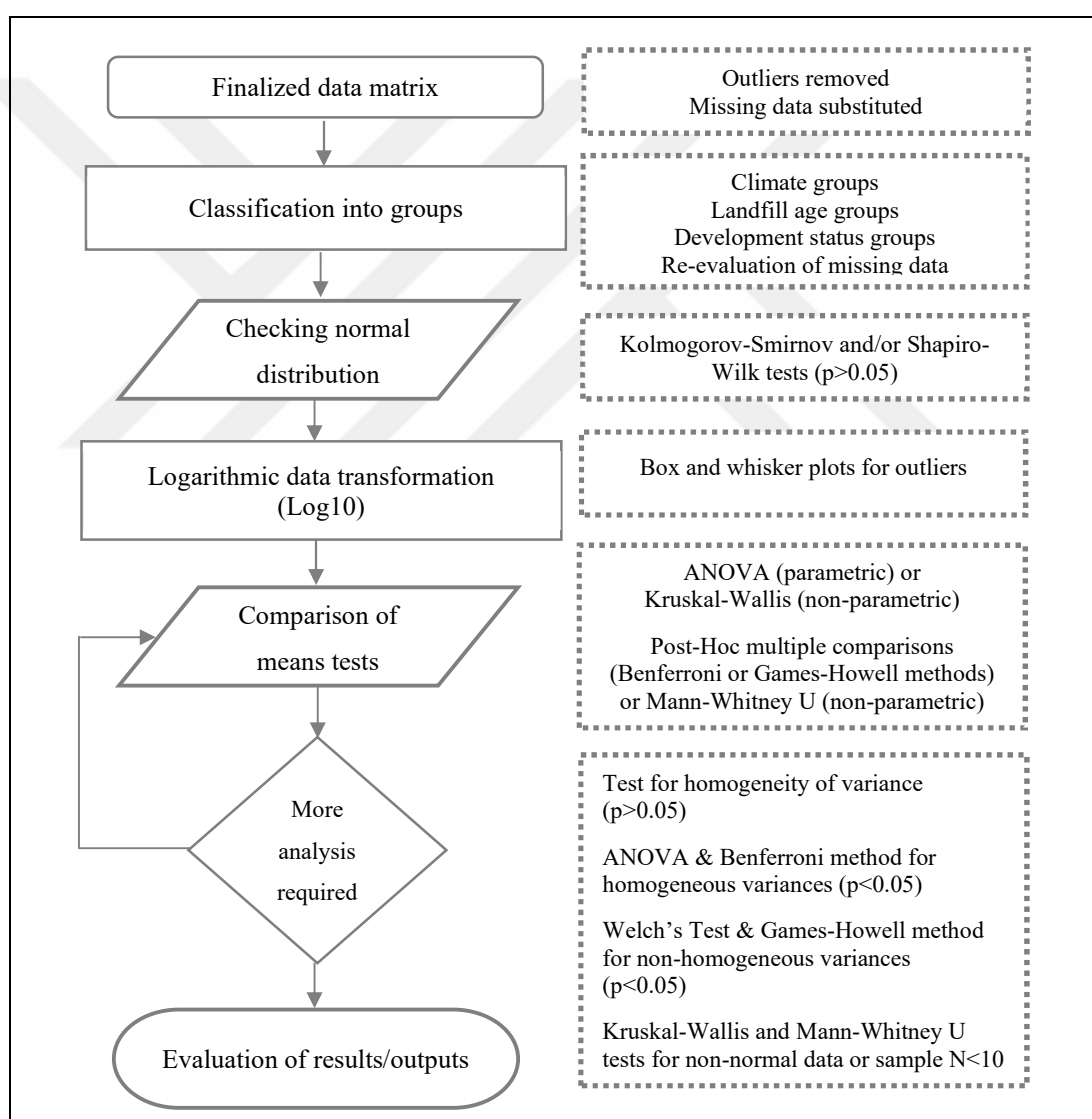


Figure 6.3. Flowchart for data preparation and analysis steps

If neither the original nor the transformed data had a normal distribution or if it required to delete too many data for normality, then other methods were applied like non-parametric tests Kruskal-Wallis and Man Whitney U (Torrance et al., 2009). Advantage of the non-parametric tests is that it could be used for the data for which normal distribution cannot be achieved. Another benefit is that small number of samples could be sufficient for the required analysis (Treister et al, 2015; Wdowczyk and Pulikowska 2020). Detailed information about the comparison tests is provided in Section 3.2.5.

6.3 Results and Discussion

6.3.1 Preliminary Examination of Climate and Age Effect on Leachate

Before a detailed analysis of leachate parameters with comparison tests, a preliminary examination of whole data set without any categorization into groups was conducted to see available trends of pollution parameters according to landfill age and precipitation rates. It was observed that without any grouping of data according to the specifications like landfill age or climate, graphical presentation do not provide any interpretable result other than the highly fluctuating leachate variables. Therefore, only some of the parameters were considered in evaluating the quality of leachate in this section by simple scatter charts. According to the given literature below, specified parameters were used for preliminary analysis of uncategorized leachate data as they allowed to have some sort of judgement about climate and landfill age effect.

In many studies it was stated that the amount of rainfall has a major impact on the leachate quality and quantity (Yang et al., 2015). High infiltration of rainwater causes mixing in the landfill leachate hence decreases concentrations of many components (Tsarpali et al. 2012; Vadillo et al., 1999). Conductivity is a good indicator of the total amount of soluble ions and inorganic materials in leachate and seasonal conditions have considerable impact on the conductivity values (Ettler et al., 2008; Mangimbulude et al., 2009). Since Cl is accepted as a conservative pollutant and generally used to determine the dilution factor in leachate studies (Christensen et al.,

2001; Renou et al., 2008), it would be useful to explore the variations in its concentration as well according to precipitation rates. In this context, EC and Cl were selected to analyse the precipitation impact on leachate quality.

Figure 6.4 presents EC and Cl values of leachate data with respect to annual precipitation rates in landfill locations. Two distinctive phases could be observed for EC and Cl values in the charts. Besides high fluctuations, the highest EC values in leachate were observed for sites with annual precipitation rates lower than 600 mm (arid/semi-arid). On the other hand, in places with annual precipitation rates higher than 1800 mm, lower EC values were seen indicating that dilution mechanism prevails over 1500-1800 mm annual rainfall on average (Figure 6.4a). For Cl concentrations, the same situation exists, but precipitation ranges are different in that case. Observed Cl values fluctuate to very high levels till 1200 mm annual average precipitation rates, but after that in moist sub-humid and humid (tropical) places, it decreases considerably (Figure 6.4b). In general, both EC and Cl values have a decreasing trend with precipitation rates. This outcome is consistent with many studies done on the leachate data belonging to a given landfill in different seasonal conditions like droughts and high rainfalls (Tatsi and Zouboulis, 2002; Vadillo et al., 1999). In the following sections, the impact of precipitation rates on leachate parameters are studied in detail.

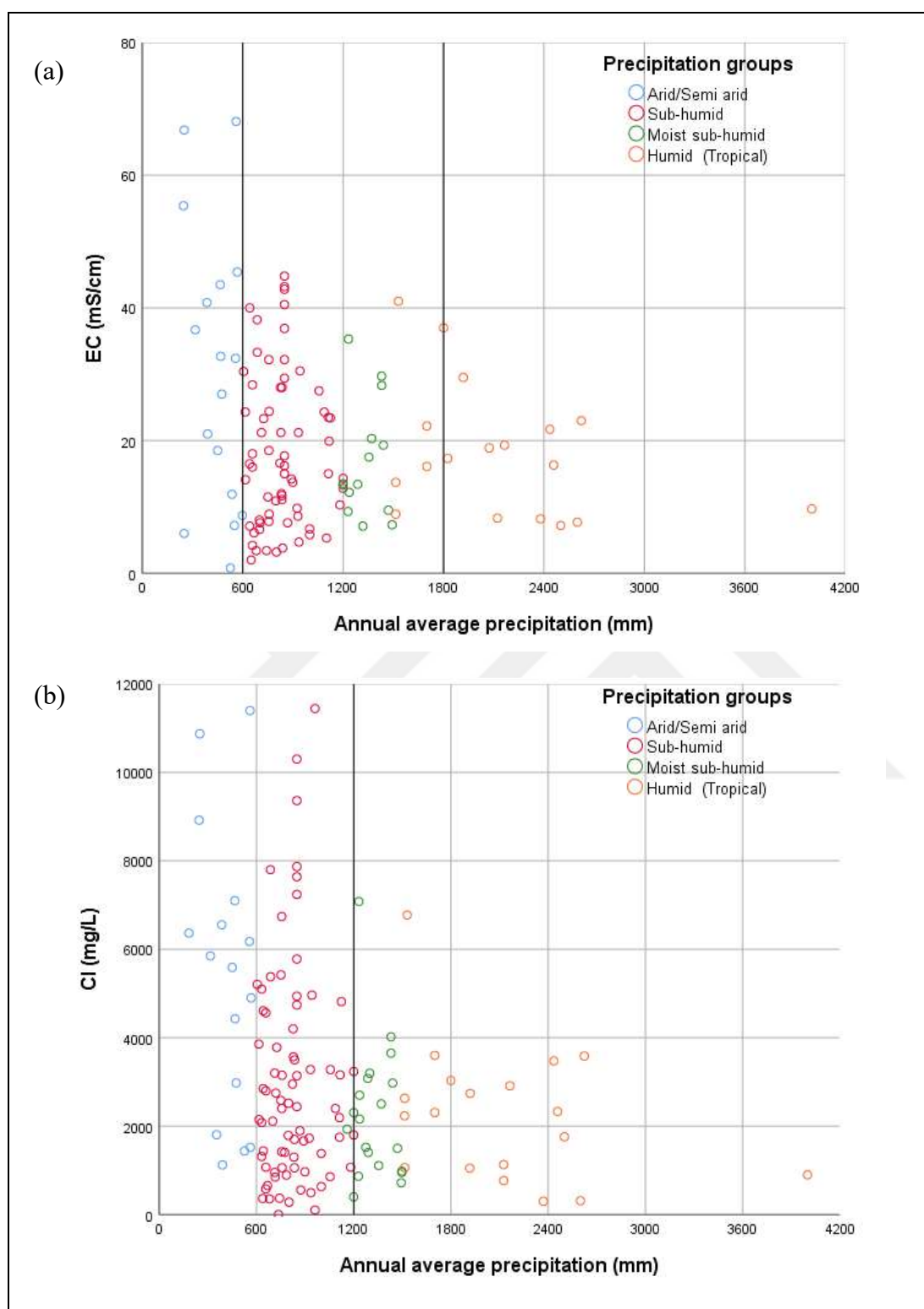


Figure 6.4. Values of leachate (a) EC and (b) Cl according to annual precipitation rates

Similar to climate effect in terms of precipitation rates on the quality of leachate, time effect in terms of landfill age was investigated by simply drawing data scatter charts. Again due to highly fluctuating character of leachate, only some parameters could be helpful to get some understanding on the impact of landfill age. In that respect, COD representing the organic content and $\text{NH}_4\text{-N}$ being an important pollutant in leachate were selected for preliminary analysis of uncategorized leachate data.

In Figure 6.5a, it can be observed that COD has a decreasing trend with landfill age. However, this decline is apparent after the first 5 years and significant in higher ages than 15 years. It means that between 5 to 15 years COD does not show a visible decreasing trend for landfills that are not operated as bioreactors. Although this issue is investigated further in the following sections, COD values get considerably lower after 15 years in many landfill leachate samples as shown for individual cases in Figure 6.5a.

$\text{NH}_4\text{-N}$ concentration in leachate fluctuates considerably with time as observed in Figure 6.5b. Only after a very long time, it can decrease to values less than 1000 mg/L. In parallel to this, Lopez et al. (2018) and Yusof et al. (2009) reported that $\text{NH}_4\text{-N}$ values do not show any declining trend with time because $\text{NH}_4\text{-N}$ remains stable due to ongoing anaerobic conditions and accumulates in the leachate with time. Chu et al. (1994) indicated that in about 10 years, ammonia nitrogen concentration reaches to around 500-1500 mg/l, and will remain at high levels for at least 50 years. As Kjeldsen et al. (2002) stated, ammonia concentrations did not show a significant decrease even 30 years after landfill closure. In the following sections the impact of landfill age on leachate parameters are studied in detail.

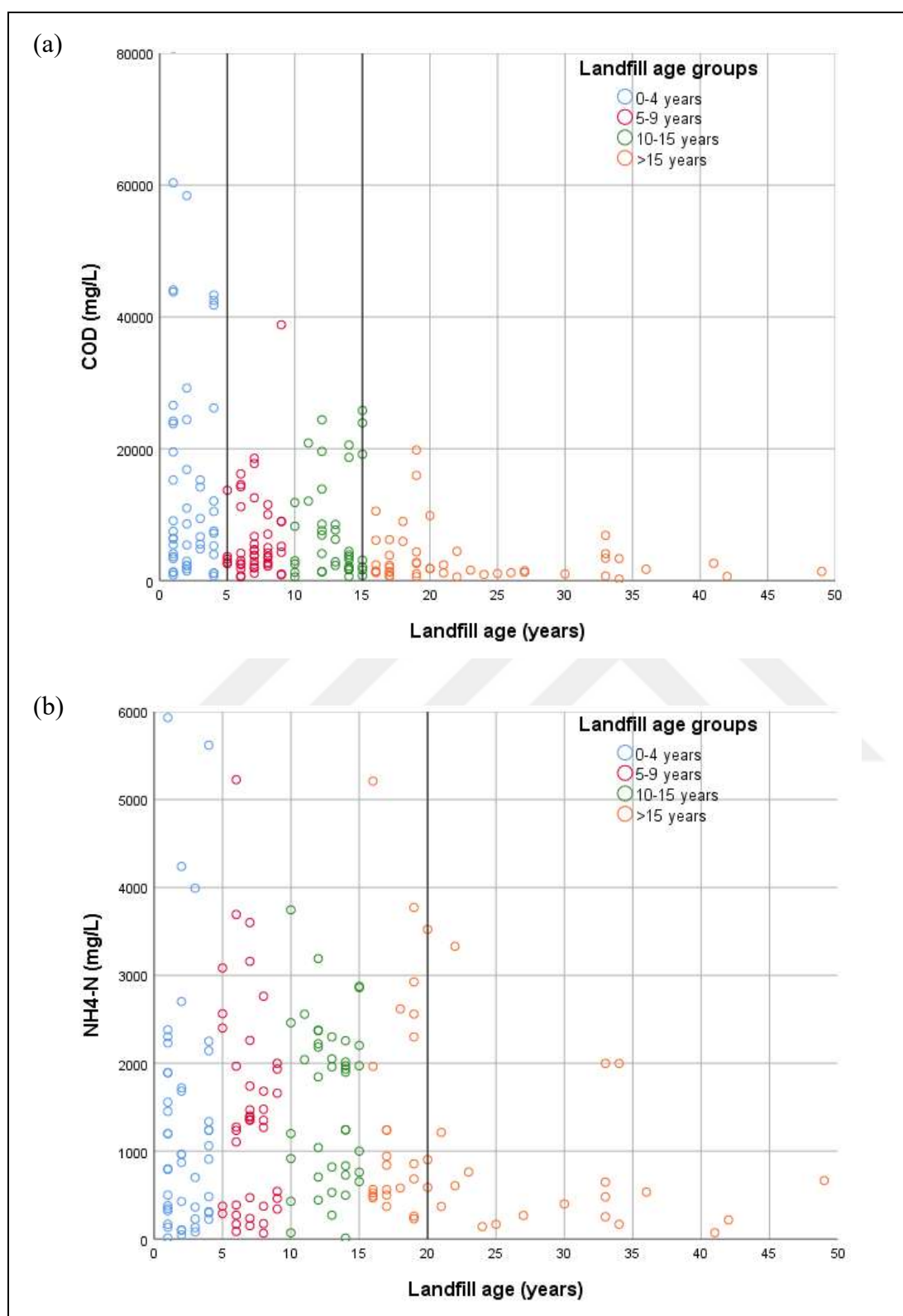


Figure 6.5. Values of leachate (a) COD and (b) NH₄-N according to the landfill age

Descriptive statistics of major physico-chemical characteristics of landfill leachate according to different landfill age and climate groups are presented in Tables 6.7 and 6.8. It could be seen that even in the same age or climate groups, most of the parameters fluctuate considerably as evidenced by high standard deviations. Moreover, a clear trend can be observed in some groups for certain parameters. For example, organics have a decreasing trend with time, whereas some inorganics (i.e., $\text{NH}_4\text{-N}$, Cl , alkalinity) do not show any specific trend with time. As Ziyang et al. (2009) stated, conductivity was found in high levels even after 10 years (Tables 6.7 and 6.8), indicating that salts are problematic in leachate over the lifetime of landfills. Regarding climate impact, besides the organic content, many other parameters are higher in climate group 1 with lower precipitation compared to climate group 2 with higher precipitation. In addition, climate group 2 has higher temperatures which may increase the rates of reactions leading to enhanced degradation and such processes. Whether all these differences are significant or not is investigated statistically in the next sections.

Table 6.7 Main physico-chemical characteristics of landfill leachate according to different landfill age groups for climate group 1

Age groups	Precipitation<1200 mm Temperature<20 Degrees															
	0-4 years				5-10 years				10-15 years				>15 years			
Parameters*	N**	Mean	Median	SD**	N	Mean	Median	SD	N	Mean	Median	SD	N	Mean	Median	SD
pH	33	7.1	7.2	0.8	30	7.7	7.9	0.8	27	8.0	8.1	0.5	38	7.8	8.0	0.7
COD	33	19688	14194	18415	30	7965	4851	7781	28	8713	4267	8398	36	3271	2053	3737
BOD	29	10838	5500	10492	26	4023	2170	5186	26	3646	931	4790	33	593	245	920
BOD/COD	29	0.49	0.50	0.15	26	0.42	0.41	0.18	26	0.32	0.32	0.22	33	0.19	0.19	0.14
NH ₄ -N	31	1381	909	1401	27	1384	1274	1301	29	1587	1934	932	36	1074	597	1211
TSS	24	1540	681	1804	18	785	406	916	13	987	650	996	10	486	131	590
TDS	14	15719	12350	12421	10	15117	16621	7310	10	16479	16182	6880	9	11686	9004	8797
Cl	21	3624	1805	3442	23	3536	2519	3047	22	3990	3850	1781	31	2492	1440	2392
EC	20	22.7	21.1	17.3	19	19.8	15.0	13.3	20	22.6	22.8	10.9	27	17.9	11.7	15.9
TA	15	6672	5823	5048	14	6596	5868	3767	18	8746	9363	3798	18	6766	3799	6811
Na	11	2596	1356	2538	13	1800	1177	1451	14	2514	2365	1374	24	1737	1255	1352
K	11	1622	1000	1618	13	1248	969	983	13	1588	1270	951	22	1076	648	1069
Fe	20	93.9	18.4	148.6	18	106.0	18.2	197.6	23	13.1	7.6	14.8	27	9.7	7.1	7.9
Ca	14	1072	430	1333	16	351	245	356	14	145	95	126	21	259	145	343
Mg	14	280	162	364	13	147	152	71	14	158	114	151	22	152	100	167
TP	20	41	29	45	15	29	17	27	11	19	11	17	10	15	8	14
SO ₄	17	542	224	721	17	187	81	266	16	290	41	510	21	226	95	367

*Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

**N represents number of data and SD standard deviation

Table 6.8 Main physico-chemical characteristics of landfill leachate according to different landfill age groups for climate group 2

	Precipitation>1200 mm Temperature>20 Degrees															
Age groups	0-4 years				5-10 years				10-15 years				>15 years			
Parameters*	N**	Mean	Median	SD**	N	Mean	Median	SD	N	Mean	Median	SD	N	Mean	Median	SD
pH	19	7.6	7.8	0.9	16	7.9	8.2	0.6	13	7.8	7.8	0.5	12	8.1	8.1	0.3
COD	18	8972	3491	15784	17	3967	2800	3680	13	5790	3000	5673	13	3689	1842	4445
BOD	17	5102	967	11379	14	815	381	1430	13	1711	396	2877	11	1518	257	2368
BOD/COD	17	0.30	0.20	0.25	14	0.15	0.14	0.08	13	0.17	0.09	0.18	12	0.20	0.16	0.21
NH ₄ -N	17	1123	962	1303	14	1530	1418	939	13	1462	1845	956	11	1205	902	965
TSS	14	326	106	413	7	1348	520	1844	3	578	480	290	9	613	271	1049
TDS	6	16756	12396	12419	9	11525	11600	5754	5	7308	5925	3959	6	8067	7630	3732
Cl	11	2381	2331	1128	11	2664	2503	1732	6	2349	1281	2418	7	2025	2234	1072
EC	9	20.2	18.9	7.3	10	21.2	19.8	10.9	6	14.6	9.2	10.7	5	11.5	9.7	5.1
TA	8	6483	6093	4258	6	7647	6794	5312	6	9479	10094	4032	5	8550	8555	3063
Na	8	1535	1377	962	5	1609	1875	672	4	1336	1367	764	-	-	-	-
K	8	1210	693	1321	4	1131	1181	623	3	956	1278	666	-	-	-	-
Fe	16	62.7	11.0	143.6	7	11.9	5.5	18.2	4	9.7	9.6	7.9	4	2.8	3.1	1.2
Ca	8	640	155	1081	3	85	38	99	3	88	43	83	2	424	424	249
Mg	7	312	248	345	3	124	158	81	3	118	86	106	2	58	58	12
TP	12	24	18	24	6	19	20	10	3	28	15	32	5	28	32	16
SO ₄	4	765	612	790	5	71	29	118	4	78	56	91	-	-	-	-

*Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

**N represents number of data and SD standard deviation

6.3.2 Comparison Analysis for Climate and Landfill Age Effect on Leachate with Two-Way ANOVA (interaction effect)

Since two different factors (climate and age) are analysed for their impact on leachate pollution parameters, interaction effect should be tested beforehand. The two-way ANOVA test was conducted for the nested groups of landfill age and climate factors presented in Table 6.9. There are 26 parameters in Table 6.9 for ANOVA analysis since TOC, TS, and TN were not included due to high missing data for those parameters. However, other parameters highly correlated with them were included such as COD, BOD, TDS, TKN, and NH₄-N. Only a limited number of leachate parameters were tested having sufficient data within all groups. It is seen that there are more than 20 data (landfills) for many parameters in the majority of the age groups, especially in the first climate group. Data is relatively low (<10) in the second climate group for many parameters. Therefore, not all the parameters given in Table 6.9 were included in two-way ANOVA. For those not tested here, comparison tests were repeated in Section 6.3.3 with one-way ANOVA.

Before conducting ANOVA, the requirement of having normal distribution in the data should be met as explained in Section 3.2.5. The check for normal distribution was done in SPSS for the raw data. Unfortunately, normal distribution could not be achieved for any of the parameters in the majority of the groups. Therefore, transformation of data into Log10 was applied and normal distribution was achieved for all of the cases after removal of one or two outliers where required. pH was used as is after removal of a few outliers to achieve normality. Two-way ANOVA was performed with pH, COD, BOD, BOD/COD ratio, NH₄-N, and Cl parameters. Only Cl parameter had less than 10 data under certain groups (Table 6.9) but it was analysed as well for representing inorganic content of leachate.

Two-way ANOVA results indicated that there is a significant main effect of both climate and landfill age ($p < 0.05$) on the pH and organic content of leachate including BOD/COD ratio (Table 6.10). Interaction effect existed for the pH, COD, and BOD parameters showing that climate and landfill age together resulted in higher difference

in those parameters between certain groups. Interaction effect of climate and landfill age on analysed leachate parameters is presented by profile plots which are the SPSS output of two-way ANOVA. Beside the profile plots, mean values of the related parameters (as antilogs taken) according to climate and landfill age groups are provided below together with case by case explanations.

Table 6.9 Number of available data for leachate parameters at each group

Climate groups	Precipitation<1200 mm temperature<20 °C				Precipitation>1200 mm temperature>20 °C			
Age groups	<5 yr	5-10 yr	10-15 yr	>15 yr	<5 yr	5-10 yr	10-15 yr	>15 yr
pH	33	30	27	38	19	16	13	12
COD	33	30	28	36	18	17	13	13
BOD	29	26	26	33	17	14	13	11
BOD/COD	29	26	26	33	17	14	13	12
NH ₄ -N	31	27	29	36	17	14	13	11
TKN	23	17	16	16	11	10	3	3
PO ₄ -P	11	12	10	14	2	3	3	-
TSS	24	18	13	10	14	7	3	9
TDS	14	10	10	9	6	9	5	6
Cl	21	23	22	31	11	11	6	7
EC	20	19	20	27	9	10	6	5
Alkalinity	15	14	18	18	8	6	6	5
Na	11	13	14	24	8	5	4	1
K	11	13	13	22	8	4	3	-
Fe	20	18	23	27	16	7	4	4
Ca	14	16	14	21	8	3	3	2
Mg	14	13	14	22	7	3	3	2
TP	20	15	11	10	12	6	3	5
SO ₄	17	17	16	21	4	5	4	-
Ni	19	17	17	21	6	3	5	1
Zn	24	19	21	24	16	8	5	3
Cu	22	15	17	21	10	5	6	1
Cr	23	17	13	19	14	6	4	1
Pb	24	18	19	21	7	6	6	1
Cd	18	18	13	20	8	6	4	1
Mn	10	11	10	20	8	3	6	1

Table 6.10 Two-way ANOVA results for the main and interaction effects between groups

Parameters tested	Climate (main effect)	Landfill age (main effect)	Interaction effect
pH	Significant (p =0.020)	Significant (p =0.001)	Significant (p =0.004)
COD	Significant (p =0.001)	Significant (p =0.000)	Significant (p =0.035)
BOD	Significant (p =0.000)	Significant (p =0.000)	Significant (p =0.015)
BOD/COD	Significant (p =0.000)	Significant (p =0.000)	Not significant (p =0.276)
NH ₄ -N	Not significant (p =0.708)	Significant (p =0.029)	Not significant (p =0.302)
Cl	Not significant (p =0.251)	Not significant (p =0.460)	Not significant (p =0.422)

All the parameters were analysed as Log10 transformations except pH
p<0.05 indicates statistically significant difference, p>0.05 no significant difference

pH value of leachate was found significantly different between both climate and landfill age groups. This difference was pronounced more in the first age group due to interaction effect (Figure 6.6a). As indicated in Figure 6.6b, leachate pH only in young landfills (<5 years) appear to be much lower than all others (age>5 years) in the first climate group. Tatsi and Zouboulis (2002) reported the same finding that as the landfill age increases over a certain period of time, pH of leachate also increases. Furthermore, pH of leachate was also found lower in low precipitation climates in comparison to high precipitation areas only for the age group of <5 years (Figure 6.6c). In general, it was stated that landfills enter into methanogenic conditions in approximately four years after closure or in 10 years after initial waste placement (Stuart and Klinck, 1998). As biological decomposition in landfills takes place mainly in two stages, leachate pH is lower in acid phase and higher in the methanogenic phase of anaerobic degradation (Ding et al., 2018). Limited moisture in landfills can delay the onset of anaerobic conditions (Stuart and Klinck, 1998). As Moody and Townsend (2017) stated due to rainfall short-circuiting in landfills and strong buffering nature of leachate, pH is not expected to change dramatically with high precipitation rates. It could be deduced that except for the first four years of landfilling, pH in leachate will be higher and constant around 7.5-8.0 regardless of climate impact for traditional operating conditions (i.e. no enhanced biodegradation). To test whether this finding is statistically significant or not, one-way ANOVA was applied and results are provided in Section 6.3.3.

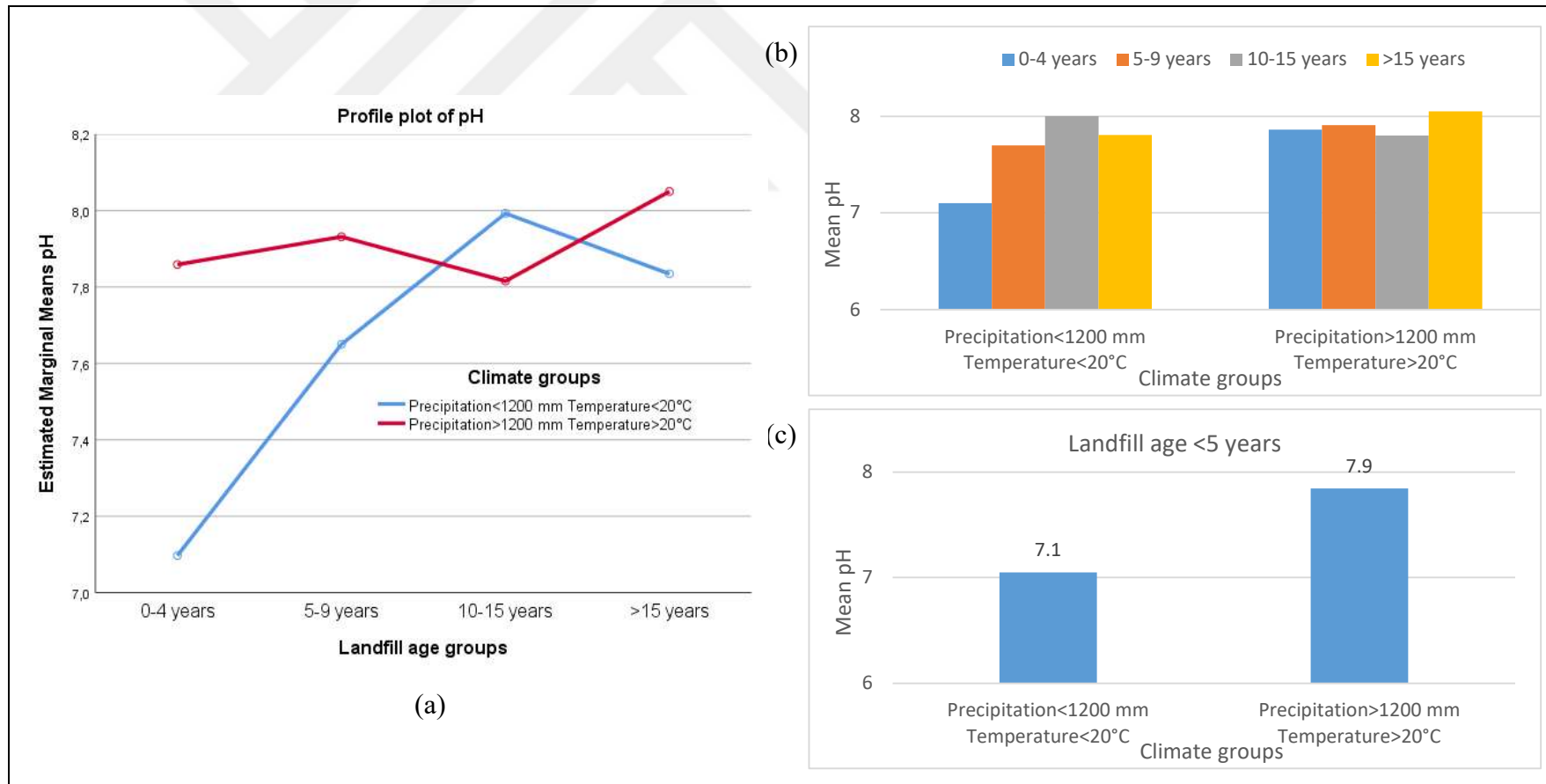


Figure 6.6. ANOVA results for leachate pH in different age/climate groups (a) Profile plot (b) Mean pH values according to climate groups (c) Mean pH values for landfill age<5 years between climate groups

For the COD and BOD cases, profile plots are given in Figures 6.7a and 6.8a, respectively. Both COD and BOD values were found significantly higher in younger leachate (<5 years) in comparison to landfills having older ages in the first climate group. Meanwhile, no significant difference with time was observed in climate group 2 (Figures 6.7b and 6.8b). Values observed are considerably higher in leachates from low precipitation areas. Tatsi and Zouboulis (2002) stated the same finding as concentrations of organics can be higher in dry climates due to limited leachate formation. This difference is more remarkable in the initial years of landfilling (<5 years) due to the interaction effect. It was observed that in low precipitation climates, both COD and BOD have a decreasing trend with time. However, this trend diminishes between ages 5 to 15 years and reappears after 15 years (Figures 6.7a and 6.8a). Paul et al. (2019) showed that physico-chemical characteristics of waste changes with the waste age. High TOC, very low nitrogen content, significantly higher microbial biomass and activity were found in aged waste. The findings of Armstrong and Rowe (1999), El-Fadel et al. (2002), and Yıldız et al. (2004) supported this conclusion. They stated that continuous dumping of new waste on top of the older ones did not increase the organic strength of leachate collected at the bottom of multi-layered waste piles. This is because older waste at the bottom acts as a bioreactor for the leachate passing through which is full of high organic matters from fresh waste parts. Therefore, after 5 or 10 years, more constant values could be obtained for parameters such as pH, COD, and BOD. Landfill age effect is analysed and discussed in detail in the Section 6.3.3.1 with one-way ANOVA.

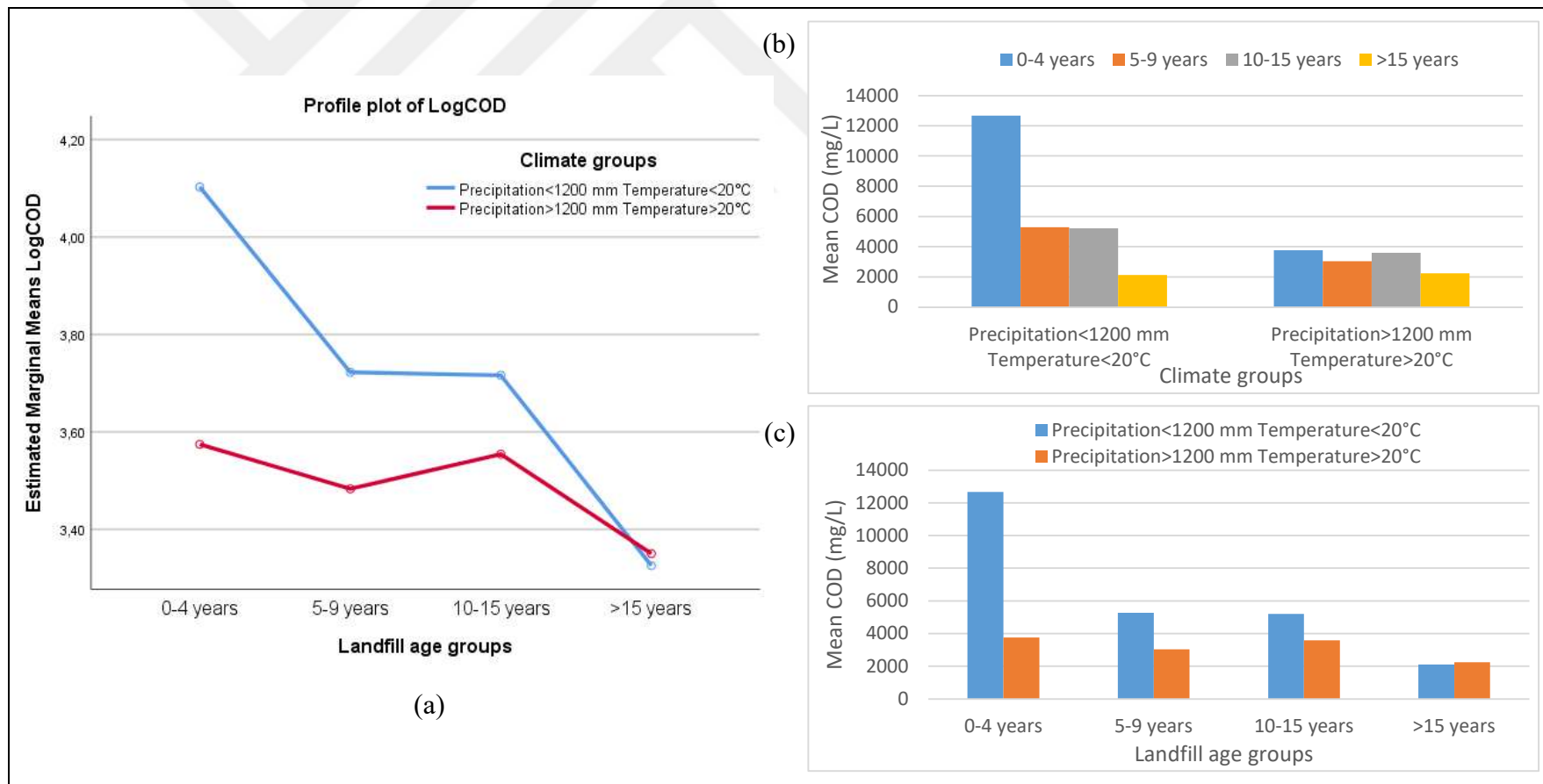


Figure 6.7. ANOVA results for leachate COD in different age/climate groups (a) Profile plot (b) Mean COD values according to climate groups (c) Mean COD values according to landfill age groups

Regarding climate factor, difference in the organic content of leachate was found statistically significant between climate groups as well. For COD, this difference is significant only in the initial years (Figure 6.7c), while for BOD it is till 15 years (Figure 6.8c). Biodegradation of waste was found very limited in high precipitation areas (Figures 6.7b and 6.8b). As Reinhart and Grosh (1998) and Bhatt et al. (2017) stated, high precipitation rates flush out both soluble organics and established microbial flora out of the landfill generating high volume of diluted leachate.

The lack of a significant trend with landfill age for organic matters in leachates of landfills in high rainfall climates indicates that biological activities and waste degradation continues in a constant manner without any time effect (Figures 6.7b and 6.8b). This is in contrast to many studies proposing considerable variation in leachate properties with time (Ehrig, 1983; El-Fadel et al., 2002; Lee et al., 2010). However, the point is that climate factor was not included in those studies. It was statistically shown here that for leachate in high rainfall and warm climate areas COD and BOD values do not decline with time and remain stable for long periods. Similarly, Reinhart and Grosh (1998) investigated leachate data from Florida landfills (heavy rainfall and warm temperatures) and stated that BOD and COD did not change with time. This is in compliance with the present study for the case of high precipitation areas. Detailed comparison of the findings with literature are provided in Section 6.3.5.

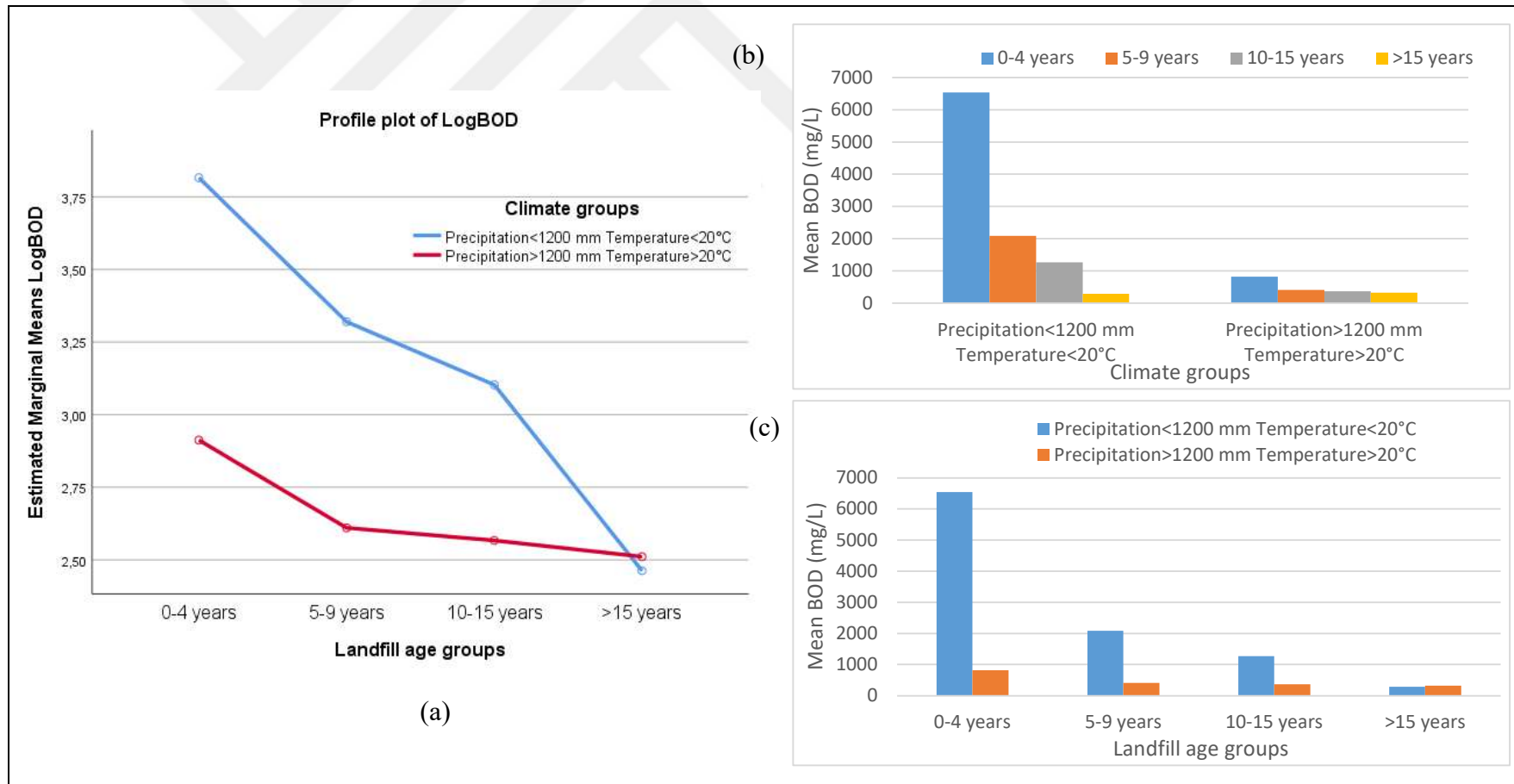


Figure 6.8. ANOVA results for leachate BOD in different age/climate groups (a) Profile plot (b) Mean BOD values according to climate groups (c) Mean BOD values according to landfill age groups

For the BOD/COD ratio showing biodegradability of leachate, two-way ANOVA found no interaction effect between climate and age groups. This means difference between BOD/COD values according to climate groups is more or less on the same range among the age groups (Figure 6.9a). BOD/COD ratio for age group of <10 years seems considerably higher than the ones having age >10 years in the first climate group (Figure 6.9b). On the other hand, BOD/COD ratio of leachate was found to be considerably lower in high precipitation and high temperature areas. It is expected that biodegradable organic components in leachate decrease as landfill age increases due to ongoing anaerobic decomposition within the waste. However, in high precipitation areas, biological activities are prohibited due to insufficient contact time of waste with microorganisms in landfills due to wash-out effect of high percolating water (Reinhart and Grosh, 1998). The wash-out effect render the expected rise in the degradability rate originating from the increased temperature in this group ineffective. Therefore, BOD/COD ratio does not exhibit significant change with time in the second climate group. This in turn indicates that leachate biodegradability is considerably low starting from the beginning of landfilling in temperate and tropical climates, whereas in dry climates, it is very high in the first 10 years and decreases considerably after that time (Figure 6.9c). It was checked from the data matrix that BOD/COD ratio is very low for majority of the landfills in climate group 2 even in their early ages.

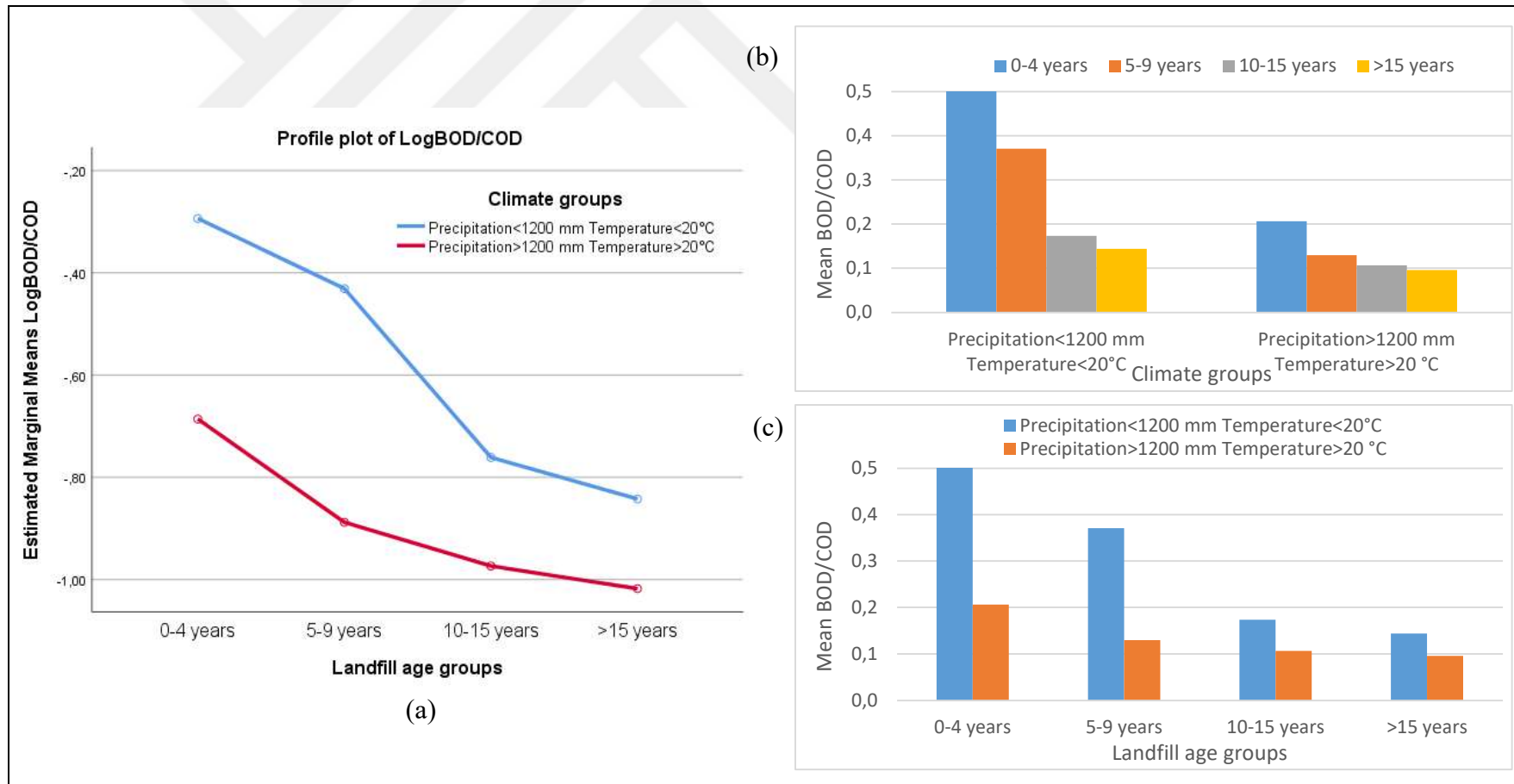


Figure 6.9. ANOVA results for leachate BOD/COD in different age/climate groups (a) Profile plot (b) Mean BOD/COD values according to climate groups (c) Mean BOD/COD values according to landfill age groups

It was stated that landfill leachate stabilization in tropical regions is achieved rapidly right after its first year of operation (Chen, 1996; Lo, 1996). Lebron et al. (2021) explained that for any given landfill age, the BOD/COD ratio of leachate in tropical climate is lower because warmer temperatures in tropical regions can enhance microbiological activity. However, this is not so if high precipitation prevails as well which is the case in many regions as high annual precipitation rates often have high annual temperatures (Table 6.4 and Figure 6.2).

Lee et al. (2010) investigated the impact of landfill age on the leachate composition from a young and old landfill site (i.e., 5 and 15 years, respectively). They found that organic matter and BOD/COD ratio decreased with landfill age. The landfill was located in a low precipitation area with low temperature as well. This is in compliance with the present study for the first climate group. Similarly, Ehrig (1983) studied 20 German landfills (low precipitation climate) for a period of 3 years. It was found that some leachate parameters like pH, COD, BOD, Fe and Ca were changing with landfill age. Except for pH, all other values were decreasing with time. On the other hand, Cl, NH₄-N, K, and Na exhibited slight increase but also high variations with time. More detailed analysis is conducted with more parameters in Section 6.3.3 by one-way ANOVA.

For the inorganic portion of leachate, Cl and NH₄-N parameters were analysed and no statistically significant differences were found between climate groups (Table 6.10). Profile plots given in Figure 6.10 indicates high difference for Cl and NH₄-N parameters between age groups of 5-10 years and >15 years. This case needs to be clarified by one-way ANOVA which can be applied by isolating the data in a specific group from other groups. This issue is detailed in Section 6.3.3.

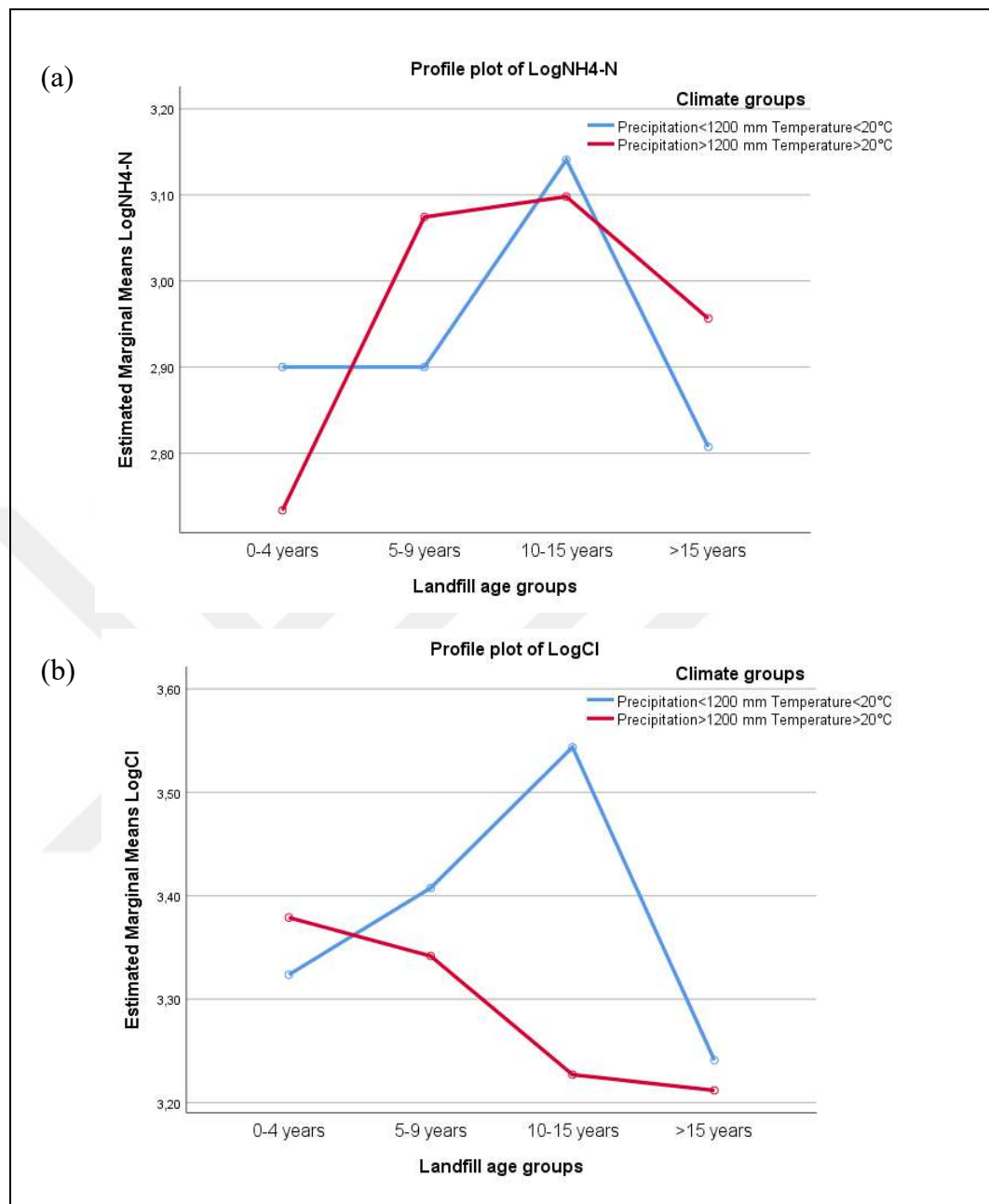


Figure 6.10. ANOVA results for different age/climate groups for (a) NH₄-N (b) Cl content of leachate

6.3.3 Comparison Analysis for Climate and Landfill Age Effect on Leachate with One-Way ANOVA (main effects)

6.3.3.1 Landfill Age Impact

Two-way ANOVA shows that some leachate parameters are affected by both landfill age and climate factors. According to the obtained results, one-way ANOVA was conducted for the selected groups (i.e., climate group 1, age group 1, etc.) for which significant differences were observed in some parameters with respect to the relevant groups.

Kjeldsen and Christophersen (2001) investigated leachate composition in a total of 106 landfills in Denmark having ages between 10-40 years. Statistical evaluations showed that the leachate constituent concentrations decreased with the age of the landfill in general. In the present study, results of two-way ANOVA, even with only few tested parameters, showed that landfill age has significant impact on the tested leachate parameters in the first climate group (low rainfall-low temperature). Therefore, all the leachate parameters provided in Table 6.9 were subjected to a comparison test within the first climate group to see the age impact. Again transformed data to Log10 was used to comply with the normal distribution criteria. For many parameters, equal number of data between groups can be observed, whereas in some groups it can not be. Welch's test and Games-Howell method as Post-Hoc test were used for non-homogeneous variances and unequal sample sizes for comparison of means (Post-Hoc, 2016). Results of the tests for the pollution parameters having statistically significant difference between landfill age groups out of 26 total parameters (Table 6.9) are presented in Table 6.11.

Table 6.11 Leachate parameters found significantly different between landfill age groups (all the samples from the first climate group)

	Landfill age group	Sample N	Means (mg/L)*	Test result**		Landfill age group	Sample N	Means (mg/L)*	Test result**
pH	<5 years	33	7.1	<5 yr is sig. lower than other age groups	TSS	<5 years	24	863	<5 yr is sig. higher than >15 yr
	5-10 years	30	7.7			5-10 years	18	401	
	10-15 years	26	8.0			10-15 years	13	552	
	>15 years	36	7.8			>15 years	10	237	
COD	<5 years	33	12666	<5 yr is sig. higher than others and >15 yr is sig. lower than others	Cl	<5 years	21	2107	10-15 yr is sig. higher than >15 yr
	5-10 years	30	5276			5-10 years	22	2556	
	10-15 years	28	5201			10-15 years	22	3497	
	>15 years	36	2116			>15 years	31	1742	
BOD	<5 years	29	6540	Same with COD	Ca	<5 years	14	418	<5 yr is sig. higher than 10- 15 yr
	5-10 years	26	2087			5-10 years	16	231	
	10-15 years	26	1266			10-15 years	14	98	
	>15 years	33	290			>15 years	21	167	
BOD/ COD	<5 years	27	0.51	<5 yr is sig. higher than >5 yr and >10 yr is sig. lower than <10 yr	Cr	<5 years	23	0.37	>15 yr is sig. lower than 5- 15 yr
	5-10 years	26	0.37			5-10 years	17	0.39	
	10-15 years	20	0.17			10-15 years	13	0.55	
	>15 years	33	0.14			>15 years	19	0.09	
NH ₄ -N	<5 years	31	794	10-15 yr is sig. higher than >15 yr	Ni	<5 years	19	0.21	>15 yr is sig. lower than 5- 15 yr
	5-10 years	27	794			5-10 years	17	0.35	
	10-15 years	28	1383			10-15 years	17	0.41	
	>15 years	36	642			>15 years	19	0.12	
TKN	<5 years	23	1088	10-15 yr is sig. higher than >15 yr	Fe	<5 years	19	26.6	<10 yr is sig. higher than >15 yr
	5-10 years	17	1310			5-10 years	18	26.5	
	10-15 years	15	2133			10-15 years	23	6.9	
	>15 years	16	794			>15 years	27	6.5	
PO ₄ -P	<5 years	10	10.5	>15 yr is sig. lower than 5-10 yr	* as antilogs were taken except pH ** “sig.” stands for “significantly” and bold values show significantly different parameters				
	5-10 years	12	15.9						
	10-15 years	10	9.0						
	>15 years	14	3.2						

As was explained in the previous section, pH, organics, and biodegradability were found significantly different between landfill ages. Confirming the aspect in Figure 6.6, pH values were found to be significantly lower only in the initial years of landfilling. The organic content of leachate has a decreasing trend with landfilling time, but it was found statistically significant only after 5 and 15 years while in between there is no major change (Figures 6.7 and 6.8). Castrillon et al. (2010) stated organic matter content of the leachate in a Spanish landfill site reached maximum values during the first years of landfilling, then decreased gradually with time. Ziyang et al. (2009) stated a similar finding that most of the chemical parameters in leachate decreased considerably in the first 4 years of landfilling.

Ding et al. (2018) indicated that methanogenic conditions can be established at the bottom of landfills after 5 years so that high strength leachate coming from fresh parts could be treated at the bottom as a result of the impact of leachate collection system. Hussein et al. (2019) stated that higher ratio of old waste compared to fresh waste or high organic acid degradation may cause lower organic content in leachate. In this regard, it could be inferred that after 5 years of landfilling, the older waste amounts will prevail in landfills so that leachate organic content could be in a stable level for longer periods of time and after 15 or 20 years stabilized waste amounts will increase further initiating the decreasing trend of organics in leachate. It should be noted also that COD measures both the organic matters and inorganic substances as oxidized by potassium dichromate solution. Therefore, exact contribution of organic or inorganic components in total COD is not known due to the complex nature of leachate. As organic matter in leachate decreases with time (Figure 6.8c), some inorganics increases (Figure 6.11), hence the share of non-carbon substances (reduced inorganics) increase in leachate COD with time (Ziyang et al., 2009) which could create the constant COD levels between ages of 5-15 years as appeared in Figure 6.7c.

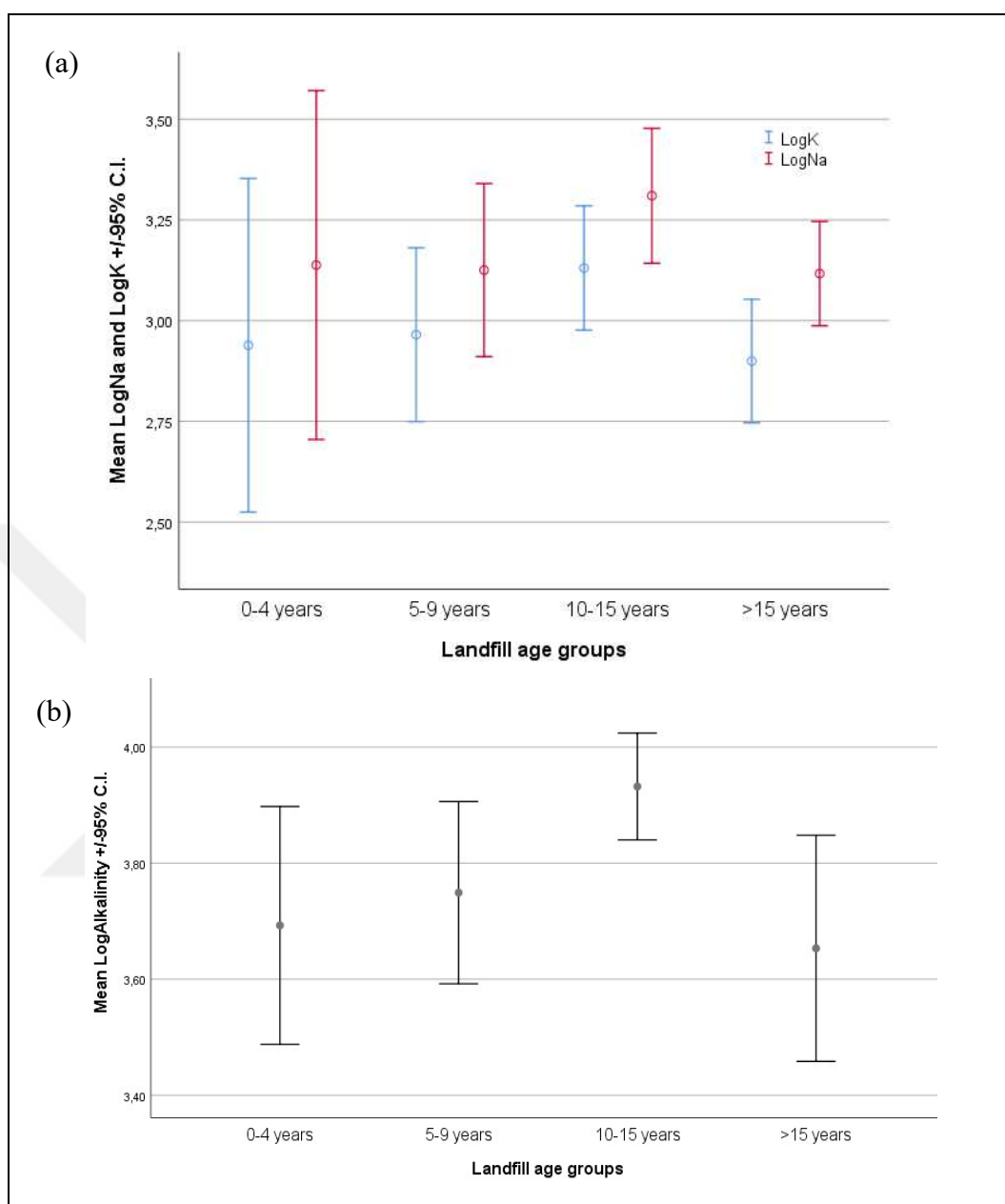


Figure 6.11. Confidence interval for mean values of (a) logK and LogNa (b) logAlkalinity at each landfill age case for the first climate group

With respect to biodegradability of leachate in terms of BOD/COD ratio, the situation was found a bit different. BOD/COD ratio decreases with time but this decrease seems more pronounced after 10 years (Figure 6.9a-blue line for climate group 1). It was also observed that after 10 years of landfilling, biodegradability of leachate becomes stable

around values of 0.1-0.3. On the other hand, in the age group 5-10 years, BOD/COD is in the range of 0.3-0.5. For younger landfills, it becomes greater than 0.5.

Nitrogen content of leachate ($\text{NH}_4\text{-N}$ and TKN) decreases significantly only after 15 years of landfilling (Figure 6.10a). In landfill ages less than 15 years, $\text{NH}_4\text{-N}$ content of leachate fluctuates considerably as observed in the scatter plot presented in Figure 6.5b. Cl concentration showed similar trend with nitrogen content of leachate. As given in Figure 6.10b, Cl concentration decreases significantly after 15 years. Similar to Cl, differences between the groups exist for other inorganics like Na, K, and alkalinity as well. However, due to the high scatter in the data, as observed from the plots of confidence intervals for the mean (Figure 6.11), they were not found statistically significant. If there is a high variation in parameter values within groups, ANOVA does not provide a significant difference, hence it is not possible to generalize the findings. This is especially relevant for inorganic content of leachate which fluctuates considerably. It could be said that majority of the inorganics in leachate increases over time in a fluctuating manner and then decreases only after about 15 years.

TSS of leachate was found as highest in the first years of landfilling and the lowest after 15 years. In between those years, TSS content of leachate has a fluctuating status. Regarding TDS content on the other hand, no statistically significant difference was found in any phase. The mass balance of TDS in leachate samples includes organic matter, ammonium, inorganic anions, metals/metalloids, and bicarbonate (Moody and Townsend, 2017). Also, results for solids in leachate analyses may not be very reliable due to its sensitivity to sampling and preservation methods (Chian and Dewalle, 1977; Kjeldsen et al., 2002). Hence comparison tests may not provide any meaning at all.

$\text{PO}_4\text{-P}$ content of leachate seems increasing in 10 years. Afterwards, it gradually decreases. Therefore, only the values between 5 to 10 were found significantly higher than the ones in >15 years. This could be due to hardly oxidizable inorganic matter rich in phosphorus so that it takes time to reach high concentrations in leachate. Also, by the metabolism of microorganisms, as well as precipitation with cations (i.e., Ca

and Mg), orthophosphates decrease with landfill age (Kapelewska et al., 2019). Regarding total phosphorus (TP), Jianguo et al. (2007) stated that anaerobic bacteria have relatively low phosphorus demand, so TP concentration in leachate could increase first and then get stable at a level of 20–30 mg/L. Nevertheless, similar to the ammonia case, there is no removal of TP in anaerobic conditions at all. As Kapelewska et al. (2019) and Ziyang et al. (2009) stated as well, TP has a decreasing trend with time, but variation between samples is too high that it revealed no significant difference in the present study.

Ca content of leachate has a decreasing trend with time but it is only significant between certain age groups. Similar to BOD and COD, Ca content of leachate was found as highest in landfills less than 5 years old for which pH values were also significantly lower. Armstrong and Rowe (1999) stated that Ca content is proportional with COD and inversely related with pH in leachate. Although high correlation exists between Ca and Mg as well (Section 5.3.3), Mg content was not found significantly different between any age groups. No specific trend was observed in its composition with time. Filho and Miguel (2017) found Ca, Mg, and Fe concentrations higher in leachate samples from experimental cells. Especially Ca concentration was found higher in the first stage where pH was low. Kjeldsen et al. (2002) stated that some of the inorganic components in leachate such as calcium, magnesium, iron, and manganese depend on the stabilization status of the landfill. They are lower in older leachate due to high pH and low organic content which they form complexes with (Ehrig, 1983).

Regarding heavy metals, only Fe, Cr, and Ni were found as significantly different between certain age groups (Table 6.11, in bold). Fe concentration was found as the highest in the first 10 years of landfilling. After that it dropped significantly. Regarding Cr and Ni concentrations, both showed slight increase with time and dropped significantly after 15 years (Figure 6.12a). This result coincides well with the Cl case in terms of having similar trends as given in Figure 6.12b. Relationship between Cl and Cr in terms of transport mechanisms in landfills is explained in Section 5.3.4.2. Anova results indicated similar patterns for Cl, Cr, and Ni co-existence in leachate

regarding formation of heavy metal complexes with inorganics. Another point was stated by Costa et al. (2019), Lo (1996), and Tatsi and Zouboulis (2002) that high degree of metal solubilization was observed due to low pH values in young landfills. Filho and Miguel (2017) studied leachate composition from experimental cells having only MSW and results showed that while high leaching of Cd, Fe, Pb, Mn, and Ca occurred in low pH stage, concentrations of Co, Zn, Ni, and Cu increased with time when the pH was above 7.0.

Paul et al. (2019) studied the change in waste characteristics in different layers of a landfill having various age conditions such as 1-8 (L1), 10-18 (L2), and 19-25 (L3) years. The studied landfill belonged to the second climate group (precipitation > 1200 mm and temperature > 20°C). The differences between the waste layers were found by analyzing mainly physical and biochemical compositions. Results showed that physico-chemical characteristics of the waste changed with the landfill age. K, Cl, and some heavy metals (Pb, Cd, and Cr) were found significantly higher in middle aged layer (10-18 years waste). In a similar study by Somani et al. (2020) regarding batch leaching tests, it was shown that TDS, sulfates, chlorides and bicarbonates were high in the water extract of mined waste. Also, significantly higher concentrations of metals were found in old waste in comparison to the fresh waste. As Öman and Junestedt (2008) stated, most of the heavy metals will remain inside the waste deposits throughout the life time of landfills since majority of them could be mobile only by sorption to inorganic and organic colloids. The ultimate conclusion of those studies is heavy metals in landfill leachate are not a major concern (Ehrig, 1983; Kjeldsen et al., 2002) but they will remain inside the landfills causing a long lasting threat for the environment. Those findings are in good agreement with the results of the present study as some inorganics and heavy metals in leachate were found increasing with time (Table 6.11). Since values for K and Pb were not found significantly different between age groups, they are not presented in Table 6.11, but their plots of confidence interval for the mean displayed a similar trend with high variation in values (Figure 6.13).

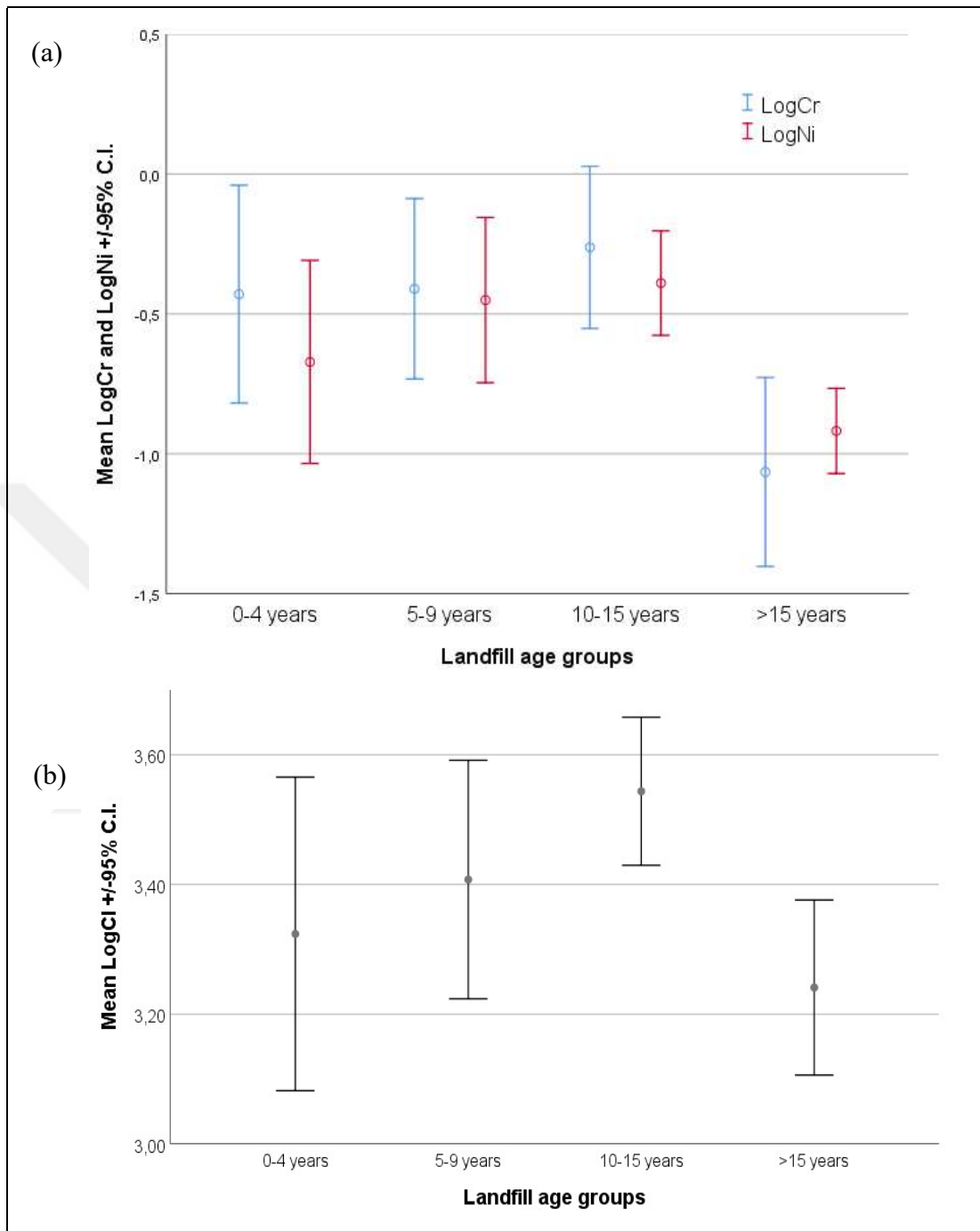


Figure 6.12. Confidence interval for mean values of (a) logCr and LogNi (b) logCl at each landfill age case for the first climate group

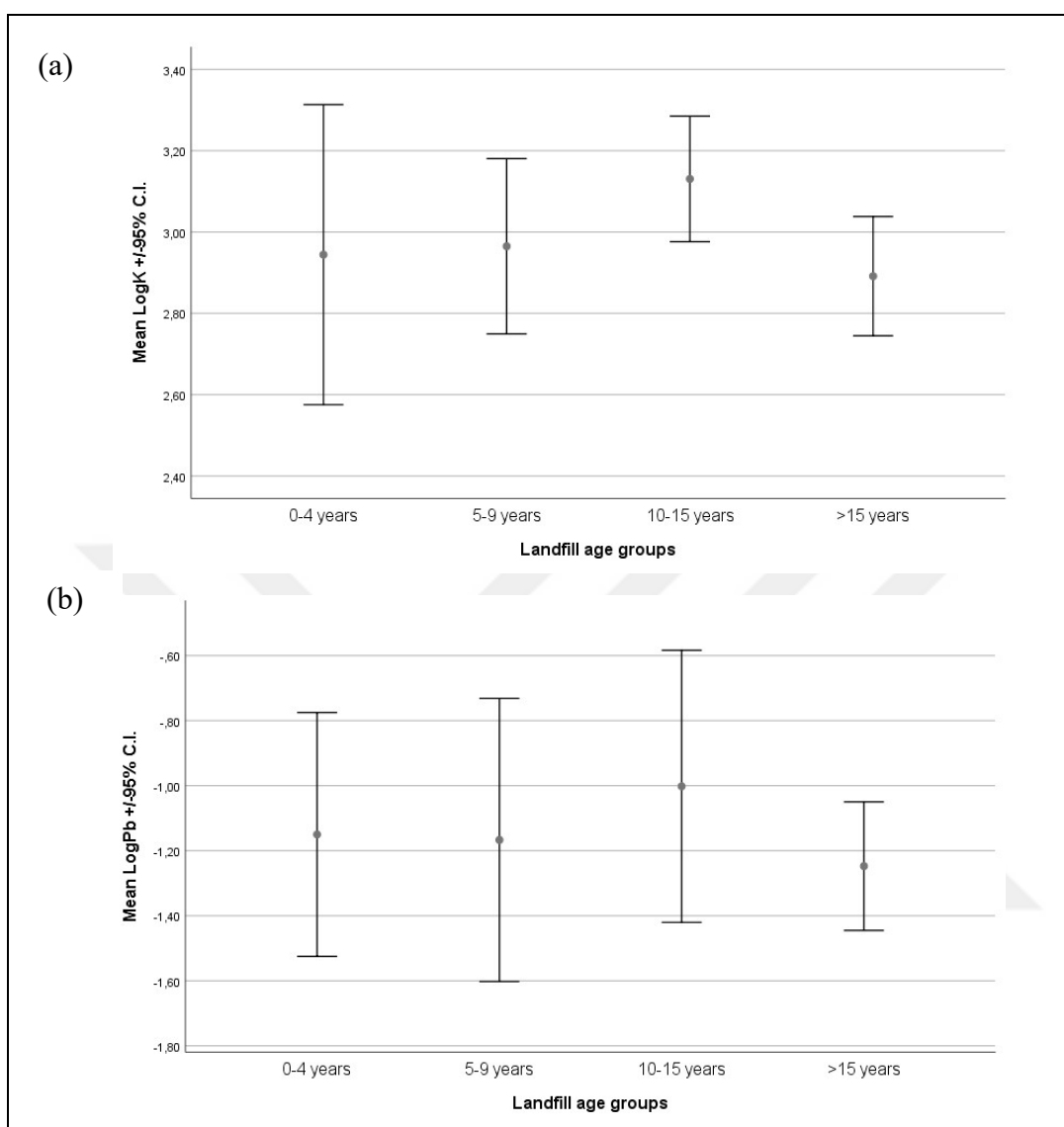


Figure 6.13. Confidence interval for mean values of (a) logK (b) logPb at each landfill age case for the first climate group

Vaccari et al. (2019) made similar analysis of leachate from both sanitary landfills and uncontrolled dumpsites. They analysed the data generally in two groups due to data scarcity. They classified landfill age into two categories as <10 years and >10 years. However, differences in leachate composition were not found as statistically significant. When compared with the results of the present study, it was found that significant difference could be observed only for landfill ages less than 5 years. Therefore, in the study of Vaccari et al. (2019), no difference was observed between

leachate parameters due to the deficiency in the data categorization (only less than/greater than 10 years). This was probably due to the limitations in the data availability for each group as faced in the present study as well for some parameters under certain groups.

Due to low number of data for certain parameters in climate group 2, one-way ANOVA was conducted only for the parameters having sufficient data ($N \geq 10$) (Table 6.9). Leachate data was isolated to include only the second climate group (high rainfall-high temperature). No statistically significant differences were found between age groups in any of the analysed parameters (pH, COD, BOD, BOD/COD, $\text{NH}_4\text{-N}$, and Cl). Results are consistent with other studies conducted in tropical or high rainfall places. Chu et al. (1994) showed that in high precipitation areas, landfills with <5 years and >10 years of age produced leachate having very similar content of organic and some inorganics. Somani et al. (2020) stated after about 2 years, the organic content of deposited waste decreases considerably and does not seem dependent on age of the landfill in India (high rainfall high temperature climate). Detailed comparisons with literature are provided in Section 6.3.5 and relevant impact flowchart is presented in Appendix E (Figure E.1).

6.3.3.2 Climate impact

Moisture condition in landfills is an important factor for hydrolysis reactions, transport of nutrients, pH buffering, dilution of inhibitory compounds, microbial activity, etc. On the other hand, excessive moisture rates can wash-out the majority of contaminants in waste in early life of a landfill. Whereas in lower moisture applications, anaerobic biodegradation is the main mechanism governing the organic content of leachate (Reinhart and Grosh, 1998). Bhatt et al. (2017) studied temperature and precipitation effect on the pH, BOD, COD, and BOD/COD in leachate from reactors including mainly organic waste. They stated that BOD and COD increase with precipitation up to some degree. Then decreases due to the dilution effect.

Komilis and Athiniotou (2014) showed the correlation between monthly precipitation rates and monthly leachate generation amounts. A significant linear correlation was found between these two parameters. In fact, the influence of climate on leachate is a complex issue. For example, in warmer climates, leachate production increases right after precipitation, as observed in a landfill case in Spain and Greece. However, in colder climates like Germany, leachate production lag behind precipitation as it occurs mostly in the form of snow (Lema et al., 1988). Furthermore, Komilis and Athiniotou (2014) found that monthly leachate generation rates were greater than the monthly infiltrating rainwater amounts into the landfill which indicates that moisture content of the highly saturated waste is also a major source of leachate in active landfills.

In this study, climate effect on leachate characteristics were investigated in two groups incorporating two individual scales for temperature and precipitation as explained in Section 6.2.1. Different parameter list was used in the analyses depending on the data availability. Leachate parameters analysed and results of one-way ANOVA for climate factors in each landfill age group are presented in Table 6.12. Except the organic content of leachate, none of the analysed parameters were found changing significantly with climate.

As the age of the landfill increases, differences among leachate parameters between climates diminishes. As given in Table 6.12, the highest number of parameters changing significantly between climates were found in the first years of landfilling as pH, organic content, solids, and biodegradability. Similarly, interaction effect was found for pH, COD, and BOD parameters in two-way ANOVA (Table 6.10). For those parameters difference between climate groups was found higher in the first age group (<5 years). Mangimbulude et al. (2009) stated that alkalinity of leachate is very high, and not significantly influenced by climate and neither Fe concentration differ significantly between low and high precipitations. The same result was obtained in this study for all the heavy metals in younger landfills (Table 6.13). Alkalinity could not be compared between climates due to low number of data for that parameter in high precipitation climate (Table 6.9).

Table 6.12 Leachate parameters analysed and results of ANOVA for climate factor

Age groups	Mean age*	Parameters tested**	Results of one-way ANOVA between climate groups
<5 years	2.3 years	pH, COD, BOD, BOD/COD, TSS, TKN, NH ₄ -N, Cl, EC, Fe, TP, Zn, Cu, Cr	pH, BOD, COD, BOD/COD, and TSS are significantly different between climate groups
5-10 years	7.0 years	pH, COD, BOD, BOD/COD, TKN, TDS, NH ₄ -N, Cl, EC	BOD, COD, BOD/COD are significantly different between climate groups
10-15 years	12.8 years	pH, COD, BOD, BOD/COD, NH ₄ -N	BOD is significantly different between climate groups
>15 years	23.7 years	pH, COD, BOD, BOD/COD, NH ₄ -N, TSS	None of the analysed parameters are significantly different between climate groups

* Mean age of the landfills in that group

** All the parameters were analysed as Log10 transformations except pH

Rainwater that percolates through the waste layers in high precipitation areas will extract soluble compounds and produce a high volume of diluted leachate (Chu et al., 1994). Vadillo et al. (1999) stated that lowest alkalinity, Cl, Na, K, NH₄-N, and EC values were observed in high rainfalls. Infact, concentration of those parameters were found only a little bit higher or lower depending on the age groups in high precipitation areas in this study without any significancy (Table 6.13). This shows that opposite to organic content, higher levels of inorganic components in leachate could be observed in climate group 2 landfills due to mainly wash-out effect causing high loadings of leachate in the short/medium term (Chen, 1996). Therefore, dilution effect may not be relevant for inorganics always as more precipitation could result in more leaching of inorganics into the leachate as seen from Table 6.13 (i.e., Cl and EC).

Table 6.13 Concentrations of parameters tested between age and climate groups

Parameters	<5 year		5-10 year		10-15 year		>15 year	
	Clm 1	Clm 2	Clm 1	Clm 2	Clm 1	Clm 2	Clm 1	Clm 2
pH	7.1	7.9	7.7*	7.9*	8.0*	7.8*	7.8*	8.1*
COD	12666	3754	5276	3041	5201*	3583*	2116*	2240*
BOD	6540	817	2087	407	1266	369	290*	324*
BOD/ COD	0.51	0.21	0.37	0.13	0.17*	0.11*	0.14*	0.10*
NH ₄ -N	794*	541*	794*	1187*	1383*	1253*	642*	904*
TKN	1088*	893*	1310*	2046*	NT	NT	NT	NT
TDS	NT	NT	13191*	9889*	NT	NT	NT	NT
TSS	863	146	NT	NT	NT	NT	237*	244*
Cl	2107*	2393*	2556*	2196*	NT	NT	NT	NT
Na	1372*	1134*	NT	NT	NT	NT	NT	NT
K	880*	717*	NT	NT	NT	NT	NT	NT
EC	16.1*	19.3*	15.8*	18.4*	NT	NT	NT	NT
TA	4929*	4732*	NT	NT	NT	NT	NT	NT
TP	20.1*	14.0*	NT	NT	NT	NT	NT	NT
Fe	26.6*	15.7*	NT	NT	NT	NT	NT	NT
Zn	0.90*	1.15*	NT	NT	NT	NT	NT	NT
Cu	0.14*	0.13*	NT	NT	NT	NT	NT	NT
Cr	0.37*	0.24*	NT	NT	NT	NT	NT	NT

Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

Clm 1: precipitation rate<1200 mm with temperature<20°C

Clm 2: precipitation rate>1200 mm with temperature>20°C

NT: Not tested due to low number of data

* Not significantly different. Bold ones are significantly different.

Confirming the aspect in Figure 6.6c, pH values were found to be significantly different between climate groups only in the initial years of landfilling. Organic content of leachate was found significantly higher in low precipitation areas (Figure 6.14). COD and BOD/COD ratio were found significantly different between climate groups only in the first 10 years while BOD was found so till 15 years of landfilling.

In fact, climate impact disappears gradually with time and does not create any big difference in leachate composition after about 10 years of landfilling (Table 6.13, in bold). Relevant impact flowchart is presented in Appendix E (Figure E.2).

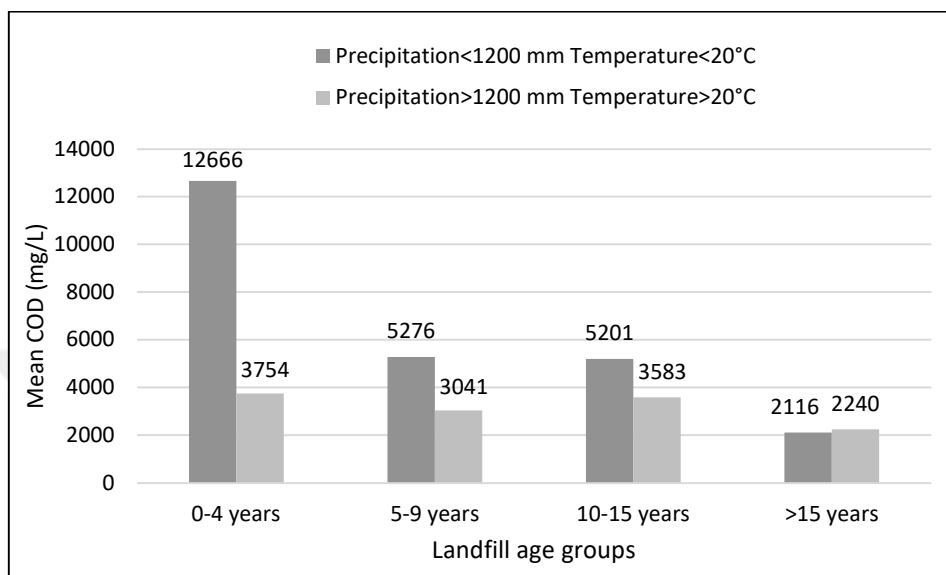


Figure 6.14. Trends of organics in leachate between different climate&age groups

6.3.4 Comparison Analysis for Development Status Effect on Leachate

Only leachate samples taken from sanitary landfills were analysed in this study, so more data exists in developed countries (H group) as seen in Table 6.14 than developing countries (UM group) where most of the waste goes to the uncontrolled (open) dumpsites (Mmereki et al., 2016; Vaccari et al., 2019).

Table 6.14 Number of data for leachate pollution parameters at each group

	High income (H) (developed countries)		Upper middle income (UM) (developing countries)	
	<10 years	>10 years	<10 years	>10 years
pH	34	40	22	13
COD	36	39	23	14
BOD	34	37	18	12
BOD/COD	34	37	18	12
NH ₄ -N	32	40	23	14
TKN	22	14	14	8
PO ₄ -P	16	14	7	5
TP	20	9	14	10
TSS	24	12	15	7
TDS	16	6	7	6
Cl	27	37	14	9
EC	22	31	14	8
Alkalinity	18	23	11	10
Na	19	28	4	3
K	19	28	4	3
Fe	25	36	10	8
Ca	23	25	5	4
Mg	20	27	5	3
SO ₄	22	27	10	6
Ni	22	26	8	7
Zn	23	31	12	8
Cu	21	25	11	8
Cr	22	22	11	6
Pb	23	27	13	8
Cd	23	24	9	6
Mn	17	24	3	4

Due to having two different factors (landfill age and development status), interaction effect was tested first to compare the leachate characteristics. Two-way ANOVA results are provided in Table 6.15 for the impact of development status and landfill age on the tested leachate parameters. Interaction effect was observed only for COD, BOD,

and NH₄-N parameters. As seen from the profile plots in Figure 6.15, difference between the concentrations of leachate parameters in developed and developing country landfills is much higher in old aged landfills (>10 years). Concentrations of NH₄-N and alkalinity were found higher in developing country landfills in both age groups. The decrease in organic content of leachate with time is more remarkable in developed countries. This could be due to lower organic content in their waste in comparison to developing countries (Mmereki et al., 2016; Vaccari et al., 2018). Therefore, organics in waste will be depleted in a shorter period of time in developed countries.

Table 6.15 Two-way ANOVA results for the main and interaction effects of development status and age groups on leachate parameters (Sample N $\geq$ 10)

Parameters tested	Development status (main effect)	Landfill age (main effect)	Interaction effect
pH	Significant (p =0.020)	Significant (p =0.001)	Not Significant (p=0.822)
COD	Significant (p =0.001)	Significant (p =0.019)	Significant (p =0.015)
BOD	Significant (p =0.000)	Significant (p =0.000)	Significant (p =0.009)
BOD/COD	Not significant (p =0.262)	Significant (p =0.000)	Not significant (p =0.153)
NH ₄ -N	Significant (p =0.000)	Not significant (p =0.263)	Significant (p =0.049)
Alkalinity	Significant (p =0.001)	Not significant (p =0.332)	Not significant (p =0.090)

All the parameters were analysed as Log10 transformations except pH and BOD/COD ratio
p<0.05 indicates statistically significant difference, p>0.05 no significant difference

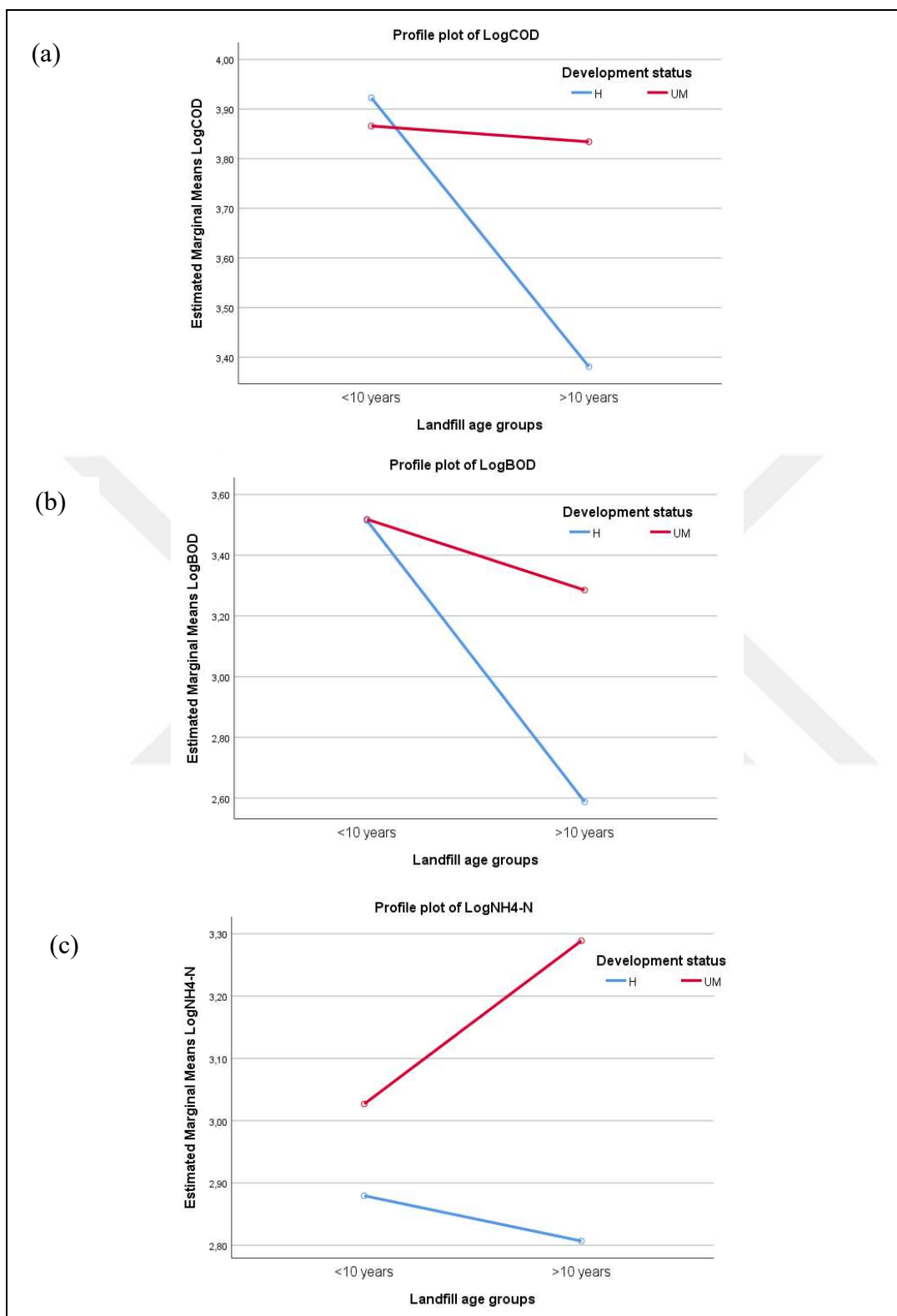


Figure 6.15. Interaction effect of development status and landfill age on leachate parameters of (a) COD (b) BOD (c) $\text{NH}_4\text{-N}$

One-way ANOVA was conducted separately for individual groups for detailed analysis and results are provided in Table 6.16. Due to limitations in the number of available data in some parameters especially for the case of developing country landfills (UM) for landfill age >10 years, the analysis focused only on limited number of parameters having sufficient data for that case ($N \geq 10$) such as pH, COD, BOD, BOD/COD, $\text{NH}_4\text{-N}$, alkalinity, and TP (Table 6.14 right-hand side).

Table 6.16 Comparison test results for the main effects of development status and age groups (Sample $N \geq 10$)

Development status	Parameters tested*	Parameters significantly different between younger (<10 yr) and older (>10 yr) landfills
H	pH, COD, BOD, BOD/COD, TKN, $\text{NH}_4\text{-N}$, alkalinity, TSS, Cl, EC, Na, K, Fe, Ca, Mg, SO_4 , Ni, Cd, Pb, Cu, Cr, Zn, Mn	pH, BOD, COD, BOD/COD, Ca, TSS, Fe, Zn, Cu, Cd, Cr
UM	pH, COD, BOD, BOD/COD, TP, $\text{NH}_4\text{-N}$, alkalinity	pH, $\text{NH}_4\text{-N}$
Landfill age	Parameters tested*	Parameters significantly different between developed (H) and developing (UM) country landfills
<10 years	pH, COD, BOD, BOD/COD, TSS, $\text{NH}_4\text{-N}$, TKN, Cl, EC, alkalinity, TP, SO_4 , Fe, Pb, Cu, Cr, Zn	Fe, Zn, Cu
>10 years	pH, COD, BOD, BOD/COD, $\text{NH}_4\text{-N}$, alkalinity	pH, COD, BOD, $\text{NH}_4\text{-N}$, alkalinity

*All the parameters were analysed as Log10 transformations except pH and BOD/COD ratio

ANOVA results for the landfill age effect for developed (H) and developing country (UM) landfills are given on the upper part of Table 6.16. It was found that only pH and ammonia content of leachate was found significantly different with time (higher in older landfills) for developing country landfills. For the case of developed countries, more parameters were tested and mainly pH, organic content, and some heavy metals were found changing significantly between younger and older landfills. Vaccari et al. (2019) stated that in developing countries, most of the pollutant concentrations were

reported as higher in landfills greater than 10 years old while BOD levels, as well as Cu, Cr, and Zn concentrations were higher in the landfills less than 10 years old. However, those differences were not found as statistically significant in their study. This finding is flawed as it ignores the climate effect. As presented in previous sections, climate has considerable impact on leachate quality and should be taken into account during analysis by treating the data accordingly (i.e., analysing the landfill age effect in the same climate group).

To evaluate the development status effect, younger and older landfills were analysed case by case with one-way ANOVA and results are given on the lower part of Table 6.16. Although many parameters were tested especially for younger landfills, only some heavy metals (Fe, Zn, and Cu) were found significantly different between developed and developing country landfills. For the older landfills more parameters became significantly different between H and UM countries such as pH, organics, NH₄-N, and alkalinity. Mean concentrations of parameters showing significant difference between groups are presented (in bold) in Table 6.17.

Rajoo et al. (2020) stated that leachate pH in landfills of developing countries is mostly neutral whereas it can be lower in developed countries. Results of the present study provided a similar finding that pH of leachate in UM countries is higher than H country landfills for both age groups (Table 6.17). Ma et al. (2022) stated that COD, BOD, and NH₄-N concentrations in leachates of high income (H) countries were much lower than those in upper middle income (UM) countries. For example, COD at young landfill leachate in UM countries were twice as high as than H country landfills. This statement coincides well with the findings of the present study only that this difference was found significant in older landfills (Table 6.17 right-hand side). Mmereki et al. (2016) stated that developing countries have rich organics and moisture content in their waste streams compared to developed countries. Because of that organics in their leachate (UM group) would be higher for longer periods of time than in leachates of developed countries. Consequently, the more fraction of organic waste in UM landfills the higher organic content in their leachate, hence it will take more time to have lower concentrations in their leachates (Table 6.17).

Table 6.17 Mean concentrations of leachate parameters between impact groups

	Landfill age effect				Development status effect			
	H		UM		<10 year		>10 year	
	<10 yr	>10 yr	<10 yr	>10 yr	H	UM	H	UM
pH	7.3	7.8	7.6	8.2	7.3*	7.6*	7.8	8.2
COD	5915	2118	7344*	7503*	5915*	7344*	2118	7503
BOD	2429	312	3297*	2210*	2429*	3297*	312	2210
BOD/COD	0.45	0.23	0.43*	0.33*	0.45*	0.43*	0.23*	0.33*
NH ₄ -N	758*	641*	1064	1944	758*	1064*	641	1944
TSS	502	215	NT	NT	502*	537*	NT	NT
Cl	2096*	1684*	NT	NT	2096*	3192*	NT	NT
EC	14.8*	11.6*	NT	NT	14.8*	15.7*	NT	NT
TA	4744*	4147*	6566*	10681*	4744*	6566*	4147	10681
TP	NT	NT	12*	16*	21*	12*	NT	NT
Ca	258	137	NT	NT	NT	NT	NT	NT
Fe	52	7	NT	NT	52	7	NT	NT
Zn	1.66	0.34	NT	NT	1.66	0.31	NT	NT
Cu	0.31	0.08	NT	NT	0.31	0.04	NT	NT
Cr	0.52	0.11	NT	NT	0.52*	0.35*	NT	NT
Cd	0.01*	0.01*	NT	NT	0.01*	0.01*	NT	NT
Pb	0.12*	0.04*	NT	NT	0.12*	0.07*	NT	NT
Ni	0.29*	0.20*	NT	NT	NT	NT	NT	NT
Mn	1.67*	0.79*	NT	NT	NT	NT	NT	NT

Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

NT: Not tested by ANOVA due to low number of data

*Not significantly different. Bold ones are significantly different

In the leachates of developed country landfills, many heavy metals (Fe, Zn, Cu, and Cr) were found significantly different between younger (higher concentrations) and older landfills (Table 6.17). On the contrary, not any big difference was observed for Mn, Cd, Ni, and Pb content. Due to low number of data for the age group >10 years in UM group (Table 6.14), heavy metals (Fe, Ni, Cd, Pb, Cu, Cr, Zn) were analysed with non-parametric Mann-Whitney U test. Results showed no significant difference between age groups in developing countries. Also, heavy metals in landfills >10 years for both developing and developed country landfills were tested with Mann-Whitney U method and no significant differences were found except Pb content (more than three times high in developing countries).

6.3.5 Discussion and Comparison of the Results with Literature

6.3.5.1 Organic Content

Mohammad-pajooch et al. (2017) explained the increase in COD and $\text{NH}_4\text{-N}$ concentrations in leachate as the increase in monthly ambient temperatures. Costa et al. (2019) stated that, the tropical climate in Brazil having high temperatures and precipitation rates, enhance the waste degradation process and leachate formation. On the contrary, Lo (1996) reported that organic matters in the waste pass through the leachate without sufficient exposure to biological activities due to excessive percolating water in Hong Kong landfills. In spite of the positive effect of high temperatures into microbiological degradation processes in landfills, accompanying high precipitation rates could negatively impact biodegradation of waste.

BOD, COD, BOD/COD ratio, and pH can be accepted as indicators of organic pollution in leachate as well as microbial activities in landfills (Bhatt et al., 2017; Moody and Townsend, 2017). Organic compounds decrease in concentration by joined effects of leaching/wash-out and biodegradation processes in landfills (Reinhart and Grosh, 1998; Ziyang et al., 2009). The most important factor affecting the rate of waste decomposition is the moisture content. Moisture content is important because it is a

critical parameter for microbial degradation and therefore has great impact on stabilization rates within the landfill (Bhatt et al., 2017; Kjeldsen et al., 2002). Reduction of the rainwater percolating through the waste decreases the water input for the biological, chemical, and physical processes in the landfills. Therefore, dry landfills could have slow stabilization rates due to lower moisture content to promote biological activities. On the other hand, high rainfall rates result in considerable infiltration through the waste that can flush soluble organics and attached microorganisms out of the landfill (Bhatt et al., 2017; Reinhart and Grosh, 1998). Excessive rainwater input into landfilled waste will generate high volume of dilute leachate by dissolving and extracting many compounds. On the other hand, in dry climates the concentrations of certain parameters like BOD, COD, $\text{NH}_4\text{-N}$ can be higher (Tatsi and Zouboulis, 2002).

In high rainfall areas, two main mechanisms affect the leachate quality. First is the dilution mechanism with high amounts of percolating water that decrease the concentrations of leachate pollutants. The second one is the wash-out effect that could cause high loadings of leachate in the short term (Chen, 1996). However, in the long term leachate parameters could be much lower in high precipitation areas. On the contrary, in dry climates concentrations of some leachate parameters were found significantly higher due to decreased leachate flows with high pollutants. Also, in dry climates many leachate parameters remain in high values for longer periods (10-15 years) due to scarcity of moisture for prolonged biological activities. Scatter plot of leachate data for the low precipitation climate group supports this hypothesis in a way that leachate parameters (i.e., COD) have higher values for longer periods of time as given in Figure 6.16a showing that degradation of waste could last longer in low precipitation areas. On the contrary, in high precipitation climates COD is found in much lower concentrations without any change with landfill age (Figure 6.16b). Those were all justified with the results of detailed statistical analyses presented in preceding sections.

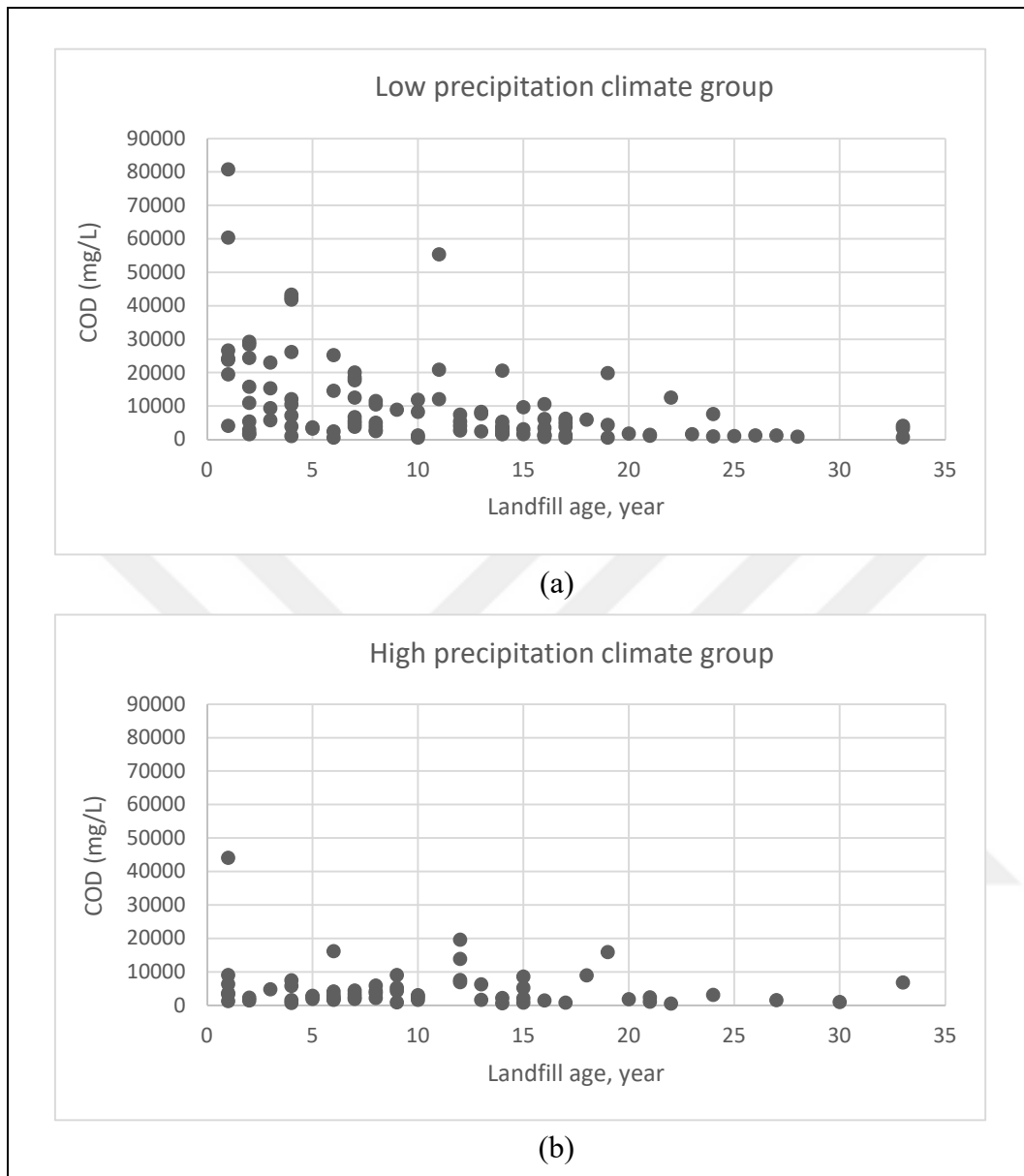


Figure 6.16. Scatter plots for the change in leachate COD content with time in (a) Low precipitation climate (b) High precipitation climate

Some authors compared their leachate samples in similar aged landfills and found different compositions. For example, Chu et al. (1994) could not explain why the young landfill leachate (<5 years) showed similar properties with old landfill leachate (>10 years). Or why methanogenic conditions are established very quickly in the studied landfills. In some references it is stated that methanogenic conditions are established very quickly in Hong Kong and Poland landfills so that very low COD

values were observed even after one year of landfill operation (Kulikowska and Klimiuk, 2008; Lo, 1996; Robinson, 2005). Chen (1996) studied the effects of landfill age and rainfall on landfill leachate in Taiwan having 2500 mm average rainfall annually. According to the results of their analysis, leachate maturation time was found far below the literature values. Bhatt et al. (2016) stated that medium temperature with low precipitation rates caused concentrated leachate. They showed that increasing water input and high temperature rates result in faster biodegradation of waste hence higher BOD and COD concentrations in leachate. From the results of this study and other examples already mentioned, the reason could be stated as the climate factor, mainly precipitation rates. Statistical tests showed that in high precipitation areas having also high temperatures, organic content of leachate and BOD/COD ratio is significantly lower. On the other hand, at landfills in arid regions or lower precipitation areas, waste remains dry due to little infiltration of rainfall, hence acidic phase or early methane production phase would be observed for a longer period of time (Kjeldsen et al., 2002). Therefore, it is possible that young leachate from a tropical region would show similar properties (i.e., low organic content) with the old leachate from arid/semi arid regions which was emerged in PCA results as well (Figure 5.9). This is the effect of dilution and/or wash-out contrary to what was thought such as rapid establishment of methanogenic conditions or quick maturation time because a significant lag time should occur prior to the start of the degradation process, especially the methanogenesis (Bhatt et al., 2017).

Regarding development status impact on landfill leachate, Ma et al. (2022) stated that the proportion of organics in waste stream can significantly impact the concentrations of COD and BOD in leachate. It was apparent in this study as well that both COD and BOD values were found higher in the leachates of developing countries (Table 6.17).

6.3.5.2 Inorganic Content

As stated above, the stabilization mechanisms for inorganic and organic components in the landfills are different. Concentration of inorganics in leachate will depend on the dissolution rate, moisture, and the dilution rate which are directly related to rainfall intrusion into the waste. Kjeldsen et al. (2002) stated that some inorganic components like Cl, Na, and K are removed from waste due to wash-out effect mostly, since leaching by sorption, complexation, and precipitation have minor importance.

As said, inorganics are removed from waste as a result of wash-out or leaching by infiltrating rainwater (Chu et al., 1994). Cl for instance is an inert substance so that its transport is mainly controlled by the fluid flow in the landfill. Highly soluble Cl will be transferred to the leachate with continuous fresh waste addition in the active landfills (Statom et al., 2004). Therefore, Cl leaching from waste will last until all the Cl is dissolved (Yıldız et al, 2004). In some studies, Cl concentrations were found as increasing with leachate age (Chu et al., 1994). On the contrary, Christensen et al. (2001) and Lee et al. (2010) reported no change for Na and Cl in young and old landfill leachates. In the present investigation Cl content of leachate samples had increasing trend with time only in the first climate group and after a long period of landfilling (i.e., 15 years) Cl tends to decrease (Figure 6.12b). Similarly, Ehrig (1983) did not state any decreasing trend for leachate Cl up to 20 years of leaching. Therefore, those studies showing a landfill age effect on Cl content of leachate and those proposing the opposite views are actually correct if the climate factor is taken into account in the statements. Statistical results demonstrated that Cl content changes with time, but only in the low precipitation climates, whereas it does not show any clear trend with time and generally remains stable in high precipitation climates. As observed from the data of Florida landfills by Reinhart and Grosh (1998), Cl concentration in leachate will be relatively stable throughout the life of the landfill. According to Ma et al. (2022) as well concentrations of $\text{NH}_4\text{-N}$ and inorganics (K, Cl, SO_4) are not changing significantly with landfill age. For other inorganics like Ca and Fe, analysis results of

present study showed that their concentrations were found significantly lower after 5 and 10 years, respectively only in the first climate group (Table 6.11, in bold).

Ammonia is released from the waste mainly by decomposition of proteins in organic matters. The only mechanism for ammonia removal in landfills is leaching, because it cannot degrade further under anaerobic conditions in the waste. Therefore, ammonia concentration will remain high even in the leachate of older landfills that are low in organic content (Kjeldsen et al., 2002). For that reason, it can be expected that there is no age effect on the ammonia content of leachate by biodegradation. This was revealed so by the results of statistical analysis in the present study (Table 6.11). Chu et al. (1994) on the other hand stated that high values of ammonia was observed in old leachate. Lee et al. (2010) reported that $\text{NH}_4\text{-N}$ concentration had no decreasing trend with time and remained high. Zhao et al. (2019) analysed young (<5 years) and old (>20 years) landfill leachates in the same area having similar climate conditions (precipitation<1200 mm and temperature<20 °C) and showed that ammonia content of leachate increased with time. Similar trends were obtained in the present study as provided in Figure 6.17 with only one difference that no significant change could be attained between groups regarding $\text{NH}_4\text{-N}$ concentration due to the fact that it fluctuates considerably. It is clear that $\text{NH}_4\text{-N}$ concentrations in leachate are more or less similar for both climate groups and there is no significant difference between landfill age groups.

Pivato and Gaspari (2006) explained the reason of ammonia release over long periods of time in landfill leachate as follows. Ammonium ion (NH_4^+) is adsorbed on negatively charged waste particles and some part of it is released with leachate during the acetogenic phase by interchange between H^+ and NH_4^+ ions. This also happens with other ions like Ca and K. High correlation between K and $\text{NH}_4\text{-N}$ (Table 5.5) could be explained by this. They both release in leachate with the same mechanisms.

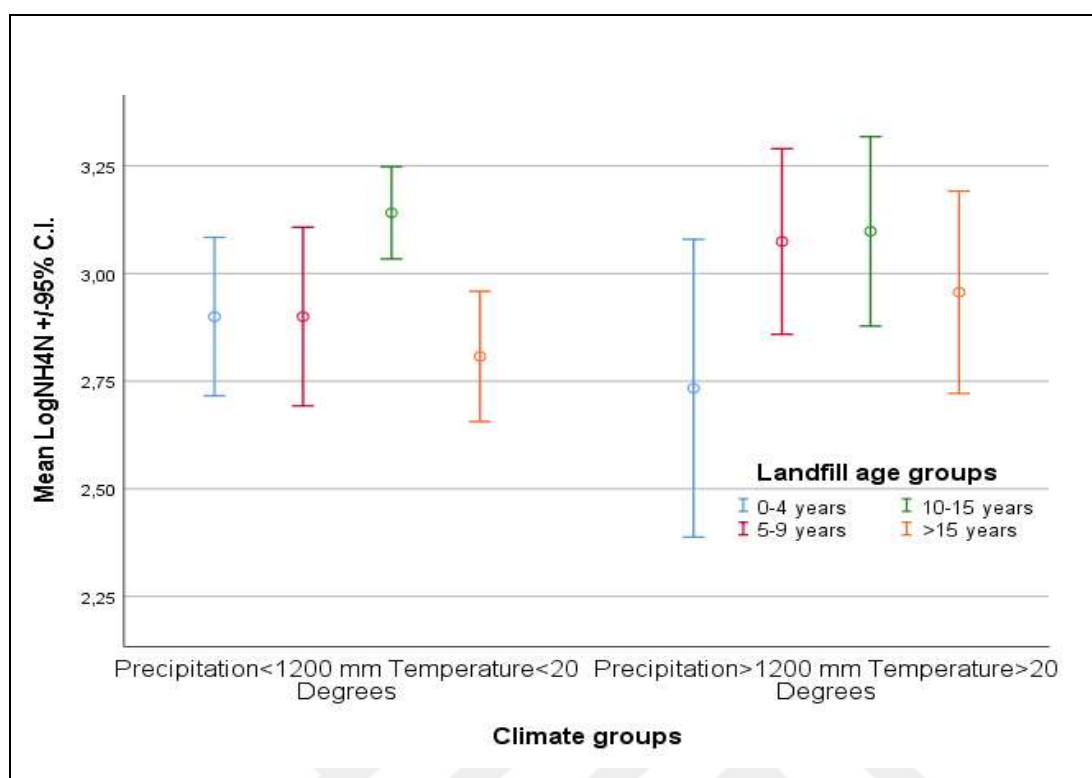


Figure 6.17. Confidence interval for mean values of LogNH₄-N for each group

As Ma et al. (2022) stated, the proportion of organics in waste stream can significantly impact the concentrations of NH₄-N and some inorganics (i.e., K, Cl, SO₄, etc.) in landfill leachate. In this study, NH₄-N and alkalinity values were found significantly higher in leachates of developing country (UM) landfills beside the organics (Table 6.17).

6.4 Conclusions

This study supplies valuable information to the waste management authorities as well as the researchers. It provides results/outputs to assess landfill leachate characterisation in different aspects. It is a very comprehensive study focusing on the impacts of climate, landfill age as well as development status on leachate. Also, the reasons behind high variations in the leachate composition of different landfills were shown as much as possible. Below conclusions could be extracted from the results of this study:

- There is a strong relationship between precipitation rates and leachate properties so that leachate categorization as frequently used in the literature as young-medium-old according to the landfill age should be evaluated meticulously by taking into account the climate effect. As the results of statistical analyses indicated, pollutant concentrations of leachate in high precipitation areas are considerably lower even for young leachate (<5 years). Therefore, comparisons made in the literature for young-old leachate in terms of pollutant concentrations should be done with caution. In tropical climates low strength young leachate could resemble the old leachate characteristics of landfills in far different climates (i.e., much lower precipitation rates).
- Some parameters do not decrease in leachate with time and they will be problematic during management of landfill activities. For example, majority of the inorganic compounds such as ammonia, salts, and EC will be high in leachate samples as long as landfilling continues. Thus, $\text{NH}_4\text{-N}$ and salts become long-term pollutants in leachate.
- Statistically different organic matter concentrations were found in landfill leachates from distinct climates and different income levels/development status. Therefore, leachate management methods need to be evaluated differently for high rainfall and low rainfall climates as well as for developed and developing countries.

- It is highly possible that after 10 years of operation, concentrations of COD and BOD as well as BOD/COD ratio of leachate will be much lower. Therefore, method of treatment would need to be re-evaluated probably from biological to more chemical/physical alternatives after that time. It could also be suggested that design life of a leachate treatment plant can be taken as approximately 10 years in average in low precipitation climates (i.e., <1200 mm) as the pollution parameters will stabilize considerably even with continuous waste disposal in active landfills.



CHAPTER 7

OVERVIEW OF RESULTS

In this study, various multivariate statistical methods were employed to investigate the landfill leachate characteristics, to get insight into the potential correlations and differences, and to find out the main reasons of variations between leachate samples originating from different landfills. Study started with data collected from landfills around the World reaching up to 511 leachate samples in total. In that respect, the results presented here come from probably the most comprehensive database ever produced. As a first screening, the original total data composed of 511 leachate samples was reduced to 360 when unsanitary landfills and closed sites were taken out. Firstly, correlations between leachate pollution parameters were sought and regression modeling were conducted between highly correlated parameters. Afterwards, PCA and cluster analysis were performed to see the status of groupings between pollutants. The PCA and Cluster tests were done with the aforementioned 360 data (Table A.1). However, depending on the selected parameters for analysis, final number of data decreases to values given in Table 7.1 due to missing data in any of the selected parameter. Finally, comparison tests (mainly ANOVA) were conducted to investigate the impact of climate (precipitation and temperature), landfill age, and development status (based on income index) on the leachate pollution parameters. For comparison tests only one data was withheld for the same landfill, so 215 data was used in total for ANOVA. Details of all the analyses performed in this study including the name of tested parameters and number of the data used are presented in Table 7.1.

Table 7.1 Summary table for the performed multivariate statistical analyses

Type of Analysis	Sample N	Parameters analysed
Pearson correlation	Depends on the compared parameters (Table 5.5)	pH, COD, BOD, NH ₄ -N, TKN, EC, TDS, Cl, alkalinity, Na, K, Fe, Ca, Mg, Ni, Zn, Cu, Cr, Pb, Cd, Mn
Regression modeling	Depends on the compared parameters (Table 5.2)	BOD, COD, TKN, NH ₄ -N, TDS, alkalinity, Cl, Na, K, EC
Cluster analysis for organics and inorganics	36	pH, COD, BOD, EC, NH ₄ -N, alkalinity, K, Cl, Na, Fe, Ca, Mg
Cluster analysis for organics and heavy metals	63	COD, BOD, pH, Fe, Mn, Zn, Cr, Cu, Cd, Pb, Ni
PCA for organics and inorganics	46	pH, COD, EC, NH ₄ -N, alkalinity, K, Cl, Fe, Ca, Mg
PCA for organics and heavy metals	50	COD (or BOD), Ni, Pb, Cd, Mn, Fe, pH, Cu, Zn, Cr
PCA for inorganics and heavy metals	46	Ca, Fe, Mn, pH, Cu, Cl, Cr, NH ₄ -N, Zn, Pb, Cd, Ni
ANOVA (two-way) for age and climate effect*	Depends on the data groups (Table 6.9)	pH, COD, BOD, BOD/COD, Cl, NH ₄ -N
ANOVA (one-way) for climate effect*	Depends on the data groups (Table 6.9)	pH, COD, BOD, BOD/COD, TKN, NH ₄ -N, TDS, TSS, Cl, Na, K, EC, alkalinity, TP, Fe, Zn, Cu, Cr
ANOVA (one-way) for age effect*	Depends on the data groups (Table 6.9)	pH, COD, BOD, BOD/COD, TKN, NH ₄ -N, PO ₄ -P, TSS, TDS, Cl, EC, alkalinity, Na, K, Fe, Ca, Mg, TP, SO ₄ , Ni, Zn, Cu, Cr, Pb, Cd, Mn
ANOVA (two-way) for age and income effect*	Depends on the data groups (Table 6.14)	pH, COD, BOD, BOD/COD, alkalinity, NH ₄ -N
ANOVA (one-way) for age effect*	Depends on the data groups (Table 6.14)	pH, COD, BOD, BOD/COD, TKN, NH ₄ -N, alkalinity, TSS, Cl, EC, Na, K, Fe, Ca, Mg, SO ₄ , Ni, Cd, Pb, Cu, Cr, Zn, Mn
ANOVA (one-way) for income effect**	Depends on the data groups (Table 6.14)	pH, COD, BOD, BOD/COD, TSS, NH ₄ -N, TKN, Cl, EC, alkalinity, TP, SO ₄ , Fe, Pb, Cu, Cr, Zn

*Analysed parameters depend on data availability (N≥10) for each group

**Development status effect

Various statistical analyses have been conducted in literature to understand the landfill leachate behavior. However, those studies were limited with a few landfill sites or from the same landfill sites in different times. Therefore, it is difficult to obtain a generalized behavior of leachate from different landfills in different climates. This study describes the statistical analysis of landfill leachate data collated with an extensive literature survey including leachate samples from 220 different landfill sites around the World. For the climate factor analysis, 68% of the data belonged to landfills in the climate group of low precipitation and low temperature regions, while 32% in the climate group of high precipitation and high temperature regions (Figure 6.1). Analysed data for the development status parsed as 65% from developed countries and 35% from developing countries. Due to limitations in the availability of data for some parameters, the analyses focused only on certain parameters having adequate number of data (Table 7.1). However, as correlation analysis showed, many leachate parameters are well-correlated with each other so that generalization of the results would be possible for untested parameters.

Multivariate statistical tests including cluster, PCA, and ANOVA were performed on the pre-treated data matrix according to the requirements of each statistical method which is presented in section 3.3. Analysis results are well-supported by each other. For example, results of cluster analysis and PCA were found highly depended on the correlation status of parameters. Highly correlated parameters were generally included in the same clusters or components, parameters in the same clusters were generally placed in the same components, and so on. PCA results revealed some reasons for leachate parameters being in the same or different components which were elaborated in the comparison tests (ANOVA). PCA scores showed that landfill age emerged as the main reason of the differences between components. The first component contained samples having high concentration of inorganic parameters belonging to aged landfills. The second component was formed by samples with high concentrations of organics and divalent ions coming mostly from younger sites. This was validated in the ANOVA as well. The inorganic content of leachate was not found

changing significantly with time as organics did. PCA suggested so as concentration of inorganic components was not found related to the biodegradation status of waste.

Ehrig (1983) was one of the earliest researchers studying variations in leachate quality in different landfills. He surveyed 20 landfills aged up to 15 years old in Germany for 3 years. He found that some parameters like EC, $\text{NH}_4\text{-N}$, Cl, K, and Na increased slightly showing a great variation with time. On the other hand, pH, COD, BOD, Fe and Ca concentrations changed considerably with landfill age. A similar finding was obtained both in PCA and ANOVA tests in the present work. In PCA, inorganic content including EC, alkalinity, Cl, K, and $\text{NH}_4\text{-N}$ were extracted as the first component explaining the highest variation in leachate while organic content with pH, Ca, and Fe were placed in the second component (Table 5.8). Armstrong and Rowe (1999) stated that variation in Cl concentration does not correlate well with COD, calcium, and pH, and may still be increasing after 14 years of operation. This will be relevant for Na and K too, as they both are conservative elements like Cl. All those findings were supported by ANOVA results as presented in Section 6.3.3.1. It can be suggested that the inorganic content of leachate will be higher in the long term and organic content and associated parameters (pH, Ca, and Fe) will cause the variation in leachate quality during early periods of landfilling.

Results of all the multivariate statistical techniques conducted in this study lead to the same conclusion that there are many leachate parameters having strong correlation with each other so that common parameters can be excluded from the sampling and analytical procedures during the monitoring activities of landfills. For example, alkalinity was found having very strong correlation with many monovalent ions in the leachate. It was a very well known issue that salts (Na, Cl, K) contribute most to the conductivity of leachate, however their high correlation with alkalinity is somewhat new and could be investigated further. There are other highly correlated parameters in landfill leachate which deserve further study. For example, conductivity and potassium is correlated with $\text{NH}_4\text{-N}$, TKN, and alkalinity almost better or in an equivalent degree than the well known parameters like Na, Cl, and TDS (Table 5.5.). This is most probably due to high concentrations of alkalinity and $\text{NH}_4\text{-N}$ in landfill leachate, hence

increasing their share in EC parameter. High correlation of K and $\text{NH}_4\text{-N}$ can be explained by exchange mechanism between NH_4^+ ions adsorbed on negatively charged waste particles with K^+ ions (Pivato and Gaspari, 2006). However, this subject is open to further investigation for any other possible mechanisms.

ANOVA results showed that climate dependent variations were statistically significant only for the concentrations of organics and related parameters such as pH and BOD/COD (Table 6.12). It was found that concentration of organics is very low during the whole landfill life in high rainfall climates (Figures 6.14 and 6.16b). Hussein et al. (2019) remarked that leachate from many sites in Malaysia (tropical climate) showed the characteristics of methanogenic phase (high pH and low organic content). Bhatt et al. (2016) presented the results of some researchers for faster waste degradation in high precipitation areas. They addressed advantages and disadvantages for introducing high moisture into the landfill. An advantage was stated as water removing the contaminants from the waste by leachate in the short-term of landfilling, while a disadvantage was high water movement flushing microorganisms out of landfills. Especially the slowly-growing bacteria (i.e., the ones responsible from methanogenesis) might be lost from the system. Therefore, it could be inferred that wash-out and dilution mechanisms prevail in high rainfall places and prevents the establishment of microbial medium in landfills for prolonged biological activities.

Similar to ANOVA results, score charts of principal components indicated that young landfill leachate in high rainfall climates could have similar properties with the ones in older landfills located in lower precipitation areas. This in turn suggests that leachate samples from landfills in different regions having distinct climates could show similar characteristics despite their different ages. On the contrary, the same aged landfill leachates could have very different composition if located in different climate conditions depending mainly on precipitation rates. Arithmetic means of organic content and biodegradability of leachates from landfills located in high rainfall regions are presented in Table 7.2.

Table 7.2 Average organic content and biodegradability for landfills in regions of high precipitation and high temperature climates

Leachate type	Landfill age	COD range (mg/L)	BOD range (mg/L)	BOD/COD range
Low strength & Moderate to low biodegradability	<5 year	2000-7000* (680-58400)**	300-2300 (43-40065)	0.12-0.34 (0.05-0.80)
	5-10 year	2100-4400 (875-16191)	200-800 (45-5668)	0.10-0.20 (0.05-0.35)
	10-15 year	1800-7000 (621-19600)	100-1300 (9-8232)	0.05-0.20 (0.01-0.52)
	>15 year	1200-4000 (550-15940)	80-1200 (36-6170)	0.03-0.25 (0.01-0.58)

*Lower and upper bounds for 95% Confidence Interval for Mean (antilogs of the values from ANOVA)

**Minimum and maximum values in the corresponding groups (antilogs of the values from ANOVA)

It was stated that continuous fresh waste addition into the landfills do not increase the organic concentration in leachate (Hussein et al., 2019). This was explained as either the result of higher ratio of old waste compared to fresh waste or high organic acid degradation. As methanogenic activities are related to pH range (Ding et al., 2018), results show that after 5 years, pH becomes constant around 7.5-8.0 in aged landfills. This suggests that the methanogenic conditions established at the bottom of landfills can treat the high strength leachate coming from fresh parts. Therefore, after 5 or 10 years, more constant values could be obtained for parameters such as pH, COD, and BOD. On the contrary, El-Fadel et al. (2002) stated that conservative contaminants like Cl in leachate would pass through the bottom layers of the landfill without any significant change in its concentration. Therefore, highly soluble Cl will continue to leach in active landfills as new waste continuously added, hence its concentration in leachate builds up accordingly (Statom et al., 2004; Yıldız et al., 2004). Cl content of leachate found increasing with landfill age in the present study as well. This finding would be relevant for Na and K as they are also conservative elements.

Classification of landfill leachate dated back to 1970s. As discussed in Chapter 4, it was found practical by many researchers to employ the landfill age criteria to define

their leachate type based on the operation time of a landfill. The reason might be that it is very difficult to estimate leachate quality in relation to other factors. Leachate classification if done properly could be helpful for researchers, decision makers, and experts working in solid waste management area. It would help to make comparisons between leachate related studies particularly treatability ones. In this study, it was shown statistically with considerable number of leachate samples that landfill age is an important factor for leachate quality up to some degree but only in specific climate conditions. This should be taken into account during defining leachate type.

According to the results of statistical analyses of present study and extensive literature review, it is clear that in low precipitation areas (dry climates), organic content of leachate has a decreasing trend with time but it is statistically significant only after 5 years and after 15 years. Between landfill ages 5 to 15 years, statistically no significant decrease were found (Table 6.11, Figures 6.7 and 6.8). Therefore, the “medium age” term for landfills having an age between 5-10 years as stated in majority of the literature studies need to be revised accordingly. Medium age leachate term could be associated to landfills between 5 to 15 years old and only in certain climate conditions, which was found as precipitation rate < 1200 mm and temperature < 20°C in this study. Following this conclusion, current old leachate terminology might be revisited as well. Old/mature leachate term could be associated to landfills over 15 years old rather than 10 years as currently employed in the literature. The results showed that even after 10 years of operation, landfills could still be in acidogenic or early methanogenic stage in dry climates. Therefore, the “medium age” term will be more relevant in this case. However, results showed that in terms of BOD/COD ratio, leachate type/biodegradability could be determined differently such as: highly biodegradable leachate in young landfills (<5 years old), moderate biodegradability in medium/intermediate landfills (5-10 years old), and low biodegradability/refractory leachate in old/mature landfills (>10 years old) (Figure 6.9 and Table 6.11). This complies with the current literature definition in terms of landfill years used. However, this definition is only relevant for low precipitation areas for which a sample list is provided in Appendix F. On the other hand, for landfill leachates from high

precipitation climates like tropical regions (i.e., India, Malaysia, Brazil, etc.), the leachate age terminology currently used or as purposed with this study will not be relevant. The leachate type from landfills in those climate conditions resembles old/mature leachate due to low organic content, high pH, and low BOD/COD ratios (Table 7.2).

In the light of findings of this study, Table 7.3 was prepared to provide detailed information for possible re-classification of leachate with associated parameters that could be considered by decision makers and researchers in the waste management area for evaluating their leachate. It should be emphasized that leachate definition could be relevant for landfills in low rainfall regions having low temperatures as well (i.e., precipitation rates < 1200 mm and temperature < 20°). A sample list of countries extracted from the leachate database that could be eligible for the suggestions given in Table 7.3 is provided in Appendix F.

There are many studies for the leachate treatment methods where evaluations and comparisons have been done according to the leachate age criteria mostly as young or old. Accordingly, young leachate was accepted as having very high COD, BOD, and BOD/COD values so that biological treatment alternatives were tested and recommended. For old leachate cases on the other hand, COD and BOD concentrations were accepted as relatively low and being mostly refractory compounds so that chemical and physical technologies were experimented more and suggested accordingly. Therefore, leachate definition as young or old is an important parameter for the researchers to plan their study for treatment alternatives and then compare their results with others studying different or the same type of leachate. In that sense, findings of this study revealed that leachate defined according to the landfill age terminology in the current literature might not be relevant for every region in terms of accepted ranges of organic content and BOD/COD ratio. This is because, young leachate from a landfill in high rainfall places would be very low in organic content (Table 7.2) in comparison to young leachate from relatively low rainfall regions (Table 7.3). Therefore, suggested treatment methods will not be relevant if it was tested for

high concentrated young leachate of low rainfall climate landfills. For that reason, leachate types and ranges for organic content and biodegradability suggested in this study (Tables 7.2 and 7.3) could be used for landfills in regions having similar climate conditions to plan and design the leachate management systems.

Table 7.3 Suggestions for definition of leachate type for landfills in regions of low precipitation and low temperature climates

1 st Alternative (according to landfill age and organic content)				
Leachate type	Landfill age	COD range (mg/L)	BOD range (mg/L)	BOD/COD range
Young	<5 year	8000-19000*	4000-10000	0.45-0.55
(High strength)		(1080-80750)**	(460-42450)	(0.34-0.81)
Medium	5-15 year	3000-8000	500-3500	0.10-0.45
(Medium strength)		(550-38850)	(81-24500)	(0.04-0.76)
Old	>15 year	1500-3000	100-500	0.10-0.20
(Low strength)		(229-19814)	(27-4946)	(0.02-0.54)
2 nd Alternative (according to landfill age and biodegradability)				
Leachate type	Landfill age	BOD/COD range	COD range (mg/L)	BOD range (mg/L)
High biodegradability	<5 year	0.45-0.55*	8000-19000	4000-10000
		(0.34-0.81)**	(1080-80750)	(460-42450)
Moderate biodegradability	5-10 year	0.30-0.45	3500-8000	1500-4000
		(0.11-0.76)	(560-38800)	(210-24500)
Low biodegradability	>10 year	0.10-0.30	1500-8000	100-2500
		(0.02-0.54)	(229-25817)	(27-16513)

*Lower and upper bounds for 95% Confidence Interval for Mean (antilog of the values from ANOVA)

**Minimum and maximum values in the corresponding groups (antilog of the values from ANOVA)

Finally, it is a well known fact that developing countries have greater proportion of food waste in their waste streams than developed countries (Ma et al., 2022). Therefore, it is expected that concentrations of COD, BOD, and NH₄-N in leachates of developing country landfills are much higher than those in developed countries.

Actually, Ma et al. (2022) found that COD values in young landfill leachate of upper middle income (developing) countries was about two times higher than that in high-income (developed) countries. In the present study, higher values were observed in developing countries (UM group) as well especially in the old aged landfills (Table 6.17). It should be emphasized that due to data scarcity in some pollution parameters especially in UM group countries, it was not possible to conduct comparison analysis for every parameter and neither for short range landfill age groups (i.e., <5 years, 5-10 years, 10-15 years, and >15 years).



CHAPTER 8

CONCLUDING REMARKS

Extensive literature review showed that previous studies covering landfill leachates included generally the same landfill or the ones in the same or similar area. In this study, leachate data from 220 different landfill sites from a total of 46 countries around the World were analysed. The main objective was to investigate the main properties and variations of constituents in MSW landfill leachate depending on different factors. Important relationships between many leachate parameters were identified. Results of regression modeling showed that there are high correlations between many leachate parameters so that even with only a few important parameters, monitoring of leachate quality would be possible. Therefore, it is highly recommended that common parameters can be excluded from the sampling and analytical procedures during the monitoring activities of landfills. Those could be interchangeably selected for monitoring: either $\text{NH}_4\text{-N}$, or TN , or TKN ; Fe or Mn ; Ca or Mg ; any of the K , or Na , or, Cl , or, EC , or TDS .

Results of ANOVA is very important to understand the behavior of leachate between different landfill age, climate, and development status groups. It was found that some leachate parameters show important variations according to different conditions such as organic content, pH , and BOD/COD ratio, while others do not have any clear trend at all being mostly inorganics and heavy metals. It was statistically proven that precipitation is the most important factor governing the leachate quality. Besides that, landfill age plays an important role especially for the organic strength but not for inorganic content. For that reason, while determining the effect of landfill age on the leachate composition and providing some ranges for pH , COD , BOD , and BOD/COD ratio, climate factor in the landfill area should be taken into account. Supporting this finding, PCA results indicated that different aged leachates may have similar composition depending on climates in their region. This means leachate

samples having different ages can group together if they come from distinct climates (i.e., arid vs tropical). On the contrary, leachates having the same or similar age may have totally different characteristics due to distinct climates where they belong.

Results showed that organic waste decomposition phases presented in the literature as number of stages may not be the same for each landfill due to the effect of climate conditions beside the reason of leachate collection method from multiple layers (in a mix of old waste and new waste). In high precipitation regions, stabilisation of leachate may not be completed in four or five phases as organic matters in the waste pass through the leachate without sufficient exposure to biological activities under high rainfalls. Therefore, low strength leachate will not necessarily include refractory organics as stated being present in low-strength old leachate. In addition to this, wash-out effect can limit the methanogenesis in high rainfall places which prevent naming low organic content leachate as old or methanogenic.

All the information gained about the factors affecting leachate quality have great value for the planning and operation of leachate management systems. It emerged that landfill leachate treatment methods will need to be modified as the leachate characterization changes with time in a landfill. Also, climate effect comes forward as an important parameter for changing the leachate quality considerably which should be taken into account during design and operation phase of leachate treatment plants. In that respect, this study supplied an extensive database to be used for the purpose of estimating the composition of landfill leachate depending on different climate, age, and development status criteria. This estimation is important when leachate treatment is to be designed and constructed in due time with landfill construction which is the case in many places. As there would be no leachate data resulted from newly constructed landfill site, a high quality leachate database would be very helpful to provide proper predictions relevant to the conditions of the landfill as much as possible. Furthermore, leachate type could be an important parameter for providing guideline to plan and design the treatment alternatives. Hence, it can be suggested that leachate age terminology should be revisited as proposed in this study.

Findings of this study about the strong relationship between precipitation rates and leachate properties comply with the literature studies as well and support the idea of categorizing leachate differently in terms of landfill age given that climate effect is taken into account as well. According to the results of ANOVA, leachate in different aged landfills in low precipitation areas (sub-humid and arid regions) could be categorized into three main phases as: young for <5 years, medium/intermediate for 5-15 years, and old/mature for >15 years old landfills with respect to COD and BOD concentrations. Alternatively, more representative terminologies could be selected such as high-medium-old strength leachate accordingly. However, results indicated that biodegradability of leachate could be defined according to the BOD/COD ratio under different ranges of landfill age as high (<5 years), moderate (5-10 years), and low biodegradability (>10 years) being only relevant for low precipitation areas. Attention should be given to high precipitation climates (moist sub-humid and humid regions) as stabilization status cannot be related directly to the chronological age of a landfill or leachate. Leachates of landfills in those regions could be defined as low strength with moderate to low biodegradability throughout the life time of landfilling.

CHAPTER 9

RECOMMENDATIONS FOR FUTURE STUDIES

ANOVA is a powerful tool for the comparison analysis of multivariate data and it provided good results to understand the impact of some important factors on landfill leachate characteristics. In this study, only leachate data from active sanitary landfills were analysed. With ANOVA it is possible to make further analyses for other subjects or impact factors as well. For instance, differences between active and closed landfills or differences between sanitary landfills and uncontrolled dumpsites, etc. could be analysed. However, categorizing leachate data into factor groups (i.e., climate, landfill age, etc.) resulted partition of the data set in sub-groups having lower number of data. Therefore, it was not possible to have sufficient number of data (accepted ≥ 10 in this study) for many leachate parameters in every group. Categorization of the data into a number of groups for each impact factor requires considerable data. This could be handled by collecting specific data as much as possible that fits to the purpose of analysis.

In this study, climate data was divided into two groups as there was no or very little data for landfills in arid/semi arid regions (precipitation < 600 mm) as well as in tropical regions (precipitation > 1500 mm). For that reason, some leachate parameters could not be analysed particularly in very high and very low precipitation climates. In the future, more data from places like Middle East, Northern Europe, Central Asia, etc. could be included for analysing the leachate composition for more than two climate or precipitation groups. Moreover, temperature effect alone as a climate parameter can be investigated in the future studies by collecting specific data in just one precipitation group (i.e., arid/semi arid places) where distinct temperatures could be observed (i.e., temperature $< 10^{\circ}\text{C}$ and between $10\text{-}20^{\circ}\text{C}$). Specific climate conditions could be tested as well according to the famous Köppen-Geiger climate classification method. Furthermore, it should be noted here that climate would have different effects on

leachate properties in the future due to great fluctuations in the meteorological events due to climate change. Increased amount of local rainfalls and/or droughts could have different impacts on landfill leachate which requires further study in the future.

Development status effect was analysed for leachate samples only from landfill areas having annual precipitation rates between 600-1200 mm and landfill ages <10 years and >10 years. It was not possible to partition the current data set into as many sub-groups as required due to scarcity of data. For future studies, development status could be investigated for younger age group as <5 years since most of the difference in leachate characteristics was observed in that age group.

Finally, it is strongly recommended that data pre-treatment steps should be followed according to the specific requirements of the statistical techniques to be used in the relevant analyses. Also, as leachate data from landfills having recirculation of leachate was not taken into account in the present study, its effect into leachate classification could be analysed separately in the future studies.

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APPENDICES

A. Leachate Data



Table A.1 Leachate database (360 data rows including active sanitary landfills)

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/COD	NH ₄ -N	TKN	TN	TP	PO ₄ -P	TSS	TDS	TS	Cl	SO ₄	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Ellouze et al. 2008	466	18.1	6	7.8	25200	13000		0.51	1750	1810				1000	34500	35500			29.0				20.6	15	32	8	8.1	1.5	0.75	0.29	0.2	18.5
Li et al. 2009	641	9.2	15	7.9	3100				1000					315			2850		16.5				7.6				0.65	0.26		0.37		
Kasıkcı and Çalh 2011	712	14.4	12	7.7	8042	3877		0.47	1681		2348			607			2549															
Sun et al. 2010	610	12.1	10	7.8	15178				1012		1114																					
Timur and Öztürk 1999	687	16.7	4	7.6	18100	10875	5550	0.60	1810	2000	2048		64.0		18835			350		9575			44	788	364	0.32	1.1	0.13	0.78	0.02	0.005	5.3
Ağdağ 2011	589	16.1	4	8.0	18034	11504		0.64	454		718			1607						9316			7.1			0.86	0.26	0.18	0.4	0.03	0.07	0.87
Wang et al. 2001	2122	22.6	3	7.9	4750	2270		0.48	1310								1890															
Calli et al. 2005	826	13.3	4	7.3	20700	12200		0.59	2330	2510			9.8	2170			3670			9850			62			0.68	0.98	0.25	0.52	0.78	1	3.8
Castillo et al. 2007	1159	23.4	18	8.0	8975	5226		0.58						282	13675	13957	1932			8555												
Liang and Liu 2008	610	12.1	11	8.5	1703				1972		2117									11898	2451	1882	1	7	626	0.2	0.02	0.02	0.12	0.01	0.006	0.01
Bohdziewicz and Kwarciak 2008	616	8.2	17	8.3	3850	400		0.10	942								2150			5050												
Kennedy and Lentz 2000	868	6	17	8.0	6200												1895	88	7.6		1210	1078	3.1	58	404	0.15	0.23	0.035	0.054	0.04	0.01	0.78
Contrera et al. 2014	1440	19.7	12	8.0	8566				2183							15330				10687			17.8			0.62	0.81	0.07	0.13	0.27	0.08	0.53
Borzacconi et al. 1999	933	16.3	7	8.0	18550	10510		0.60	1470	1640		17.0				21700	3280	91		9280			41.0	408		0.67	3.8		2.6	0.51	0.05	10.2
Kettunen et al. 1999	638	5.1	8	7.0	2500	1550		0.62	175		195	1.8					365	40					75.0	625								
Im et al. 2001	1116	13.5	2	7.3	24400	10800		0.44	1682	1766		31.2		2400			3160	162		6700			76			0.16	2.8	0.78	2.4	0.72	0.02	16.4
Yang and Zhou 2008	1720	22.2	19	7.5	15940	6170		0.39	2300			14.5		3345						13070												
Kheradmand et al. 2010	316	16.8	10	6.3	55351	49400		0.81	1460					1962						12430			386		1432	1.06	1.2	0.05				14.4
Sadri et al. 2008	519	2.1	33	8.3	4079	730		0.18	648	1150													7			0.14	0.5	0.017	0.061	0.013	0.001	0.66

[illegible]

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/ COD	NH4 -N	TKN	TN	TP	PO4 -P	TSS	TDS	TS	Cl	SO4	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Primo et al. 2008a	1200	14	15	7.7	1750	120	1300	0.07	1225					295			1876	500	9.1		1364	845	2	155	73							1.25
Fang et al. 2005	2122	22.6	9	7.8	12900	5062	3240	0.39	2100																							
Kennedy et al. 1988	1452	6.3	15	5.5	19560				260	337						13000				4550			401		212		51					49
Chen 1996	1700	22.1	1	8.3	3340	296		0.09	1892	2119		15.1		239					16.1				4.5				0	1.22				
Chen 1996	1794	21.9	1	6.7	1214	967		0.8	133	162		6.7		102									29				1		0.3			
Chen 1996	1488	22.5	1	7.8	3641	743		0.2	1452	1605		27.5		66									12				0.01		1.3			
Chen 1996	1488	22.5	1	7.7	1311	247		0.19	379	452		8.9		54									1				0.01		0.005			
Chen 1996	1488	22.5	1	8	825	43		0.05	13	75		21.7		10									10.0				0.01		0.3			
Chen 1996	1488	22.5	2	8.5	1456	70		0.05	47	544		23.5		20									22				0.6		0.5			
Chen 1996	1488	22.5	2	8.4	2282	386		0.17	962	1029		7.9		110									8				0.01		0.3			
Chen 1996	1700	22.1	5	8.2	3447	620		0.18	2505	2648		15.4		194									8				3.4		1.1			
Kang et al. 2002	1319	12.5	7	7.9	5348	2684		0.5	1826	2192				143	12952	13095				7928												
Kang et al. 2002	1319	12.5	3	6.6	41507	32790		0.79	1896	2482				1873	30812	32685				9130												
Maranon et al. 2008	900	13	1	8.2	19250	9960		0.52	2470										27													
Garcia et al. 1996	1200	14	2	7	37000				1046				3.4	1690				631		4275												
Huo et al. 2009	1232	18.4	12	7.8	19600	8232		0.42	2370				3				7080	95	35.3													
Lopez et al. 2004	567	16	11	8.2	10540	2300	3900	0.20	5210				32	1666	18589	20255	4900		45.4	21470	3970	3460	2.7	16	24	0.31	0.16		2.21	0.015	0.01	0.04
Amokrane et al. 1997	751	9.5	11	8.2	4100	200	1430	0.05	1040				8.4	200			5420	550			3000	880	0.9	68	110	0.81	0.73	0.39		0.46	0.1	
Rodriguez et al. 2004	900	13	14	8.3	3895	931		0.24	2013	2213	2239									8887			5.5			0.43	0.35	0.28		0.2	0.035	
Chen et al. 2008	562	3.4	4	7.3	12075	4900	3500	0.40	298			12.1		412			1520	277					10.7				0.93	0.05		0.035	0.035	
Kalyuzhnyi and Gladchenko 2004	680	4.7	41	6.8	15110				722		930	35	12					284					125				15.6	0.13		0.07	0.004	
Kalyuzhnyi et al. 2003	680	4.7	41	6.6	2620				72	125	126	29	5.6					95					8.8				1.5	0.14		0.07	0.003	
Wang et al. 2016	610	12.1	17	8.3	2305	105		0.05	1240	1366	1370	4.5								12470												
Durmusoglu and Yilmaz 2006	835	14.5	5	7.9	1760							2.5		670												0.25	1.55	1.6	0.11	0.45	0.02	
Zhang et al. 2013	1497	17.5	21	7.8	1105	876		0.79	667		956	13.5					820	270					15.5			0.2		0.74	0.31	4.56	0.24	2.39

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/COD	NH ₄ -N	TKN	TN	TP	PO ₄ -P	TSS	TDS	TS	Cl	SO ₄	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Zhang et al. 2013	1497	17.5	4	8	1491	1000		0.67	468		775	15.4					1430	55					1.9			0.06	17.2	0.18	0.17	0.23	0.01	0.54
Zhang et al. 2013	1497	17.5	5	7.7	1906	834		0.44	677		1163	19.9					3154	73					38.7			0.51		1.85	0.78	11.39	0.6	5.98
Ziyang et al. 2009	1112	16.1	2		7125		2868		4251		4368	35	34.3						41.5	18162												
Ziyang et al. 2009	1112	16.1	4		1543		625		1564		1754	18	17.6						17.9	8049												
Ziyang et al. 2009	1112	16.1	6		2424		420		1274		1490	14.8	12.4						15	5573												
Ziyang et al. 2009	1112	16.1	9		1015		410		807		1092	3.4	2.4						12.3	5227												
Ziyang et al. 2009	1112	16.1	11		1523		210		715		980	7.2	4.8						12.1	4649												
Ziyang et al. 2009	1112	16.1	12		695		75		238		428	0.6	0.2						6.4	1754												
Derco et al. 2010	1290	10.4	19	8.8	2755	217		0.08	856	1143	1144						1405															
Martinen et al. 2002	650	4.5	6	7.6	560	240		0.50	85	86									2													
Rivas et al. 2003	523	16.8	2	8.3	2300	850	928	0.37		85													16.2			1.36	0	0.25	0.67		0.001	0.71
Rivas et al. 2003	523	16.8	3	8.4	15273	460	5367	0.03		475													19.4			0.42	0.12	0.04	3.3	0	0	1.67
Boumechhour et al. 2013	736	17.1	7	8.1	3848	388		0.11	3159	5236	5238		37.3	10			3	995	0.5				21.5				1.5				1.7	
Modin 2012	668	7.8	57	7.4		827	611		176		333	9.3		380			657	103	6.1				11			0.07	0	0.049	0.071	0.03	0	2.1
Robinson 2005	1918	22.7	5	8.6	2560	167		0.07	2563	2760			27.6				2740		29.5	11500	2100	1000	5.5	19	31		2					
Robinson 2005	2125	22.7	4	7.4	680	43		0.06	1240				3.2				1130				2420	430	8.4	38	50		1.2					
Robinson 2005	758	9.6	13	8	8260	3020	3350	0.37	2110				14.2				5200	3	28.5	11500	4030	1690	30.2	123	95	2.81	0.33	0.04	2.17	0.11	0.005	0.55
Robinson 2005	758	9.7	13	8.1	8510	495	2500	0.06	2050				19.7				3150	3	24.4	11500	2480	1270	11.2	67	76	1.7	0.19	0.025	1	0.19	0.005	0.32
Robinson 2005	822	9.4	14	8.2	3140	112	878	0.04	1240				9.3				2950	3	16.6	8200	1960	860	3	53	50	0.14	0.02	0.01	0.11	0.02	0.005	0.13
Robinson 2005	828	20.9	16	7.5	2488	300		0.12	1917				8.4				2133	0	16.2	4802	1160	721	2.7	47	78	0.09	0.08	0.025	0.115	0.056	0.005	0.01
Robinson 2005	828	20.9	21	7.7	1382	300		0.22	886				4.8				2481	0	17.1	5665	1291	658	4.6	75	150	0.08	0.03	0.025	0.1	0.002	0.0005	0.02
Robinson 2005	828	20.9	2	7.6	2021				957				4.6				2437	15	18.1	5118	1712	1153	2.3	112	222			0.025	0.182	0.007	0.001	
Robinson 2005	828	20.9	4	7.8	3949				1060				6.7				3569	36	21.2	5823	1905	1060	2.8	93	196		0.13	0.025	0.263	0.02	0.0005	
Robinson 2005	925	19.8	2	7.6	1605	695		0.43	675				5				1197	96	12.7	5524	914	940	18.6	208	355	0.12	0.17	0.025	0.08	0.002	0.0005	0.86
Robinson 2005	925	19.8	6	7.9	733	300		0.41	387				1				1728	26	9.8	2654	826	541	12	161	152	0.12	0.12	0.005	0.079	0.007	0.0005	

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/COD	NH ₄ -N	TKN	TN	TP	PO ₄ -P	TSS	TDS	TS	Cl	SO ₄	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Robinson 2005	475	16.9	33	8.1	3382				1998				12.8				2977	94	27	9950	2320	1615	7.3	136	141		0.06					
Robinson 2005	1916	21.8	1	7.5	6332	3496		0.55	346								1050			2385							2.18		0.038			
Robinson 2005	1916	21.8	2	7.8	4593	1832		0.40	1218								2641										1.66		0.09			
Robinson 2005	1916	21.8	5		4925	1300		0.26	1138								3932			7155							2.46		0.26			
Robinson 2005	1054	10	26	7.2	1181	76		0.06									859	1			669	471	0.2		95	0.11	1.24	0.05	0.06	0.07	0.01	0.4
Robinson 2005	1430	25.6	7	7	1980		990		1350			14					3650	2	28.3	15970	2179	1819	3.1	199	182	0.5	0.24	0.025	0.16	0.5	0.025	1.65
Centre for Advanced Engineerin g 2000	1367	15.1	6	7.3	2587	1959		0.76	168					954			402	26	13.3		628	281	99.8	342	128	9.78	12.2			0.089	0.005	2.98
Centre for Advanced Engineerin g 2000	1240	12.7	11		1200	530		0.44	290								410	32		1860	290	150	178			0.03	1.6		0.08	0.02	0.003	4.6
Centre for Advanced Engineerin g 2000	750	14.1	10	8.5	1260	100	378	0.08	428								2584	23	11.5	3500	1360	720	2	95	140	0.35	0.6		0.1	0.005	0.001	0.55
Centre for Advanced Engineerin g 2000	1364	14.6	19	7.5	1040	230		0.22	684								867	85	9.3		780	306	2.9	93	88	0.12	0.23		0.13	0.09	0.003	0.89
Eldyasti et al. 2011	834	9.1	31	8.4	1259	565		0.45	360	392	395	6.2	3.4	263						1619												
Pastore et al. 2018	642	13.9	23	8	5300	1325		0.25	1600		1750	9.9		295			3600	1300	18		1800	1500	1.2		300	0.5	0.01		0.03			0.01
Ferraz et al. 2013	1440	19.7	21	8	2393	427	718	0.18	1213	1421						11430			19.3	7770												
Wallace et al. 2015	899	3.5	16	7	1223	445		0.36	524	600	604	2.8	2.3	116	5310	5426	967	93.4	13.7	4290	918	417	22.6	255	150	0.2	0.4	0.1	0.1	0.1	0.1	2
Site data	615	14.3	1		4585	2200		0.48			1253			115																		
Site data	615	14.3	3	8.2	5181	2017	1175	0.39	1014	1285		15.4		260					17	8581												
Site data	615	14.3	4	7.9	5267	3450	1773	0.66	1335	1483		13.4		215					24.3	9800												
Site data	703	11.3	1	7.2	7478						456	8.1		151					7.6										0.06	0.005		
Site data	337	11.3	1	6.5	60333					1905		43.7		2933				2055							0.49	2.61	0.125	3	0.026	0.004		
Pala and Erden 2004	687	16.7	11	8.2	20865	1325		0.06	2040			7.5		156				360	33.3				15	130		0.5	0.8	0.5		0.7	0.1	
Pala and Erden 2004	687	16.7	9	8.2	38950							4.3		1442					27.4								3.1	0.6		0.7	0.2	
Yu et al. 2010	1720	22.2	6		16191	5668		0.35	3692	5087	5096	28.4		5281																		
Yalılı Kılıç et al. 2007	712	14.4	2	7.1	32957	22904		0.69	73			38.4		1825									100				49.8	15.2	7	56.7	2.8	

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/ COD	NH ₄ - N	TKN	TN	TP	PO ₄ - P	TSS	TDS	TS	Cl	SO ₄	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Yahli Kılıç et al. 2007	712	14.4	3	7.1	19048	11894		0.62						1429									42.5				49.6	11.6	6.6	7	0.06	
Renou et al. 2008a	834	9.1	2	5.8	13800	9660		0.7	42	212		0.8				12730																
Hippen et al. 1997	796	10	14		3561	1512		0.42	833	1083	1084						1790													0.5		
Castrillón et al. 2010	838	13.5	3	7.98	13478	8275		0.62	1478																		0.79	0.16	0.59	1.18	0.07	
Castrillón et al. 2010	838	13.5	5	8.3	5132	2414		0.45	1485																		0.49	0.04	0.27	0.23	0.02	
Castrillón et al. 2010	838	13.5	8	8.3	3893	800		0.20	2156																		1.06	0.1	0.54	0.65	0.04	
Castrillón et al. 2010	838	13.5	13	8.2	2838	700		0.24	2300																		0.26	0.05	0.41	0.04	0.05	
Castrillón et al. 2010	838	13.5	17	8.37	3757	858		0.23	2132																		0.42	0.15		0.03	0.02	
Jianguo et al. 2007	1962	20.3	12	7.4	13890	7249	4445	0.52	2376		2499	64.0															13.4	0.82		0.636		2.35
Mertoğlu and Çalıh (2011)	942	14.1	15	8.0	20414	11250		0.49	3355	4212		37.9		920	21573	23238	4275	10	35.4	14650			43.7			0.64	0.48	0.07	2.025	0.42		
ÇOB 2010	942	14.1	15	8.1	9105	5925	2215	0.65	1480	1890		68		880	12390	14870	2200		21.4	8400												
Mertoğlu and Çalıh (2011)	826	13.3	15	7.8	25817	16513		0.60	2202	2561		18.2		1971	16220	24851	4200	10	28	10408			10.4			0.95	0.93	0.063	1.582	0.707		
ÇOB 2010	826	13.3	14	7.8	28716	17230	10774	0.60	2560	2670		27		3440	16480	24200	5950		27.7	10100												
Mertoğlu and Çalıh (2011)	712	14.4	13	7.98	7682	4004		0.49	1959	2673		32.0		829	12058	13391	3200	88	21.2	9500			7.56			0.52	0.58	0.073	1.347	0.447		
Mertoğlu and Çalıh (2011)	712	14.4	14	7.8	8225	4543		0.56	1460	2109		19.2		795	10295	11575	2400	10	18	6725												
Mertoğlu and Çalıh (2011)	1056	18.8	1	6.92	43783	24634		0.59	2380	2908		40.6		2902	15983		3279	670	27.5	11358			94.2			1.12	1.71	0.113	2.048	0.728		
Mertoğlu and Çalıh (2011)	1056	18.8	2	8.2	12976	5488		0.45	2463	2724		29.4		1834	16835	18669	4725	10	23.2	11900			4.39			0.59	0.5	0.129	1.578	0.488		
Mertoğlu and Çalıh (2011)	891	14.4	2	7.58	8586	4976		0.57	965	1091		17		735	7723	10069	1667	58	14.2	5458			12.1			0.22	0.83	0.136	0.87	0.292		
Mertoğlu and Çalıh (2011)	891	14.4	3	7.86	2957	1590		0.48	1194	1362		19.6		461	8546	9340	1978	10	15.8	8056			2.78			0.39	0.44	0.103	0.888	0.326		
Mertoğlu and Çalıh (2011)	557	15.5	14	8.05	20564	10821		0.52	2256	2630		43.5		2058	24168	27409	6175	10	32.4	9225			3.78			0.99	0.23	0.118	1.983	0.743		
Mertoğlu and Çalıh (2011)	725	13.8	1	7.6	15263	8925		0.61	1893	2227		27.9		1170	14498	18109	3781	10	23.3	9993			16.5			0.67	0.37	0.105	1.387	0.46		

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/ COD	NH4 -N	TKN	TN	TP	PO4 -P	TSS	TDS	TS	Cl	SO4	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Mertoğlu and Çalli (2011)	725	13.8	2	7.92	16205	8713		0.48	2148	2651		32.8		2004	15741	20034	4552	10	27.5	10542			6.04			0.56	0.36	0.09	1.307	0.562		
Mertoğlu and Çalli (2011)	942	14.1	2	6.6	50050	26000		0.52	1689	3060				2475						12325			95			0.98	0.6	0.35	1.35	0.74	0.1	
Akkaya et al. 2011	942	14.1	11	8.1	12063				2558	2868		25.3			20300		4962		30.5	17234	2315	2433	24.1	85	264	1.01	0.87	0.28	1.1	0.94	0.15	2.51
İmen et al. 2009	466	18.1	6	7.5	14950	4894		0.33	2570	3740	4060								43.5				7.4	371	258	0.3	0.44	0.37	1.05	0.48		
Peixoto et al. 2018	1318	20	30	8.1	1013		286	0.00	398	427		8.7		39	4663	4702				7.1			1.1			4	0.45	0.14	0.086	0.01	0.006	0.42
Zin et al. 2012	4000	27.3	17	8.1	770	109		0.14	500			42		271									2.7									
Stuart and Klinck 1998	450	15.9	1	8.4			864		773								2935	71		7081	2465	1205	2.2	66	175	0.02	0.13	0.01	0.33	0.02	0.008	0.63
Stuart and Klinck 1998	450	15.9	8	8.9			1070		66								5590	538		6578	4040	2200	1.4	23	146	0.41	0.09	0.02	0.52	0.02	0.008	0.04
Stuart and Klinck 1998	2370	23.3	3	7.9			32		81			0.5					301	7		567	128	165	0.01	50	24	0.01	0	0.003	0.005	0.05	0.003	2
Stuart and Klinck 1998	1430	25.6	7	6.8	6800		2398		284			1.1					3068	359	12.7	1501	1680	499	14.6	616	237	0.5	0.15	0.025	0.05	0.5	0.025	11.1
Morris et al. 2018	891	9.4	10		550	141		0.26	915				5.4																			
Zakaria and Aziz 2018	2379	27.5	15	8.1	3852	196		0.05	1241						6237																	
Aziz et al. 2015	2599	27.5	17	8.0	1295	285		0.20	562			32.3		553	4279	4832	313		7.7	14667			3.8	600	66.0		0.33					
Aziz et al. 2015	4000	27.3	16	8.7	1456	257		0.18	564			44.6		693	5922	6615	899		10	16000			3.4	248	49		0.22					
Al-Wabel et al. 2011	111	25.4	2	6.2	17003									5604			8972	1504	52.8		6109	3684	168.0	7413	1963	0.51	0.18	0.18	0.26	0.02	0.001	10.8
Henry et al. 1987	767	8.1	14	6.6	1870	930		0.50	10	75		0.2		90	2980	3070				1070			36.0				0.19	0.05			0.008	
Chu et al. 1994	1274	8.7	3	5.8	5518	3670		0.67	130	157							1522				904	195	242.0		40	0.05	4.90	0.13	0.05	0.05	0.05	40
Fan et al. 2006	1825	22.5	10	7.6	2525	164	2160	0.06		123	138	5.0		905	5925	6830			17.3		1787	1278	4.2	38	86	0.15	0.35	0.46	0.65	0.10	0.005	0.39
Ding et al. 2018	1287	18.3	8	8.2	3676	570		0.16	1682		1824	14.0		360															0.35	0.42	0.05	
Zainol et al. 2012	4000	27.3	15	8.1	855	158		0.19	857					233			1800	91	11.9				2.2			0.16	0.26	0.08				0.08
Zainol et al. 2012	2599	27.5	16	7.6	117	29		0.24	210					47			243	142	2.9				0.4			0.07	0.09	0.03				0.09
Xaypanya et al. 2018	1430	28.1	6	7.7	3055	755		0.25	1105	2110	2122				17600		4020		29.7								0.60		0.38	0.07	0.004	
Yılmaz 2000	942	14.1	3	7.3	25975	16625		0.64	1875	2313	2316		3.3	1134						10063			75.0			0.68	2.0			1.70		39
Costa et al. 2018	1299	23.2	35	8.2	2194	45	896	0.02	2980								2512															

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/ COD	NH4 -N	TKN	TN	TP	PO4 -P	TSS	TDS	TS	Cl	SO4	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Costa et al. 2018	1354	23.5	2	7.8	1931	863	653	0.44	2185								1597															
Dia et al. 2018	1101	4.8	15	7.8	1619	460	317	0.29	653			4.9							5.3	4301			24.7				1.5					
Lima et al. 2017	1173	23.2	27	8.1	1515		555																									
Demirci 2017	589	16.1	8	8.3	7050	3200		0.45			521		71.2													0.5	1	0.07	0.4	0.3	0.01	
Demirci 2017	589	16.1	14	8.5	5300	1266		0.24						260												8.6	0.51	11.9	1.95	0.25	0.01	
Aghasa et al. 2017	2164	23.3	8	8.1	7200	3139		0.440	1721	2988	3021	25.3								7808												
Aziz et al. 2011	2599	27.5	13	7.9	1635	373		0.22	600			37.0		689	4951	5640	324		8.3	15350			4.1	640	71		0.25					
Rowe 1995	834	9.1	1	6.3	9536				90								521						144	989								
Rowe 1995	834	9.1	2	6	9785	7462		0.76	131								568						192	1049								
Rowe 1995	834	9.1	3	5.8	15130	7151		0.47	241								982						487	1520								
Rowe 1995	834	9.1	4	6.1	8960	7865		0.88	149	187							489						145	985								
Yildiz 2001	834	9.1	6	6	14222	8716		0.61	271	307	339			381			1700	10	11.1	6051	974	431	265	1435	325	0.27	1.75	0.073	0.062	0.016	0.009	7.8
Yildiz 2001	834	9.1	7	6.3	9076	5390		0.59	194	212	221			937			1441		9.3	4300	897	348	40	954	284	0.19	1.23	0.023	0.054	0.04	0.002	8.3
Yildiz 2001	834	9.1	8	6.3	16271				249	282	321			425			2060	0	14	7094	1376	537	243	1579	429	0.28	0.94	0.03	0.21	0.04	0.011	9.4
Yildiz 2001	834	9.1	12	7.1	5425	3778		0.70	430	464	472			166			1471	16	10.8	3715	976	456	54	487	227	0.22	0.77	0.035	0.12	0.013	0.007	3.8
Yildiz 2001	834	9.1	14	7.1	13372	8115		0.61	971	1114	1114			750			3050	153	17.8	8403	2011	925	225	632	429	0.69	7.9	0.014	0.17	0.011	0.026	6.7
Akyol 2005	712	14.4	2	7.1	30798	18404		0.60	1586		4536	42.7						756	38.3	8710			46				76.7	11.1	9	6.8	0.03	
Akyol 2005	712	14.4	4	7.5	22734	14381		0.61				23.5		1986					43.8				35.2				24.7	14.41	7.974	7.489	0.143	
Noerfitriyani et al. 2017	3332	26	33	7.8	6860	3960		0.58			373			72																		
Silva et al. 2013	1178	14.4	12	8.1	7426	1325		0.18	3864	3940	4080	35		126			3549	42		17100			4.8									
Indelicato et al. 2018	605	18.4	16	8.03	15039	4460		0.30	3720			7.3		1377			5540		25.7				4.96			3.08	4.55	1.18	0.005	0.005	0	1.23
Indelicato et al. 2018	605	18.4	18	8.31	10624	3790		0.35	3219			13.3		1214			5087		29.1				6.43			0.3	0.87	0.384	0.005	0.012	0.003	0.12
Indelicato et al. 2018	605	18.4	20	8.13	9860	4946		0.50	3523		3886	28.2		1266			5205		30.4				7.11			0.26	0.48	0.106	0.005	0.05	0.003	0.12
Indelicato et al. 2018	605	18.4	22	8.4	12519	5392		0.36	2634			15.1		1194			3317	50	24.8				6.79			0.2	0.4	0.149	0.005	0.027	0.0005	0.16
Sang et al. 2010	413	10.1	20	7.2	1800	970	443	0.54	587	753	933										2224	1323	9.6			0.02	4.86	0.54	0.36	0.38	0.02	0.4
Zhong et al. 2009	961	16.4	15	7.9	19170				1971			8.2								11693												
Reis et al. 2017	1370	21.3	8	8.6	4184				1476	1922	1925	26.6					2503		20.3	7370												

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/ COD	NH4 -N	TKN	TN	TP	PO4 -P	TSS	TDS	TS	Cl	SO4	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Peng et al. 2008	610	12.1	8	7.6	10525	4920		0.47	1550		1950	12	10.5							9500			1.8			0.74			0.8			
Zakaria et al. 2015	2379	27.5	14	7.9	2180	227		0.10	1897						4855				8.2													
Abu Amr et al. 2018	2073	27.2	2	8.6	2300	110		0.05	870					88					18.9													
Cheibub et al. 2014	1299	23.2	34	8.3	3332	141		0.04	1998					53	9337	9390	3196			8857												
Bu et al. 2010	562	3.4	4	6.7	41800	19400	14060	0.46	2250		2520			5400	28800	34200																
Di Iaconi et al. 2006	642	13.9	12	8.4	24400		8870		3190	3380			14	1540			4610	300	40	12000	3930	3380	7.5		118	0.4	0.80			0.03	0.002	
Di Iaconi et al. 2010	642	13.9	15	8.5	9700	1500		0.15	2600	2700		27		390			6000	1800	21	16100	4200	1800		240	212	0.6	0.5	1	1.2	0.7	0.1	0.2
Ganigue et al. 2009	560	15.7	19	8.1	4357	810	1946	0.19	3772	4058	4060								68.1	7170												
Vilar et al. 2011	1087	14.7	10	7.5	11839	8580		0.8	2460	2870	2890	10.5	2.5	1260	20200	21460	2400	2000	24.3				61.4									
Khattabi et al. 2002	936	9.6	24	6.7	932	106		0.09	141								496	187	4.7	1564			5.8			4.41	1.08	0.885				
Frascri et al. 2004	719	13.7	4	8.3	3800	1300		0.34	950			10					2650		18.0													
Frascri et al. 2004	719	13.7	7	8.4	4700	1600		0.34	1400			26					2750															
Ntsele et al. 2000	1000	20.5	19	10	600	245		0.41	230	262	294		0.2	77	3389	3655	1383	33	6.7	1272	658	371	4.8	91	65	0.1	0.13	0.075	0.05	0.075	0.025	1.68
Kurniawan and Lo 2009	2122	22.6	12	8	7587	493	2286	0.07	2221										8.3	11523						2		9	11.4	5.2	0.22	
Istirokhatun et al. 2018	2439	25.4	6	8.4	1781									178	4305								9.8					0.11				
Tulun and Bilgin 2017	354	11.8	9	7.4	18000	11000	4511	0.61			3367									9520												
Statom et al. 2004	1493	23.6	4	7.3	820	73		0.09	460	610		0.97			3800		790	1.8	9.5				2.14				0.04		0.021			
Statom et al. 2004	1493	23.6	5	7.6	1200	34		0.03	630	660		3			4100		760	0.7	10.5		920		3.4				0.09		0.027			
Statom et al. 2004	1493	23.6	9	7.2	875	45		0.05	340	575		3.5			3215		720	37	7.3	2495	565		3.2			0.06	0.1		0.024			0.14
Statom et al. 2004	1493	23.6	7	6.84	741	84.1		0.11	403.5	640.5		3.23			4367		1090	42	9.2				2.81			0.05	0.12		0.037			
Ai et al. 2019	1236	17.2	15	8.5	2080	180		0.09	2875								2700			9500												
Erabee and Ethaib 2018	1987	27.1	3	8	4800	1260		0.26						273	12440	12713						19					3.4	0.09		0.14	0.11	0.67

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/COD	NH ₄ -N	TKN	TN	TP	PO ₄ -P	TSS	TDS	TS	Cl	SO ₄	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Erabee and Ethaib 2018	1987	27.1	9	8.1	6400	1125		0.18						230	11300	11530							79				6.5	0.37		1.2	0.31	1.9
Raghab et al. 2013	183	20.6	10	8.2	8250	3400		0.41	3745					3330	26612	29942	6365															
Çeçen and Gürsoy 2000	557	15.5	1	7.8	14807	7928		0.54	857	1044							4983			9843			9.4			2.5	0.5	0.16	0.47	0.82	0.14	0.46
Bila et al. 2005	1299	23.2	24	8.2	3096	130		0.05	775								4635				2950	1800	6.8	280	85	0.18	0.3	0.09	0.15	0.05	0.005	0.13
de Moraes and Zamora 2005	1390	17.1	15	8.4	5200	720	1058	0.14		1114			11.3			1212	2590			1240	1512	1480	13.2	11	9	1.4	1.1	0.36	0.45	0.28		0.3
Ahn et al. 2002	1219	11.7	8	7.5	3500	1625		0.46	1350	1715	1715	6.5		1050						4560												
Park et al. 2001	1200	12.5	2	7.2	22500	5180		0.23	2205	2940	2940	64					3500	320			2400	1640	142	1410	294	1	3		1		2	27
Tüylüoğlu 2001	942	14.1	1	6.3	49025	24067		0.49	842	903		5.3		2285			4160	1283						115		0.45	0.7	0.4	1.3	0.29	0.3	
Tüylüoğlu 2001	942	14.1	3	6.8	33567	24767		0.74	1361	1855		39		872	24933	25805			30.2				40				1.38		1.68	0.67	0.09	
Tüylüoğlu 2001	942	14.1	4	7.5	17353	12317		0.71	1631	1963		30		503	17516	18019			30.5				36				0.76		0.61	0.54	0.08	
Tüylüoğlu 2001	826	13.3	3	7.4	22974	15156		0.66	1795	2109		8.3		577	16867	17444	2987		27.8	9155			70			0.87	1.1	0.17	0.51	0.78	0.18	4.3
Tüylüoğlu 2001	826	13.3	4	7.6	18578	11816		0.64	2180	2370		16		1066	21288	22354	4402		34.4	10658			35			0.58	1	0.15	0.29	0.64	0.08	
Tüylüoğlu 2001	687	16.7	1	7.5	14900	6900	4550	0.46		2580							6330	142		13050												
Tüylüoğlu 2001	687	16.7	2	7.8	19070	11000	5770	0.58	1470	1610	1765		48		22570		5620	352		7040			14.2	97	640	0.45	0.38	0.02	0.01	0.02	0.005	0.11
Şahinci 2014	600	13.5	2	8.1	2930					551		1.6		722									2.5				0.69	0.02	0.05	0.165	0.002	
Şahinci 2014	600	13.5	3	8.0	1731					803		2.4		70									0.9				2.13	0.023	0.05	0.025	0.002	
Shehzad et al. 2016	2766	27.2	22	8.4	550	37		0.08	3330					207																		
Steiner et al. 1979	1124	11.8	6	7.1	11210	4460		0.40	1966				2.8	1994	11190	13184	4816	114		5685	1177	969	245	260	158	0.53	8.7	0.44	0.16	0.52	0.04	
Steiner et al. 1979	1124	11.8	8	7.3	21836	11359		0.52	883				3	1730	13181	14911	3101	428		4830	1457	968	176	290	60.7	1.27	11	0.32	0.22	0.45	0.1	
Steiner et al. 1979	1124	11.8	7	6.6	20032	13000		0.65	724	760			2.6	549	14154	14703	4395	683		5620	1386	950	378	358	110	1.98	31	0.39	0.43	0.81	0.09	
Hilles et al. 2016	386	19.9	19	8.2	19814	1326		0.07	2925						23826		6552	818	40.8	23250	6000	4346		1620	785	4.6	5.8	0.44		0.14	0.26	0.08
Bashir et al. 2017	2435	27.6	3	8.0	6650	1260		0.24	700					410					21.7				3			0.18	1.2	0.18		0.03	0.001	0.31
Irene Lo 1996	2125	22.7	15	7.9	750	81	230	0.11	760	960			2.5	480			770	1			470	190	15	43	31	0.06	0.29	0.08	0.35	0.1	0.01	0.44
Irene Lo 1996	2125	22.7	2	6.4	6610	1600	1565	0.24	1500	2000			10.0	1000			3400	39								0.1	1	0.02	0.12	0.04	0.01	
Zhong et al. 2017	1402	17.7	12	7.8	6912	565	3415	0.08	1845												2139	1399	1.7	184	236	0.2		0.003		0.021		0.3

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/ COD	NH4 -N	TKN	TN	TP	PO4 -P	TSS	TDS	TS	Cl	SO4	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Wang et al 2014	1402	17.7	10	8.4	1888	143	803	0.08	2100										21.2													
Heavy 2003	873	9.1	42	7.9	625	190		0.30	218				2.1				557															
Renou et al. 2008b	756	12.9	11	8.6	1339	80	480	0.06	468	470							2400	398	18.5	5700	1934		2.5	117	99							
Renou et al. 2008b	756	12.9	11	8.4	1342	88	509	0.07	370	373							1420	104	7.8	3500	942		23	104	45							
Renou et al. 2008b	756	12.9	11	8.6	1634	81	652	0.05	727	730							6740	564	32.2	9900	5840		2.7	43	65							
Secil 2007	826	13.3	1	5.9	45000	22000		0.49	150					3000																		
Secil 2007	826	13.3	2	6.7	31000	18000		0.58	850					700																		
Silva et al. 2004	1299	23.2	23	8.4	3465	150	910	0.04	775								4635				2950	1800	6.8	280	85.0	0.18	0.30	0.09	0.15	0.05	0.005	0.13
Kargi and Catalkaya 2011	700	17.6	1	7.8	5440				168		352	1.7		215	5872	6087			8.0													
Batarseh 2006	1514	23.7	12	7.2	1313	9		0.01	443								1061			3200												
Batarseh 2006	1514	23.7	20	7.6	1842	36		0.02	902								2234			4500												
Batarseh 2006	1514	23.7	17		2019	51		0.02	1240								2630															
Batarseh 2006	1514	23.7	10		1641	32		0.02	904								1520															
Batarseh 2006	1514	23.7	6		1646	34		0.02	667								1298															
Park et al. 2005	1200	12.5	7	7.5	4601	582			1740			14.0		824			1800	41	14.3	8035	1707	1123		146	156							
Park et al. 2005	1319	12.5	9	7.9	3898	682	3322	0.17	860			11		815			2156	6	10.2	4523	1445	892		41	178							
Park et al. 2005	1319	12.5	8	7.4	2373	371	2138	0.16	504			10		323			257	21	4.7	2199	380	250		155	54							
Park et al. 2005	1319	12.5	7	7.7	3372	659		0.2	1462			13		1910			3070	56	15.5	5306	1957	1239		194	187							
Park et al. 2005	1319	12.5	4	7.7	4038	385	1715	0.10	168			10		2350			2478	15	10.5	6125	2470	1345		107	157							
Park et al. 2005	1319	12.5	2	8.3	1840	342	1490	0.19	1075			8		2320			1294	80	17.5	10113	1117	712		297	142							
Park et al. 2005	1319	12.5	9	7.4	649	64	465	0.10				1		270			1620	70	8.4	3713	1528	968		179	118							
Park et al. 2005	1319	12.5	2	7.5	753	45		0.06	470			8		240			458	8	5.5	1743	346	226		143	47							
Park et al. 2005	1319	12.5	5	8	492	86		0.17	689			9.0		150			1431	90	8.5	2905	1582	1395		450	116							
Park et al. 2005	1319	12.5	4	8	4109	1058		0.26	1176			11		1790			667	11	15.7	3757	550	436		87	79							
Kulikowsk a and Klimiuk 2008	712	7.4	2	7.8	1800	720		0.40	98		127	8.5	6.1	405	4576	4981	954	224						342	281	0.04	0.29	0.03	0.06	0.92	0.009	

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/COD	NH4 -N	TKN	TN	TP	PO4 -P	TSS	TDS	TS	Cl	SO4	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Kulikowska and Klimiuk 2008	712	7.4	5	8.1	610	79		0.13	364		401																					
W12A landfill	834	9.1	1	6		6360			168	202	203				7705		478	354	7.1		450		247									8
W12A landfill	834	9.1	3	6		12100			337	450	451				11759		778	476	10.6		604		450									15
W12A landfill	834	9.1	4	6		10200			518	847	848				12671		819		9.9		635		389									18
W12A landfill	834	9.1	28	7.3	810	115		0.14	530	701	702	1.9					1099	327	8.9	3478	692		14.2	154	179	0.13	0.47	0.02	0.03	0.015	0.01	0.33
W12A landfill	834	9.1	10	7.2		2932			731	781							1377		11.6				95									2
W12A landfill	834	9.1	16	7.3		846			880	1000							1295	18	11.1				33.5	175	284	0.25	0.92	0.085	0.07	0.314	0.025	1.01
W12A landfill	834	9.1	22	7.4		1145			607			2.9					1058	588	11.7		850		21	387	237	0.2	0.48	0.043	0.07	0.185	0.019	0.51
W12A landfill	834	9.1	25	7.6	1173	358		0.31	530			5.2					940	860	9.5	3755	759		35	492	258	0.06	0.36	0.045	0.03	0.045	0	0.47
W12A landfill	834	9.1	30	7.5	760	79		0.10	551	616	617	1.5					899	30	9.4	3482	645		13.9	139	179	0.08	0.16	0.02	0.020	0.02	0.005	0.22
W12A landfill	834	9.1	34	7.2	232	95		0.41		221	227	1.0					692	38	6.4	1743	359		7.1	151	95.0	0.04	0.03	0.01	0.010	0.01	0.01	0.31
Cataldo 2012	798	15.7	7	8.7	5550								31.6				2519	109	10.9		300	450	5.9	154	45.0	0.18	0.31	0.03	0.350	0.045	0	0.75
Robinson and Barr 1999	680	9.9	34	6.9	229	50		0.22	168				0.1				356		3.4				17.8				0.10					
Robinson and Barr 1999	1181	9.3	9	7.1	1000	210	299	0.21	541								1070		10.3	4540	750	469	10.4	249	206		0.53					
Robinson and Barr 1999	1000	10.1	25	7.5	1070	232	393	0.22	167								634		5.8	2565	621	329	25.0	200	134	0.08	0.74	0.05	0.03	0.02	0.005	2.33
Gelaziene 1999	632	6.1	33	8.0	690	50		0.07	480			1.5					5100									0.550		0.6	1.70	0.2		
Ivan et al. 2019	985	25.8	12	8.3	5346	1098	2857	0.21	1210	1419		37.0		95	12450	12545	3156		17.9	4305	11850	10252	64.1			0.350	3.20	0.21	6.98	0.24	0.007	0.81
El Ouair et al. 2017	466	18.1	15	8.4	23926	2841	10920	0.12	2859	4144	4559								43.5				7.7	371	267	0.3	0.47	0.37	0.92	0.48		
leachless project 2018	849	14	<5	8.2	29219	12250		0.42	4239	5320	5324	148	23.1	3105	26619	29724	7870	9.6	42.8		5618	3173	6.4	543	161	0.37	0.99	0.51	7.2	0.11	0.004	0.24
leachless project 2018	849	14	<5	8.2	13533	12690		0.94	5246	5706	5709	74	24.1	159	25235	25394	6310	213	44.7		4832	2514	5.6	48	195	0.67	1.27	4.13	1.6	0.1	0.003	0.15
leachless project 2018	849	14	<5	7.6	26592	11912		0.45	2302	3573	3608	75	56.7	639	36222	36861	10300	30	44.8		6869	2960	6.8	184	113	0.25	2.68	3.18	0.85	0.07	0.029	0.21
leachless project 2018	849	14	<5	8	14194	4874		0.34	3991	4435	4438	97	29.2	399	28217	28616	7640	2250	43.2		4694	3129	17.3	79	162	0.89	1.16	0.65	2.1	0.15	0.076	0.36
leachless project 2018	849	14	<5	8.7	16849	2310		0.14	2702	3261	3264	149	58.7	186	33324	33509	9360	511	40.5		5511	4854	30	9	13	0.55	1.58	1.35	4	0.13	0.075	0.15

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/COD	NH ₄ -N	TKN	TN	TP	PO ₄ -P	TSS	TDS	TS	Cl	SO ₄	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
leachless project 2018	849	14	5-10	7.7	12554	5550		0.44	1386	2099	2102	45.4	10.7	595	14166	14761	3140	66	16.2		1435	1430	14.9	810	187	0.46	10.7	3.28	1.1	0.09	0.003	0.75
leachless project 2018	849	14	5-10	7.8	13701	4100		0.30	3084	3640	3643	94.4	48.6	118	27372	27489	7240	81	36.9		4802	3197	9.5	70	66	0.55	1.7	1.93	2.7	0.13	0.002	0.13
leachless project 2018	849	14	5-10	8.3	10026	1184		0.12		3351	3386	64.2	25.1	198	20361	20558	4940	30	32.2		3451	1932	9.2	97	90	0.31	1.6	2.57	0.47	0.04	0.028	0.31
leachless project 2018	849	14	5-10	8	8938	2307		0.26	2000	2546	2550	56.9	29.3	154	19076	19229	5780	8	29.4		2541	2764	9.6	175	99	0.39	0.86	1.07	2.1	0.09	0.075	0.75
leachless project 2018	849	14	5-10	8.4	13646	2450		0.18	2217	2925	2928	90.2	20.2	257	31923	32180	8970	824	39.3		4847	4892	4.4		223	0.58	0.38	0.53	13.8	0.08	0.072	0.36
leachless project 2018	849	14	>10	8.1	2725	632		0.23	757	1506	1541	38.7	19.6	97	9004	9100	2440	99	15		1625	715	4.1	121	66	0.21	1.13	5.36	0.44	0.05	0.029	0.41
leachless project 2018	849	14	>10	8.2	3460	164		0.05	486	1366	1401	37.8	55.8	106	11089	11195	2480	30	17.1		1900	1601	15.8	49		0.34	2.17	3.32	1.03	0.11	0.079	0.27
leachless project 2018	849	14	>10	8.4	4777	443		0.09	378	978	981	50.4	18.1	111	15333	15443	4740	627	17.7		2328	2262	34.7	95	39	0.51	1.77	1.36	1.45	0.15	0.076	2.7
Septiariva et al. 2019	2164	23.3	9	8.8	5200	1071		0.21	1659	2324			2.8	1950	11600	13550			19.3													
Shouliang et al. 2008	1232	18.4	3	6.7	53200	39900		0.75	2760				1.6				6150	163	31.5													
Abuayyash et al. 2018	250	19.5	2	6.1	11000	4000		0.36	105				8	2500	2000	4500			6		5700	1000		3500	300	5.2	3.4	0.64	5.2	0	3.7	
Mendez et al. 1989	1325	13.6	20	7.6	830				280				120	200						1900												
Mendez et al. 1989	1025	14.2	1	7.3	4100				800					1200						3160												
Somani et al. 2019	766	26.7	17	7.4	18184										16809		9169	693	15.2	11652			13.2			1.02	4.2	0.35	0.32	0.46	0	0.59
de Almeida et al. 2019	1354	23.5	6	7.8	4137	290	2154	0.07	1236					520	15110	15630	1109		17.5													
Rizkallah et al. 2013	686	15.2	7	7.3	17760	10935		0.62	3600		4200	39.5	25.5	635	19100	19735	7800		38.2	15460						0.48	0.05	0.007	0.46	0.007	0.008	0.19
Zhao et al. 2019	961	16.4	1	6.6	24200				500								104							4004	1015							
Zhao et al. 2019	961	16.4	24	8	7580				2000								510							2433	967							
Lee et al. 2010	785	8.3	5	6.5	3641	2032		0.31	289	475	478						894	197			764		32.1	421	154							
Lee et al. 2010	785	8.3	15	5.7	875	196		0.20	260	304	306						735	82			688		11.2	182	133							
Visvanathan et al. 2007	1530	28.1	9	8.4	4300	418		0.10	1934	2186					18900				41				52.7				2.28	0.11		0.1	0.01	
Kiddee et al. 2014	536	16.4	6	7.9			414								6600				11.9				1.2			0.01	0.01	0.022	0.026	0.0005	0	

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/COD	NH ₄ -N	TKN	TN	TP	PO ₄ -P	TSS	TDS	TS	Cl	SO ₄	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Lacerda et al. 2014	1056	23	4	7.7	1153				1235					101	6396	6497	2305		13.5	5908	1230	1114		134				0.07	0.04		0.05	
Lacerda et al. 2014	1056	23	3	7.6					606								1650		7.8	3670	700	395										
Yusof et al. 2009	2458	27.1	1	5.3	44081	29426	16828	0.67	1197	1413	1430	20					2331			6278	700	629	133	133 ₃	498	0.46	3.21	0.163	0.843	0.221	0.011	23.1
Bakraouy et al. 2017	555	17.9	8	8.4	11520	6710		0.58						3126																		
Tsarpali et al. 2012	700	17.3	5	8.4	3248	1114		0.34	374	485	498		9.4	249	4118	4367	2113	265	6.6								0.69	0.36	1.75	1.67	0.1	0.84
Jensen and Christensen 1999	642	8	19	6.7			120		260				2.5				1440	15	7.1	1657	665	705	1.5	145	63	0.08	0.36	0.007	0.003	0.016	0.0004	0.25
Jensen and Christensen 1999	758	7.6	18	7.3			190		580				1.5				1060	7	8.9	2981	560	590	22	130	100	0.05	0.09	0.034	0.056	0	0.0003	0.5
Jensen and Christensen 1999	831	8.3	13	7.5			540		820				6				1300	11	12	4786	720	870	21	315	125	0.06	5.31	0.002	0.19	0.002	0.004	2
Baig et al. 1999	774	9.7	23	8.2	1585	113		0.07	762					57			1407				970	280	9	155	144							
Baig et al. 1999	715	12.7	16	7.6	782	117		0.15	353	406				131			1100				720	333	2.5	115	80							
Baig et al. 1999	715	12.7	5-10	7.9	2915	758		0.26	235					136			850							240								
Baig et al. 1999	715	12.7	4	7.6	5940	3623		0.61	460					1224										620								
Baig et al. 1999	715	12.7	17	7.5	1057	106		0.10	630					20										114								
Kromann et al. 1998	658	7.6	>10	7.5	2390	150		0.06	490								2800	9.7	16		1490	715	3	330	115							1.3
Kromann et al. 1998	658	7.6	>10	7.1	2310	790		0.34	530								4560	42	18		1940	855	0.3	400	140							0.1
Kromann et al. 1998	658	7.6	>10	6.9	1400	630		0.45	705								580	11.5	4.2		530	595	9.1	195	110	0.23	0.05	0.22				4.4
Kromann et al. 1998	658	7.6	>10	7	615	27		0.04	840								1070	15	28.4		2840	2645	7.8	200	250	0.08	0.1	0.04				1.2
Slomczynska and Slomczynski 2004	742	5.8	3	5.9	9425	5250	1700	0.56	227	250	250	7.7		466	3694	4160	370	100	3.4	1960	206	187	234	400	54.0	0.03	0.65	0.022	0.06	0.01	0	
Slomczynska and Slomczynski 2004	969	10.9	9	5.4	38800	24500		0.63			630	11.3		310						6510			810			1.2		1.3	1.1	1.4	0.03	
Slomczynska and Slomczynski 2004	801	4.9	4	6.9	1080	870	250	0.81	225	254	254	1.7		397	2333	2730	280	10	3.2	2050	229	172	78	198	96	0.02	0.1	0.011	0.035	0.001	0	

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/ COD	NH4 -N	TKN	TN	TP	PO4 -P	TSS	TDS	TS	Cl	SO4	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Slomeczyns ka and Slomeczyns ki 2004	969	10.9	7	6.4	3800	2460		0.65			56	5.9		220						1280			245			0.1	5.3	0.18	0.05	0.05	0.01	
Slomeczyns ka and Slomeczyns ki 2004	550	7	2	6.5	19721	8979		0.46	693		769	8.3		600			1330															
Slomeczyns ka and Slomeczyns ki 2004	550	7	17	7.5	4981	1941		0.39	1673		1850	13.6		263			6000															
Cabeza et al. 2007	1200	14	14	8.4	4434	640	2782	0.14	1934				55	317			3235	39	12.8		2402	1476	4.6	108	88	0.19	0.15		0.61	0.01		1.17
Yu 2007	526	6.9	27	8	1258	580		0.46	268		296		7.9	146			1440	150	0.8	2020				228	2	0.1	0.3	0.139	0.092	0.015	0.001	
Mishra et al. 2016	2623	27	1	7.8	6444	3391		0.53				82.4		1356	18367	19723	3584	854	23	10922	1524	192	166	176	272	0.38	7.59	3.62	0.84			1.6
Mishra et al. 2016	2623	27	7	7.7	3425	1458		0.43				57.2		378	7298	7676	912	1183	13.7	6052	2586	186	159	461	384	0.05	6.32	2.81	0.31			2.96
Hermosilla et al. 2009	468	12.3	16	8.3	6119	100	1481	0.02	1965						16360		4430		32.7	14443			6.6									
Reinhart and Grosh 1998	1497	23.6	14	6.9	621	28		0.05	498								983										0.2					0.38
Reinhart and Grosh 1998	1497	23.6	7	7.2	1120	200		0.18	471								952	7.3									0.29					
Jia et al. 2011	1236	17.2	7	8.2	2440	225	914	0.09	1359					164	10856	11020	2160	280	12.2	2326	1324	1362	2	38	158	0.22	0.68	0.06	0.93	0.56	0	0.35
Zhang et al. 2011	1236	17.2	6	8.1	2400	370		0.15	2470								2900			10500												
Boateng et al. 2018	1145	25.9	13	7.8	6249	396		0.06	271				349		4558			17														
Kapelewsk a et al. 2019	550	7	36	8.1	1731	195	385	0.11			718	7.4	3.7			292			7.2													
Karimipou rfarid et al. 2019	316	16.8	19	8	2600	980		0.38	2560								5850	1555	36.7		1946	1004		676	64							
Baettker et al. 2020	1500	17.5	6	8	3162	916		0.29	2628							12858			32.2	12590			5.6			0.42	0.5	0.05	0.05	0.05	0.08	0.32
Baettker et al. 2020	1500	17.5	7	8.1	3078	1400		0.45	1792	1856		7.9		148	8127	8275	1890	25	26.3	7133			8.1			0.1	0.5	0	0.2	0.005	0.001	0.5
Baettker et al. 2020	1500	17.5	8	8.1	8621	5000		0.56	2574	2680		32.6		731	18360	19091	2330	38	26.6	10818			49			0.18	1.2	0.084	0.31	0.016	0.002	1.87
Corsino et al. 2020	605	18.4	4	7.5	11137				923			3.2							20.1													
Zegzouti et al. 2019	250	19.6	1	6.8	19523	11600		0.59	5931	6335	6370	65.1		1927					66.8							0.12	0.17		0.06	0.19		
Zegzouti et al. 2019	246	18.6	6	8.1	14561	5300		0.36	5226	6149	6177	33.6		430					55.4							0.23	0.21		0.21	0.08		
Pereira et al. 2018	1354	23.5	6	7.4	3947				1452								3948		20.4													

Reference	AAP	AAT	Landfill age	pH	COD	BOD	TOC	BOD/COD	NH4 -N	TKN	TN	TP	PO4 -P	TSS	TDS	TS	Cl	SO4	EC	TA	Na	K	Fe	Ca	Mg	Ni	Zn	Cu	Cr	Pb	Cd	Mn
Chen et al. 2020	961	16.4	8	7.9	5001				1269	2191	2204						11443	535														
Weijin et al. 2018	636	14.5	4	6	43305		15490		2142																							
Ramirez-Sosa et al. 2013	985	25.8	14	8.3	10434	942		0.09						357	18770	19128																
He et al. 2009	1112	16.1	1	6.3	80750	42450	19800	0.53	2230			35					1750	1050		12750												
Tizaoui et al. 2007	466	18.1	6	8.7	5230	500		0.10				8.8					4870	76		18725						0.53	0.94	0.14	2.24			
Robinson and Maris 1985	632	9.8	1	6.0	5028	3035	1663	0.60	76	152	152		0.4	204			233				140	117	102	348	37	0.18	17.6	0.08	0.14	0.11	0.003	22.8
Robinson and Maris 1985	632	9.8	1	6.2	23800	11900	8000	0.5	790	800	803		0.7	539			1315				960.0	780	540	1820	252	0.57	21.5	0.12	0.56	0.4	0.031	26.5
Robinson and Maris 1985	632	9.8	21	7.5	1160	260	465	0.22	370	380	381		1.4	101			2080				1300	590	22.5	250	185	0.09	0.37	0.03	0.04	0.14	0.008	2.2
Knox and Jones 1979	838	7.1	4	6.9	7180														3.8	3190												
Kim and Lee 2009	1247	11.7	7		1552	926		0.60	887																							
Keenan et al. 1993	1108	9.3	2	6.3	5400	2340		0.43												1440			400	460		0.63	0.41	0.15	0.06	0.45	0.18	37.5
Adhikari et al. 2014	1505	18.1	8	7.8	2195	344		0.16																								
Adhikari et al. 2014	3474	20.6	9	6.6	8999	952		0.11																								
Chang 1989	1700	22.1	2	5.5	58400	40065		0.69	1720	1887		63				43840	3600	1830		8530	2820	2470	570	3090	1000		9.5	0.32	5	1.3	0.2	5.1

AAP: Annual Average Precipitation (mm) AAT: Annual Average Temperature (C°)

Table A.2 List of references for leachate database (360 active sanitary landfills)

Reference	Number of data from that reference	Citation
Abood et al. 2014	1	Abood, A. R., Bao, J., Du, J., Zheng, D., Luo, Y., 2014. Non-biodegradable landfill leachate treatment by combined process of agitation, coagulation, SBR and filtration. <i>Waste Management</i> . 34(2), 439–447.
Abu Amr et al. 2018	1	Abu Amr, S.S., Alkarkhi, A.F.M., Alslaibi, T.M., Abujazar, M.S.S., 2018. Performance of combined persulfate/aluminum sulfate for landfill leachate treatment. <i>Data in Brief</i> . 19, 951-958.
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Adhikari et al. 2014	2	Adhikari, B., Dahal, K.R., Khanal, S.N., 2014. A Review of Factors Affecting the Composition of Municipal Solid Waste Landfill Leachate. <i>International Journal of Engineering Science and Innovative Technology (IJESIT)</i> . 3(5), 273-280.
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Aghasa et al. 2017	1	Aghasa, Helmy, Q., Chaerul, M., 2017. Influence of Organic Loading and Mixing to the Stabilized Leachate COD Removal Using Circulating Anaerobic Reactor. <i>Reaktor</i> . 17(2), 59-66.
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Ai et al. 2019	1	Ai, J., Wu, X., Wang, Y., Zhang, D., Zhang, H., 2019. Treatment of landfill leachate with combined biological and chemical processes: changes in the dissolved organic matter and functional groups. <i>Environmental Technology</i> . 40(17), 2225-2231.
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Reference	Number of data from that reference	Citation
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Reference	Number of data from that reference	Citation
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Site data	1	Kastamonu, Türkiye
Site data	1	Konya, Türkiye
Slomczynska and Slomczynski 2004	6	Slomczynska, S., and Slomczynski, S., 2004. Physico-Chemical and Toxicological Characteristics of Leachates from MSW Landfills. <i>Polish Journal of Environmental Studies</i> . 13(6), 627-637.
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Uygur and Kargı 2004	1	Uygur, A. and Kargı, F., 2004. Biological nutrient removal from pre-treated landfill leachate in a sequencing batch reactor. <i>Journal of Environmental Management</i> . 71, 9-14.
Veli et al. 2008	1	Veli, S., Öztürk, T., Dimoglo, A., 2008. Treatment of municipal solid wastes leachate by means of chemical- and electro-coagulation” <i>Separation and Purification Technology</i> . 61, 82-88.
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Reference	Number of data from that reference	Citation
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Yalılı Kılıç et al. 2007	2	Yalılı Kılıç, M., Kestioğlu, K., Yonar, T., 2007. Landfill leachate treatment by the combination of physicochemical methods with adsorption process. <i>Journal of Biological & Environmental Sciences</i> . 1(1), 37-43.
Yang and Zhou 2008	1	Yang, Z. and Zhou, S., 2008. The biological treatment of landfill leachate using a simultaneous aerobic and anaerobic (SAA) bio-reactor system. <i>Chemosphere</i> . 72, 1751–1756.
Yıldız 2001	5	Yıldız, E.D., 2001. Development of modeling approaches for landfill leachate management. <i>Environmental Engineering, METU</i> , Ph.D. Thesis.
Yılmaz 2000	1	Yılmaz, G., 2000. Katı Atık Sızıntı Sularından Biyolojik Azot Giderimi. <i>ITU</i> , Ph.D. Thesis (in Turkish).
Yong et al. 2018	1	Yong, Z.J., Bashir, M.J.K., Ng, C.A., Sethupathi, S., Lim, J.W., 2018. A sequential treatment of intermediate tropical landfill leachate using a sequencing batch reactor (SBR) and coagulation. <i>Journal of Environmental Management</i> . 205, 244-252.
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Zakaria et al. 2015	1	Zakaria, S.N.F., Aziz, H., Abu Amr, S., 2015. Performance of Ozone/ZrCl4 Oxidation in Stabilized Landfill Leachate Treatment. <i>Applied Mechanics and Materials</i> . 802, 501-506.
Zegzouti et al. 2019	2	Zegzouti, Y., Boutafda, A., El Fels, L., El Hadek, M., Lebrihi, A., Bekkaoui, F., Hafidi, M., 2019. Quality and quantity of leachate with different ages and operations in semi-arid climate. <i>Desalin. Water Treat.</i> 152, 174-184.
Zhang et al. 2011	1	Zhang, H., Ran, X., Wu, X., Zhang, D., 2011. Evaluation of electro-oxidation of biologically treated landfill leachate using response surface methodology. <i>Journal of Hazardous Materials</i> . 188 (1-3), 261–268.
Zhang et al. 2013	3	Zhang, Q-Q, Tian, B-H, Zhang, X., Ghulam, A., Fang, C-R, He, R., 2013. Investigation on characteristics of leachate and concentrated leachate in three landfill leachate treatment plants. <i>Waste Management</i> . 33, 2277–2286.
Zhao et al. 2019	2	Zhao, R., Wang, X., Chen, X., 2019. Impacts of different aged landfill leachate on PVC corrosion. <i>Environ Sci Pollut Res.</i> 26, 18256-18266.

Reference	Number of data from that reference	Citation
Zhong et al. 2009	1	Zhong, Q., Li, D., Tao, Y., Wang, X., He, X., Zhang, J., Zhang, J., Guo, W., Wang, L., 2009. Nitrogen removal from landfill leachate via ex situ nitrification and sequential in situ denitrification. <i>Waste Management</i> . 29(4), 1347-1353.
Zhong et al. 2017	1	Zhong, H., Tian, Y., Yang, Q., Brusseau, M.L., Yang, L., Zeng, G., 2017. Degradation of landfill leachate compounds by persulfate for groundwater remediation. <i>Chemical Engineering Journal</i> . 307, 399–407.
Zin et al. 2012	1	Zin, N.S.M., Aziz, H.A., Adlan, M.N., Ariffin, A., 2012. Characterization of Leachate at Matang Landfill Site, Perak, Malaysia. <i>Academic Journal of Science</i> . 1(2), 317-322.
Ziyang et al. 2009	6	Ziyang, L., Youcai, Z., Tao, Y., Yu, S., Huili, C., Nanwen, Z., Renhua, H., 2009. Natural attenuation and characterization of contaminants composition in landfill leachate under different disposing ages. <i>Science of the Total Environment</i> . 407, 3385-3391.
Total number of data	360	

B. Outputs for Outlier Detection Process

Table B.1 Permanently removed data references from leachate database (bold ones are outliers)

Landfill ID	Reference	Country	Income Index	Ann. Avg. Precip. mm	Ann. Avg. Temp. C	Landfill name	Place	Landfill age	NH ₄ -N	TDS	Cl	EC	TA	Na	K
37	Dolar et al. 2016	Croatia	H	930	11	Jakuševac landfill	Zagreb	49			10484	8.6			
81	Boumechhour et al. 2013	Algeria	UM	736	17.1	Ouled Fayet landfill	Ouled Fayet	7	3159			0.5			
150	Aziz et al. 2015	Malaysia	UM	2599	27.5	Kulim Landfill	Simpang Empat	17	562			7.7	14667		
151	Aziz et al. 2015	Malaysia	UM	4000	27.3	Kuala Sepetang landfill	Taiping	16	564			9.7	16000		
168	Aziz et al. 2011	Malaysia	UM	2599	27.5	Kulim Landfill	Simpang Empat	13	600			8.3	15350		
196	Ganigue et al. 2009	Spain	H	560	15.7	Corsa landfill	Reus	19	3772			68.1	7170		
277	Ivan et al. 2019	Mexico	UM	985	25.8	Merida Yucatan landfill	Merida	12			3156	17.9		11850	10252
294	Abuayyash et al. 2018	Palestine	LM	250	19.5	Al-Menya Landfill	Bethlehem	2		2000				5700	

Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

Table B.2 Pearson correlation analysis of leachate parameters without removal of outliers ($r \geq 0.7$ is given in bold)

Parameters*	COD	BOD	TOC	NH ₄ -N	TKN	TN	TDS	TS	TA	pH	Cl	EC	Na	K	Fe	Ca	Mg
COD	1	0.948	0.966				0.695	0.799									
BOD	0.948	1	0.914														0.731
TOC	0.966	0.914	1				0.783	0.878									
NH ₄ -N	0.272	0.184	0.336	1	0.974	0.962			0.749			0.825					
TKN	0.329	0.223	0.462	0.974	1	0.999	0.761		0.787			0.811					
TN	0.355	0.200	0.344	0.962	0.999	1	0.829	0.832	0.769			0.854	0.880	0.769			
TDS	0.695	0.525	0.783	0.651	0.761	0.829	1	0.984			0.884	0.871					
TS	0.799	0.677	0.878	0.587	0.689	0.832	0.984	1			0.811	0.868					
TA	0.304	0.238	0.330	0.749	0.787	0.769	0.522	0.397	1			0.697					
pH	-0.452	-0.509	-0.440	0.317	0.312	0.338	0.114	-0.096	0.236	1							
Cl	0.327	0.243	0.139	0.589	0.639	0.687	0.884	0.811	0.615	0.281	1	0.788	0.775				
EC	0.547	0.410	0.512	0.825	0.811	0.854	0.871	0.868	0.697	0.192	0.788	1	0.699				
Na	0.314	0.071	0.082	0.522	0.633	0.880	0.583	0.486	0.516	0.305	0.775	0.699	1	0.883			
K	0.290	0.075	0.147	0.555	0.503	0.769	0.506	0.439	0.431	0.287	0.583	0.635	0.883	1			
Fe	0.456	0.500	0.357	-0.173	-0.215	-0.306	-0.107	0.147	-0.118	-0.648	-0.137	-0.104	-0.119	-0.083	1		
Ca	0.472	0.608	0.672	-0.116	-0.106	-0.165	-0.231	0.105	0.132	-0.494	0.071	0.201	0.301	0.205	0.488	1	0.841
Mg	0.566	0.731	0.628	0.066	0.059	-0.007	0.103	0.327	0.317	-0.381	0.128	0.282	0.300	0.285	0.417	0.841	1

*Total alkalinity (TA) is in mg/L as CaCO₃, EC is in mS/cm, and the other parameters are in mg/L

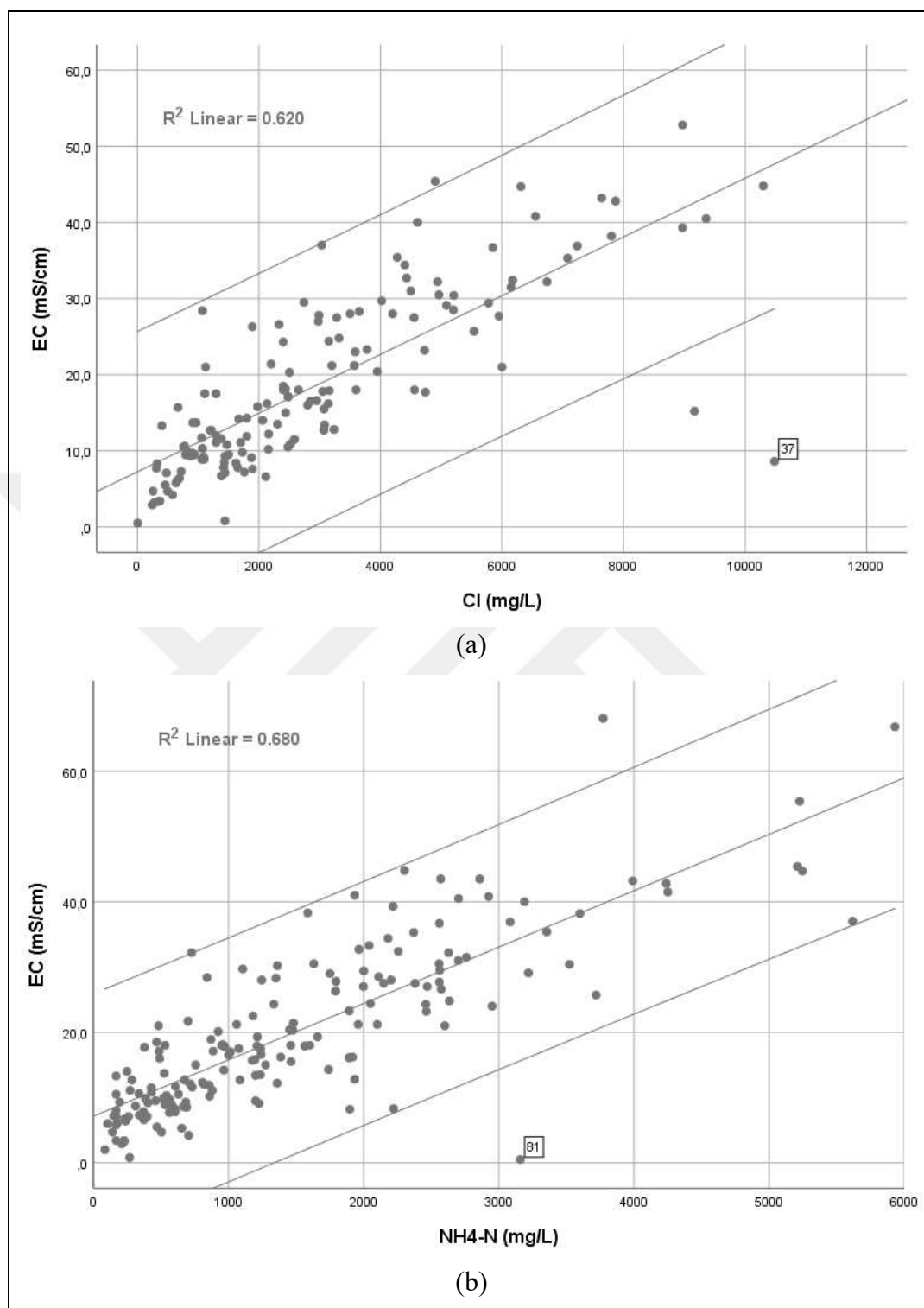


Figure B.1. Outlier data (circled) with scatter plots for (a) Cl vs EC (b) NH₄-N vs EC (the upper and lower lines show 99% prediction interval; the numbers within boxes indicate landfill ID)

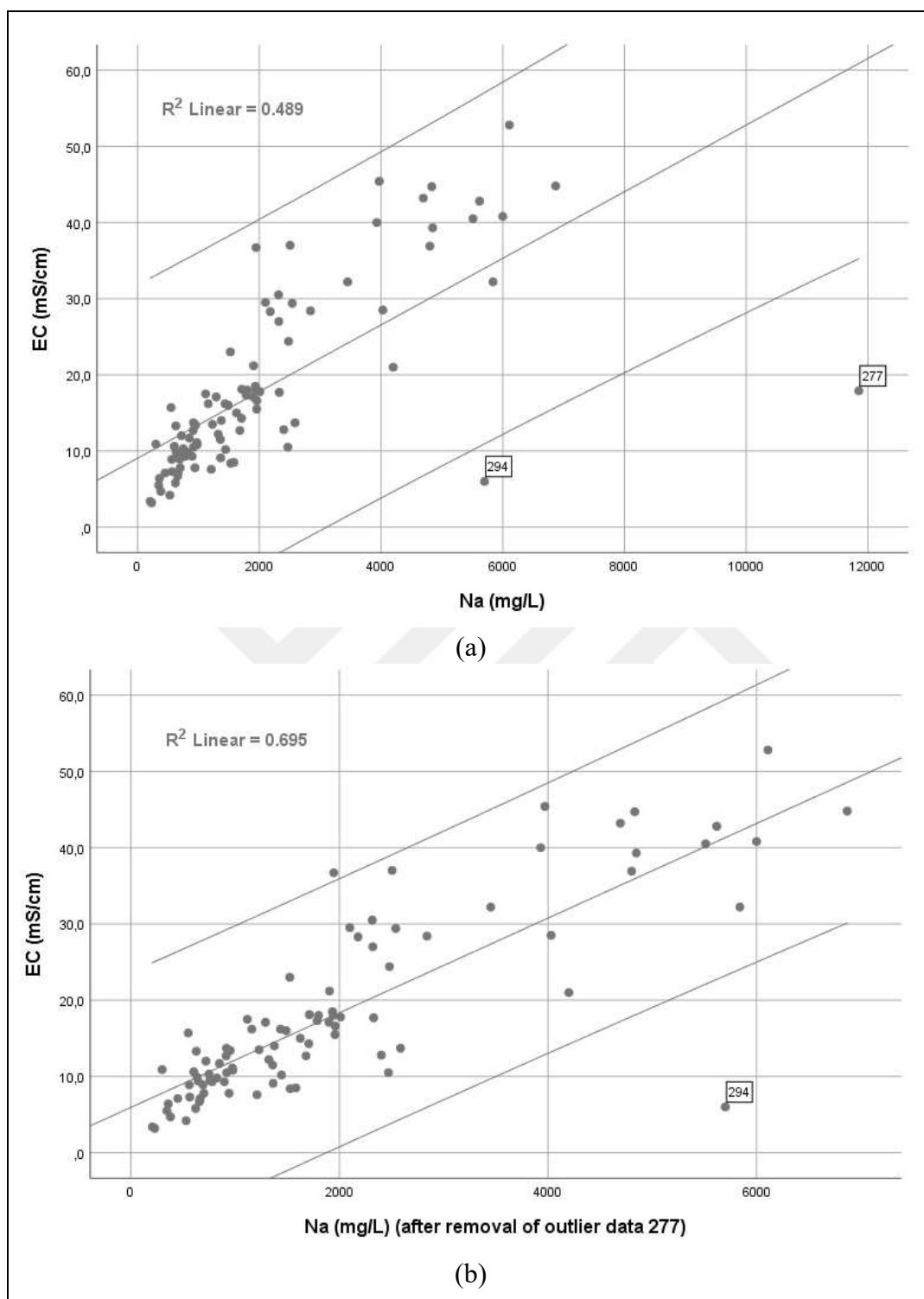


Figure B.2. Outlier data (circled) with scatter plots for (a) Na vs EC (b) Na (after removal of outlier data 277) vs EC (the upper and lower lines show 99% prediction interval; the numbers within boxes indicate landfill ID)

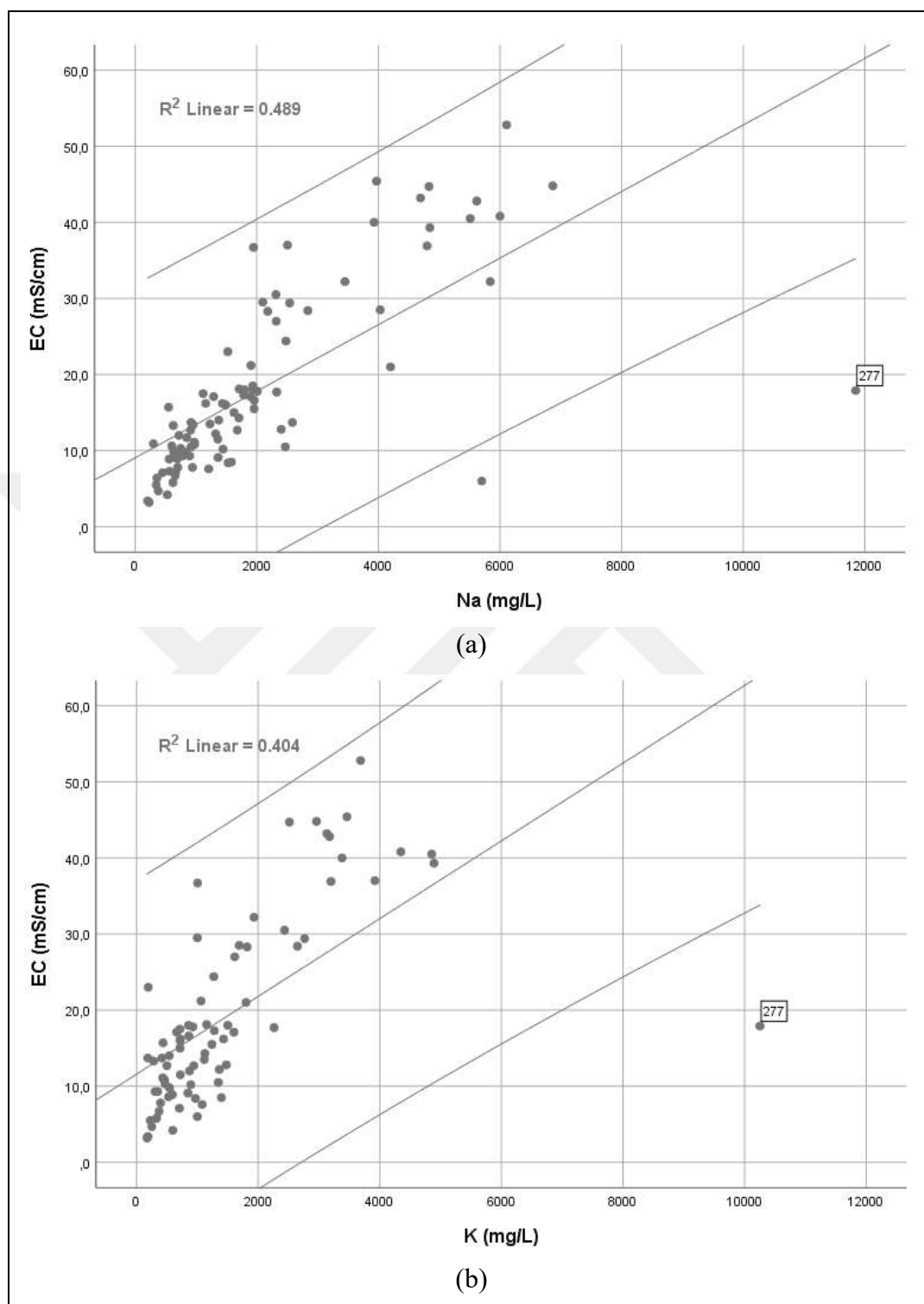


Figure B.3. Outlier data (circled) with scatter plots for (a) Na vs EC (b) K vs EC (the upper and lower lines show 99% prediction interval; the numbers within boxes indicate landfill ID)

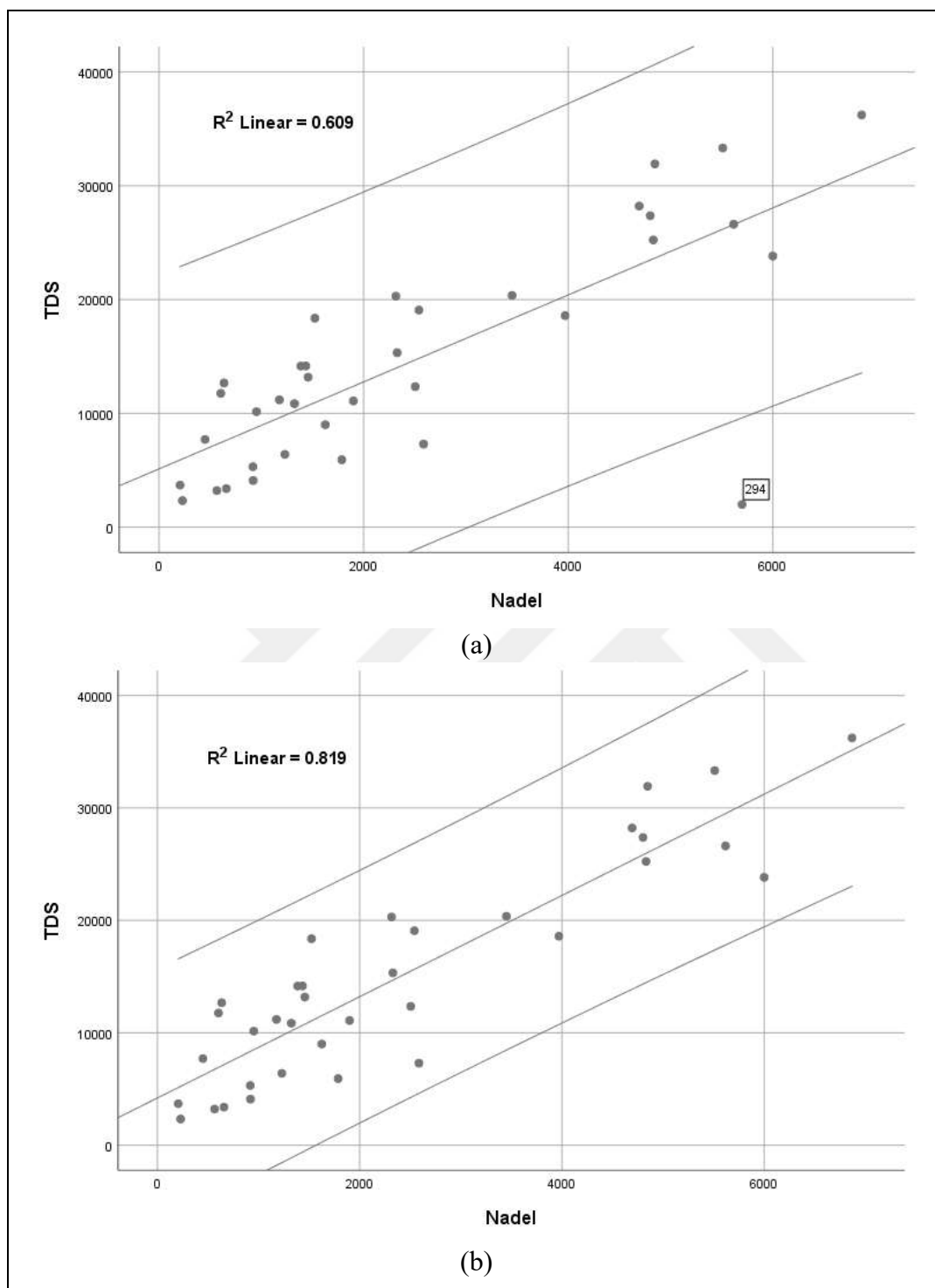


Figure B.4. Outlier data (circled) in scatter plot for Na vs TDS (a) with outlier data (b) After removal of outlier data (the upper and lower lines show 99% prediction interval; the numbers within boxes indicate landfill ID)

C. Plots Related to Regression Modeling

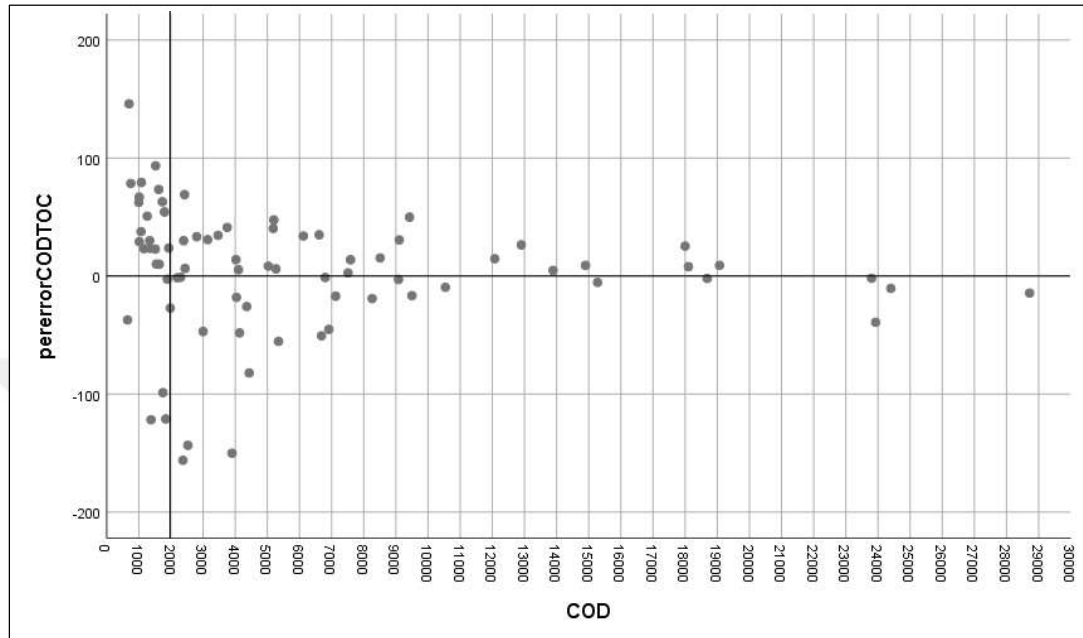


Figure C.1. Percent error chart for COD and TOC regression model

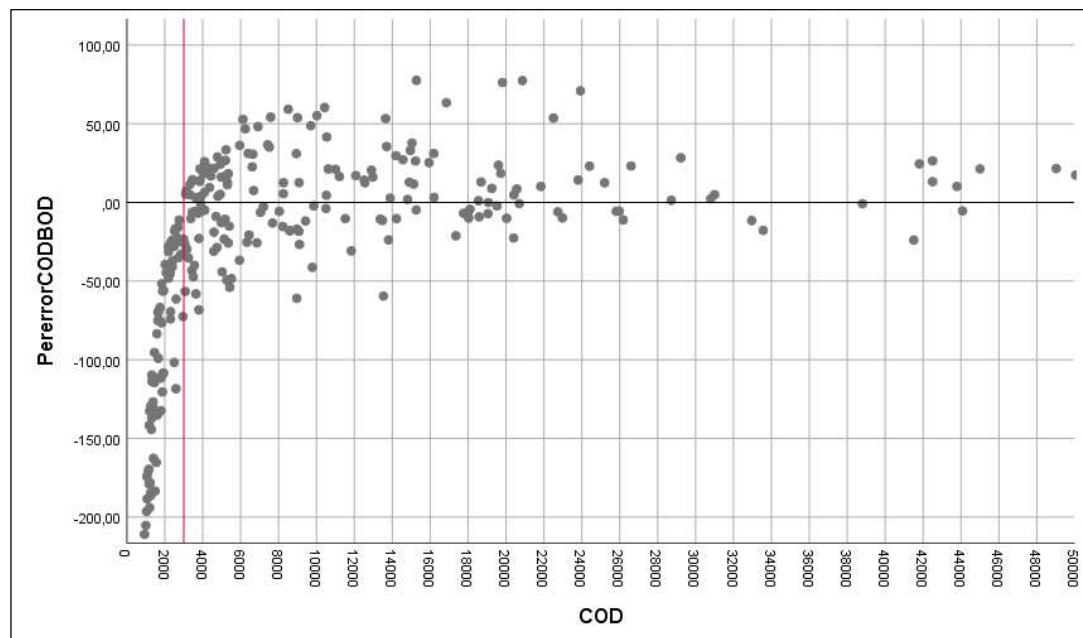


Figure C.2. Percent error chart for COD and BOD regression model

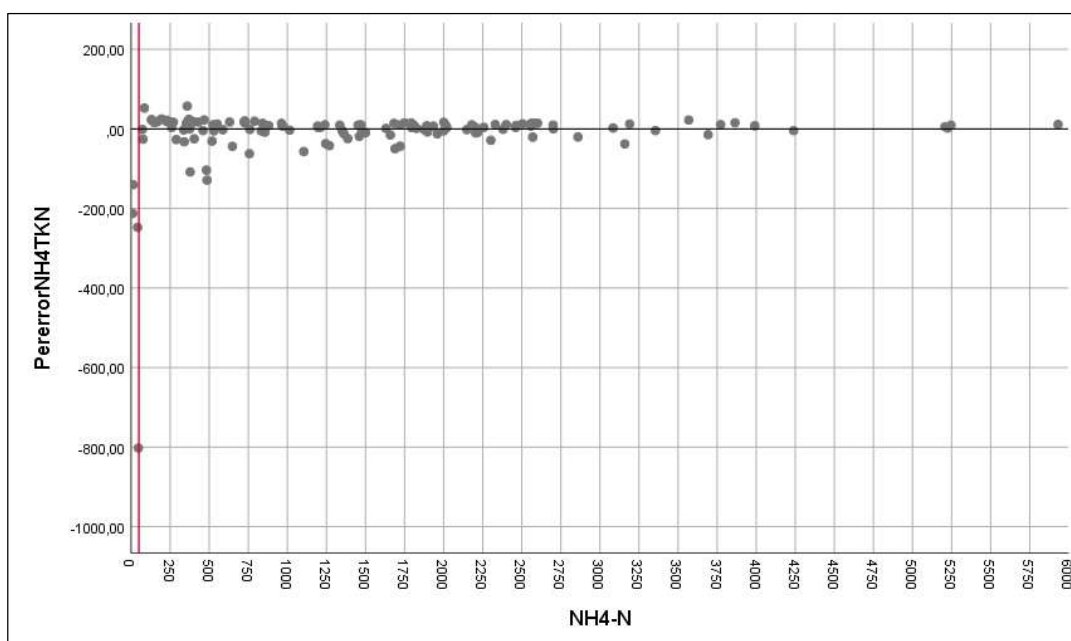


Figure C.3. Percent error chart for NH₄-N and TKN regression model

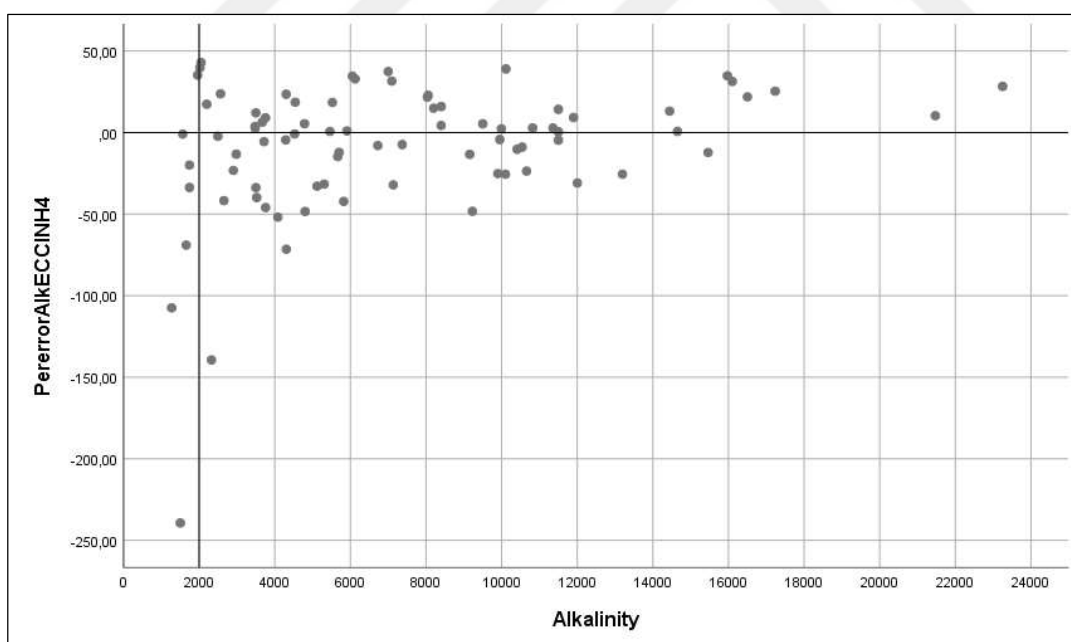


Figure C.4. Percent error chart for Alkalinity and EC-Cl-NH₄-N regression model

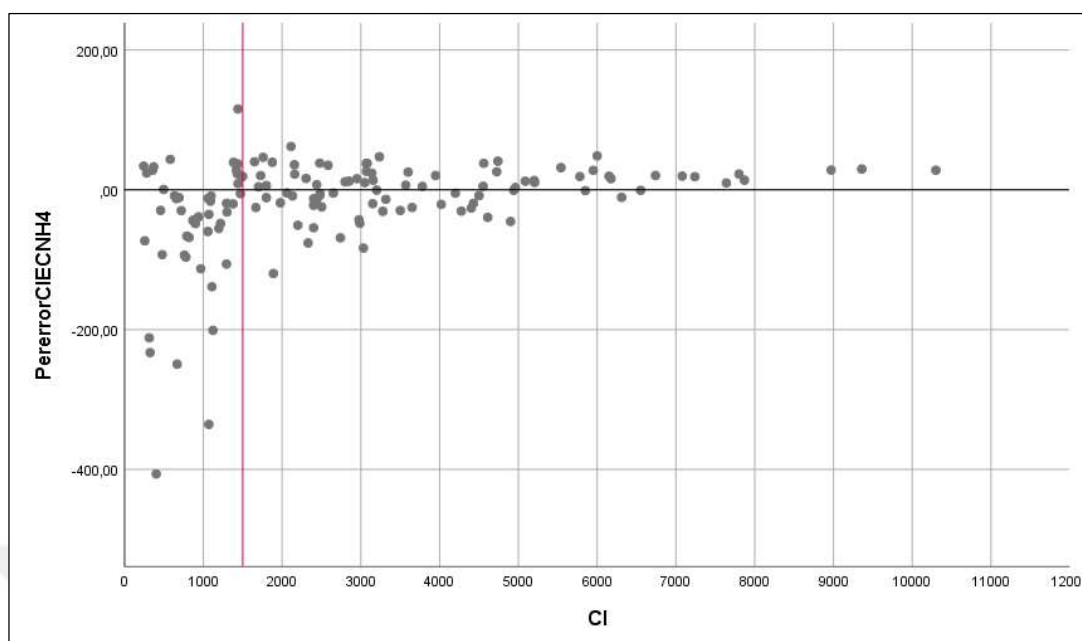


Figure C.5. Percent error chart for Cl and EC-NH₄-N regression model

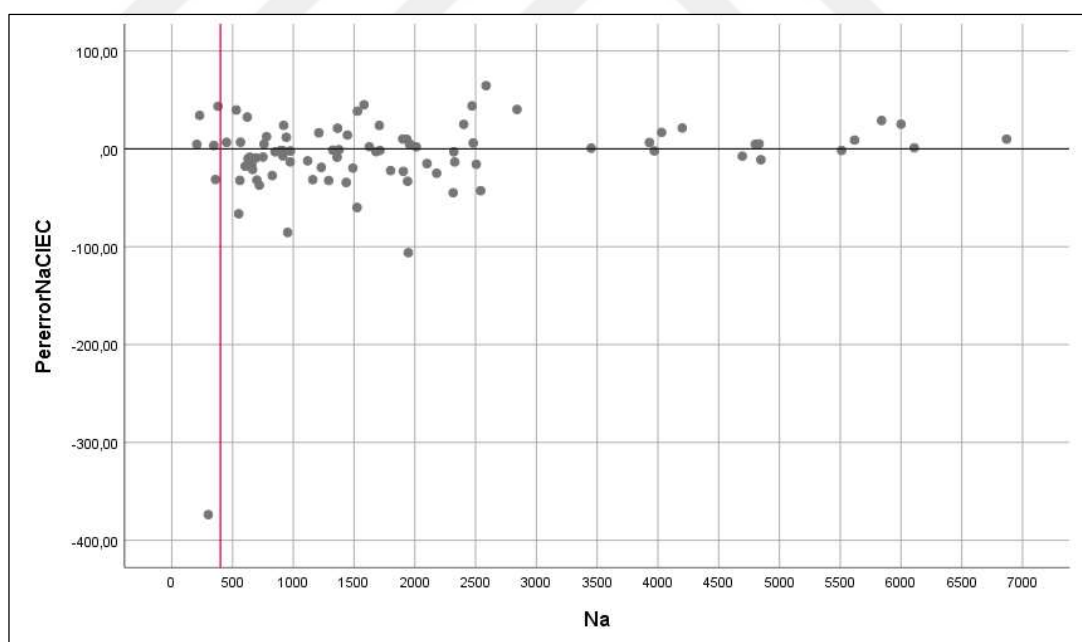


Figure C.6. Percent error chart for Na and Cl-EC regression model

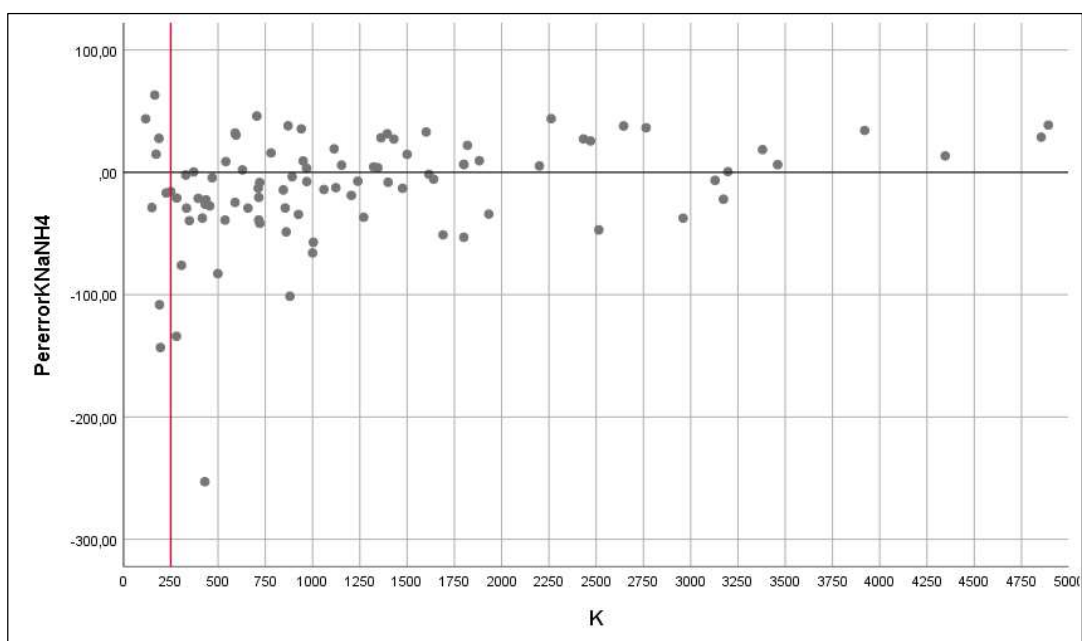


Figure C.7. Percent error chart for K and Na-NH₄-N regression model

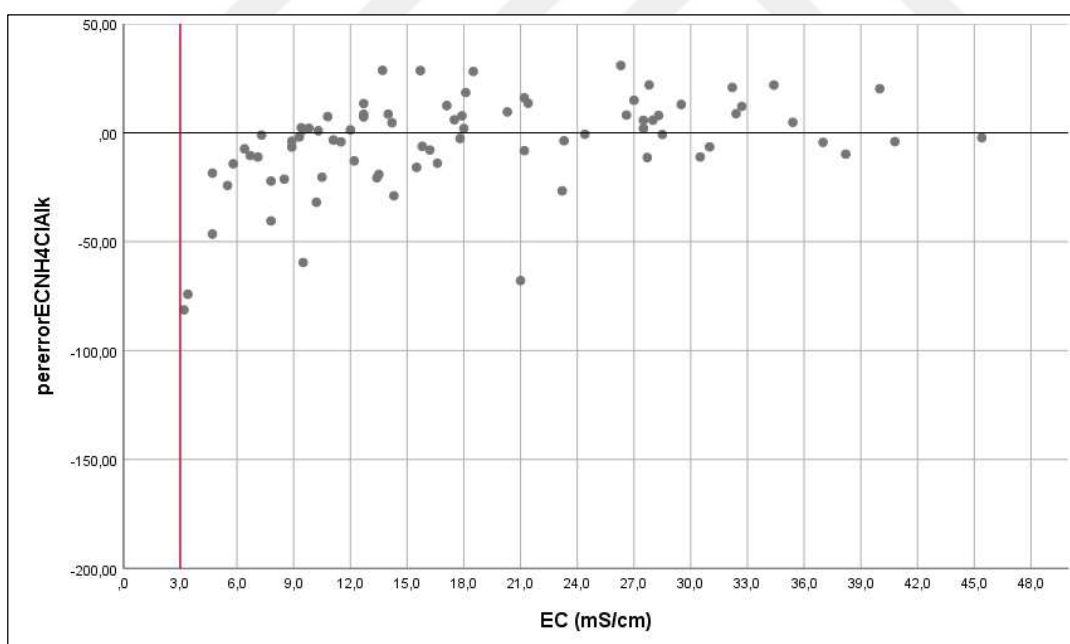


Figure C.8. Percent error chart for EC and NH₄-N-Cl-Alkalinity regression model

D. PCA Tables

Table D.1 Principal components (PC) for 10 leachate parameters with three components (Sample N=46, KMO=0.756)

Parameters	PC1	PC2	PC3
ZEC	.951	-.041	-.043
ZAlkalinity	.908	.029	.066
ZCl	.891	-.018	-.090
ZK	.889	-.201	-.064
ZNH ₄ -N	.725	.056	-.325
ZFe	-.108	.913	.135
ZCOD	.303	.878	.114
ZpH	.323	-.652	-.406
ZCa	-.204	.646	.638
ZMg	-.026	.257	.912
Eigenvalue	4.4	3.0	0.8
% variance	40.9	25.6	15.6
Cumulative %	40.9	66.5	82.1

Table D.2 Principal components (PC) for 10 leachate parameters with four components (Sample N=46, KMO=0.756)

Parameters	PC1	PC2	PC3	PC4
ZCl	.929	.078	.211	-.132
ZK	.919	-.114	.233	-.107
ZEC	.749	-.022	.595	-.005
ZFe	-.106	.917	-.101	.158
ZCOD	.221	.891	.153	.152
ZpH	.461	-.570	-.056	-.480
ZNH ₄ -N	.286	-.003	.896	-.201
ZAlkalinity	.642	.019	.671	.129
ZMg	-.030	.206	-.057	.929
ZCa	-.147	.625	-.216	.644
Eigenvalue	4.4	3.0	0.8	0.7
% variance	30.6	24.1	17.9	16.4
Cumulative %	30.6	54.7	72.6	89.0

E. Impact Flowcharts

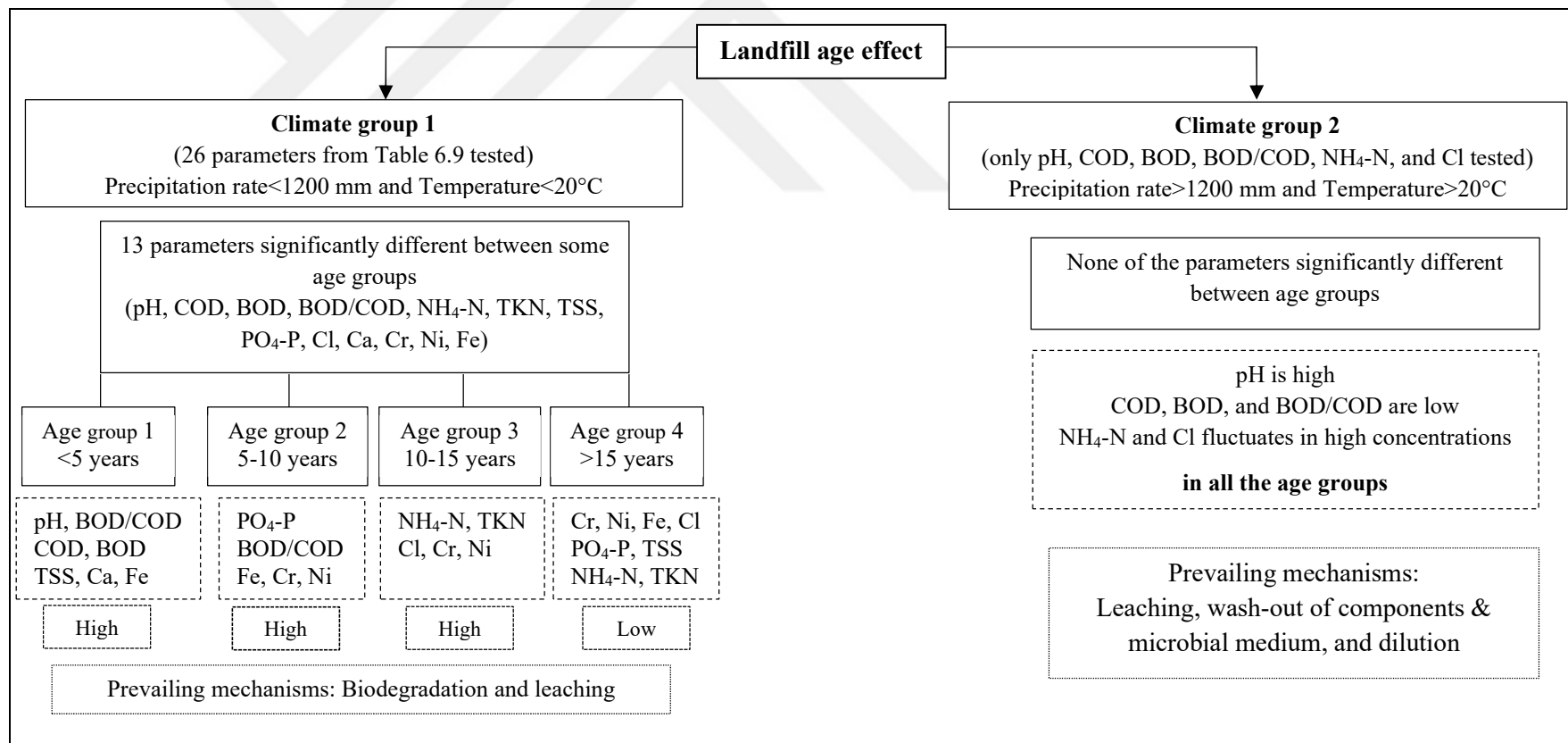


Figure E.1. Flowchart of landfill age impact

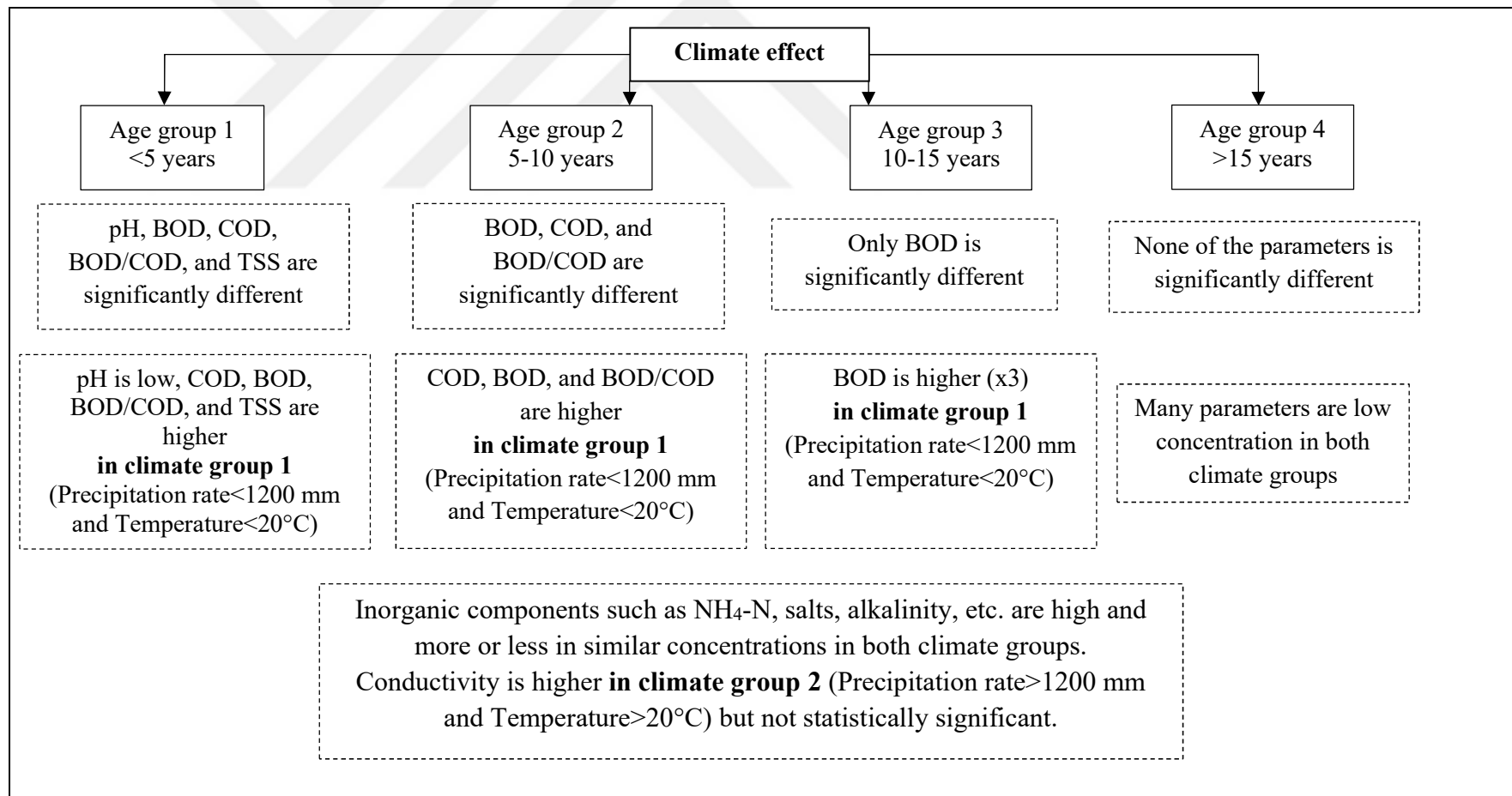


Figure E.2. Flowchart of climate impact

F. Sample List of Countries

Table F.1 Sample list of countries having relatively low annual precipitation rates and temperatures

Country	Location	Income Index	AAP	AAT	Country	Location	Income Index	AAP	AAT
Algeria	Algiers	UM	540	17.5	Mexico	*	UM	538	15.9
Australia	Adelaide	H	536	16.4	Morocco	Tangier	LM	435	18.3
Canada	*	H	834	6.9	New Zealand	Rosedale	H	1155	13.3
Canada	Winnipeg	H	519	2.1	Norway	*	H	772	5.4
Canada	Quebec	H	1101	4.8	Palestine	*	LM	318	19.7
China	*	UM	555	10	Poland	*	H	606	7.5
China	*	UM	1140	17	Portugal	Porto	H	1133	14.6
Croatia	Zagreb	H	930	11	Russia	Moscow	UM	680	4.7
Denmark	*	H	702	8	Slovenia	Ljubljana	H	1290	10.4
Egypt	Alexandria	LM	183	20.6	South Africa	*	UM	918	20
Ethiopia	Addis Ababa	L	1143	16.3	South Africa	*	UM	495	17
Ethiopia	Mekelle	L	581	19.1	South Korea	*	H	1230	12
Finland	*	H	643	4.4	Spain	*	H	1113	14
France	*	H	793	11	Spain	*	H	556	15
Germany	Kolenfeld	H	720	9.6	Sweden	*	H	645	7.6
Greece	*	H	420	17	Tunisia	Tunis	LM	466	18.1
Greece	Diakopto	H	700	17.3	Turkey	*	UM	785	14.6
Iran	Rasht	UM	1360	16	Turkey	*	UM	470	13
Iran	*	UM	329	15.6	UK	*	H	716	9.8
Iraq	Erbil	UM	420	20.2	UK	*	H	1200	9
Ireland	*	H	946	9.4	Uruguay	Montevideo	H	933	16.3
Italy	*	H	666	15.5	USA	*	H	1027	10.7
Lebanon	Beirut	UM	893	20.5	USA	Jacksonville	H	1268	20.5
Lebanon	Zahle	UM	686	15.2	Yemen	Ibb city	L	879	17.1
Lithuania	Siauliai	H	632	6.1					

AAP: Annual Average Precipitation (mm) AAT: Annual Average Temperature (C°)

* Arithmetic average of different locations having similar AAP and AAT values

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BS	Marmara Univ. Environmental Engineering, İstanbul	2000

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PUBLICATIONS

1. Ergene D., Aksoy A. and Sanin F.D. “Comprehensive analysis and modeling of landfill leachate”, Waste Management, 145, 48-59 (2022).
2. Ergene D. and Alp E. “Planning for the closure of uncontrolled landfills in Turkey to reduce environmental impacts”, Waste Management & Research, 34, 1173-1183 (2016).
3. Coptý N.K., Ergene D. and Onay T.T. "Stochastic model for landfill gas transport and energy recovery", Journal of Environmental Engineering, 130 (9), 1042-1049 (2004).
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