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WATER QUALITY EVALUATION

HEAVY METALS EVALUATION IN SUPERFICIAL WATERS OF FAN RIVER IN RUBIK, ALBANIA

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Water quality evaluation – Evaluation of Heavy Metals in Superficial Waters of Fan River in Rubik, Albania.

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Abstract

This study is a practical research about the water quality evaluation of the Fan River that flows through Rubik city in Albania. It is mostly focused in heavy metals concentration of superficial water of Fan River. Rubik was an industrial area for heavy metals extraction and for many years industrial solid wastes have been thrown out of mines into environment without any treatment or proper management. The main objective is finding the value of heavy metals in Fan River waters. The research in this study is conducted by qualitative and quantitative methods. Sampling method issued in fieldwork research about the contaminant elements into Fan River waters. The results of the research provide a clear understanding of the water classification into classes according to its quality. The research outcome is useful for evaluating the current status of the Fan River water quality according to each heavy metal. At the end, the results will give a clear frame of contamination levels development through years after the landfill construction for industrial solid wastes, and the gaps of environmental monitoring in Rubik. The result of this study is the contamination of Fan River waters from Pb, Zn and Ni. The waters of the river cannot be used without treatment for scaling down the concentration of those three heavy metals.

Language: English **Key words:** Heavy metals, water classes, industrial wastes, landfill, superficial waters, Albanian law.

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1 Introduction

This study is developed as a case study which aims to gather evidences about the water quality of Fan River for assessing the impact of the heavy metals industry. Rubik has been an industrial area for copper, chromium, iron-nickel and copper extraction. After passing from communism system to democracy system, the main plant and mines were exploit from private entrepreneurs. The overusing of the mines and the uncontrolled process of industrial wastes, have made Rubik city the hot-spot area regarding environmental issues in Albania.

1.1 Background and Problem Statement

Nowadays, the main sources of environment pollution are heavy metals and oil extraction and refining, mobile sources, district heating plants, domestic heating, cement production, and irregular garbage burning (Ejupi, Korça, Durmishi, Musliu, Ismail, Jashari & Shabani 2016, 99). Humans` living conditions are directly dependent on the ecosystem where they live, so it is necessary to protect and preserve the environment as well as to ensure a lasting quality of life for present and future generations. The environment pollution is one of the major issues that worry local and national authorities for its impact on the population health. Thus, environment pollution management is a current problem area but also a future challenge.

This study is focused in one of these environment pollution sources, the heavy metals extraction and refining. The problem area of this study is about the environment destruction from this pollution source in one of the richest areas of Albania with heavy metals, named Rubik.

Acccording to the head of IGEWE (personal communication, 06.02.2018), Albania is a rich country regarding mineral resources, like chromium, cooper and iron-nickel. Before 1990, Albania used to have an intensive exploitation of mineral resources especially referred to copper mineral, for which was ranked the world`s third largest producer. Several exploitation mines were built for those heavy metals, and have been followed by building smelting and enrichment plants. All three of these heavy metal industry elements were present in the Rubik area, and have been functional for over a period of 40 years. Like most of Albania`s industry, the mining sector used to have lack of management expertise, outdated equipment and technology. Furthermore, technological waste has been discharged without any kind of control mainly on river banks and sometimes directly on the river of Albania.

According to the Law on environmental management of solid wastes (2003/9010) for undertaking environmental issues carried from those past actions, in Albania, several projects have been undertaken for the rehabilitation of contaminated areas, or for storing technological waste in specific landfills. The main issue with construction of a landfill is the considerable harm caused to the quality of soil, water, and air in its surroundings (Kristo 2007, 107-109). For this reason, all aspects of water, soil and air pollution are considered in this study in a general context, as interactive elements of the environment. This study evolves in more specific context by focusing on quantitative assessment of water quality spoilage from the heavy metals industry in the past. More precisely, the focus is about the water quality of the Fan River, the closest river to the new landfill of Rubik area in Albania. This study is developed as a case study which aims to gather evidences about the water quality of Fan River for assessing the impact of the heavy metals industry.

1.2 Research Question and Objectives

The Rubik area has been for many years an area threatened by water, air and land pollutants due to metal mining. These mines have been operating out of environmental compliance standards for many years. (Law on environmental management of solid wastes 2003/9010.) The aim of this study is to define the current situation about the water quality in Fan River at Rubik area. Therefore, the research question of this study is as follows:

What is the state of Fan River waters regarding heavy metals concentration?

The main objective is to analyze the Fan River water, by getting the values of 9 heavy metals. From these values I will show which the contaminators of the water are and what is the class has qualifications from each of them. Another objective of this study is to see the actual state the landfill, the landscape, compare the differences of air pollutants limits according EU standards and Albanian standards for air pollutants limit for a year.

1.3 Framework, Timeline, and Process

The framework shown in Figure 1 gives a visual summary of the topics where this study is focused the most. The study main focus is about heavy metals pollution in Fan River. Research work of this study is evolved around the Rubik area and Fan River, environmental standards, environmental issues, surface and groundwater, water classes, Albanian and EU environmental standards and law, and so on.

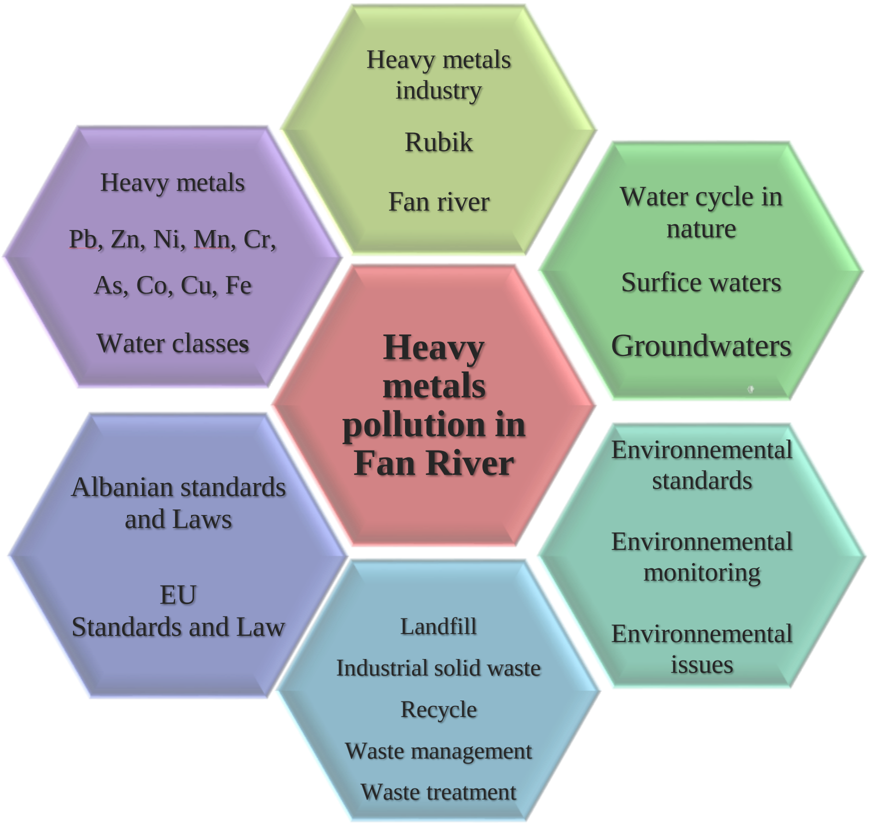

Figure 1. A visualized framework of the study.

The Figure 2 shows a visual timeline of the study work. This study started on 1 February 2018 with literature review. The information sources were books, articles, and research papers, provided as a print-out and as online information. Qualitative research was conducted by interview with doctor regarding health issues consequences from heavy metal pollution. Afterwards, sampling method applied in Rubik site was the next phase in the process. Besides, interviews with residents of Rubik regarding past and present environmental issues were conducted. In the same day, samples were taken to the laboratory and after a week the results were ready. Statistic work is performed to evaluate the values taken from three replicates for each site about the nine heavy metals concentration. The results of these data analysis consist in calculating the mean value of heavy metals concentration from three replicates from two sites, their variable deviation, and their standard deviation. The study process continues with generating conclusion and visualizing them into tables and charts. Thesis structuring and thesis presentation were the last steps before finalizing this study.

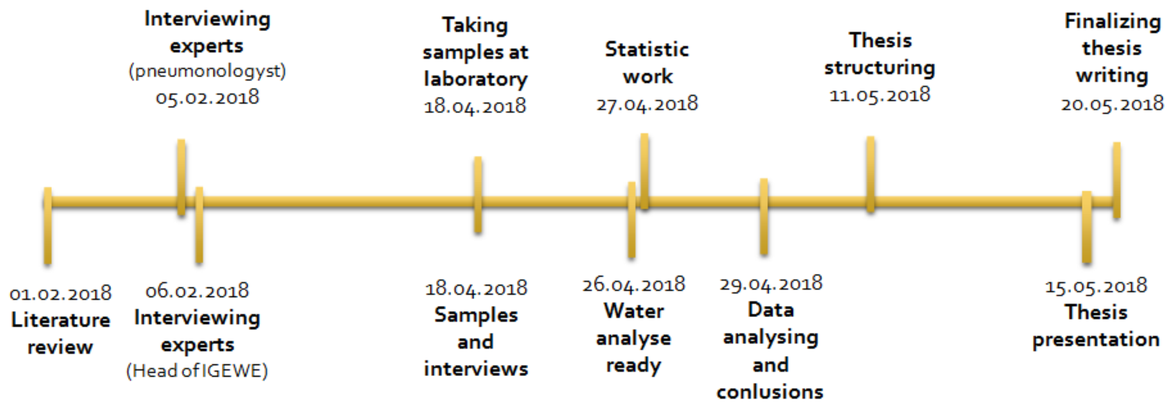

Figure 2. Timeline of the study.

Figure 3 shows a visualization of the whole process work in this study. The study process includes seven steps, described as follows.

Step1: Stating the problem

Landfill area of the industrial solid waste is close to Fan River flow. It is a big risk of heavy metals contamination of its water from the lixiviants coming from underground waters to the river.

Step 2: The research question

Defining the research question: *What is the state of Fan River waters regarding heavy metals concentration?*

Step 3: Identify the methods

To have a study on the concentration of heavy metals in surface waters, you must first choose the place of study. In this case, Rubik was chosen as a hot-spot for the environment in Albania (UNEP 2000, 36). Meanwhile, I gathered information from various publications and studies, personally meeting people who could provide me with valuable information. An important work is the research on Albanian laws related to the preservation of environmental quality. The final data were processed using statistical methods and laboratory equipment, with quantitative and qualitative data.

Step 4: Identifying boundaries of the study

It was helpful to know the difficulties and boundaries of the study, so that can be considered beforehand in case of a similar project in the future. The boundaries were about the expensive laboratory prices for water analyses, the long distance and the high travel costs to reach the sampling site (Rubik area was far away from Tirana), the limited access of Albanian Law and other relevant information regarding environmental issues.

Step 5: Developing measurements protocol

There were three replicates taken from two sites which were randomly chosen. To take water samples were used laboratory polyethylene bottles, sterilized and labeled. The laboratory equipment used to determine heavy metals concentration was the Atomic Absorption Specter. An online calculator named *Statistic How To* was used for fast and accurate data calculation of standard deviation.

Step 6: Determination of tolerable limits on decision error.

Regarding the sampling values from the three replicates taken from the two sites, the mean value of each element on both sides and the standard deviation has been applied to evaluate their variance. Relative standard deviation of the data will give how many percentage points the results will vary.

Step 7: Making more effective the design for obtaining data

Due to the high prices for laboratory analyzes of the heavy metals in water, the sampling method in this study was restricted in the number of the replicates and the samplings sites. Also, the long distance from Rubik to Tirana was the reason of taking the water samples within one day only. For obtaining more accurate and reliable data, the best approach would have been by taking ten replicates and choosing more than two sites. The heavy metals concentration also depends on climatic and meteorologist factor. Thus, taking replicates in different days through a changing climate would have increased the quality of this study results.

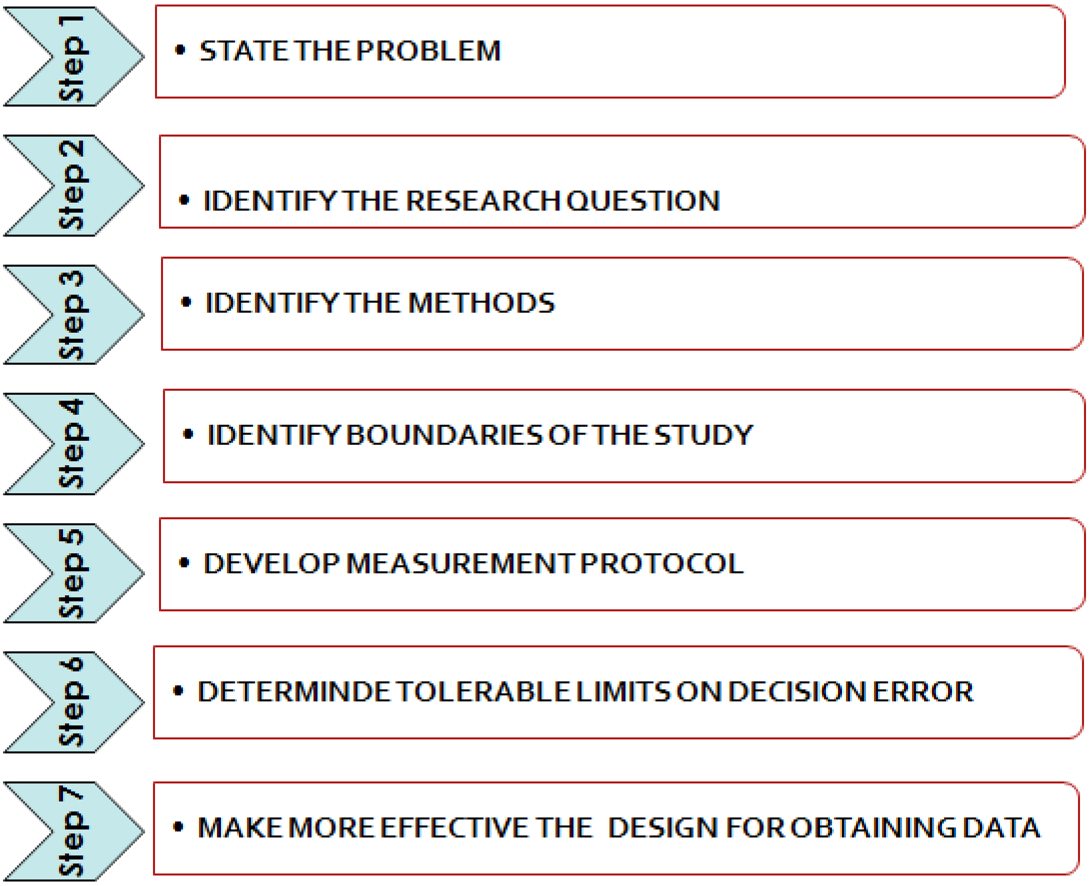

Figure 3. Process of the study.

1.4 Methodology

The scope in this study is to evaluate parameters of heavy metals in water of Fan River, Rubik, for defining their impact in water quality. The research process in this study includes primary research and secondary research (Elbasani 2003, 109). The primary research is about fieldwork research involving people by using survey methods such as *interviews*, and research involving measurements of heavy metal concentration on surface water. The secondary research includes *literature review*. These qualitative and quantitative research methods adopted to undertake this project has provided valuable knowledge, information and data.

Literature Review

The secondary research includes literature review which consisted in deep theory research about concepts, definitions and facts related to the problem area. Secondary research consists also in collecting and synthesizing existing data which were relevant to this study, and also about defining the right methods to be used. Gathered insights from literature review have essentially contributed in the understanding of the problem area under investigation and was the groundwork to aid with the fieldwork research.

Interviews

The empathic approach of this research had in focus the impact of environment pollution due to these heavy metals in the resident health. To get a deep understanding of such impact, it was conducted interviews with specialist, doctor and residents of Rubik.
The interview method has been applied in two ways, *interviewing the experts* of the problem area and interviews with residents living in the problem area.

Sampling Design

For the measurements of heavy metal concentration, it is used *simple random methods,* that means that the sites but were chosen by chance with no purpose behind (Elbasani 2003, 161). For taking the samples, there were provided six polyethylene bottles a laboratory in Tirana. These bottles were sized of 1 liter each, were distillated and labeled, and filled with a little quantity of nitric acid for preventing the cations of metals to be absorbed from the inner layer of the bottle or to precipitate (Sulce 2005, 66). The samples were taken to the same laboratory for analyses with *Atomic Absorption Specter*.

Analysis Methods

To determine the final values of heavy metals concentration *statistical methods* have been used. The methods were used to analyses the data received from the laboratory. The goal of using these methods was to determine the final values of concentration in water for each of nine heavy metals concentration by finding out the mean of these three individual replicates from each site. The classification of waters into classes for each heavy metal concentration values is made by using *Albanian Standards* of heavy metals concentration into superficial waters, which has been adapted with the EU standards. These quantitative results are presented not only as table data but also by using grouped bar chart visualization. The data of air pollution is performed by using evidence-based method of 2017 provided by the Municipality of Mirdita which has Rubik city under administration. These values are compared with EU and Albanian standards for nitrogen dioxide (NO_2), sulfur dioxide (SO_2) and particulate matter 10 micrometer diameter (PM10).

2 Literature Review

The literature review is a secondary research method used to gather information from published sources, from previous research or projects, with the aim of providing essential information for the current project. The aim of using this method was to extract information from precedent research, studies, and projects to get in-depth knowledge of the problem area of the study. Internet resources have expedited literature searches significantly in this study, by allowing to access libraries from around the world using online tools, digital journals, digital research papers, e-books and e-libraries, and statistical data. Nevertheless, during this researcher, a special attention was given to the sources themselves for ensuring that the research and literature included in the study are relevant and credible sources. The gathered information from this research was necessary to provide some direction to this study and a certain frame for its project.

In this study, in order to understand the impact of the heavy metals in river waters, in environment, and in humans' health, conducting in-depth literature review has been considered a crucial starting point of the whole process. To accomplish this purpose was used literature review method by reading books, articles and studies which tell and explain what is the role of heavy metals and which is their limit of concentration that separates them from being necessary and useful to being dangerous and toxic. Several articles, journals and research papers have been found as online sources, such as "European rivers and lakes", "Selected water quality standards", "Parameters of water quality", and more. The books were found from personal library and from university library.

2.1 Country Profile - Albania

This sub-chapter of the study is about an understanding of geographic and relief of Albania, its clime and hydrology features and Albanian Environmental Legislation. Climate and relief are features that have a significant impact on the concentration of water elements and substances. Metals can reduce the hydrogen in the water by producing hydrogen molecules. As a consequence, water pH can affect the heavy metals concentration into the water. Excessive rains can raise river water levels by decreasing heavy metals concentration and the reverse can increase their concentration (Karanxha

2001, 59). A large matrix of these components taken into account in detailed studies to have results as accurate.

Albania is a rich country from water point of view. In Albania there exist 136 rivers and streams with a total length of 50,000 km. The water of this network collected by 10 main rivers flowing down to the Adriatic Sea. The territory of Albania is divided into six river basins (Brin, Avramovski, Vermoote, Bassi, Callebaut, Lust & Hunt 2007, 21). The uses of water are founded in agriculture, aquaculture, tourism, economy, food industry, transport etc. Being so important, the evaluation of their quality and monitoring is of particular importance.

Geographic Location and Climate

The Republic of Albania is located on the Balkan peninsula, in its southwestern part. The relief of Albania prevails in 2/3 of the mountains. Its territory covers an area of 28748 km^2 (Islami et al. 2002, 14.) The site where the landfill is built is part of the Middle Albania, in its northern part, located in the outskirts of Rubik City. The relief is flat and the place is located on the edge of the Fan River at a distance of about 80 m.

Albania, from the climatic point of view, is part of the subtropical belt with humidity, of the west coast of the continents of the earth's surface. In this belt, a special place occupies the subtropical Mediterranean area of Southern Europe, where the peninsulas Pyrenees, Alps and Balkan are inserted. In the last one, the subtropical Mediterranean area includes almost all of Albania and the NW part of Greece. In Albania climatic regionalization, as basic factors are taken solar radiation, latitude, the general circulation of the atmosphere and local factors and the differences of those climatic elements are the result of mutual interaction of these factors.

Albania is part of subtropical Mediterranean climate, which is categorized by mild winters with abundant rainfalls and dry, hot summer has the highest value of the factual monthly extension of sun regime is observed in July. The annual amount ranges from 2841 hours to 2000 hours. The smallest amounts of sunshine are observed in December (Islami et al. 2002, 14.) The annual air temperature ranges from 7 °C to 15 °C. On the coastal zone and even in the southwest these temperatures can reach up to 16 °C (Islami et al. 2002, 33).

Winds in Albania are present during the all year and change frequently direction and speed. This situation is particularly determined especially by cyclones passing through the Mediterranean Sea and the Balkan Peninsula as well as by other local factors such as sea nearness, ridges direction and rivers valleys.

The highest values of relative air humidity are observed in the cold time of the year, which is explained by the cyclone activity observed over our country during this period of the year. The air humidity amplitude, as one of the important indicators of the scale of the continental climate, shows the Mediterranean climate character of Albania. The annual precipitation in total over Albania is 1,485 mm/year and 70% of the highest precipitations are during October-March that is the coldest months of the year. The northern part of Albania, which is part of the Alps, has the highest average annual rainfall where the annual rainfall reaches 2800-3000 mm/year. The seaside lowlands are characterized by annual rainfall ranging from 900-1250 mm. (Islami et al. 2002, 34.)

Environmental Legislation

Albania has begun to develop a framework regarding environmental issues that are problematic during last decades. This was one of the priorities of Environmental Strategy and NEAP (National Environmental Action Plan) by Government of Albania in 2002, which is one important instrument for the application of the environmental policies in Albania.

During 1997-1998 were developed major legal documents and the amendments of the law on “Environmental Protection”.

In 1998 the law Nr.8364 addresses the full spectrum of the environmental policy problems. During this year, was established the National Environmental Agency as the highest state body which was depended directly from Council of Ministers. In 2001the Ministry of Environment was created which. Its role is to define the nation`s environmental strategy, to develop and implements national environmental protection efforts, approve admissible limits for polluters emissions and coordinate activities with other governmental body responsible for the protection of environment.

Some governmental entities that have important roles in environmental policy are: Ministry of Agriculture and Food, Ministry of Transport and Telecommunications, Ministry if Industry and Energy, Ministry of Health, National Water Council, Public Health Institution, Institute of Hydrometeorology Institute, Council on Territorial Adjustment and Institute of Soil Research. (Islami et al. 2002, 49.)

Albania in 1990 changed from the communist system to a democratic system and many laws, articles, decisions, standards, norms and regulations related to the environment were changed, and every year they adopt to the EU policy. The EU policy framework is mainly focused on two main points: one is the identification of the limit value for air pollutants oriented to the protection of human health and ecosystems and on the other hand the development of a coordinated controls plan and air quality management. EU Air Quality Directives, geared towards national legislation of Albania, according to the Law on the protection of ambient air quality 2014/162. The implementation of this Law will decrease the problem of air pollution as a dynamic phenomenon that is needed to be managed.

Some important legislation for air quality protection are the Law for air protection from contamination 2002/8897, the Decision on approval of air emission rates 2002/435 and DCM for the adoption of provisional norms of emissions in the air and their application 2003/248.

For river waters quality, DCM on permissible rates of urban discharges from treatment plants for coastal areas and river 2005/177 the evaluation of contaminants norms parameters, is adopted from the EU Water Framework Directive 98/83/EC. According to this DCM, the water of rivers is classified into five classes.

2.2 Landfill and Components

The landfill is defined as the place of final disposal of waste inside or on the ground in a controlled manner, according to various sanitary requirements, environmental and security (Potera 2002, 25).

The waste management process is one of the most difficult problems in Albania, which is one of the main priorities of the Albanian government in the field of environment, which are the National Strategy and National Waste (DCM, 115/2012). Drafting of the Draft National Strategy for Solid Waste Management and Draft Law for Integrated Waste

Management has had a significant impact on the creation one of the most advanced and professional concept for waste management in Albania. The main purpose and importance is their full implementation. This begins by starting to minimize wastes, recycle, treat for use of heat or energy for industrial purpose, household, etc., and depositing them at landfill. The MoEFWA (Ministry of Environment, Forest, Water and Air) is in the process of adopting the National Strategy and the Law on Administration Integrated Waste for the period 2010-2025. Implementing them and a series of new DCMs will do give a boost before integrating waste treatment by avoiding them instead of depositing by implementing the objectives of the EC Directive on Landfill. (DCM, 115/2012.)

2.2.1 Surface-waters and Groundwater / Waters Cycle

In landfill projects**,** one of the most important parts is the protection of groundwater and surface water from the pollution that it can cause. Superficial waters are all the natural waters and all sources, wells or other watersheds that directly from surface waters. (Potera 2002, 84.)

Water circulation in nature is a continuous process. In the water balance of a water collector basin, the components of the nutrient part are the atmospheric precipitation, the condensation of water vapor into the soil and the soil surface as well as the supply from the groundwater of neighboring aquifers. On the other hand, the reduction in the quantity of groundwater is caused by the increase of the evapotranspiration, the discharge through resources, the increase of the discharge into rivers etc., always leading to the reduction of the groundwater level. Groundwater of a basin are always in a continuous movement (water circulation in nature). For different time periods, feeding and discharging of the water basin is variable, whereas for very large time, are observed equalization tendency of the amount of discharge, by establishing a condition called a hydrodynamic equilibrium in which the levels fluctuate around the average values and are simply seasonal.

Lixiviants are liquids used to extract the assigned metal from the mineral. The metal can be recovered from it in a concentrated form after leaching. Factors which affects the production of lixiviants:

- Climatic conditions;
- Planimetric characteristics of the place and surrounding areas;
- Humidity of wastes and production of fluids from biodegradation processes;
- Infiltrations from the substrate;
- Operations in the storage center;
- Calculation of lixiviants for an urban waste storage center.

Groundwater is or can be obtained from either through an underground formation. Groundwater bodies are distinguished by superficial water bodies from the relatively slow movement of water through the soil, which means that the stand time in groundwater, are generally in collocation of higher magnitude than in surface water. A body of groundwater can remain so for decades or even hundreds of years because the natural processes of rinse are very slow. There is a considerable rate of physical-chemical and chemical interdependence between water and filler material. (Potera 2002, 83.)

In all areas where the surface and the underground water meet, the interconnection between the two must be evaluated. Infiltration on rainfalls into high lands happens in the filling area where the hydraulic fill is reduced by the depth and the flow of the saturated network is lower than the water base (Kristo 2007, 67). The natural chemical quality of the groundwater is generally good, but elevated concentrations of a number of components can cause problems for water use.

Water from the depths of the earth has been used since ancient times for home use for livestock and irrigation and the use of groundwater has increased since that time. According to the statistics, in USA water uses are for watering in agriculture sector 39%, power plant 39%. For urban purpose 12 %, and other purposes 6%. In Europe, also groundwater has always played a key role in water supply. (Sulce 2005, 58.)

A number of countries receive a significant proportion of their freshwater resources as external inflow, as shown in Figure 4. Hungary and the Netherlands had the highest dependency on transboundary water resources. Freshwater resources per inhabitant are considered an important indicator for measuring the sustainability of water resources. Croatia, recorded the highest freshwater resources. Finland and Sweden had the next highest volumes of freshwater resources per inhabitant; low levels were recorded in the six

EU Member States: France, the United Kingdom, Spain, Germany, Italy and Poland. (European Commission 2017a.)

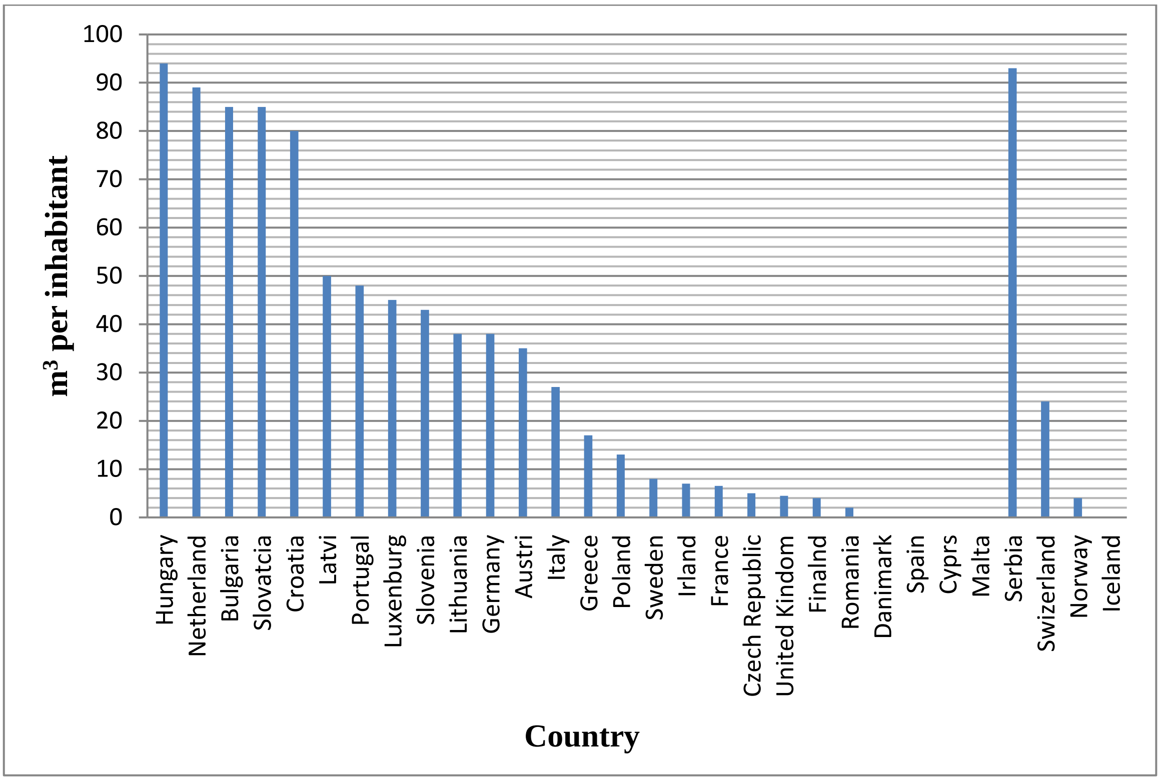

Figure 4. Total freshwater abstraction by public water supply, 2015.

Albania can be considered as a rich country regarding water sources. The major of water sources are surface waters like rivers, lakes and lagoons. Groundwater sources represent 23% of total water sources. Water sources play a main role in Albanian economy by producing about 97% of total electricity production from hydro-power plants (IBRD 2003, 6).

The origin of fresh natural water is from atmospheric precipitation or direct precipitation of rain or indirect from rivers, lakes or canals, as shown in Figure 5. Underground water is the origin of many streamlining and an important component of lakes and oceans, and is an integral part of the hydrological cycle (Sulce 2005, 57).

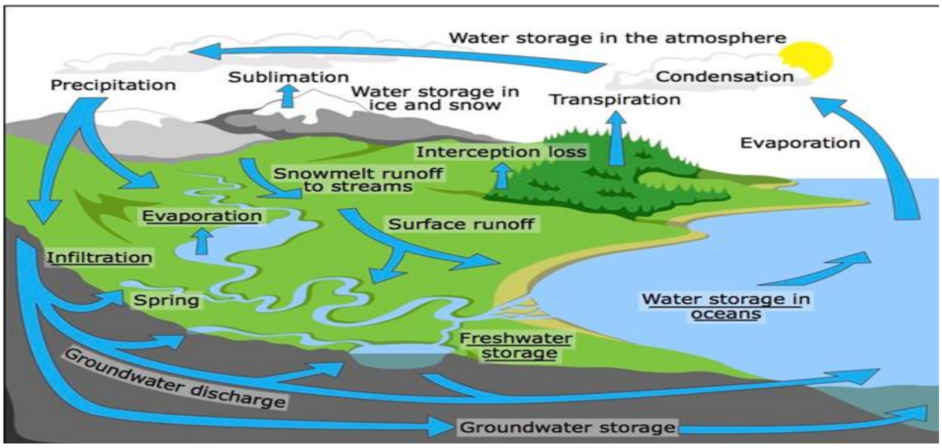

Figure 5. Water Cycle in Nature.

2.2.2 Heavy Metals and Impact in Human`s Health

Heavy metals, are called as "a treasure" for daily life but after useful use they are returned to a dangerous substance because they can return to reactive or toxic and in high concentration they are a hazard for and deadly for flora and fauna. Their tendency is to accumulate in the food chain, sediment and fauna (Potera 2002, 46).

Acccording to the pneumologist (personal communication, 05.02.2018) the heavy metals, such as Pb, Zn, Mn, Ni, Co, Hg, Fe, have an importance in the physiological functions of the human cell. They are also found in nature and in animals. But at high levels they lead to death and contamination of the environment. Contamination from heavy metals to humans can come from aspiratory track by breathing contaminated air, food, water and from touching them without precaution rules.

Chromium (Cr) – The Chromium toxicity includes liver, nephrites and gastrointestinal irritation and can cause dermatitis by skin contact. Chromium has potent carcinogenic effects on human and animals.

Lead (Fe) – is an important metal for the life of plant and animals. It is essential component in the hemoglobin structure and its luck in blood can cause anemia but in high doses it is toxic.

Cobalt (Co) – Cobalt deficiency is not common in humans, but its toxicity is we well-documented. Cobalt is acutely toxic in large doses, and in cumulative long-term, low level exposure. Most of what is known about cobalt toxicity comes from cases of ingestion or inhalation of excess cobalt.

Nickel (Ni) – it induces embryo toxic, allergic reactions and contact dermatitis. Nickel sensitization also occurs in general population from exposure to coins, jewelry, watchcases, clothing and fasteners. Nickel is a potential carcinogen for lung and may cause skin allergies, lung fibrosis and cancer of respiratory system.

Manganese (Mn) – high doses of inhalation can cause death. Miners have been found to have chronic poisoning if they are exposed to vapors of manganese compounds, but also individuals who drink water from contaminated wells.

Arsenic (As) – Intake of 70 to 80 mg of trivalent arsenic (III) oxide has been reported to be fatal for man. Inorganic arsenic produces acute, sub acute and chronic toxic effects.

Lead (Pb) – is poisonous metal. Pb poisoning is caused by environmental exposure that causes mental retardation in many children, while mild poisoning results in anemia. Organic compounds of Pb easily reach human body through the skin and endanger the central nervous system. Kidney and liver are considered potential targets of Pb toxicity before storage in bones.

Zinc (Zn) – does not accumulate in the body. It is the enzyme activator. It affects bone growth, development and functioning of reproductive organs, etc. There are very rare cases of poisoning with Zn. Its action in aquatic life depends on its strength, saturation with oxygen and temperature. Zinc is an essential micronutrient that affects several metabolic processes of plants and has a long biological half-life.

Copper (Cu) –Intake of excessively large doses of Cu leads to severe mucosal irritation and corrosion, hepatic and renal damage and central nervous system irritation followed by depression.

Metals in groundwater are more mobile in waters with low pH, most of the mass is as an ion charge. Over the pH limit, the majority of natural underground waters, the concentration of most metals is small and as result, complexes of these metals in these circumstances can intensify their solubility. River water pollution is one of the consequences of the rapid growth of the population, industries and agrarian practices and urbanization. Surface waters are an important part of health, society and economy, so preventing them contamination is an important aspect that requires regulatory involvement from the regulators such as laws and their implementation. The heavy metals do not decompose in nature and they enter into the water by becoming part of the food chain and sediment. The life span of heavy metals is long and this makes their presence in the environment and in long-term and dangerous aquatic organisms.

Surface and underground water can be contaminated by unprocessed liquid discharges remains of them. Composition of liquid discharges depends on the characteristics of the waste, their composition, size of the area, the landfill's, humidity and filtration rate of rain water, temperature etc. (Ejupi et al. 2016, 100.)

2.3 Environmental Issues in Rubik area

One of the major problems that has disturbed the Albanian society especially after the 90`s are waste from the mining industry, spread in several regions and cities of Albania.
Near the valley of the river Fan has existed copper processing industry for more than 40 years. All the remains of this industry, created waste piles on the banks of the Fan River, damaging flora and fauna of this valley. In 1998, all production in mines, enrichment and smelting factories was stopped. A reduction in the concentration of metals discharged from the running water of mines and enrichment factories was noticed. After the 1990`s, this problem was addressed by local and foreign institutions and specialists who, through their studies, have identified the presence of heavy metals and environmental hazards in this region. Part of those studies was, the Department of Analytical Chemistry Albanian Geological Survey Laboratory, "Fan River Environmental Rehabilitation Program" studies - an Albanian - Swedish Project (1999). (Daci 2013, 35.)

The concentrations of some elements measured at some monitoring stations are significantly increased or decreased depending on the month of the month they were taken, which is related to the amount of water intake and its acidity. In cases where monitoring of these studies is done in completely dry weather or low precipitation, then Cu and Zn concentrations in mining water are found to be relatively low. It is also found that solid waste that is not properly managed constitutes a major environmental risk, with negative implications for human life and environmental sustainability. Worldwide, landfills are known for the release of a wide range of harmful pollutants such as gases, liquid streams and particulate matter that can cause disease to humans and cause air, soil and soil pollution of water. Air monitoring is also important for the Rubik area, having a history of dust and gas emissions from the factory and the copper mining industry. These norms are defined by Albanian law. (DCM 2003/803.)

The reason for the construction of the landfill in Rubik was the creation of a storage site of solid waste materials from the copper processing plant which was a threat to the ecosystem and the people of that area. The project centered on the creation of a safe and functional structure, the non-flow of hazardous and polluting gases and water, the land sludge and security for many years, and the environment with little contamination of heavy metals. Cazizni, Vergerio and Cargnelutti (2008, 12) found that twelve features can be distinguished in the landfill structure, as listed below and as shown in Figure 6.

1. Underground water;
2. Compressed clay;
3. Plastic layer;
4. Drainage fluid collection tubes;
5. Textile insulating membrane;
6. Gravel;
7. Drainage layer;
8. Soil layer;
9. Old plots with waste;
10. New plots with waste;
11. The lixiviants movement stream;
12. Drainage basin.

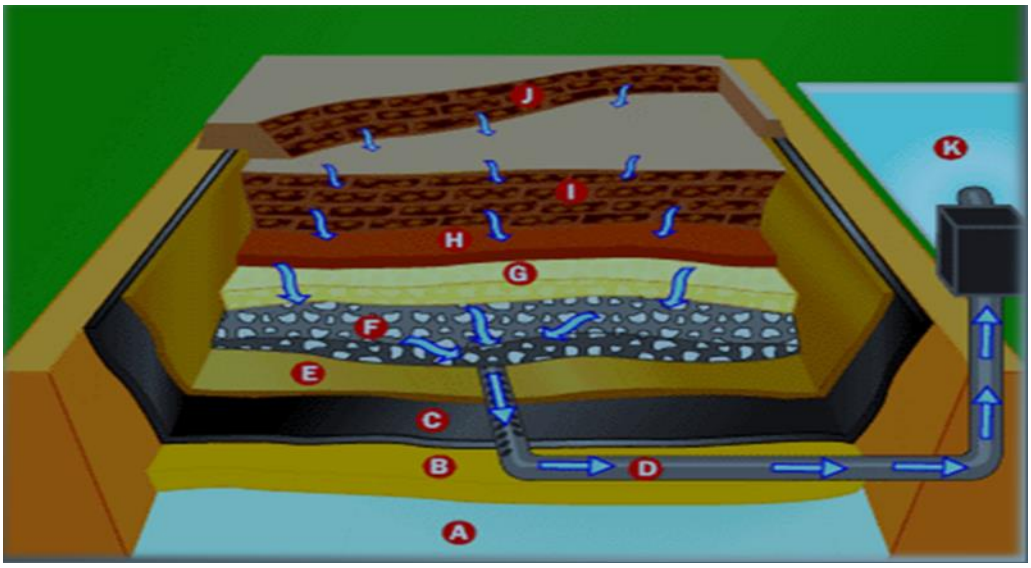

Figure 6. Rubik landfill structure.

From sources till near to Klos, Mat River passes through narrow gorges and after the gorges of the Shkopet, Mat begins to widen and opens in the coastal plain. Fan River itself is formed by the union of Big Fan River and Little Fan River. The maximum flow is associated with rainfall and melting of the snow.

The Mat Basin has a great extension. It starts in South-East with the massive Martanesh massif from where the Mat River flows and continues to the Munelles massif in the North. The Mat River from originates from the basin with the same name. It derives from Martanesh Peak Mountain. It has a length of 144 km, an aquifer water collection surface of 2441 km2 and an average aquifer altitude of 746 m. His branches are Fani River and Uraka River. (Preka et al. 2014, 52.)

The landfill implemented in Rubik is part of the Mat River Basin, which also includes the Big Fan and Small Fan Rivers, as shown in Figure 7. These are also the main hydrographic network that is directly linked to landfill construction. The wasted waters coming out form the process of metal refinery plants are rich in heavy metals, and they are directly discharged into the river, causing pollution of its waters. The acidity with the value pH = 3 of Fan River water, creates a favor conditions and increase the dissolving of heavy metals such as; Cu, Zn, Pb, Fe, Co, Ag, Mn, Se, Sb etc, which are characteristic of the enrichment factories and the copper smelting and refining plant in Rubik. (Preka et al. 2014, 54.)

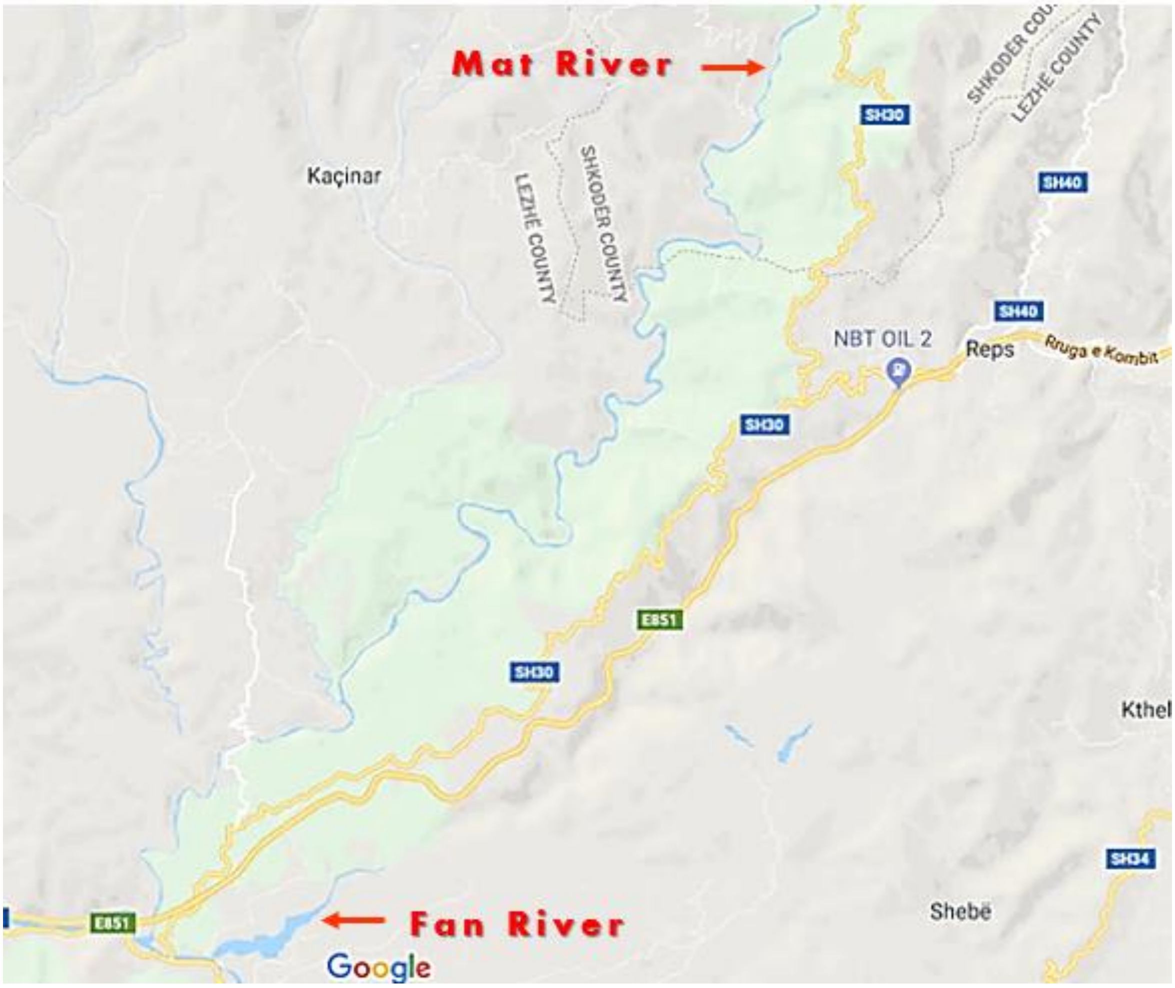

Figure 7. Mat and Fan River.

In 2011, after the completion of landfill construction, monitoring of groundwater was carried out by monitoring wells located near the landfill. These data were handled by the Municipality of Mirdita in the hope that every year continuous monitoring would be carried out to see if there was leakage of wastewater from the landfill. However, as result of the fieldwork carried out in the current study was observed that one of these wells was damaged by the vandalism over the years after the construction, and the second well was closed and in bad functional structure conditions, modified and not maintained. For these reasons, it was unfeasible to take samples of these two wells. However, the data of these water samples after the landfill construction were available, and so, the heavy metal content in the groundwater was included in the current study, and compared with the permissible norms.

After the construction of the landfill in Rubik in 2011, the superficial land is cleaner as removal and collection of waste away from this area continues to be made at assigned points away from landfill. The land above the landfill is covered with grass and allows residents of the area to have a green space for leisure, as shown in Figure 12. There are also grazing livestock animals in the area. Residents around the landfill areas have begun to use land for agricultural purpose by planting grapes, onions, tomatoes, peppers, etc. (personal communication with Rubik area residents, 2018.)

2.3.1 Water quality in Rubik / Heavy Metals in Fan`s River 2018

Years before, this river was used as a collector of untreated waters of the plant. Subsequently, after the closure of the plant activities, the landfill was constructed, which removed the solid waters of the copper processing industry from the river bank also.

Land protection is of great importance as one of the aims of landfill. A non-appropriate landfill structure can result in contamination of the groundwater and indirectly would damage the flora and fauna of the area, which is rich and diverse. In the plant part, wastes are located and are assembled to be ready for transportation to the landfill. Land protection has a major importance as this can cause groundwater pollution and indirectly damage the flora and fauna of the area that is rich and diverse. Wastes of the plant were collected and transported to the landfill. The polluted surface of the soil was removed from the area, because the bad smell and the dust coming from it were dispersed by wind. One part of the waste was treated with lime to perform heavy metal bonding. Then these wastes were placed in the grove prepared for this purpose. Waterproofing materials were used for the structure of the landfill. Such as: clay and high-density and polyethylene geo-membrane 2mm thick (Cazizni et al. 2008, 43). Figure 8 shows moments during the process of geo-membrane and geo-textile placement.

Figure 8. The process of geo-membrane and geo-textile placement (Mirdita Municipality, 2010).

2.3.2 Air Quality in Rubik

In the period when Rubik's copper smelting activity was in activity, considerable amount of sulfur dioxide was discharged into the atmosphere (UNEP 2000, 36). In the workers at that time, these concerns were noted: cough with secretions and blood, redness, tonsillitis, chest pain, watering, blood on the nose, bruising, paralysis of the respiratory apparatus and often "shock". (Daci, 2013, 88.) The Landfill of Rubik city and its location close to Fan River is shown in Figure 9.

The major sources of air pollution have been industries for chromium smelting, copper, cast-iron, cement and steel metallurgy and thermos plants. However, after 1992 those industries were closed.

Figure 9. Landfill of Rubik city and its location close to Fan River.

Some legal initiatives related to the quality of ambient air and fuel taxation have been undertaken during 2014-2015. DCM no. 594, dated 10.09.2014, adopted the National Strategy of Environmental Air Quality and Law no. 162/2014, dated 04.12.2014 "On quality protection environmental air ". This protects the air quality and is based on Directive 2008/50 / EC and 2004/107 / EC. The law came into force in December 2017. (REC 2014, 10.) According to those Directives, EU daily rate for SO_2 µg/m^3 is 40 (not to be passed more than three times per year), for NO_2 is 40 µg/m^3 and PM10 is 50 µg/m^3 (European Commission 2017b). According to DCM for the adoption of provisional norms of emissions in the air and their application 2003/248, the annual rate of SO_2, NO_2 and PM10 were 50 µg/m^3. As it is obvious, annual norms of air emissions in Albania Law are higher than those provided by EU Directives.

The Albanian National Environment Agency carried out air monitoring in 2017 to assess the air quality in the Rubik area. These data have been taken into account in the current study.

Figure10. Air monitoring site for 2016 in Rubik.

These data were collected from the Municipality of Mirdita, which includes this area. The measurement point is in southern part of landfill as shown in Figure 10.

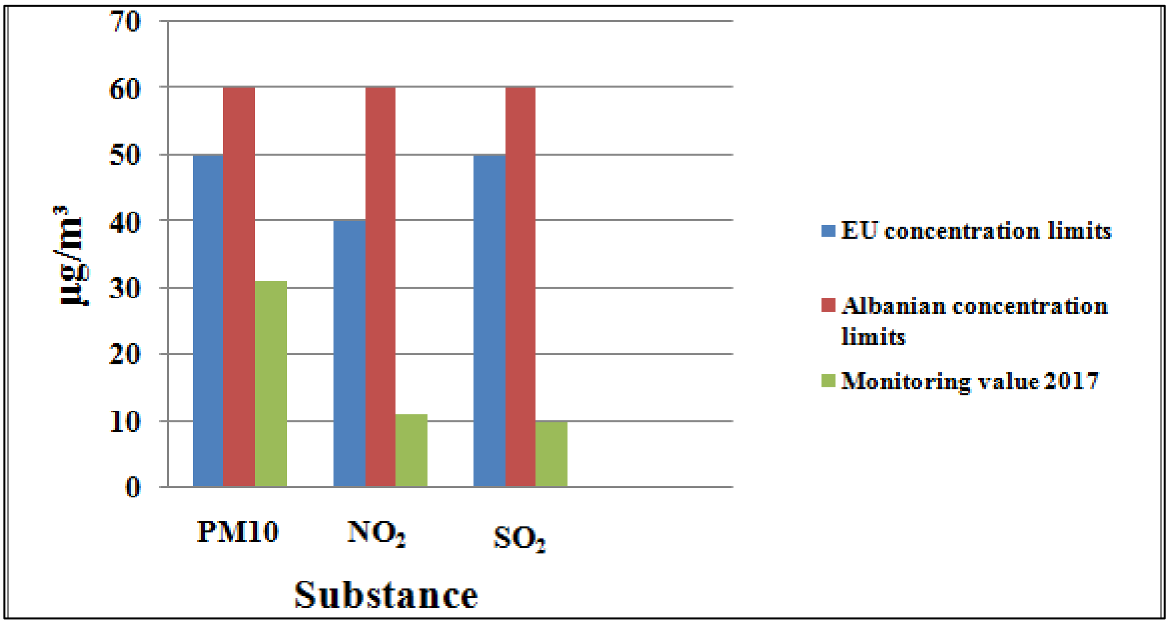

Figure11. Comparing the pollutant concentration standard under EU law, Albanian Law and values of 2017.

Based on the results of 2017 in the Rubik area, the air quality is very good and the parameters of the three elements analyzed are below the maximum allowed norms from EU standards and Albanian standards. Respectively, PM10=31 $\mu g/m^3$, NO_2=11$\mu g/m^3$, SO_2=10 $\mu g/m^3$, as shown in Figure 11. A table is created in addition and shown in Appendix I.

2.3.3 Soil Quality in Rubik

Lack of solid waste management is a highly sensitive issue in Albania. With the transition of the Albanian society to the market economy, the solid waste production in the country grew fast. Rapid growth of the population, urbanization and flourishing of activities, rising imports seemed to be an inefficient waste management system. Solid waste was discharged in inappropriate places and banned, thus spotting the entire space around, residents and animals with dangerous poisonous substances. Such areas are often found near rivers or other aquatic facilities that are used for drinking water. Another problem was the dumping of industrial and hazardous waste. (Tola et al. 2017, 164.)

In the Rubik area, soil contamination was reduced after the landfill was built because the majority of the contaminated material was removed from the former factory and during the excavations has reached its natural level, which is gravel land.

Figure 12. Landfill area covered with grass,2018.

After the implementation of the landfill, there are no more wastes into the surface of this area. There is not polluting discharges in the area where the landfill will be built. Only the groundwater of the plant in the rainy season are polluted by the material collected time ago, from the time when the Rubik Plant was put into operation until its closure. All the materials that have been assembled to date such as: bricks, building waste, etc. are arranged and in some cases, those of asbestos are placed in the container. Also, all materials on site such as grease, oils, etc. are arranged side by side and divided by categories and apart from each other. The place is very clean and the landscape is pleasant compared to the previous situation where there was rubbish dumped by residents and Rubik, like wood, metal waste, etc. Now, can be said that the entire surface where all the contaminated plant material will be collected, is total isolated. It is obvious that ground protection measures are in maximum and guarantee long-term protection from heavy metal waste. Figure 13 shows view of the former copper plant in Rubik.

Figure 13. Photos showing the view of the former copper plant in Rubik, 2018.

3 Fieldwork Research

This part of the study is about conducting fieldwork research aiming to know the concentration of heavy metals in Fan River waters. Rubik area is a hot-spot area regarding environmental issue, due to its industrial activities for more than 40 years. (UNEP 2000, 36.) The landfill construction raised also the warning of environmentalist specialist and different NGO because it was constructs close to Fan River. After 1990 in some areas along bank river, private company of constructing materials toke place and proclaimed their area as private ownership. Due to lack of legislation and fines for illegal activities, they continued to work with any precaution regarding environmental pollution and regression. Many industrial wastes of the past industry activities were thrown into the bank river without any treatment or selection.

Those critical environmental issues for Rubik, urged me to make a study regarding heavy metals concentration in Fan River. I tried to find other studies or materials regarding water contamination of Fan River, but it was a big gap of data sources. So, I stared to gather general information regarding Rubik area and incorporate them with my data and conclusions.

3.1 Sampling Design / Environmental Sampling Design

The sampling design is a crucial part in the process of data collection aiming to reach scientific conclusions. In order to ensure the data sufficiency for accurate conclusions, a well-developed sampling design is a key factor. A sound result in any study depends on data accuracy. When applying sample design in environmental studies about contamination, to generating accurate data about the level of contamination, the following should be considered:

1. The appropriateness and accuracy of the sample collection and handling method;
2. The effect of measurement error;
3. The quality and appropriateness of the laboratory analysis;
4. The representativeness of the data with respect to the objective of the study.

A sample design is made up of two elements: *sampling method* and *estimator*. *Sampling method* are the followed rules and procedures when elements of the population under investigation are included in the sample. The most common methods are: simple random sampling, stratified sampling, and cluster sampling. The estimation process for calculating sample statistics is called *the estimator*. Different sampling methods may use different estimators. Similarly, the formula for the standard error may vary from one sampling method to the next.

3.1.1 Sampling Design Used in Environment Research

Sampling design was the first method which was conducted in the fieldwork research. Sampling design is a roadmap to this research and the right method for obtaining results about the study. By following the steps of sampling design, I had a clear frame of my study work structure. (Elbasani 2003, 408.)

The main goal of sampling design was to support the conclusion about whether surface water contamination levels from heavy metals were exceeding the acceptable threshold of risk. The first objective of sampling method was to characterize the nature and extent of heavy metals contamination at two random sites in the Fan River, and then to estimate the mean characteristics of this population. Statistical methods to determine the final data of concentration for each element have been used. Sampling design also supported on detecting the boundaries for an elaborated analyses of heavy metals concentration and other environmental issues.

Defining the target population, the sampled population, and the sampling unit is an important step in developing a sampling plan. The **target population** (population - a group of individual persons, objects, or items from which samples are taken for statistical measurement) is the set of all units that comprise the items of interest in a scientific study. The **sampled population** is that part of the target population that is accessible and available for sampling. A **sampling unit** is a member of the population that may be selected for sampling, such as a specific volume of air or water. (Elbasani 2003, 410.)

3.1.2 Create Sampling Design to Collect Environmental Measurement Data

The target population in sampling design of this study is the landfill in Rubik area, and the sampled population is the surface water of Fan River in this area. The sampling unit in this study is water samples, token with polyethylene bottles of 1 liter. The bottle was treated with distillated water and nitric acid, labeled and closed. Before taking the water, the bottles were filled and pour out three times with river water. The bottle sample was filled 90% to allow the water to mix before analyzing with Atomic Absorption Specter.

To determine the concentration of heavy metals that the surface waters of the Fan River, water samples were drawn from two randomly located sites 1 and 2. Site 1 in the south of the landfill area and site 2 in the north of the landfill area. The sites were chosen randomly, without taking into account any consideration for the time, place or weather, named as simple random sampling method.

The objective of this study was to address whether the heavy metals are present contaminant of the river water, thus the data collected from only two sites of Fan River are thought to be enough representative for the analytical results of water samples (for instance, if the objective would have been to estimate the average concentration level of the entire river then random sampling locations should be generated from the entire site of the river, different depth, by including much more samples and during a long period of time). For a better representativeness of the data, water samples were collected at two different points of the Fan River apart from each other.

The objective of choosing those sites, were to see from the results if the water was more contaminated after passing the landfill area, which is 80 m in distance with the river flow. The Site 2 was chosen under the bridge on the part of landfill area where the river flows. The Site 1 was chosen under the bridge in south part of landfill area where the river flows. The process of choosing sampling design consisted in three factors: the area of concern, data quality, and constrains. A chart of this process and the description of these three relevant factors are shown in Figure 13.

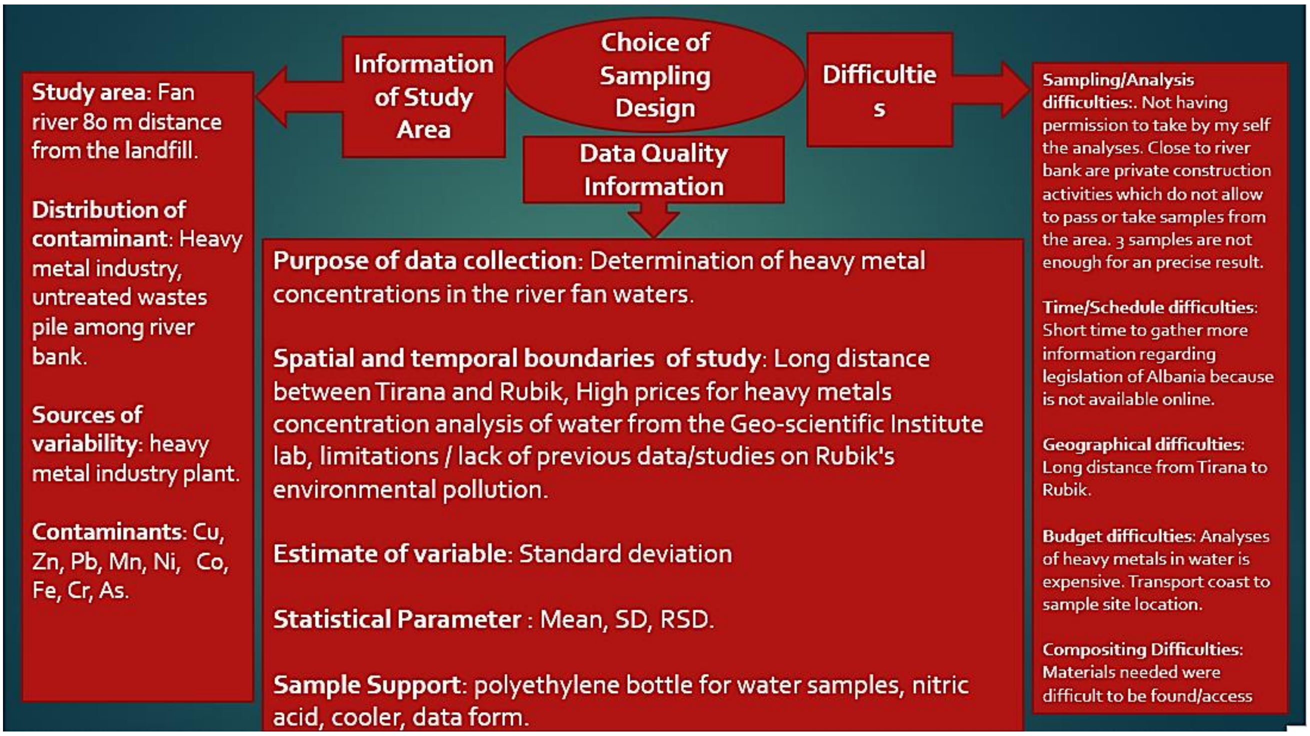

Figure 14. The process of selecting the sampling design and its factors.

3.1.3 The measurement Protocol

Measurement protocol is the next step applied in sampling design after the sampling unit is selected. A measurement protocol is a specific procedure for performing analyses to determine the characteristics of interest for each sampling unit. The measurement protocol includes the procedures for collecting a physical sample, handling and preparing the physical sample, applying an analytical method (including the sample preparation steps) to obtain a result (that is, to obtain the data for the sample), and protocol for resampling if necessary. (Elbasani 2003, 310.)

The random sampling by taking three replicators for each of two sites has been applied in this study. Afterwards, a measurement protocol has been applied as described in the following paragraphs.

Sampling

High-density polyethylene plastic bottles were used for the sampling of water samples and acidified with pH <2 with nitric acid. This was done to avoid suction of cations from the inner layers of the bottle and to minimize the perception of cations during the conservation. The container should be 90 % filled with the water sample to allow mixing before the analysis.

The samples were stored at a temperature of <4 ºC in a freezer container token from the laboratory in advance. Previously, they were purified in the laboratory with nitric acid, washed 2-3 times with distilled water and labeled. Each sample was taken with 1-liter bottles, labeled, and sealed with cork. (Sulce 2002, 66.)

Applying an analytical method to obtain results

For analysis of the samples were used techniques of atomic absorption spectrometry (A.A.S) in Laboratory of Geosciences Institution. A.A.S is thus called because this is based on the study of emission spectrum of various substances. Realization of spectrum becomes possible only if the analyzed substance is heated in the flame of a gas bulb up temperature to 2000-3000 ºC or in an electric arc where the temperature ranges from 5000 to 7000 ºC. The substance in these conditions vaporizes and dissipates in the atoms and the ones which release the prism spectrum of the spectrum meter. In the experiment, a number of light lines of different colors appear to form the linear spectrum. This spectrum of each element is characterized by continuous spectrum lines having a defined wavelength and in accordance with the nature of these lines judging the presence of the element and the concentration of the subordinate element with the intensity of the spectral line. (Karanxha 2001, 181.)

Statistic Analyze

The data for site 1 and 2, were calculated by using the main and standard deviation of the three replicates, for each element. By following the steps as below:

1 - Calculation of the mean of the samples, for each element from three samples, for each site. The formula for mean is:

$\bar{x} = \sum x_i/n$, x_i are the data value and n is the number of replicates. (Elbasani 2003, 55.)

2 - Standard Deviation of the three values received by the replicators for each element, for both sites, is calculated by using online calculator *Statistic How To*. SD shows how far from the mean are the data.

3 - Relative Standard Deviation is expressed in % and is calculated with the formula: (SD*100)/n. (Elbasani 2003, 58.)

4 - The values obtained are presented as above:

$\bar{x}$ ± RSD% or $\bar{x}$ ±SD. (Elbasani 2003, 74.)

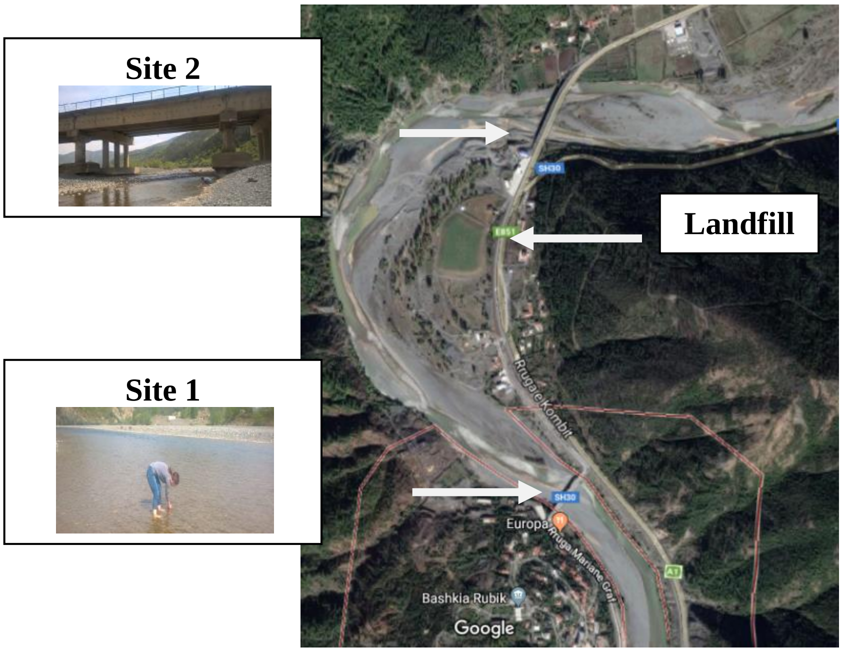

Figure 15. Location of Site 1 and Site2 on Fan River.

3.1.4 Heavy metals concentration and classification of Fan River waters

To evaluate more precise data, three samples were taken from Site 1 and 2 of water river. Data for concentration of heavy metals in each sample, where organized into a table and calculated according to mean, standard deviation and relative standard deviation formulas.

The data were registered into tables to have clear data information for each element token in analyze. The range of the mean values of each element concentration is presented on the last column of the tables 1 and 2.

Table 1. Site 1 heavy metals concentration, standard deviation and relative standard deviation values.

Site 1						
Element	**Sample**	**mg/l**	**Mean**	**SD**	**RSD (%)**	**Element concentration**
Cu	S1. R1	0.001	0.001	0	0	0.001
	S1. R2	0.001				
	S1. R3	0.001				
Zn	S1. R1	0.051	0.054	0.0035	0.116667	0.054±0.0035
	S1. R2	0.058				
	S1. R3	0.054				
Pb	S1. R1	0.078	0.074	0.014	0.466667	0.074±0.014
	S1. R2	0.085				
	S1. R3	0.058				
Mn	S1. R1	0.004	0.004	0.001	0.033333	0.004±0.001
	S1. R2	0.003				
	S1. R3	0.005				
Ni	S1. R1	0.032	0.034	0.002	0.066667	0.034±0.002
	S1. R2	0.035				
	S1. R3	0.036				
Co	S1. R1	0.007	0.007	0.0006	0.02	0.007±0.0006
	S1. R2	0.008				
	S1. R3	0.007				
Fe	S1. R1	0.072	0.078	0.007	0.233333	0.078±0.007
	S1. R2	0.085				
	S1. R3	0.076				
Cr	S1. R1	0.008	0.008	0.001	0.033333	0.008±0.001
	S1. R2	0.007				
	S1. R3	0.009				
As	S1. R1	0.011	0.012	0.0015	0.05	0.012±0.0015
	S1. R2	0.014				
	S1. R3	0.012				

Table 2. Site 2 heavy metals concentration, standard deviation and relative standard deviation values.

Site 2						
Element	**Sample**	**mg/l**	**Mean**	**SD**	**RSD (%)**	**Element concentration**
Cu	S2. R1	0.001	0.001	0	0	0.001
	S2. R2	0.001				
	S2. R3	0.001				
Zn	S2. R1	0.042	0.047	0.004	0.133333	0.047±0.004
	S2. R2	0.049				
	S2. R3	0.05				
Pb	S2. R1	0.009	0.007	0.0015	0.05	0.007±0.0015
	S2. R2	0.007				
	S2. R3	0.006				
Mn	S2. R1	0.004	0.004	0.0006	0.02	0.004±0.0006
	S2. R2	0.004				
	S2. R3	0.003				
Ni	S2. R1	0.033	0.037	0.005	0.166667	0.037±0.005
	S2. R2	0.042				
	S2. R3	0.035				
Co	S2. R1	0.008	0.008	0.0006	0.02	0.008±0.0006
	S2. R2	0.008				
	S2. R3	0.007				
Fe	S2. R1	0.089	0.095	0.007	0.233333	0.095±0.007
	S2. R2	0.097				
	S2. R3	0.1				
Cr	S2. R1	0.004	0.003	0.001	0.033333	0.003±0.001
	S2. R2	0.003				
	S2. R3	0.002				
As	S2. R1	0.012	0.011	0.0015	0.05	0.011±0.0015
	S2. R2	0.011				
	S2. R3	0.009				

The mean values of each element concentration were presented as grouped bar chart. The chart shows a high concentration of iron and nickel on site 2. Cu, Pb and Mn have the same concentration values at both study points in the Fan River. As and Co have a very small difference between their concentrations on site 1 and site 2, and shows their higher concentration on site 2. These differences of Heavy metal concentration in site 1 and site 2 are shown in Figure 16.

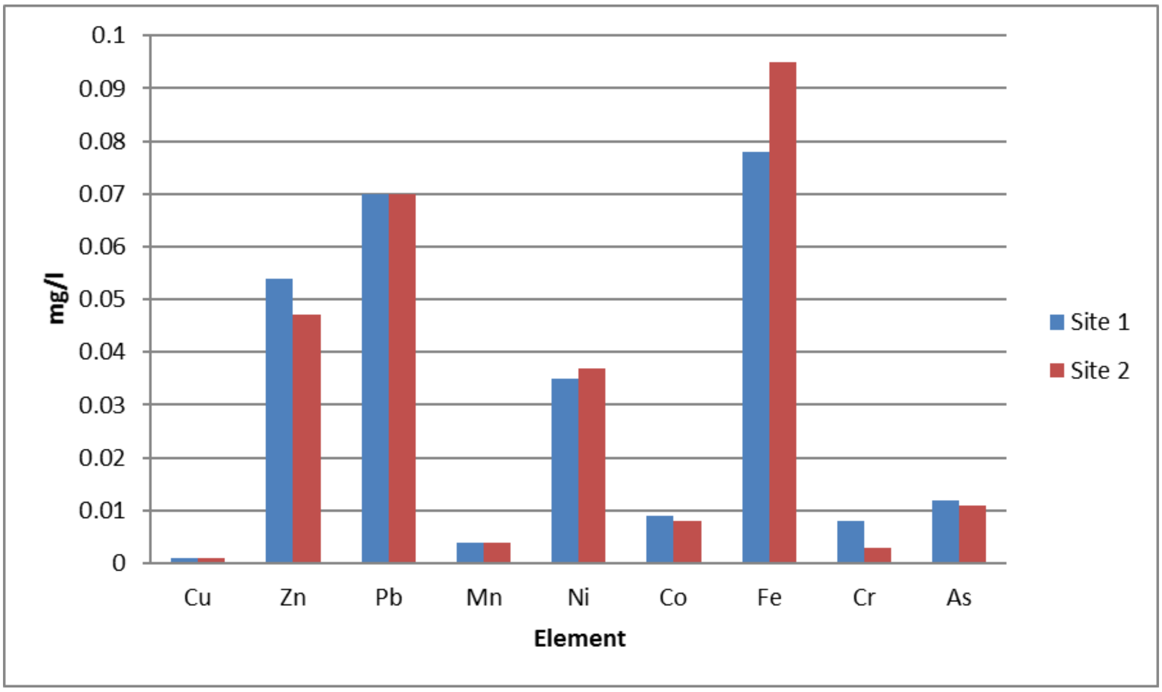

Figure 16. Defining the differences of Heavy metal concentration in site 1 and site 2.

As mentioned, waters are classified into five classes. To determine the water quality according to the content of each of the heavy metals, the Table 5 shows the concentration of the element for each water class category. The most common indicators for assessing the water quality in rivers are: the content of organic and inorganic concentration, nutrients, inorganic nitrogen and the content of micro-nutrients.

According to the parameters in DCM 2005/177, river waters are classified in five classes: *class 1A*, *class 1B*, *class 2*, *class 3* and *out of class*. This classification is shown in Table 3. The moderate status of *class 2* is considered to be a minimum acceptable water quality class. According to the head of IGEWE (personal communication, 06.02.2018) each class has its own description as follows.

Class 1A - very pure oligotrophic water, which in natural state with eventual disinfection can be used for drinking, for the production and processing of food products and presents an environment for the growth and cultivation of fish. The buffer capacity of the water is very good. It is constantly focused on oxygen, low in nutrients and bacteria. Their color is blue.

Class 1B - Less polluted water, mesotrophic water, which in natural state can be used for swimming and recreation, for the growth of the fish, can be used for drinking, for the production and processing of food products. The buffer capacity and oxygen saturation throughout the year are good. Those waters have green color.

Class 2 - average eutrophic water, which can be used for irrigation in natural state, and can also be used in industries where water quality is not needed. The buffering capacity is poor, but it is still suitable for growing of the fish. There is evidence of loads of harmful substances and microbial contamination. The color of the water is yellow.

Class 3 - powerful eutrophic water, polluted water that can be used only after proper processing. The buffer capacity is exceeded, leading to the light levels of acidity, by affecting fish growth. There is the "bloom" of alga by causing anaerobic conditions and starvation of fish. Microbiological pollution does not allow this water to be used for recreation. Concentration of harmful substances can vary from chronic water life levels to acute aquatic toxicity. The water color is red.

Out-of class - highly polluted hypertrophic water, which in natural condition cannot be used for any use. Water has not buffering capacity and its acidity is harmful for the fish. Concentration of harmful substances reaches the acute levels of toxicity to aquatic fauna. Major problems occur in the lack of the oxygen, leading so to anaerobic conditions. Terminology used for these waters would be waters of a “bad status” and its color is black.

Table 3. Water Classes based on concentration of heavy metals (mg/l). (according to DCM No. 177, dated 31.03.2005).

No.	Element	Class 1A(mg/l)	Class 1B(mg/l)	Class 2(mg/l)	Class 3(mg/l)	Out of class (mg/l)
1	Cu	< 0.05	0.05	0.1	1	>1
2	Zn	< 0.3	0.3	1	5	>5
3	Pb	< 0.05	0.05	0.05	0.05	>0.05
4	Mn	0.1	0.1	1	2	>2
5	Ni	<0.01	0.02	0.05	0.1	>0.1
6	Co	<0.01	0.02	0.05	0.1	>0.1
7	Fe	<10.5	1	3	5	>5
8	Cr	0.025	0.05	0.1	0.25	>0.25
9	As	≤ 0.01	0.02	0.05	0.1	>0.1

Taking into consideration the water analyzes obtained for this study; I set the values in the tables according to the respective limits. For Site 1 and Site 2, the table content of the classes is the same feature. By integrating the results from the sampling sites (Figure 14) and the corresponding classes (Table 3), the following results have been derived for the sampling sites of the present study (Table 4).

Table 4. Classes for the water quality for the Fan River (monitoring values) according to each element concentration.

Element	SITE 1	SITE 2
Cu	Class 1A	Class 1A
Zn	Class 1B	Class 1B
Pb	Out of class	Out of class
Mn	Class 1A	Class 1A
Ni	Class 1B	Class 1B
Co	Class 1A	Class 1A
Fe	Class 1A	Class 1A
Cr	Class 1A	Class 1A
As	Class 1A	Class 1A

From Table 5 is observed that concentration of Cu, Mn, Co, Fe, Cr, As - are categorized in values of the first class of water quality, for both sites. Zn is categorized in second class, which means that it is close to the limit of becoming a risky polluter. Pb is categorized in values of the fifth class of water quality.

According to water analyses undertaken from Albanian Geologic Service Laboratory, the main polluter of the water was iron (Fe). According to water qualification regarding its intended use, iron concentration sort Fan River as Out of Class category. The water can`t be used for any purpose and the rehabilitation from this contaminant requires an immediate plan. Another polluter was zinc (Zn) and nickel (Ni), which categorized Fan River as Class 1B. The water can be used for a restricted purpose, but still it needs some treatment before use. The other elements are relatively in small concentration by categorizing the Fan River in class 1A and waters can be used for any purpose without treatment.

Mirdita Municipality did a monitoring of air quality in Rubik area in 2017. The monitoring went throughout a year regarding three pollutants element: NO_2, SO_2 and PM10. Their concentration was showing good air quality from those pollutants. I did cooperation between air pollutants standard limit concentration from Albanian Legislation and EU policy. Even if Albanian legislation has a higher level for limit concentration of air pollutants comparing with EU standards, Rubik air is clean and under those limits.

3.2 Interviews

Qualitative research was conducted by interviewing experts regarding health issues consequences from heavy metal pollution and residents of Rubik regarding past and present environmental issues were conducted. These interviews are described as follows.

Interview with expert – Head of IGEWE

An additional method of primary research performed in this study was the in interview with the head of the Institute of Geosciences, Energy, Water and Environment. With a long experience in the field of hydrology and hydrology, I was given the opportunity to familiarize myself with the necessary and valuable information for my study with reliable sources and references. In IGEWE, access to information was moderated but with the support of the head of this institution I could provide information about the hydrology of Albania, climate, environment, water economy, mineral resources of Rubik etc. The interview with the Head on Institute of Geo-science, Energy, Water and Environment, helped me to better understand the climate of Albania, how the environmental issues were connected with climate factors, information about mines in Rubik area etc.

Interview with expert – Pneumologist at the Trauma Hospital in Tirana

The study took into consideration the professional knowledge of the pneumologist at the Trauma Hospital in Tirana. During the interview with him, he explained how heavy metals, from important elements to human’s life, could turn into endangered life threats.

Interview with problem area residents

Part of my study was interviews with residents of Rubik, how they had experienced the decision to build a landfill and how they now looked at the situation. One of the oldest changes in the environment that they have observed is cleaning the area from heavy metal industry waste and urban waste left untreated. Now they have a clean and unsafe environment for their children who often find themselves in the mining waste piles. The dangers of building a safe landfill were great for residents of the area.

At the beginning, they had expected the construction of the landfill with protests and with skepticism. But designers and engineers do the works by explaining the construction steps from time to time so that residents knowing step by step with the waste disposal process. Upon completion of the landfill until today, they have a high degree of assurance that the landfill facility is within any kind of security measure for non-use of the environment and groundwater, and most importantly for the Fan River flow.

3.3 Conclusions

The area chosen to implement the construction of the landfill is not the most suitable place because it is very close to the Fan River. The location is selected at the former copper Plant due to land ownership. From a geological point of view, the site consists of kaolin - type clay. A regular monitoring of the structure parts of landfill is required. With the construction of the landfill in Rubik, was achieved the following:

- Prevention of water pollution from heavy metals, except lead which is still is in very high concentration;
- Preventing air pollution;
- Creating a clean area from the urban wastes;
- Promotion of responsible land management and preservation;
- Prevention of risks and losing a nice landscape.

The air is unpolluted from SO_2, NO_2 and PM10, but data about their concentration in 24 hours and a week are missing. The superficial land appears clean from urban waste. Two wells of landfill monitoring appear out of order. The lack of online legal resources of the Republic of Albania is a major obstacle to obtaining accurate and updated information and data for the current environmental legislation in power. The high cost of laboratory analysis from the IGEWE Laboratory limits researcher and scientific research that require a large number of samples to get accurate data. The presence of private enterprises along the Fan River can create conflicts with people who want to do measurements and studies in that area. It is necessary to equip anyone who is interested for study reasons to visit the area with a permission approved by Mirdita Municipality.

Iron and nickel are found at higher levels in the river flow area to the north of the landfill compared to the flow to the south of the landfill. Zn, Cr, Co, and As are located at high levels in the River Fan River south of the landfill, compared to the flow to the land of the landfill. It is recommended to prohibit gravel exploitation on the river bed which affects the breakdown of hydrodynamic and hydrochronic equilibrium in the Fan River. It is recommended that competent authorities exercise authority over the protection of surface water, subsoil and ecosystem as a whole. Updating the monitoring system, recording data and information and making them public and accessible to the public.

Fan River waters are characterized by very high contamination of lead and its water is classified as outside class category. The Fan River waters are characterized by zinc and nickel pollution, which categorize the water as category 1B class. The heavy metals Cu, Mn, Co, As, Cr and Fe are found in small concentrations on the Fan River by categorizing it according to these concentrations in the category 1A class. Should be found the source and cause of concentrations above the permissible norm of Fan River, lead, nickel and zinc. Fan River waters need immediate treatment and rehabilitation from high concentration of Pb. Long-term monitoring of heavy metals in the Fan River is necessary to create an updated framework for its environmental state.

4 Discussion

A healthy environment status is related to the health of population, strong economy, good social life, rich flora and fauna. One of the environment elements is the water found in the nature. The scope of this thesis, represented in the main research question, was to find out what is the state of Fan River waters according to heavy metals concentration.

The answer to this question is provided from both primary and secondary research conducted in this study. Primary research methods such as interviews and sampling design has been used by having in focus. Tools such as sample bottles, cooler, and Atomic

Absorption Specter have been used for gathering insights and analysis. Furthermore, it was proven that was a considerable lack of data provided from governmental institution for systematic monitoring of environmental status in Rubik. These results have also provided an evaluation of the actual status of landfill structure, like the monitoring well out of function status.

For monitoring the maintenance of the landfill structures and features, I took evidences of monitoring wells which are not functioning as they are supposed to be. Those wells were supposed to be part of monitoring underground waters coming from the lixiviants of the solid wastes. There are some data of the concentration for heavy metal taken from those wells, but the last date of its monitoring is 2011. Now those wells are out of order, because of vandalism.

In cases where monitoring of these studies is done in completely dry weather or low precipitation, then Cu and Zn concentrations in mining water are found to be relatively low, compared to rain-track monitoring. It is also found that the concentrations of some elements measured at some monitoring stations are significantly increased or decreased depending on the month of the month they were taken, which is related to the amount of water intake and its acidity.

Solid waste that is not properly managed constitutes a major environmental risk, with negative implications for human life and environmental sustainability. Worldwide, landfills are known for the release of a wide range of harmful pollutants such as gases, liquid streams and particulate matter that can cause disease to humans and cause air, soil and soil pollution of water. (Daci 2013, 90.) Air monitoring is also important for the Rubik area, having a history of dust and gas emissions from the factory and the copper mining industry. The limits of air pollutants concentration in the air are defined by Albanian law.

One of the dangers of the construction of the landfill in Rubik was the flow of water from landfill burial waste, their joining with underground waters and filtering on the ground to pour into the Fan River.

For me as a student, the importance of this thesis was to know better the actual status of Rubik area and heavy metals concentration of Fan River flow close to landfill of industrial waters. I have learned how to choose the right methods, how to arrange them in a certain sequence through the process, and sometimes how to combine them with each-other, for accomplishing specific objectives. The most important lesson was about gathering the right data and to calculate them on the right methods, to have at the end reliable results. To know boundaries for the right of being inform ac cities, as a student, regarding environmental issues and accessing easily into legislation and government institution. In the end, I benefited from this thesis by improving my knowledge and improved my academic writing skills.

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Table 4. Classes for the water quality for the Fan River (monitoring values) according to each element concentration. Created by Totaj, O. 2018.

Appendices

Appendix I – Albanian standards versus EU standards

Comparison between EU Limits - Albania limits and monitoring values in 2017 carried out by Mirdita Municipality, for air quality standards.

Element	EU Limits	Albanian Limits	Air Monitoring 2017
PM10	50 (μg/m^3)	60 (μg/m^3)	31 (μg/m^3)
NO_2	40 (μg/m^3)	60 (μg/m^3)	11 (μg/m^3)
SO_2	50 (μg/m^3)	60 (μg/m^3)	10 (μg/m^3)

Appendix II – Laboratory determination of heavy metals into water

The following paragraphs describes how the concentration of heavy metals into water is determined in laboratory.

Determination of copper (Cu) with A.A.S.

Principle of the method: Cu determination method with A.A.S. is based on the measurement of the amount of electromagnetic radiation emitted by Cu atoms with wavelength "λ" 327.4 nm. The content of the elements on request is expressed in% or p.m (parts per million).

Determination of zinc (Zn) by A.A.S.

Principle of method: The method of determination of Zn by A.A.S is based on the measurement of the amount of electromagnetic radiation emitted by Zn atoms with wavelength "λ" 213.9 nm. Highly purified chemical reagents and deionized water are used for analysis. The content of the elements on request is expressed in% or in p.p.m (parts per million).

Determination of Pb with A.A.S.

Principle of method: The method of determination of Pb with A.A.S is based on the measurement of the amount of electromagnetic radiation emitted by Pb atoms with wavelength "λ" 405.8 nm. Highly purified chemical reagents and deionized water are used for analysis. The content of the elements on request is expressed in% or in p.p.m (parts per million).

Determination of arsenic (As) with A.A.S

Principle of method: The volumetric determination method of arsenic is based on the titration of potassium aromatic solution of arsenic after its reduction from As + 5 in the sample diluted with sulfuric acid.

Determination of Manganese (Mn) with A.A.S

Principle of the method: The determination method of Mn with A.A.S is based on the measurement of the amount of electromagnetic radiation emitted by atoms of Mn with wavelength "λ" 403.1 nm, highly pure chemical reagents and deionized water are used for analysis. The content of the elements on request is expressed in% or in p.p.m (parts per million).

Determination of Nickel (Ni) with A.A.S

Principle of the method: The determination method of Ni with AAS is based on the measurement of the amount of electromagnetic radiation emitted by the atoms of Ni with wavelength "λ" 341.5 nm, highly pure chemical reagents and deionized water are used for analysis. The content of the elements on request is expressed in% or in p.p.m (parts per million).

Determination of Cobalt (Co) with A.A.S

Principle of Method: Co-determination method with AAS is based on the measurement of the amount of electromagnetic radiation emitted by Co atoms with wavelength "λ" 345.4 nm Highly pure chemical reagents and deionized water are used for analysis. The content of the elements on request is expressed in% or in p.p.m (parts per million).

Definition of Iron (Fe) with A.A.S

Principle of the method: The spectrophotometric method of iron determination is based on the radiation measurement absorbed by the yellow complex forming the $Fe + 3$ ion with sulfosalicylic acid in the ammonia environment (pH = 8-11). Measurements are performed at the wavelength spectrophotometer "λ" = 440 nm.

Among the elements that are considered as potential toxic pollutants coming from mining waste are generally separated Zn, Pb, Cu, Ni, Cd, Hg, Mo and As. But even such elements as Cr, Co and Se may constitute significant contamination in specific areas. (Karanxha 2001, 186.

Appendix III – Transcription of interviews

Citizen 1

Me: What are this planting?

Citizen 1: Green onions.

Me: Is the soil good?

Citizen 1: Yes. We take a lot of production.

Me: You do not worry about being contaminated?

Citizen 1: No. Removing the remains of the plant has saved us and has opened us up to the surrounding area.

Me: How to look at the construction quality of the landfill?

Citizen 1: Very good. We have seen ourselves with our eyes as they have built it. They have secured it in the best way.

Citizen 2

Me: What are the watering?

Citizen 2: Grapes.

Me: Is the water from Fan River flow?

Citizen 2: No, the private business does not permit us to set up the water supply system. They declare it as private areas. Also, we don't know the water quality of the river. No one came to take analyses or inform us.

Me: Where do you find the water?

Citizen 2: From the well. It's good water.

Citizen 3

Me: How does the environment created by the landfill look like from your point of view?

Citizen 3: We are free of the bad small of waters thrown here without any criterion or control. There are no more piles of industrial waters. We have no information what they were, if they were radioactive, toxics or explosions materials. There are children who play around and explore, touching everything. We were afraid from diseases caused by those wastes

Me: What about landscape?

Citizen 3: Is beautiful. Is like our eyes are opened. Kids are playing here in the field that was created.

Me: Does anyone want to control any damage to the landfill?

Citizen 3: Jo. The one who build it left and no one has in mind now to come and see. Neither the government, neither the private groups.

The pneumologist at the Trauma Hospital in Tirana

Me: Does the body of human beings have heavy metals without being contaminated by an external source?

The pneumologist: Yes, it does.

Me: Are the people who breathe heavy metal contamination exposed to life threat?

The pneumologist: Yes, they are. Health problems can even cause life threatening

Me: Are people in life danger if eating food contaminated by heavy metals?

The pneumologist: Yes, they are. They are also found in nature and in animals. But at high levels they lead to death and contamination of the environment

Me: Are people at risk of contaminating contaminated material or heavy metal?

The pneumologist: Yes, they are. The heavy metals have an importance in the physiological functions of the human cell but also can cause death.

Me: What are the most common health issues as consequence of heavy metals contamination?

The pneumologist: Vomiting, losing conscience, skin disease, respiratory illness, cirrhosis, digestive tract disease, toxicity, ophthalmic problems.

Me: Can you underline some main health issues regarding chromium, lead, cobalt, nickel, manganese, arsenic, lead, zinc an copper?

The pneumologist: Chromium is toxic for liver, nephrites and gastrointestinal irritation and can cause dermatitis by skin contact. Chromium has potent carcinogenic effects on human and animals.

Lead is an important metal for the life of plant and animals. It is essential component in the hemoglobin structure and its luck in blood can cause anemia but in high doses it is toxic.

Cobalt deficiency is not common in humans, but we know cases from its toxicity. Cobalt is acutely toxic in large doses, and in cumulative long-term, low level exposure. Most of what is known about cobalt toxicity comes from cases of ingestion or inhalation of excess cobalt.

Nickel it induces as embryo toxic, allergic reactions and contact dermatitis. Nickel sensitization also occurs in general population from exposure to coins, jewelry,

watchcases, clothing and fasteners. Nickel is a potential carcinogen for lung and may cause skin allergies, lung fibrosis and cancer of respiratory system.
Manganese in high doses of inhalation can cause death. Miners have been found to have chronic poisoning if they are exposed to vapors of manganese compounds, but also individuals who drink water from contaminated wells.

Arsenic intake of 70 to 80 mg of trivalent arsenic (III) oxide has been reported to be fatal for man. Inorganic arsenic produces acute, sub acute and chronic toxic effects.
Lead is poisonous metal. Pb poisoning is caused by environmental exposure that causes mental retardation in many children, while mild poisoning results in anemia. Organic compounds of Pb easily reach human body through the skin and endanger the central nervous system. Kidney and liver are considered potential targets of Pb toxicity before storage in bones.

Zinc does not accumulate in the body. It is the enzyme activator. It affects bone growth, development and functioning of reproductive organs, etc. There are very rare cases of poisoning with Zn. Its action in aquatic life depends on its strength, saturation with oxygen and temperature. Zinc is an essential micronutrient that affects several metabolic processes of plants and has a long biological half-life.

Copper intake of excessively large doses of Cu leads to severe mucosal irritation and corrosion, hepatic and renal damage and central nervous system irritation followed by depression.

The Head of the Institute of Geosciences, Energy, Water and Environment (IGEWE)
Me: Is Albania rich with surface waters?
The Head of IGEWE: Yes, quite a lot.
Me: Is there a qualification of surface waters according to heavy metals concentration?
The Head of IGEWE: Yes, of course. Class 1A is very pure as named oligotrophic water; you can drink without any treatment. Can be used for the production and processing of food products and the fish can live and grow in this class of water. The buffer capacity of the water is very good. It is constantly focused on oxygen, low in nutrients and bacteria. Those are blue waters.
Class 1B is less polluted water, mesotrophic water, can be used for swimming, for the growth of the fish, can be used for drinking, for the production and processing of food

products. The buffer capacity and oxygen saturation throughout the year are good. Those waters have green color.

Class 2 are average eutrophic water, can be used for irrigation, and can also be used in industries where water quality is not needed. The buffering capacity is poor, but it is still suitable for growing of the fish. Harmful substances and microbial contamination are present in those waters. The color of the water is yellow.

Class 3 is for powerful eutrophic water, polluted water that can be used only after proper treatment process. The buffer capacity is exceeded, and the water has light levels of acidity. This affects the fish growth. There is the "bloom" of alga by causing anaerobic conditions and starvation of fish. Concentration of harmful substances can vary from chronic water life levels to acute aquatic toxicity. Microbiological pollution does not allow this water to be used for recreation. Those waters are red.

Class Out of class is for highly polluted hypertrophic water, which in natural condition cannot be used for any use. Water has not buffering capacity and its acidity is harmful for the fish. Concentration of harmful substances reaches the acute levels of toxicity to aquatic fauna. Major problems occur in the lack of the oxygen, leading so to anaerobic conditions. Those waters are named as “bad status” and its color is black.

Me: Is Rubik's area rich in minerals and heavy metals?

The Head of IGEWE: Yes, very much. Albania is a rich country regarding mineral resources, like chromium, cooper and iron-nickel. Albania during communism system used to have an intensive exploitation of mineral resources especially referred to copper mineral, for which was ranked the world`s third largest producer. Mines were built for heavy metals, plants for smelting and enrichment. All three of these heavy metal industry elements were present in the Rubik area and operated for 40 years or more. The mining sector used to have lack of management expertise, outdated equipment and technology. It is a nature crime that technological waste has been discharged without any kind of control mainly on river banks and sometimes directly on the river of Albania.

Me: Does the climate affect the quality of water?

The Head of IGEWE: Yes, in a considerable way.

Me: Can river water be contaminated from wastes from mines?

The Head of IGEWE: Yes, definitely.