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Degradation of the Pharmaceutical and Medical Waste on Landfills Due to Rainwater Infiltration

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ABSTRACT

In Bosnia and Herzegovina, there is a large number of non-sanitary landfills and landfills where pharmaceutical waste from households is being disposed, as well as the medical waste from different health institutions. Expired pharmaceuticals are mostly being disposed there, because there is very little attention paid on waste sorting at the place of its origin. Now days, there are thousands of different medicine, and mainly, along with the other household waste, expired antibiotics are disposed of too. Degradation of expired pharmaceuticals in the environment very often leads to formation of products with completely different properties in regards to the original compound, but also it is not uncommon for those newly formed products to be more toxic than the original ones. This paper analyses the impact of rainwater infiltration on the body of the landfill where expired pharmaceuticals are being disposed of, as well as other medical waste at municipal landfills in Bosnia and Herzegovina. Field studies on the size of the rainwater infiltration as a function of different parameters have been conducted, and also degradation analysis of different expired drugs that are being disposed of in landfills has been performed.

Keywords- pharmaceutical and medical waste, landfill, infiltration, degradation.

I. INTRODUCTION

Disposal of pharmaceutical waste, mostly expired pharmaceuticals from households, as well as the medical waste from health institutions, could cause a big problem, especially in those countries where that issue is not covered by legislation. It is not uncommon case that pharmaceutical and medical waste from health institutions (hospitals, health centers. regional dispensaries in rural and suburban settlements) are being transported, devoid of any special markings, with utility vehicles, and then disposed of on city landfills together with all other types of waste, and without any previous treatments.

Medical waste is a special type of waste, which is generated mostly in health institutions. It is a heterogeneous mixture of municipal, infectious, pathological, pharmaceutical and laboratory waste, disinfectants and packaging, as well as radioactive and hazardous chemical waste. Harmful and hazardous medical waste contains toxic, carcinogenic and infectious substances and according to the properties and place of its origin is divided into: pathological waste, infectious waste, sharp objects, pharmaceutical waste, and chemical waste.

Pharmaceutical waste is generated in the process of production of medicines, also those are medicines and chemicals that have been returned from the department where they had been spilled, spoiled, prepared, but unused and also expired drugs, or those medicines that should have been thrown away for any reason in health facilities and households.

In total environmental pollution of Bosnia and Herzegovina, medical waste does not occupy a large part, but it is potentially among the most dangerous types of waste because it can cause infections and poisoning. Pollution coming from different health facilities can be very dangerous for public health and ecosystem in which that waste is being stored and disposed off.

Groundwater and surface water pollution due to pharmaceutical and medical waste in solid and liquid state are listed global hazards, present both in industrialized countries and in developing countries such as Bosnia and Herzegovina.

According to the previous assessments, liquid waste from health facilities, which is very often ejected through wastewater, makes up around 75% of total waste in those health facilities. In Bosnia and Herzegovina, certain part of different medical and pharmaceutical waste (mostly from small rural and suburban settlements and private doctors' offices) is disposed of on communal landfills as well, which most often are not arranged according to basic European standards. According to some estimates, 20% od dangerous medical waste had ended up in ordinary waste collection containers [1]. During rainfall, such disposed pharmaceutical and medical waste on landfill could, by its complete or partial degradation and infiltration of storm water, very significantly pollute ground and surface waters, especially in those landfills which are not sanitary, of which there are large number in Bosnia and Herzegovina [6] [7].

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II. DISPOSAL OF PHARMACEUTICAL AND MEDICAL WASTE

The most common practice of pharmaceutical waste disposal in European countries is throwing it away together with other household waste. Disposing medicine in sewerage system is the second most common practice in some countries. It is a very little number of those consumers who return unused medicine in the pharmacies. Certainly, these numbers depend on how strict the laws for medical and pharmaceutical disposal are in different countries.

Medicine like antibiotics and antidepressants are considered to be the most dangerous for the environment, and their disposal on the landfills represents a significant problem due to infiltration and possible pollution of groundwater around the landfill.

In Bosnia and Herzegovina, preliminary estimates of the amount of medical waste generated in hospitals and health centers have been made, and according to WHO standards. Based on those preliminary estimates, around 10.000 to 15.000 tons of medical waste is generated per year, and that is without expired drugs. According to the Law on Waste in Bosnia and Herzegovina, the obligations and responsibilities of institutions and individuals on waste management are given. Healthcare institutions where the dangerous waste is generated are obligated to dispose it on ecologically acceptable way. Complexity and risk of the medical waste that is generated in the health facilities demand a complex waste management, both inside and outside the facility[1].

Waste from the health facilities is most often gathered without separation on its source, and mainly mixed with other municipal waste. Only 18% of waste from health facilities has been disposed of on the landfill, and 0,5% of the waste has been exported for harmless disposal, while the rest of the waste has been inadequately disposed of, and mostly with other municipal waste. Only in some healthcare facilities are used needles separated and disposed of in empty plastic or glass infusion bottles. The rest of the infectious material (used syringes, infusion and transfusion systems, urinary bags, gauze, etc.) is most often inserted into open and often rusty bins. Collection bins from individual hospital wards and other healthcare facilities are often taken out by hand to the ground floor, and then transported by trolley to a place for central disposal of waste, which is neither adequately fenced nor secured. Municipal waste is not being separated from dangerous medical and pharmaceutical waste to a greater extent.

On the landfills in Bosnia and Herzegovina medical waste separation from municipal waste is not performed. Current practice of such waste mixing on the landfills with inadequate work system can lead to a rapid spread of infectious diseases and to the pollution of groundwater due to degradation and solubility of different medicaments. https://doi.org/10.31033/ijrasb.8.1.16

Disposal of medical and pharmaceutical waste in Bosnia and Herzegovina is one of the very important problems. In the last eight to ten years, there has been an improvement in the legislation related to the disposal of waste from health care institutions. Although it is precisely prescribed how medical waste must be collected, sorted and disposed of, most health care institutions, especially in suburban and rural areas, have not started to apply the regulations yet.

New regulations have been adopted in Bosnia and Herzegovina about the waste disposal, but mostly the authorities do not monitor their implementation and do not apply punitive measures [8]. Disposal of medical and pharmaceutical waste on the municipal landfills is the cheapest way to take care of it. The impression is that health care institutions, in order to save money, use the lack of inspection supervision and do not apply the new legal regulations [1].



Figure 1: Medical and pharmaceutical waste on municipal landfills

III. IMPACT OF THE PHARMACEUTICALS ON THE ENVIROMENT

Significant impact of the pharmaceuticals in groundwater in Bosnia and Herzegovina had been spotted 25 years ago. It is important to state that on of the sources of pollution of the groundwater are landfills. Research to date has established that pharmaceuticals reach the groundwater by leaching from the landfills.

When we look at surface waters (rivers) then the retention time is a crucial parameter when measuring the concentration of pharmaceuticals in rivers. When the water from smaller tributaries flows in, elevated concentrations decrease very quickly below the dilution detection limit, while at higher flows the dilution effect is negligible. From this it can be concluded that river hydrology plays a key role in dilution for a particular location [7]. The main conclusion of many studies is that wastewater is the main source of pharmaceuticals in water systems, but their concentrations are drastically

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reduced by moving away from the source of pollution, that is, downstream [2].

The presence of pharmaceuticals in the aquatic environment depends on the treatment of wastewater, the type of pharmaceuticals, pharmacological and physicochemical properties of pharmaceuticals. In households, frequently used aspirin is metabolized to carbon dioxide, making it difficult to detect. Some other hydrophobic components will be sorbed into the sludge and broken down there into smaller molecules that can later be spotted and detected. The biggest problem is the development of bacterial resistance due to constant exposure to antibiotics. Some of the most active pharmaceuticals have shown poor biodegradability in wastewater treatment, namely ciprofloxacin, ofloxacin and metronidazole [2] [3] [5].

Pharmaceutical compounds have a great variety, and antimicrobial agents are of particular importance. Their occurrence in the environment is worrying due to the possible spread and development of bacterial resistance through permanent exposure. These effects can occur as a result of the discharge of wastewater from pharmaceutical plants and hospitals. Cases of resistant bacteria have already been reported in rivers. Consumption of large amounts of the antibiotics has led to their high prevalence in the environment. Antibiotics enter the environment through watercourses, by discharging wastewater into rivers. Other major sources of antibiotic occurrence are hospital and industrial effluents and effusions from pharmaceutical plants.

Pharmaceuticals differ significantly in molecular structure, molar mass and other physico-chemical properties, even though they may belong to the same group. Different functional groups of molecules affect their activity. The main properties of pharmaceuticals, important for dissolution at the landfill, are photostability, sorption on solid materials, biodegradability and solubility in water.

Biodegradation of pharmaceuticals is an important aspect of assessing their environmental fate and environmental risks. Biodegradation may result in partial decomposition or complete mineralization of the compounds in the aquatic environment and in the landfill due to the infiltration of rain into the body of the landfill. Biodegradation depends on the temperature measured during field testing, so low temperature values reduce the rate of degradation. This knowledge should be cautionary, especially for countries with cold climates, and during winter days in Bosnia and Herzegovina when temperatures fall below 0°C, because the air decomposition lasts longer and can lead to freezing of the soil and easier penetration into the environment waste [7] [8].

Photolytic degradation involves the degradation of a molecule by its exposure to light. On the surface of the landfill, photolysis can be direct and indirect. In direct photolysis, a certain pharmaceutical compound absorbs solar radiation, which leads to the decomposition of molecules, while in indirect photolysis, the compound does not have to or cannot absorb sunlight [5]. In indirect photolysis, the compound present in the sample absorbs light and then reacts directly with the pharmaceutical or a reactive intermediate is formed, which reacts with the pharmaceutical. The main species that absorbs solar radiation is dissolved organic matter present in surface waters (rainfall on the body of the landfill). Excitation caused by dissolved organic matter leads to the formation of various photochemically reactive intermediates (Photochemically produced reactive intermediates PPRI), including hydroxyl radicals, singlet oxygen, peroxy radicals and superoxide.

Pharmaceuticals and the expired drugs that absorb radiation, are subjected to direct photolysis. The rate of degradation is due precisely to the absorption of light. The half-life of pharmaceuticals during direct photolysis can last from a minute to several tens of days. Direct photolysis has been studied for just over 40 compounds, which is still a very small number considering the total number of pharmaceuticals that are currently disposed of in municipal landfills in Bosnia and Herzegovina.

IV. ANTIBIOTIC RESISTANCE DUE TO THEIR PRESENCE IN SOIL

Antibiotic resistance has become an important problem in recent years. Due to resistance, antibiotics are not competent to show their effect in certain infections, because certain bacteria have become "resistant" to certain antibiotics, due to frequent exposure to the antibiotics. It is not only thought that a person consumes antibiotics often and without adequate need (although this is a very common problem), but resistance can also occur due to unconscious intake of antibiotics due to their inappropriate disposal, so they can be found in drinking water, soil, and environment. Thus, a person can 'saturate' its body with certain antibiotics, and these bacteria find a way to overcome their effects and so they become resistant to them.

Antibiotics can get into the environment in a variety of ways. They mainly enter the environment through wastewater. Also, it is often the case that they are found in wastewater treatment plants, which indicates that they are very likely to fail to be completely removed during treatment, but can also be found in landfills [4].

Antibiotics are usually metabolized in liver, and excreted from the body in the form of their metabolites, through the kidneys, that is through urine or through the feces. As such, they then reach the environment through sewers, and can be found in the soil and water around us. The problem is often that although antibiotics may not be detected as such in the waters or soil where they are tested, although they are present in unchanged form, they are often present in the form of these metabolites and other degradation products that are difficult to detect and completely remove from the wastewater.

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The soil, and thus the body of the landfill, receives a large portion of the secreted antibiotics through fertilizer and sewage sludge. As a result, the soil becomes an inevitable hotbed of antibiotics. Some antibiotics are less, while some are more soluble. Penicillins are more soluble, while tetracyclines and fluoroquinolones are more stable, which allows them to survive longer in the environment (in the soil and body of the landfill if they are disposed of in municipal landfills), and to accumulate in higher concentrations [3]. In Bosnia and Herzegovina, there are no legal restrictions that would limit the discharge of the antibiotics and other pharmaceuticals, and their concentrations in surface and groundwater. Namely, the most common entry of antibiotics into the environment is through the effluents of municipal devices, where it is not possible to completely remove them from wastewater. This leads to significant pollution of soil as well as groundwater and surface water.

V. INFILTRATION IN THE LANDFILL BODY

This paper provides data on the measured infiltration for one period in 2016. Measurments of the infiltration had been conducted on the municipal landfill near Tuzla.

In order to obtain the infiltration model as well as the infiltration pattern, data on air temperature, relative humidity, temperature in the landfill body at a depth of 30 cm, as well as data on the compressibility of the landfill body were analyzed.



Figure 2: Rainfall height in 2016 near infiltration measurements

During 2016, several infiltration measurements were performed at a landfill near the city of Tuzla in Bosnia and Herzegovina. Figure 2 shows the levels of rainfall by months in 2016 in relation to the average precipitation levels for the period 1960-1990. From Figure 2 it can be concluded that during March, May, June and October the highest rainfall was near the experimental field of measuring the infiltration of the municipal landfill near Tuzla, and during September and December there was the least rainfall. Disposed expired medicines as well as medical waste were noticed at that municipal landfill.

Measurement of infiltration in the experimental field of the municipal landfill near the city of Tuzla was performed during the months of May, June and September 2016 using an infiltrometer. It was during these months that precipitation was above and below average rainfall heights.





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Figure 3 shows the amount of infiltrated water as a function of time during the month of May 2016. During the infiltration measurements in May 2016, the precipitation was higher than the average rainfall, and the air temperature ranged from 22° C to 28° C, and the relative humidity ranged from 45% to 57%. The measured dynamic compressibility modulus at the experimental infiltration measurement points at the landfill ranged from $5.34 \text{ MN} / \text{m}^2$ to $7.24 \text{ MN} / \text{m}^2$. The temperature in the body of the landfill at a depth of 30 cm ranged from $19.1 \,^{\circ}$ C to $19.5 \,^{\circ}$ C and was measured with a geothermometer. For the given measured infiltration data and the given data of air temperature, previous

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precipitation, relative humidity, temperature in the landfill body and compaction of the landfill body, an exponential equation is obtained which can be a good indicator of the amount of infiltrated water in the landfill body during rainy months, especially from the acpect of dissolution of expired medicines and medical waste and significant pollution of leachate entering the soil and groundwater, especially in unregulated landfills. This term is an indication of the size of the infiltration in the month when the rains were higher than the average rainfall by about 15%.



Figure 4: The amount of the infiltrated water in time during dry month

Figure 4 shows the amount of infiltrated water as a function of time during the month of September 2016. During the infiltration measurement in September 2016, the precipitation height was significantly lower than the average rainfall, and the air temperature ranged from 24° C to 29° C. The measured dynamic compressibility modulus at the experimental infiltration measurement points at the landfill ranged from 5.55 MN / m² to 7.41 MN / m². The temperature in the body of the landfill at a depth of 30 cm ranged from 20.4° C to 20.9° C measured with a geothermometer.

For the given measured infiltration data and the given data of air temperature, previous precipitation, relative humidity, temperature in the landfill body and compaction of the landfill body, an exponential equation is obtained which can be a good indicator of the amount of infiltrated water in the landfill body during dry months, especially from the aspect of dissolution of expired medicines and medical waste and significant pollution of leachate entering the soil and groundwater, especially in unregulated landfills. This term is an indicator of the size of the infiltration in the month when the rains were less than the average rainfall by about 45%.

Based on the measured data of infiltration that affects the dissolution of pharmaceutical waste,

exponential equations were obtained and those could serve as an approximate value of infiltration only into the body of the landfill during the previous rainy days and previous dry days. For infiltration of rainfall during the previous rainy days, the infiltration that dissolves old drugs in the landfill can be represented by the expression:

$$y = 1,1798e^{0.9658x} \tag{1}$$

Also, for infiltration of rainfall during the previous dry days, infiltration that dissolves old drugs in the landfill can be represented by the expression:

$$y = 0.8428e^{0.8754x} \tag{2}$$

y - infiltration time (minutes)x - amount of water (liters).

Expressions (1) and (2) represent exponential equations of the time of rain infiltration into the body of the landfill depending on the amount of infiltrated water. Both terms are directly related to the relative humidity, the compaction of the landfill body and the temperature in the landfill body. It is these indicators that affect the size of the infiltration and its duration.

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VI. CONCLUSION

The paper analyzes the measured infiltration data on the body of the landfill in Bosnia and Herzegovina from the aspect of dissolving discarded pharmaceutical and medical waste. Of particular importance are the expired drugs, which, depending on their chemical composition and structure, dissolve in water. It is known that water is the best solvent in nature, and in Bosnia and Herzegovina, expired medicines that end up in landfills are very often discarded from households. In general, landfills are largely unregulated, unsanitary, and pollution due to the dissolution and decomposition of the pharmaceutical and medical waste leads to a large pollution of soil and groundwater, because such landfills do not have a drainage system or sealing system in its foundation.

The exponential equations, given in this paper, represent the approximate value of rain infiltration time on the landfill body as a function of previous precipitation in relation to the field measurement day, relative humidity, air temperature, landfill body temperature and landfill body compaction at the measurement site. These equations can be used to obtain the time of infiltration of certain precipitation heights, that is, the volume of rain falling per m² of body surface area of the landfill.

There is currently little or insufficient information available on pharmaceuticals in the environment that would help to reach a final conclusion on the importance and impact of resistant bacteria in the environment and to enable the assessment of the risks to human health and the ecosystems around the landfills. Detection of pharmaceutical sources as contaminants represents the greatest potential for their removal. Reducing environmental pollution by pharmaceuticals includes a wide range of measures and actions, such as reducing prescribed doses in medicine and appropriate disposal of unused pharmaceuticals and those that have expired.

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