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## Municipal leachates health risks: Chemical and cytotoxicity assessment from regulated and unregulated municipal dumpsites in Lebanon



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### HIGHLIGHTS

### GRAPHICAL ABSTRACT

- Leachates analysis identified numerous organic compounds and heavy metals.
- High levels of Mn, Cr, Ni, Cd, nitrates and phosphates identified.
- Leachates triggered significant cytotoxic and genotoxic damages.
- Significant damages in blood, bone marrow and mice organs.
- Compounds identified posed significant health and carcinogenic risks.

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#### ABSTRACT

The proper management of municipal waste is critical for resource recovery, sustainability and health. Lebanon main approach for managing its municipal waste consisted of landfill disposal with minimal recycling capacity. This approach contributed to exceeding the holding capacity of existing landfills leading eventually to their closures. The closure of a major landfill (Naameh landfill) servicing Beirut and Mount Lebanon areas led to municipal wastes piling in the streets and forests for more than a year in 2016. The main problem identified in the municipal wastes consisted of untreated leachates (from regulated and unregulated dumpsites) going straight into the Mediterranean Sea. Therefore leachate samples were collected and subjected to chemical characterization followed by biological assessment. The chemical characterization and profiling of the Lebanese leachates were compared to results reported in Lebanon, Europe and United States as well as to the toxicity reference values (TRV). The biological assessment was conducted in vitro using human derived immortalized cell cultures. This strategy revealed significant alarming cellular organelles and DNA damages using in vitro cytotoxicity assays (MTS and comet assay). The significant damages observed at the cellular level prompted further animal model investigations using BALB/c mice. The animal data pointed to significant upregulation of liver activity enzymes coupled with significant damage expression in liver spleen and bone marrow DNA. The presented research clearly indicated that there is an urgent need for development of national waste strategies for proper treatment and disposal of municipal waste leachates in Lebanon.

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### 1. Introduction

\* Corresponding author. E-mail address: Christian.khalil@lau.edu.lb (C. Khalil). Municipal waste disposal into landfills is the primary option adopted by numerous countries worldwide (Baderna et al., 2011) (Mukherjee et al., 2015) (Gong et al., 2014). This disposal method poses numerous challenges due to the unplanned development of landfill areas lacking adequate engineering controls or even improper oversaturation of existing landfills. This leads to the potential formation of toxic gases and leachates that can escape containment and find their way to the soil and groundwater (Mukherjee et al., 2015).

Leachates consists of the liquid effluent generated from municipal waste consolidation into landfills. This liquid is a cocktail of numerous chemicals that are the result of water passing through the waste and saturating it with organic and inorganic matter. The produced leachate poses significant disposal challenges for landfill operators worldwide due to its potential to contaminate soils, surface water, aquifers and sea water.

Leachate composition is affected by numerous factors as outlined in Johansen and Carlson 1976 (Johansen and Carlson, 1976). Briefly these factors include landfill age, the geological conditions present in addition to local weather affecting the hydrogeological conditions in the landfills. Other important parameters to be considered within the landfill includes interaction of various chemicals, the internal temperature and pH. Landfill consolidated breakdown can occur under aerobic and anaerobic conditions. This breakdown contributes to stabilizing the organic component leading to lower leachates organic and inorganic concentrations (Jędrczak, 1994).

Landfill operators worldwide tend to compact the waste into landfills. This practice creates anaerobic conditions which results in methane gas formation. The formation of methane is observed in old landfills and is usually the result of acidogenic in young landfills followed by methanogenic reactions in old landfills. It is also important to note that the composition of the leachates together with climate and technology used in compacting the waste can play an important role in altering the leachates profile (Slomczyńska and Slomczyński, 2004).

Many studies were conducted on landfill sites worldwide with a focus on mutagenicity (Deguchi et al., 2007) and toxicity mainly in plants (Bhat et al., 2016). Across northern Spain for example, four municipal solid waste landfill sites have been monitored for eleven perfluoroalkyl carboxylates (PFCAs) and five perfluoroalkyl sulfonates (PFSAs). PFSAs importance stems from their C–F bonds making them highly stable in long alkyl chains (Prevedouros et al., 2006). This property led to their extensive production and various applications including coating materials, water repellent surfactants and fire retardant. (Busch et al., 2010) (Dauchy et al., 2012) (Yan et al., 2015). PFSAs high stability makes them non-biodegradable, persistent and extremely difficult to remediate using non-conventional methodologies (Quiñones and Snyder, 2009) such as membrane bioreactors (Fotakis and Timbrell, 2006) with limited remediation success (Fuertes et al., 2017).

The cytotoxicity and DNA damage induction in four simulated landfill soil leachates from Nigeria and India were also evaluated. Theses assessments were conducted using the MTT cell proliferation assay for cell number determination assay and alkaline comet assay for the DNA damage assessment (Swati et al., 2017). Heavy metals (Cadmium, Iron and Zinc) (Ratzinger et al., 2002), Polycyclic Aromatic hydrocarbons (PAH), polycyclic chlorinated biphenyls (PCBs) and organic chemicals detected in samples were higher than allowable exposure limits. The researchers also reported significant cytotoxic and DNA damage induction in exposed cells leading to significant morphological alterations and apoptosis. All these results clearly indicated the significant health risks posed by leachates exposure (Alimba et al., 2016). The assessment of leachates from India also showed a similar pattern with reports of high level of organics (158 times allowable limits) and lower heavy metal content. The paper highlighted the significant health risks posed by low concentrations of PAH especially when these PAH interact synergistically to cause cytotoxic and genotoxic damages (Ghosh et al., 2015).

Leachates from landfills in Turkey were also assessed. Researchers reported varying pH (4–8), temperatures (2.8 and 24.5  $^{\circ}$ C) and organic compounds (33 in all) such as phthalates and naphthalene and alkanes (Banar et al., 2006).

Lebanon leachates were previously analyzed and reported in a 2002 study by El-Fadel et al. (2002). The leachates were analyzed for a number of parameters such as pH, COD, Total organic Carbon (TOC), Total Dissolved Solid (TDS), chlorides, sulfates, orthophosphates, nitrates, ammonia nitrogen, hardness, and heavy metals.

Recently in 2016, municipal waste has been an emerging concern in our Lebanon. The regulated dumping sites could no longer manage the tremendous quantity of municipal garbage generated and therefore numerous unregulated dumping sites were created throughout the country. This large number of unregulated waste disposal areas could be correlated to environmental pollution and human diseases. To minimize the impact of dumpsites on human health and the environment, a qualitative and quantitative research into leachate production, toxicity and potential management was needed. It is important to note that waste leachate was typically released to the environment without any treatment, increasing the risk of environmental and human damages. Consequently, we must improve our knowledge of such matrices to be able to develop reliable treatment processes.

The main challenges for leachates management were mainly toxicological in nature (Kalka, 2012) (Ghosh et al., 2017) (Slomczyńska and Slomczyński, 2004). The leachates chemical composition was anticipated to vary between different dumping sites depending on the nature of the waste as well as the climate. Establishing toxicity profile predictions for these leachates is challenging due to the unique geochemical nature of each landfill and the variation in soil layers and water table (Koshy et al., 2007).

The uncontrolled disposal of these leachates into the soil and waterways poses significant challenges due to the composition of these leachates mainly in the form or organic and inorganic contaminants (Nagarajan et al., 2012) (Raghab et al., 2013). Leachates also poses pressures on Biochemical and Chemical Oxygen Demand (BOD and COD), TOC, ammonium and sulfur compositions and heavy metals in soil and groundwater (Gajski et al., 2012). The mixture of compounds generated by landfill is complex and more than 200 compounds of hazardous nature have been previously identified in landfill leachates. These compounds ranges from aromatic to phenols, halogenated and other compounds as described in the literature (Öman and Junestedt, 2008) (Baun and Christensen, 2004) (Adar and Bilgili, 2015).

The various chemical compounds in municipal leachates can lead to significant damages in ecological systems, food chains and ultimately human population. These effects can range from toxicity to carcinogenicity as reported by many researchers (Mukherjee et al., 2015) (Moraes and Bertazzoli, 2005) (Gajski et al., 2012).

A map showing the municipal dumps leachates collection sites can be referred to in Fig. 1.

The main motivation behind the research was to highlight the significant health and environmental risks posed by disposal of untreated municipal leachates into the natural environment. We anticipate that the findings will guide decision makers in the country to adopt better waste management policies and strategies. The novelty of this study is at two levels. Firstly, the research aims at establishing a linkage between the Lebanese leachates chemical compositions and the biological impacts observed at the cellular and animal levels (not previously reported in the literature). Secondly, we could not find any published data on the genotoxic impacts of Lebanese municipal leachates and this is a very important

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